CBE Attachments 1 through 15

Attachment 1

Source Inventory of Bay Area Greenhouse Gas Emissions

Updated: February 2010

BAY AREA
AIR QUALITY
MANAGEMENT
DISTRICT

939 Ellis Street
San Francisco, California 94109

SOURCE INVENTORY OF BAY AREA
GREENHOUSE GAS EMISSIONS

Base Year 2007

BAY AREA AIR QUALITY MANAGEMENT DISTRICT

February 2010

Prepared by
Emission Inventory Section
Tirlochan S. Mangat, Manager

Sukarn J. Claire
Tan M. Dinh
Amir K. Fanai
Michael H. Nguyen
Stuart A. Schultz

Approved by
Henry D. Hilken, Director of Planning, Rules & Research

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Bay Area Greenhouse Gas Emissions Inventory: 2007

Introduction

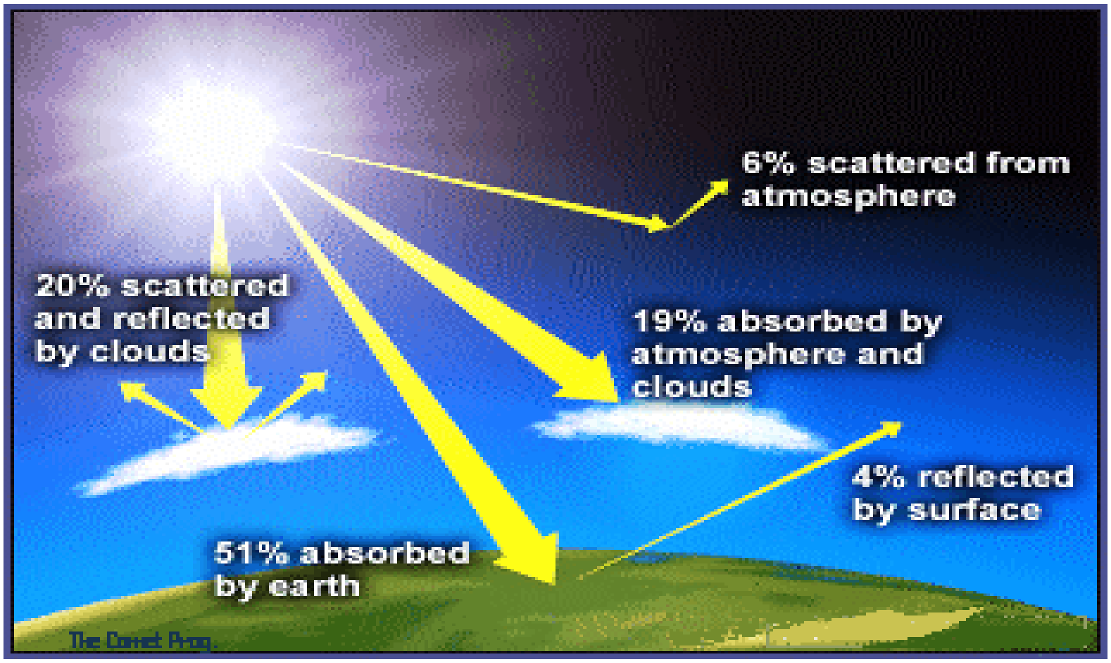
The Bay Area Air Quality Management District (Air District or BAAQMD) is the regional public agency responsible for the protection of the public's health and welfare from airborne pollutants, primarily by promulgating and enforcing regulations to reduce air pollution as provided by the Federal Clean Air Act, California Clean Air Act and State legislative mandates. The District's jurisdiction encompasses all of seven counties- Alameda, Contra Costa, Marin, San Francisco, San Mateo, Santa Clara, Napa- and the southern portions of Solano and Sonoma counties. The Air District is governed by a 23-member Board of Directors composed of locally elected officials from each of the nine Bay Area counties. In addition to its regulatory program, the Air District issues permits, operates an extensive incentives program, and conducts public outreach and education.

The Air District established a climate protection program in 2005 to explicitly acknowledge the link between climate change and air quality. Rising temperatures as a result of climate change threaten to undermine years of progress in improving air quality in the San Francisco Bay Area. At the same time, many longstanding air quality strategies such as programs to promote alternatives to the automobile, improve energy efficiency and encourage cleaner technologies also reduce greenhouse gases. The Air District regularly prepares inventories of criteria and toxic air pollutants to support planning, regulatory and other programs. This greenhouse gas inventory is based on the standards for criteria pollutant inventories and is intended to support the Air District's climate protection activities.

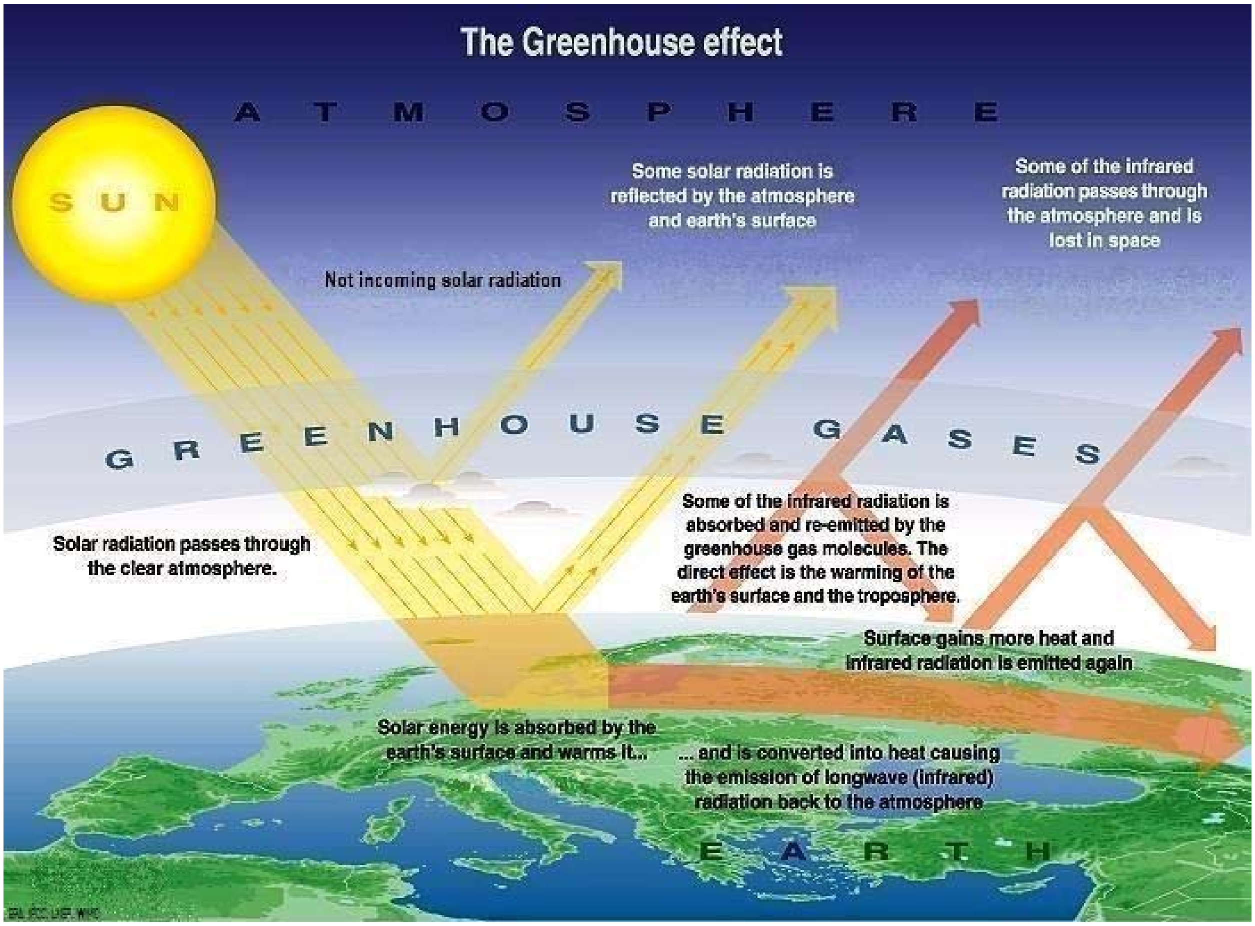
Climate Change and Greenhouse Gas Emissions

The greenhouse effect is a natural process by which some of the radiant heat from the Sun is captured in the lower atmosphere of the Earth, thus maintaining the temperature and making Earth habitable. The gases that help capture the heat are called greenhouse gases (GHG). All of these gases have been identified as forcing the earth's atmosphere and oceans to warm above naturally occurring temperatures.

Once, all climate changes on Earth occurred naturally. However, during the Industrial Revolution, we began altering our climate and environment through changing agricultural and industrial practices. Before the Industrial Revolution, human activity released very few gases into the atmosphere, but now through fossil fuel burning, deforestation and growing population (e.g. waste disposal), we are affecting the natural mixture of gases in the Earth's atmosphere. Increased concentration of these gases is upsetting the natural balance of incoming and outgoing solar energy. Emissions of carbon dioxide are the leading cause of global warming, with other pollutants also contributing. Carbon dioxide concentrations, which ranged from 265 ppm to 280 ppm over the last 10,000 years, only began rising in the last two hundred years to current levels of 365 ppm, a 30% increase.



The natural balance of solar energy received by Earth



Some greenhouse gases occur naturally in the atmosphere, while others result from human activities. Naturally occurring greenhouse gases include water vapor, carbon dioxide, methane, nitrous oxide, and ozone. Certain human activities, however, add to the levels of most of these naturally occurring gases. The most common greenhouse gases, and those which are identified in AB32, the Global Warming Solutions Act of 2006 and are discussed in this report, are carbon dioxide, methane, nitrous oxide, hydrofluorocarbons, perfluorocarbons, and sulfur hexafluoride.

Carbon Dioxide (CO_2) is released to the atmosphere when solid waste, fossil fuels (oil, natural gas, and coal), and wood and wood products are burned.

Biogenic Carbon Dioxide (Bio-CO_2) emissions are a subset of total CO_2 emissions which are emitted from materials that are derived from living cells, excluding fossil fuels, limestone and other materials that have been transformed by geological processes. Bio-CO_2 originates from carbon that is present in materials such as wood, paper, vegetable oils and food, animal, and yard waste.

Methane (CH_4) is emitted during the production and transport of coal, natural gas, and oil. Methane emissions also result from the decomposition of organic waste in municipal solid waste landfills and the raising of livestock.

Nitrous oxide (N_2O) is emitted from agricultural and industrial activities, as well as during combustion of solid waste and fossil fuels and during production of adipic acid, and nitric acid.

Very powerful greenhouse gases, also known as *high global warming potential* (GWP) *gases* that are not naturally occurring, include hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), and sulfur hexafluoride (SF_6). These gases are emitted from industrial processes such as semiconductor manufacturing, use as refrigerants and other products, and electric power transmission and distribution.

Each greenhouse gas differs in its ability to absorb heat in the atmosphere. High GWP gases such as HFCs, PFCs, and SF_6 are the most heat-absorbent. Methane traps over 21 times more heat per molecule than carbon dioxide, and nitrous oxide absorbs 310 times more heat per molecule than carbon dioxide. Often, estimates of greenhouse gas emissions are presented in carbon dioxide equivalents, which weight each gas by its global warming potential. Table A shows the global warming potentials for different greenhouse gases for a 100 year time horizon. The global warming potentials used in this report are in accordance with the Second Assessment Report (SAR) of the Intergovernmental Panel on Climate Change (IPCC).

Table A: Global Warming Potentials (GWPs) for Greenhouse Gases

Gas	GWP
CO_2	1
CH_4	21
N_2O	310
HFCs/PFCs	90- 11,700
SF_6	23,900

Greenhouse Gas Emissions Inventory

An emissions inventory is a detailed estimate of the amount of air pollutants discharged into the atmosphere of a given area by various emission sources during a specific time period. This GHG emissions inventory builds on the Air District's many years of experience preparing inventories of criteria and toxic air pollutants.

This emission inventory includes direct and indirect GHG emissions due to human activities. The emissions are estimated for industrial, commercial, transportation, residential, forestry, and agriculture activities in the San Francisco Bay Area region of California. Both direct greenhouse gas emissions from locally generated electricity in the Bay Area and indirect emissions from out-of-region generated electricity for consumption in the region are reported.

Emissions of CO_2, Bio-CO_2, CH_4, N_2O, HFCs, PFCs, and SF_6 are estimated using the most current activity (e.g., cubic feet of natural gas burned or vehicle miles traveled) and emission factor data from various sources. Activity data used in preparing this GHG inventory is the same as is used in preparing the Air District's criteria and toxic inventories. Emission factor data was obtained from the U.S. Department of Energy's (DOE's) Energy Information Administration (EIA), the California Energy Commission (CEC), and the California Air Resources Board (CARB).

Methodology

Emission sources can be broadly divided between stationary and mobile sources. Stationary sources can be further divided between point and area sources. Stationary emission sources identified on an individual basis or as a single source are called point sources. Electric power generating plants and oil refineries are examples of point sources. Based on Air District permits for stationary sources, the Air District maintains a computer database with detailed information on operations and emission characteristics for nearly 4,000 facilities, which include roughly 25,000 different sources, throughout the Bay Area. Activity data on the sources are collected at the process level from each facility and are updated regularly as part of permit renewal. The greenhouse gas emissions from these sources are calculated by multiplying activity data by standardized emission factors for each greenhouse gas. These emission factors take into account fuel-specific carbon content and the percent of carbon that oxidizes to convert to carbon dioxide emissions. Some of the combustion emission factors for various fuels used for this emissions inventory are shown in Table B. Examples of activity data used to develop the inventory are shown in Table C.

Stationary emission sources that are not identified individually are called area sources. Area sources are groups of numerous small emission sources, which individually do not emit significant amounts of pollutants but together make an appreciable contribution to the emission inventory. Many area sources do not require permits from the Air District, such as residential heating, restaurants, and the wide range of consumer products such as paints, solvents, and cleaners. Some facilities considered as area sources do require permits from the Air District, such as gas stations and dry cleaners. Emissions estimates for area sources are developed based on estimated activities and emission factors for various categories.

Mobile sources consist of on-road motor vehicles and other mobile sources. Examples of on-road motor vehicles are cars, trucks, buses and motorcycles. Other mobile sources include boats, ships, trains, aircraft, and garden, farm and construction equipment. Greenhouse gas emissions for on-road motor vehicles were calculated using CARB's EMFAC2007 model together with vehicle miles travelled (VMT) and other activity data by county from the Metropolitan Transportation Commission's (MTC) Regional Transportation Plan (RTP2030). Other off- and on- road mobile source emissions were calculated based on estimated fuel used and emission factors in Table B. GHG emissions for ships are calculated for ship travel within 100 miles of the San Francisco coastline. Aircraft emissions are calculated for air travel within the Air District boundaries.

Table B: Generalized GHG Emission Factors (Lbs. /Usage Unit)

Fuel	CO_2	CH_4	N_2O	Unit
Liquid Fuels				
Distillate Fuel (Fuel Oil, Diesel)	22.4	0.00053	0.00019	Gallon
Jet Fuel	21.1	0.00052	0.00019	Gallon
Kerosene/Naphtha	21.5	0.00050	0.00018	Gallon
Liquified Petroleum Gases (LPG)	12.8	0.00025	0.00002	Gallon
Motor Gasoline	19.6	0.00055	0.00020	Gallon
Residual Fuel (Bunker C Fuel Oil)	26.0	0.00022	0.00021	Gallon
Aviation Gasoline	18.4	0.00052	0.00019	Gallon
Bio-diesel	20.7	0.00049	0.00018	Gallon
Propane	12.7	0.000003	2.3E-07	Gallon
Butane	14.7	0.000003	2.3E-07	Gallon

Table B (continued)

Fuel	CO_2	CH_4	N_2O	Unit
Gaseous Fuels				
Natural Gas	120.6	0.00020	0.00020	1000 ft^3
Landfill Gas	110.5	0.21050	0.00024	1000 ft^3
Digester Gas	104.7	0.02997	0.00030	1000 ft^3
Carbon Monoxide	116.1	0.00270	0.00019	1000 ft^3
Refinery Waste Gases	139.0	0.00320	0.00022	1000 ft^3
Solids				
Refuse/Waste	2,000	0.29790	0.08980	Ton
Wood and Other	3,814	0.29790	0.08980	Ton
Agriculture Waste Burning	174	0.14000	0.35000	Ton
Petroleum Coke	6,769	0.44920	0.10630	Ton

Table C: 2007 General Statistics

County	Population (1000's)	Daily Electricity Usage (Megawatt hours)	Daily Natural Gas Usage (Million cu.ft.)	Daily Gasoline Sales (1000's gal.)	Daily Vehicle Miles Traveled (Millions)
Alameda	1,532	31,395	150	1,759	38.0
Contra Costa	1,039	23,204	636	1,150	25.7
Marin	255	4,033	24	337	6.2
Napa	136	2,679	13	169	4.5
San Francisco	801	17,720	118	961	12.4
San Mateo	729	12,909	71	947	19.4
Santa Clara	1,805	44,114	212	2,160	40.1
Solano*	308	6,443	44	353	7.2
Sonoma*	431	6,977	31	489	10.6
Total	**7,036**	**149,474**	**1,300**	**8,327**	**164.1**

* Portion within Bay Area Air Quality Management District

Revisions to the Previous GHG Inventory

This emissions inventory estimates greenhouse gas emissions produced by the San Francisco Bay Area in 2007. This inventory updates the Air District's previous GHG emission inventory for base year 2002 (published November 2006). All activity data has been updated to reflect more current industrial activity, motor vehicle travel, and economic and population growth. Most of the methodologies for calculating emissions remain the same, with certain exceptions: 1) emissions from electricity consumed in the Bay Area but generated outside the region are now included; 2) emissions of ozone depleting substance (ODS) substitutes such as hydrofluorocarbons and perfluorocarbons used as refrigerants etc. are now included; 3) more complete oil refinery process emissions are included in this inventory; 4) certain off-road equipment (e.g., construction and industrial) previously was included in the transportation sector and is now reported separately; 5) ship emissions are now calculated for travel within 100 nautical mile of the California coastline rather than 3 nautical miles to be consistent with the Air District's criteria pollutant inventory and; 6) biogenic CO_2 emissions are calculated but not included in the total CO_2 equivalent estimates for the region.

Because of these revisions, caution should be used in comparing this 2007 GHG emission inventory to the previous 2002 inventory. These revisions have resulted in different estimates of Bay Area greenhouse gas emissions. Consequently, the percentage contributions from individual sectors may be affected. For example, estimates of transportation emissions have not changed significantly between the two inventories, but the percentage from the transportation sector has changed because the estimated total emissions are greater in this inventory (due mainly to increased estimated emissions resulting from the revisions summarized above). Such ongoing updates are typical of emission inventories. Examining emissions forecasts and backcasts in a single emission inventory is more useful in determining trends than comparing one inventory against another.

Summary of Bay Area GHG Emissions

In 2007, 95.8 million metric tons of CO_2-equivalent ($MMTCO_2E$) greenhouse gases were emitted by the San Francisco Bay Area (88.7 $MMTCO_2E$ were emitted within the Bay Area Air District and 7.1 $MMTCO_2E$ were indirect emissions from imported electricity). A breakdown of Bay Area greenhouse gas emissions by pollutant is shown in Figure 1 and Table D.

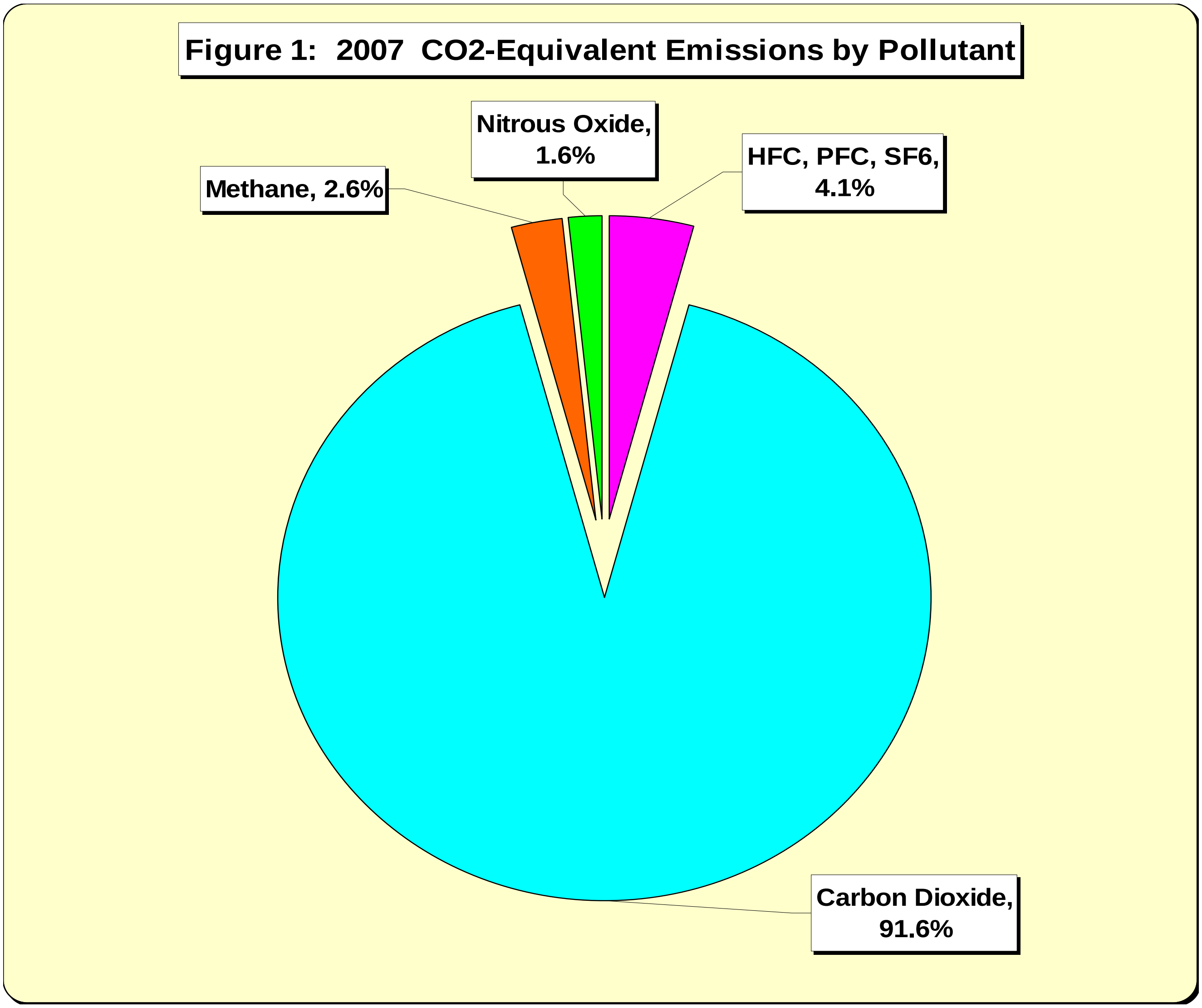


Figure 1: 2007 CO2-Equivalent Emissions by Pollutant

Table D: 2007 CO2- Equivalent Emissions by Pollutant

Pollutant	**Percentage**	**CO2-Equivalent (Million Metric Tons / Year)**
Carbon Dioxide	91.6%	87.8
Methane	2.6%	2.5
Nitrous Oxide	1.6%	1.5
HFC, PFC, SF6	4.1%	4.0
Total	**100%**	**95.8**

The greenhouse gas with the greatest emissions is carbon dioxide (CO_2). Carbon dioxide emissions from various activities in the Bay Area represented about 91.6 percent of total greenhouse gas emissions in 2007. Carbon dioxide emissions are mainly associated with combustion of carbon-bearing fossil fuels such as gasoline, diesel, and natural gas used in mobile sources and energy-generation-related activities. Other activities that produce CO_2 emissions include oil refining processes, cement manufacturing, waste combustion, and waste and forest management.

Bio-CO_2 emissions are from materials that were grown through the process of photosynthesis and thus the carbon they contain was relatively recently in the atmosphere. Burning of these materials does not add any net CO_2 to the atmosphere. Bio-CO_2 emissions are tracked and shown separately in Tables K through T and Table V and are not counted in the anthropogenic emissions inventory directly. This is consistent with CARB's methodology for GHG inventories. Landfills, fireplaces, and wastewater treatment plants are the largest sources of Bio-CO_2 emissions.

Methane (CH_4) emissions from various sources represent 2.6 percent of Bay Area's total CO_2-equivalent GHG emissions. Landfills, natural gas distribution systems, agricultural activities, stationary and mobile fuel combustion, and gas and oil production fields categories are the major sources of these emissions.

Nitrous oxide (N_2O) emissions represent 1.6 percent of the overall greenhouse gas emissions inventory. Municipal wastewater treatment facilities, fuel combustion, and agricultural soil and manure management are the major contributors of nitrous oxide emissions in the Bay Area.

Emissions from high GWP gases such as hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), and sulfur hexafluoride (SF_6) make up about 4.1 percent of the total CO_2-equivalent emissions. Industrial processes such as semiconductor manufacturing, use as refrigerants and other products, and electric power transmission and distribution systems are the major sources of HFCs, PFCs and SF_6 emissions in the Bay Area.

GHG Emissions by Sector

Greenhouse gas emissions by end-use sectors are shown in Figure 2 and Table E. Fossil fuel consumption in the transportation sector was the single largest source of the San Francisco Bay Area's greenhouse gas emissions in 2007. The transportation sector contributed about 36.41 percent of greenhouse gas emissions in the Bay Area. Categories included in this sector are on-road motor vehicles, locomotives, ships and boats, and aircraft.

Figure 2: 2007 Bay Area GHG Emissions by Sector

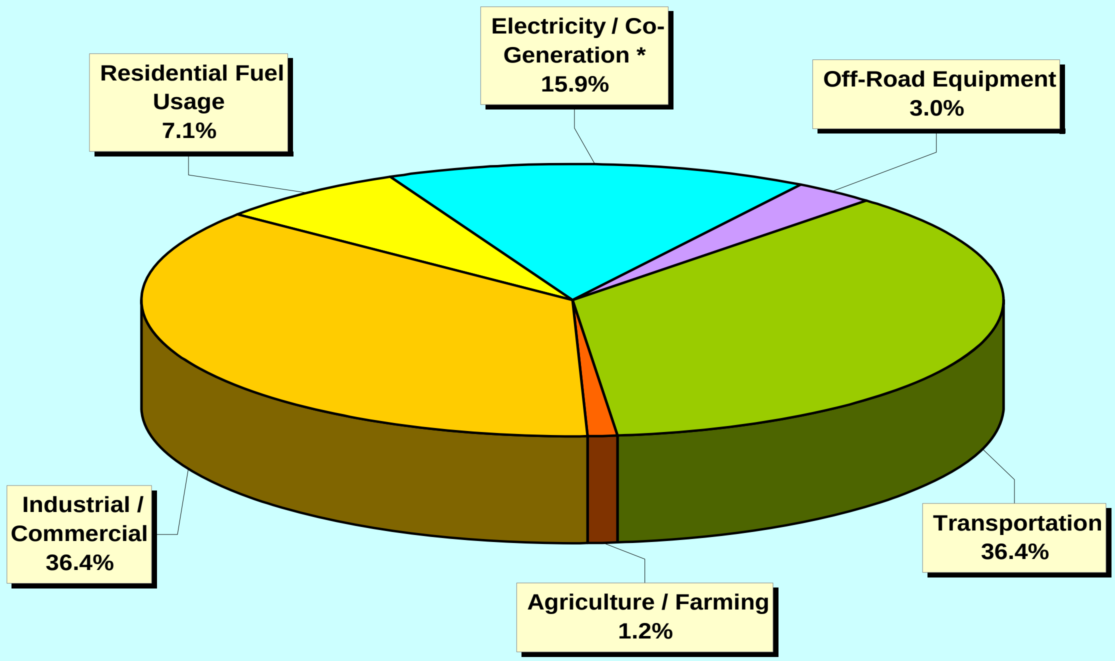


Table E: 2007 Bay Area GHG Emissions by Sector

End-Use Sector	% of Total Emissions	CO2- Equivalent (Million Metric Tons / Year)
Industrial / Commercial	36.40%	34.86
Residential Fuel Usage	7.12%	6.82
Electricity / Co-Generation *	15.87%	15.20
Off-Road Equipment	3.05%	2.92
Transportation	36.41%	34.87
Agriculture / Farming	1.16%	1.11
Total	**100%**	**95.8**

* Includes Imported Electricity emissions of 7.1 MMTCO$_2$ E

The industrial and commercial sector (excluding electricity/co-generation and agriculture/farming, which are reported separately) was the second largest contributor, closely following the transportation sector, with 36.4 percent of total GHG emissions. Industrial and commercial sources include industrial processes such as oil refining, natural gas and other fuel combustion, waste management, cement manufacturing, fuel distribution, refrigerant usage, and some other small sources.

Energy production activities such as electricity generation and co-generation were the third largest contributor with 15.9 percent of the total GHG emissions (including imported electricity emissions). California imports about one-fifth to one-third of its total electricity usage, mainly from the northwestern and southwestern states. The Bay Area used about 54.6 million megawatt hours of electricity in 2007, about one-third of it was generated outside of the Bay Area. Electricity and co-generation facilities within the Bay Area Air District emitted about 8.1 million metric tons of CO_2-equivalent ($MMTCO_2E$) emissions in 2007 and emissions from electricity imports were estimated to be 7.1 $MMTCO_2E$. While imported electricity is a relatively small share of the Bay Area's electricity mix, out-of-region electricity generation sources contribute a larger share of GHG emissions. This is due to the fact that electricity generation in the Bay Area is mainly from natural gas-fired and other cleaner burning power plants. Out-of-state electricity generation is also from coal-fired power plants which have higher carbon intensity.

The contribution from residential fuel combustion was the fourth largest with 7.1 percent of the total GHG emissions. Residential fuel combustion emissions are primarily from space heating, cooking and water heating.

Off-road equipment such as construction, industrial, commercial, and lawn and garden equipment contributed 3.0 percent of GHG emissions.

Agriculture and farming was the smallest sector with 1.2 percent of the total greenhouse emissions in the Bay Area.

More detailed information on greenhouse gas emissions by source category, for the region and for each county, is provided in Tables K through U. Table V contains the list of the 200 largest greenhouse gas emission point sources/facilities in the San Francisco Bay Area.

An emissions breakdown for the two largest greenhouse gas emitting sectors in the Bay Area, industrial/commercial and transportation, is shown in Figure 3, Table F and Figure 4, Table G respectively.

Figure 3: 2007 Industrial/ Comm. Sector Emissions Breakdown

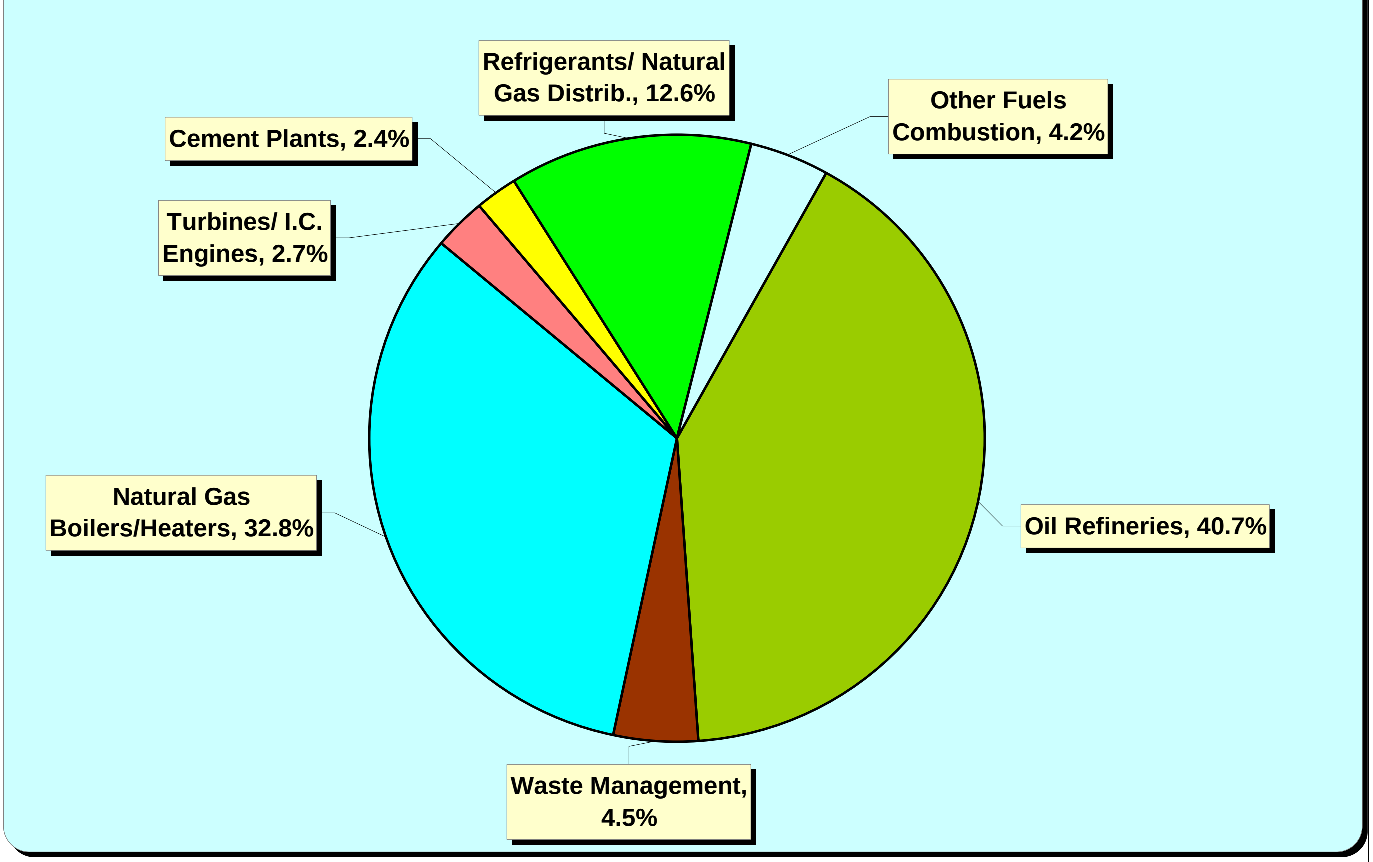


Table F: 2007 Industrial/ Commercial Sector Emissions Breakdown

Source Category	% of Total Emissions	CO2-Equivalent (Million Metric Tons / Year)
Oil Refineries	40.7%	14.2
Waste Management	4.5%	1.6
Natural Gas Boilers/Heaters	32.8%	11.4
Turbines/ I.C. Engines	2.7%	1.0
Cement Plants	2.4%	0.8
Refrigerants/ Natural Gas Distrib.	12.6%	4.4
Other Fuels Combustion	4.2%	1.5
Total	**100%**	**34.9**

Figure 4: 2007 Transportation Sector Emissions Breakdown

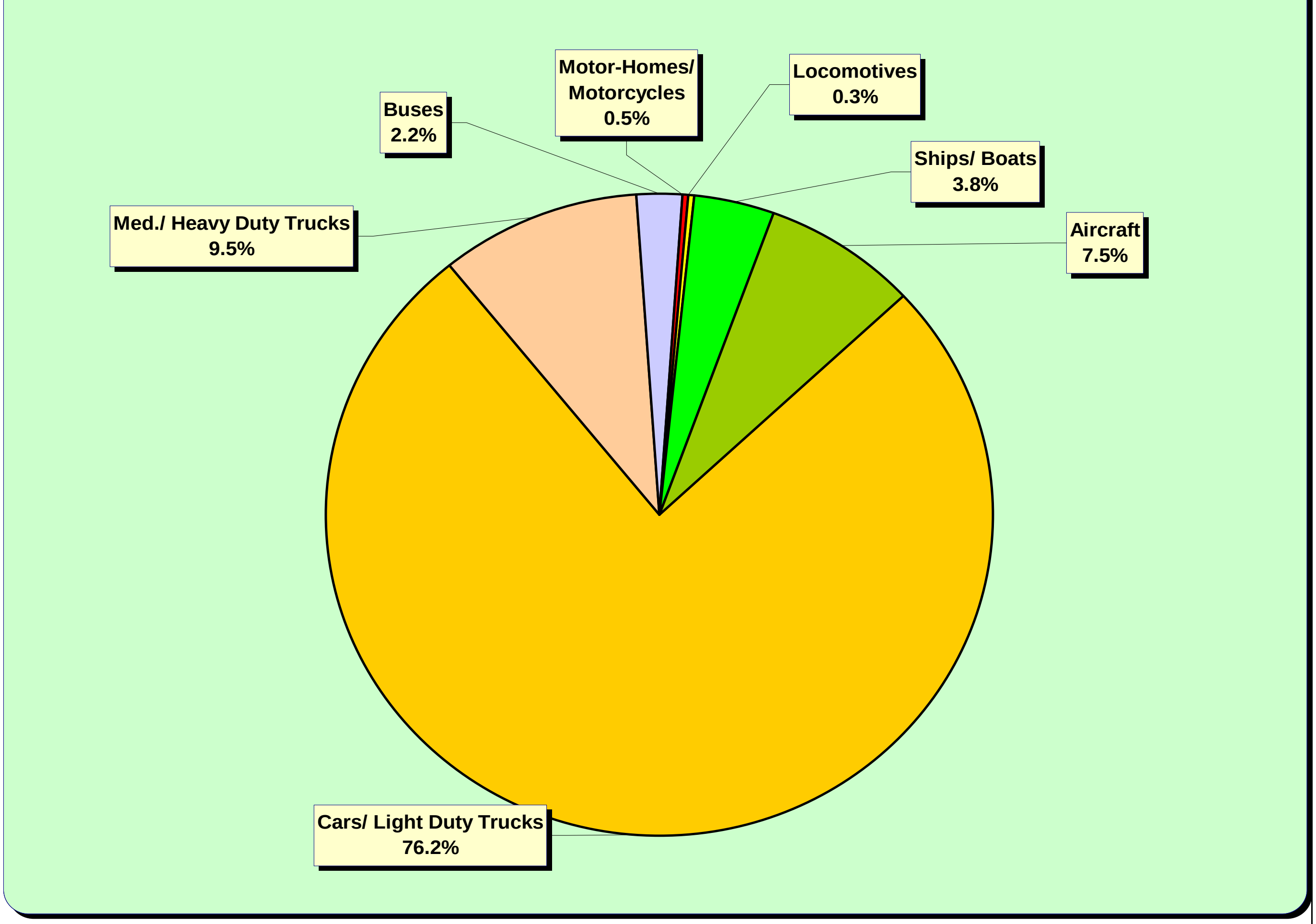


Table G: 2007 Transportation Sector Emissions Breakdown

Source Category	% of Total Emissions	CO2-Equivalent (Million Metric Tons / Year)
Cars/ Light Duty Trucks	76.2%	26.6
Med./ Heavy Duty Trucks	9.5%	3.3
Buses	2.2%	0.8
Motor-Homes/ Motorcycles	0.5%	0.2
Locomotives	0.3%	0.1
Ships/ Boats	3.8%	1.3
Aircraft	7.5%	2.6
Total	**100%**	**34.9**

GHG Emissions by County

GHG emissions for the nine Bay Area counties under the Air District's jurisdiction are summarized in Figure 5 and Table H. See Tables K-T for detailed emissions information.

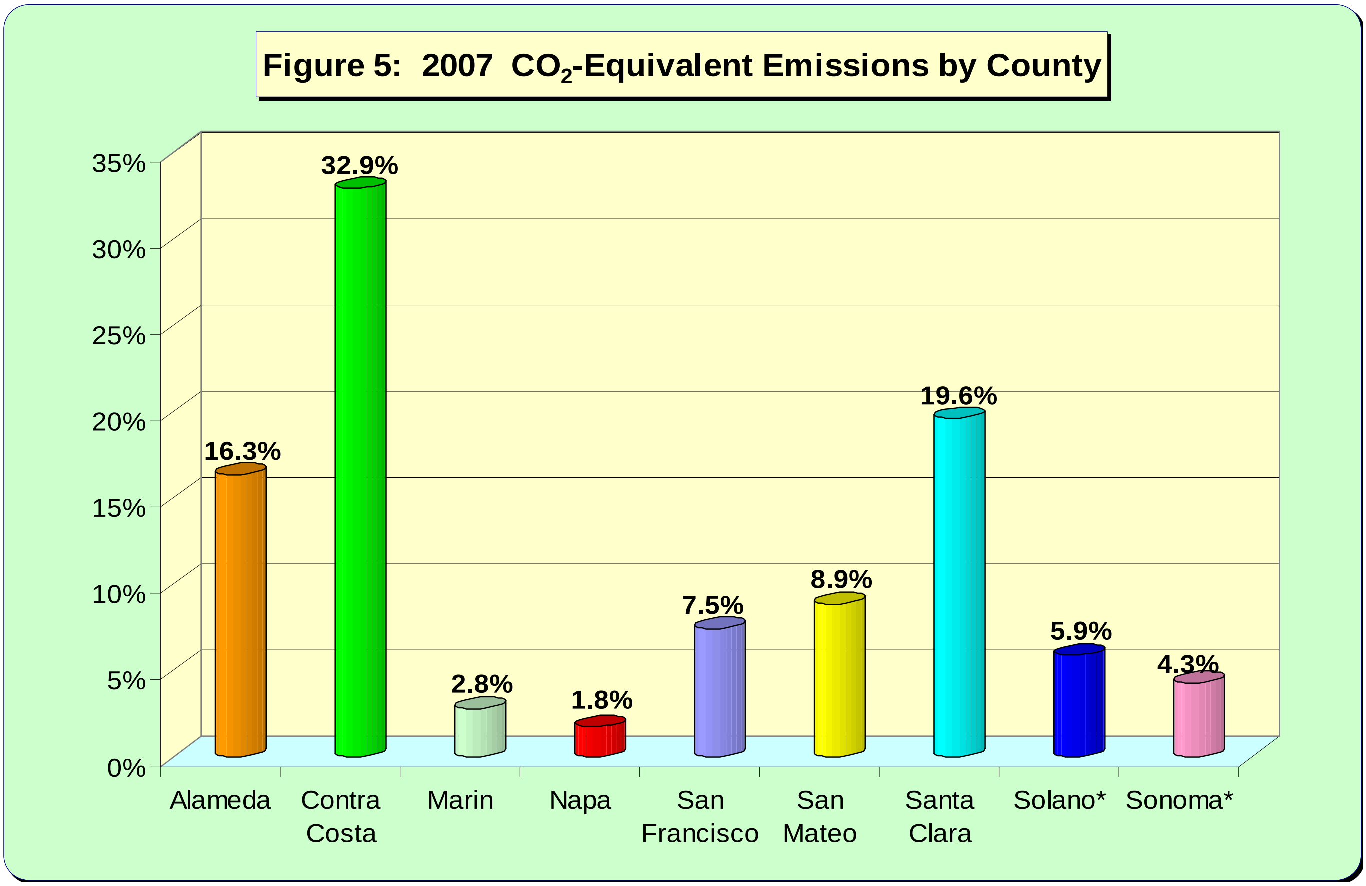


Table H: 2007 CO2- Equivalent Emissions by County

County	% of Total Emissions	CO2- Equivalent (Million Metric Tons / Year)
Alameda	16.3%	15.7
Contra Costa	32.9%	31.5
Marin	2.8%	2.7
Napa	1.8%	1.7
San Francisco	7.5%	7.1
San Mateo	8.9%	8.5
Santa Clara	19.6%	18.8
Solano*	5.9%	5.7
Sonoma*	4.3%	4.1
Total	**100%**	**95.8**

* Portion within BAAQMD

A breakdown of emissions by end-use sectors for each county is shown in Figure 6 and Table I. This figure and table show relatively higher industrial/commercial sector emissions in Contra Costa and Solano Counties due to the oil refining industry. All other counties show the largest contribution from the transportation sector.

Figure 6: 2007 County Emissions Breakdown by Sector

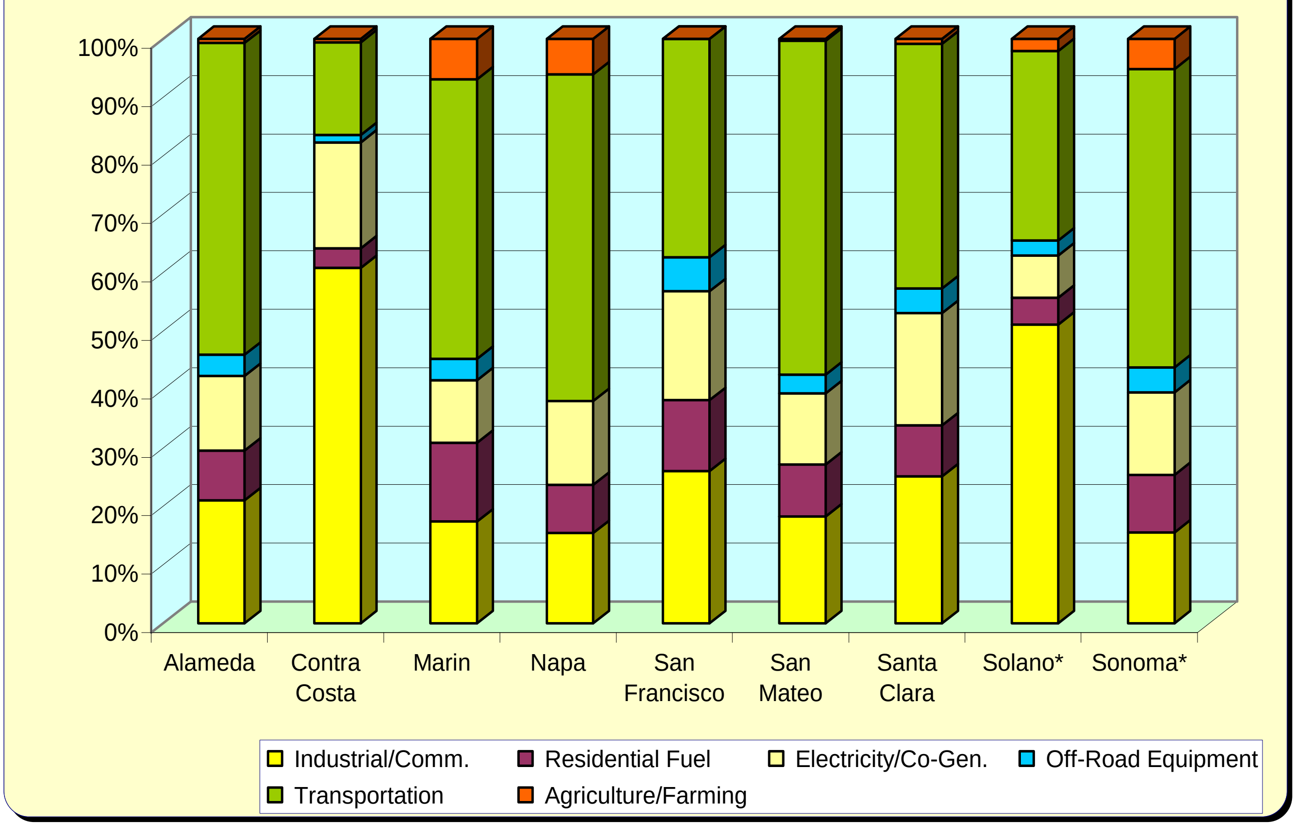


Table I: 2007 County Emissions Breakdown by Sector (Million Metric Tons CO2-Equiv./Yr.)

Sector	Alameda	Contra Costa	Marin	Napa	San Francisco	San Mateo	Santa Clara	Solano*	Sonoma*
Industrial/Comm.	3.3	19.2	0.5	0.3	1.9	1.6	4.7	2.9	0.6
Residential Fuel	1.3	1.1	0.4	0.1	0.9	0.8	1.6	0.3	0.4
Electricity/Co-Gen.	2.0	5.7	0.3	0.2	1.3	1.0	3.6	0.4	0.6
Off-Road Equipment	0.6	0.4	0.1	0.0	0.4	0.3	0.8	0.1	0.2
Transportation	8.4	5.0	1.3	0.9	2.7	4.8	7.9	1.8	2.1
Agriculture/Farming	0.1	0.2	0.2	0.1	0.0	0.0	0.2	0.1	0.2
Total	15.7	31.5	2.7	1.6	7.1	8.5	18.8	5.7	4.1

* Portion within BAAQMD

GHG Emission Trends for Bay Area

Under “business as usual” conditions, greenhouse gas emissions are expected to grow in the future due to population growth and economic expansion. Figure 7 and Tables J and U show emissions trends by sectors for the period 1990 to 2029.

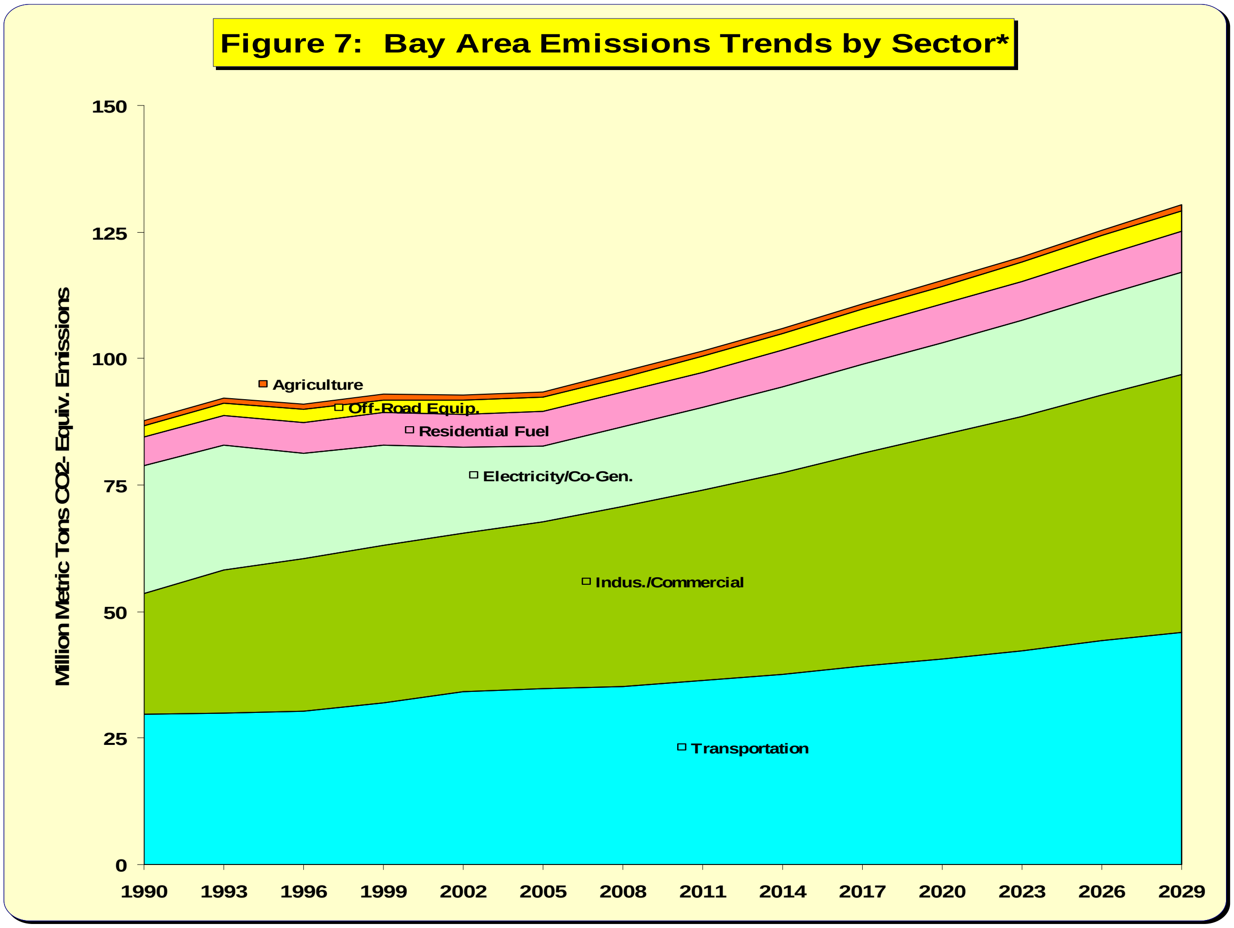


Figure 7: Bay Area Emissions Trends by Sector*

Table J: Bay Area Emissions Trends by Sector (Million Metric Tons CO_2-Equiv.)*

Category	1990	1993	1996	1999	2002	2005	2008	2011	2014	2017	2020	2023	2026	2029
Transportation	29.8	29.8	30.4	32.0	34.1	34.8	35.3	36.3	37.6	39.3	40.7	42.2	44.2	46.0
Indus./Commercial	23.9	28.5	29.9	31.1	31.4	32.8	35.6	37.7	39.9	42.0	44.2	46.4	48.6	50.8
Electricity/Co-Gen.	25.1	24.6	20.9	19.8	17.0	15.1	15.6	16.3	16.9	17.6	18.3	18.9	19.6	20.4
Residential Fuel	5.8	6.0	6.2	6.4	6.6	6.7	6.9	7.0	7.2	7.4	7.5	7.7	7.9	8.0
Off-Road Equip.	2.2	2.3	2.5	2.6	2.7	2.8	3.0	3.1	3.3	3.4	3.6	3.8	3.9	4.1
Agriculture	1.0	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1
Total	87.7	92.2	91.0	93.0	92.8	93.4	97.4	101.5	106.0	110.8	115.4	120.2	125.3	130.4

* “Business as usual” projection

Figure 8 shows the San Francisco Bay Area region's overall greenhouse gas emissions trends. More details on emissions trends are provided in Table U.

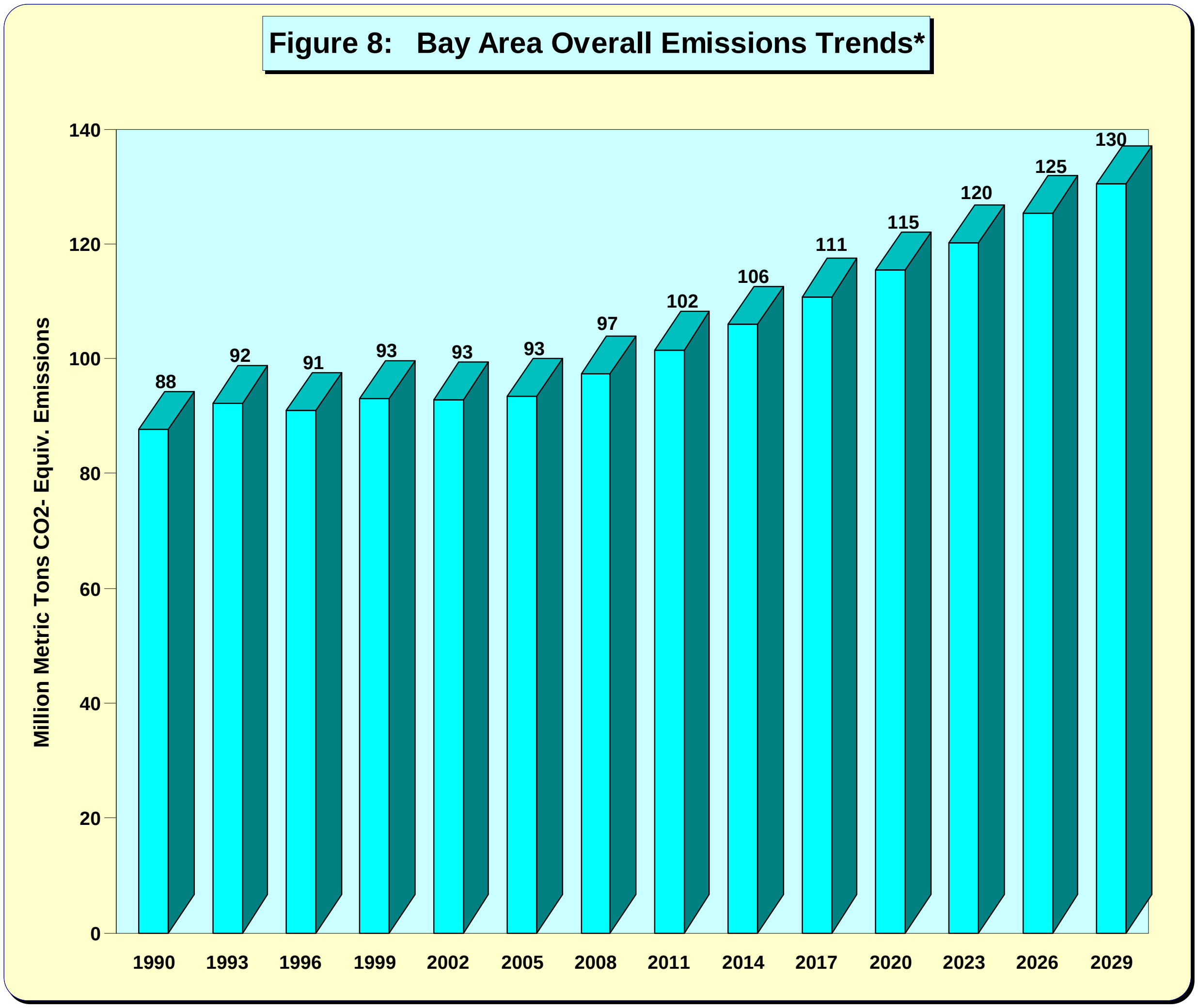


Figure 8: Bay Area Overall Emissions Trends*

* "Business as usual" projection

These projections reflect regulatory programs in place as of 2007 (benefits of CARB GHG regulations for motor vehicles adopted in 2004, the Pavley regulations, are not included in this inventory, pending U.S. EPA approval of a waiver under the Clean Air Act to implement the regulations). If current trends continue, Bay Area GHG emissions are expected to increase at an average rate of approximately 1.4 percent per year. The long term GHG emissions trends are expected to go upwards *absent policy changes*. Year-to-year fluctuation in emissions trends is due to variation in economic activity and the fraction of electric power generation in this region. Power generation in the Bay Area varies year-to-year depending on various factors including the availability of hydroelectric and other imported power.

Greenhouse gas emissions are projected based on estimated growth in various source categories. For example, CARB's EMFAC2007 computer model was utilized to project GHG emissions from transportation sources. In these models, fuel consumption estimates were based on the anticipated change of fleet mix and the growth of various types of on-road and off-road vehicles. Growth in VMT is based on the MTC's Regional Transportation Plan (RTP2030). For aircraft categories, the fleet mix and the growth data from the MTC's Regional Airport System Plan were incorporated into the GHG projection models.

The projected GHG emissions from power plants operating in the Bay Area were based on the California Energy Commission's 2003 Fuel Usage Forecasts for the Bay Area.

Emission projections for the oil refineries were based on the California Energy Commission's report on California's Petroleum Infrastructure (2007).

The GHG projections from other major sources such as landfills, natural gas fuel distribution, and cement manufacturing were estimated by using Association of Bay Area Government's employment and population data. California Integrated Waste Management data were also considered in the landfill projection process.

Climate protection activities in California are occurring at the state, regional and local level. CARB and other state agencies are identifying measures to achieve the AB32 emission reduction goal of meeting statewide 1990 GHG emissions levels by 2020. Specifically, in December 2008 CARB adopted the AB32 Scoping Plan which outlines a strategy to achieve AB32 goals. The Scoping Plan does not set emission reduction goals for regions within the state, but it is expected that all California communities will need to contribute to statewide reductions. The Air District and our regional agency partners are implementing climate protection programs. Bay Area cities and counties are preparing local climate action plans. These efforts are expected to reduce future Bay Area GHG Emissions below the projections presented here.

This GHG emissions inventory will be updated as climate protection programs are implemented and as additional information about activity data, emission factors and other inputs becomes available.

Table K:

Annual GHG Emissions: Bay Area Year 2007 (Metric Tons / Year)

SOURCE CATEGORY	CO_2	CH_4	N_2O	PFC/HFC	SF_6	Total GHG CO_2-Equivalent	Biogenic CO_2
INDUSTRIAL/ COMMERCIAL							
Oil Refineries							
Refining Processes	3,445,064	79	--	--	--	3,446,782	--
Refinery Make Gas Combustion	4,772,971	97	6	--	--	4,776,959	--
Natural Gas and Other Gases Combustion	4,860,268	267	18	--	--	4,871,495	--
Liquid Fuel Combustion	89,450	1	1	--	--	89,760	--
Solid Fuel Combustion	1,000,216	29	6	--	--	1,002,637	--
Waste Management							
Landfill Combustion Sources	--	1,241	1	--	--	26,455	584,565
Landfill Fugitive Sources	--	56,747	3	--	--	1,192,596	154,411
Composting/POTWs	--	2,773	965	--	--	357,224	--
Other Industrial/ Commercial							
Cement Plants	841,350	13	3	--	--	842,475	--
Commercial Cooking	134,612	--	--	--	--	134,612	--
ODS Substitutes/Nat. Gas Distrib./Other	--	16,356	--	2,184	0.13	4,390,999	21
Reciprocating Engines	550,270	1,989	1	--	--	593,584	234,013
Turbines	354,697	78	1	--	--	356,663	66,141
Natural Gas- Major Combustion Sources	2,400,044	51	3	--	--	2,402,179	--
Natural Gas- Minor Combustion Sources	8,979,100	172	165	--	--	9,033,745	--
Coke Coal	989,442	28	6	--	--	991,823	--
Other Fuels Combustion	349,171	132	2	--	--	352,485	85,346
Subtotal	28,766,652	80,052	1,181	2,184	0.13	34,862,465	1,124,497
RESIDENTIAL FUEL USAGE							
Natural Gas	6,456,173	124	118	--	--	6,495,464	--
LPgas/Liquid Fuel	166,508	3	11	--	--	169,911	--
Solid Fuel	--	6,242	67	--	--	151,742	628,550
Subtotal	6,622,682	6,369	196	--	--	6,817,118	628,550
ELECTRICITY/ CO-GENERATION							
Co-Generation	5,292,826	1,261	4	--	--	5,320,398	89,512
Electricity Generation	2,730,973	163	1	--	1.18	2,762,968	3,525
Electricity Imports	7,102,311	59	33	--	--	7,113,680	--
Subtotal	15,126,111	1,483	37	--	1.18	15,197,047	93,037
OFF-ROAD EQUIPMENT							
Lawn and Garden Equipment	105,742	192	77	--	--	133,803	--
Construction Equipment	1,785,078	289	11	--	--	1,794,433	--
Industrial Equipment	729,035	433	41	--	--	750,852	--
Light Commercial Equipment	226,118	111	42	--	--	241,375	--
Subtotal	2,845,974	1,025	171	--	--	2,920,462	--
TRANSPORTATION							
Off-Road							
Locomotives	88,092	5	35	--	--	99,152	--
Ships	731,679	74	28	--	--	742,064	--
Boats	509,165	252	168	--	--	566,451	--
Commercial Aircraft	1,877,665	91	68	--	--	1,900,661	--
General Aviation	231,066	46	8	--	--	234,642	--
Military Aircraft	478,178	31	15	--	--	483,454	--
On-Road							
Passenger Cars/Trucks up to 10,000 lbs	26,070,815	2,783	1,480	--	--	26,587,907	--
Medium/Heavy Duty Trucks > 10,000 lbs	3,232,949	181	208	--	--	3,301,335	--
Urban,School and Other Buses	722,698	21	192	--	--	782,755	--
Motor-Homes and Motorcycles	156,636	147	42	--	--	172,846	--
Subtotal	34,098,941	3,629	2,246	--	--	34,871,276	--
AGRICULTURE/ FARMING							
Agricultural Equipment	183,929	34	2	--	--	185,364	--
Animal Waste	--	25,860	254	--	--	621,761	--
Soil Management	15,954	--	899	--	--	294,758	43,110
Biomass Burning	--	97	8	--	--	4,363	3,145
Subtotal	199,883	25,991	1,163	--	--	1,106,246	46,255
GRAND TOTAL EMISSIONS	**87,660,281**	**118,549**	**4,993**	**2,184**	**1.3**	**95,774,635**	**1,892,340**

Table L:
Annual GHG Emissions: ALAMEDA Year 2007 (Metric Tons / Year)

SOURCE CATEGORY	CO_2	CH_4	N_2O	PFC/HFC	SF_6	Total GHG CO_2-Equivalent	Biogenic CO_2
INDUSTRIAL/ COMMERCIAL							
Oil Refineries							
Refining Processes	--	--	--	--	--	--	--
Refinery Make Gas Combustion	--	--	--	--	--	--	--
Natural Gas and Other Gases Combustion	--	--	--	--	--	--	--
Liquid Fuel Combustion	--	--	--	--	--	--	--
Solid Fuel Combustion	--	--	--	--	--	--	--
Waste Management							
Landfill Combustion Sources	--	304	--	--	--	6,475	143,077
Landfill Fugitive Sources	--	17,818	1	--	--	374,448	48,018
Composting/POTWs	--	280	196	--	--	66,683	--
Other Industrial/ Commercial							
Cement Plants	--	--	--	--	--	--	--
Commercial Cooking	26,922	--	--	--	--	26,922	--
ODS Substitutes/Nat. Gas Distrib./Other	--	1,659	--	479	0.13	945,746	--
Reciprocating Engines	73,600	129	--	--	--	76,331	16,585
Turbines	31,710	71	--	--	--	33,325	66,141
Natural Gas- Major Combustion Sources	497,257	18	1	--	--	497,927	--
Natural Gas- Minor Combustion Sources	1,141,243	22	21	--	--	1,148,189	--
Coke Coal	25,266	--	--	--	--	25,313	--
Other Fuels Combustion	88,518	25	--	--	--	89,133	9,320
Subtotal	1,884,516	20,325	220	479	0.13	3,290,493	283,142
RESIDENTIAL FUEL USAGE							
Natural Gas	1,286,715	25	24	--	--	1,294,546	--
LPgas/Liquid Fuel	24,050	--	2	--	--	24,543	--
Solid Fuel	--	710	8	--	--	17,255	71,930
Subtotal	1,310,766	735	33	--	--	1,336,344	71,930
ELECTRICITY/ CO-GENERATION							
Co-Generation	111,703	394	--	--	--	120,085	55,974
Electricity Generation	26,465	1	--	--	0.26	32,715	396
Electricity Imports	1,846,601	15	8	--	--	1,849,556	--
Subtotal	1,984,769	411	9	--	0.26	2,002,356	56,370
OFF-ROAD EQUIPMENT							
Lawn and Garden Equipment	23,536	43	17	--	--	29,781	--
Construction Equipment	360,793	58	2	--	--	362,691	--
Industrial Equipment	122,557	75	7	--	--	126,281	--
Light Commercial Equipment	46,774	23	9	--	--	49,913	--
Subtotal	553,660	199	35	--	--	568,666	--
TRANSPORTATION							
Off-Road							
Locomotives	24,977	1	10	--	--	28,113	--
Ships	108,727	6	2	--	--	109,479	--
Boats	293,593	35	106	--	--	327,032	--
Commercial Aircraft	482,494	20	17	--	--	488,135	--
General Aviation	64,925	11	2	--	--	65,881	--
Military Aircraft	3,677	--	--	--	--	3,694	--
On-Road							
Passenger Cars/Trucks up to 10,000 lbs	5,744,125	621	336	--	--	5,861,250	--
Medium/Heavy Duty Trucks > 10,000 lbs	1,209,381	54	60	--	--	1,229,050	--
Urban,School and Other Buses	186,711	4	57	--	--	204,517	--
Motor-Homes and Motorcycles	30,728	29	9	--	--	34,017	--
Subtotal	8,149,332	782	598	--	--	8,351,165	--
AGRICULTURE/ FARMING							
Agricultural Equipment	15,487	3	--	--	--	15,607	--
Animal Waste	--	2,814	52	--	--	75,340	--
Soil Management	954	--	40	--	--	13,305	1,146
Biomass Burning	--	58	3	--	--	2,269	1,199
Subtotal	16,441	2,875	96	--	--	106,522	2,345
GRAND TOTAL EMISSIONS	**13,899,491**	**25,326**	**991**	**479**	**0.4**	**15,655,546**	**413,788**

Table M:

Annual GHG Emissions: CONTRA COSTA Year 2007 (Metric Tons / Year)

SOURCE CATEGORY	CO_2	CH_4	N_2O	PFC/HFC	SF_6	Total GHG CO_2-Equivalent	Biogenic CO_2
INDUSTRIAL/ COMMERCIAL							
Oil Refineries							
Refining Processes	2,910,760	71	--	--	--	2,912,287	--
Refinery Make Gas Combustion	3,455,837	70	5	--	--	3,458,717	--
Natural Gas and Other Gases Combustion	4,773,383	258	18	--	--	4,784,246	--
Liquid Fuel Combustion	89,450	1	1	--	--	89,760	--
Solid Fuel Combustion	1,000,216	29	6	--	--	1,002,637	--
Waste Management							
Landfill Combustion Sources	--	83	--	--	--	1,761	38,904
Landfill Fugitive Sources	--	6,168	--	--	--	129,622	17,044
Composting/POTWs	--	65	87	--	--	28,228	--
Other Industrial/ Commercial							
Cement Plants	--	--	--	--	--	--	--
Commercial Cooking	13,461	--	--	--	--	13,461	--
ODS Substitutes/Nat. Gas Distrib./Other	--	8,604	--	282	--	686,738	--
Reciprocating Engines	81,825	391	--	--	--	90,082	18,464
Turbines	3,781	--	--	--	--	3,788	--
Natural Gas- Major Combustion Sources	633,351	7	1	--	--	633,758	--
Natural Gas- Minor Combustion Sources	4,137,569	79	76	--	--	4,162,750	--
Coke Coal	964,175	28	6	--	--	966,509	--
Other Fuels Combustion	213,592	26	1	--	--	214,332	34,769
Subtotal	18,277,402	15,880	199	282	--	19,178,675	109,181
RESIDENTIAL FUEL USAGE							
Natural Gas	994,896	19	18	--	--	1,000,951	--
LPgas/Liquid Fuel	18,887	--	1	--	--	19,288	--
Solid Fuel	--	1,666	18	--	--	40,504	167,246
Subtotal	1,013,783	1,686	37	--	--	1,060,743	167,246
ELECTRICITY/ CO-GENERATION							
Co-Generation	4,245,060	372	2	--	--	4,253,633	--
Electricity Generation	1,449,714	73	--	--	0.17	1,455,466	1,620
Electricity Imports	--	--	--	--	--	--	--
Subtotal	5,694,774	444	3	--	0.17	5,709,099	1,620
OFF-ROAD EQUIPMENT							
Lawn and Garden Equipment	16,040	29	12	--	--	20,296	--
Construction Equipment	303,191	49	2	--	--	304,760	--
Industrial Equipment	52,636	29	2	--	--	53,996	--
Light Commercial Equipment	25,132	12	5	--	--	26,861	--
Subtotal	397,000	120	21	--	--	405,913	--
TRANSPORTATION							
Off-Road							
Locomotives	24,562	1	10	--	--	27,646	--
Ships	63,340	3	1	--	--	63,758	--
Boats	69,323	44	21	--	--	76,609	--
Commercial Aircraft	--	--	--	--	--	--	--
General Aviation	19,607	11	1	--	--	20,037	--
Military Aircraft	--	--	--	--	--	--	--
On-Road	--	--	--	--	--	--	
Passenger Cars/Trucks up to 10,000 lbs	4,178,993	432	234	--	--	4,260,682	--
Medium/Heavy Duty Trucks > 10,000 lbs	401,364	23	27	--	--	410,348	--
Urban,School and Other Buses	102,211	2	15	--	--	106,770	--
Motor-Homes and Motorcycles	29,368	27	8	--	--	32,309	--
Subtotal	4,888,771	543	316	--	--	4,998,160	--
AGRICULTURE/ FARMING							
Agricultural Equipment	19,349	4	--	--	--	19,500	--
Animal Waste	--	4,440	44	--	--	106,901	--
Soil Management	913	--	204	--	--	64,034	589
Biomass Burning	--	10	1	--	--	431	274
Subtotal	20,262	4,454	249	--	--	190,867	863
GRAND TOTAL EMISSIONS	**30,291,977**	**23,126**	**825**	**282**	**0.17**	**31,543,450**	**278,910**

Table N:
Annual GHG Emissions: MARIN Year 2007 (Metric Tons / Year)

SOURCE CATEGORY	CO_2	CH_4	N_2O	PFC/HFC	SF_6	Total GHG CO_2-Equivalent	Biogenic CO_2
INDUSTRIAL/ COMMERCIAL							
Oil Refineries							
Refining Processes	--	--	--	--	--	--	--
Refinery Make Gas Combustion	--	--	--	--	--	--	--
Natural Gas and Other Gases Combustion	--	--	--	--	--	--	--
Liquid Fuel Combustion	--	--	--	--	--	--	--
Solid Fuel Combustion	--	--	--	--	--	--	--
Waste Management							
Landfill Combustion Sources	--	131	--	--	--	2,788	61,597
Landfill Fugitive Sources	--	5,278	--	--	--	110,927	14,093
Composting/POTWs	--	39	37	--	--	12,390	--
Other Industrial/ Commercial							
Cement Plants	--	--	--	--	--	--	--
Commercial Cooking	4,038	--	--	--	--	4,038	--
ODS Substitutes/Nat. Gas Distrib./Other	--	273	--	87	--	161,358	--
Reciprocating Engines	16,158	4	--	--	--	16,237	401
Turbines	1	--	--	--	--	1	--
Natural Gas- Major Combustion Sources	22,929	--	--	--	--	22,950	--
Natural Gas- Minor Combustion Sources	132,891	3	2	--	--	133,699	--
Coke Coal	--	--	--	--	--	--	--
Other Fuels Combustion	4,472	6	--	--	--	4,602	1,173
Subtotal	180,489	5,733	40	87	--	468,992	77,263
RESIDENTIAL FUEL USAGE							
Natural Gas	331,202	6	6	--	--	333,217	--
LPgas/Liquid Fuel	14,330	--	1	--	--	14,630	--
Solid Fuel	--	587	6	--	--	14,264	58,869
Subtotal	345,532	593	13	--	--	362,111	58,869
ELECTRICITY/ CO-GENERATION							
Co-Generation	2,581	31	--	--	--	3,224	1,776
Electricity Generation	--	--	--	--	0.04	1,022	--
Electricity Imports	284,092	2	1	--	--	284,547	--
Subtotal	286,673	33	1	--	0.04	288,793	1,776
OFF-ROAD EQUIPMENT							
Lawn and Garden Equipment	6,259	11	5	--	--	7,920	--
Construction Equipment	66,234	11	--	--	--	66,583	--
Industrial Equipment	11,942	6	1	--	--	12,230	--
Light Commercial Equipment	11,588	6	2	--	--	12,345	--
Subtotal	96,023	34	8	--	--	99,079	--
TRANSPORTATION							
Off-Road							
Locomotives	725	--	--	--	--	817	--
Ships	53,605	6	2	--	--	54,497	--
Boats	20,077	48	6	--	--	22,818	--
Commercial Aircraft	--	--	--	--	--	--	--
General Aviation	17,302	3	1	--	--	17,559	--
Military Aircraft	--	--	--	--	--	--	--
On-Road							
Passenger Cars/Trucks up to 10,000 lbs	1,008,598	108	59	--	--	1,029,239	--
Medium/Heavy Duty Trucks > 10,000 lbs	80,905	6	8	--	--	83,376	--
Urban,School and Other Buses	64,651	1	20	--	--	71,011	--
Motor-Homes and Motorcycles	5,642	6	2	--	--	6,283	--
Subtotal	1,251,506	177	98	--	--	1,285,600	--
AGRICULTURE/ FARMING							
Agricultural Equipment	7,486	1	--	--	--	7,544	--
Animal Waste	--	7,352	44	--	--	168,172	--
Soil Management	695	--	34	--	--	11,178	--
Biomass Burning	--	3	--	--	--	120	104
Subtotal	8,181	7,356	79	--	--	187,014	104
GRAND TOTAL EMISSIONS	**2,168,404**	**13,927**	**239**	**87**	**0.04**	**2,691,589**	**138,012**

Table O:

Annual GHG Emissions: NAPA Year 2007 (Metric Tons / Year)

SOURCE CATEGORY	CO_2	CH_4	N_2O	PFC/HFC	SF_6	Total GHG CO_2-Equivalent	Biogenic CO_2
INDUSTRIAL/ COMMERCIAL							
Oil Refineries							
Refining Processes	--	--	--	--	--	--	--
Refinery Make Gas Combustion	--	--	--	--	--	--	--
Natural Gas and Other Gases Combustion	--	--	--	--	--	--	--
Liquid Fuel Combustion	--	--	--	--	--	--	--
Solid Fuel Combustion	--	--	--	--	--	--	--
Waste Management							
Landfill Combustion Sources	--	31	--	--	--	651	14,380
Landfill Fugitive Sources	--	535	--	--	--	11,245	1,669
Composting/POTWs	--	149	43	--	--	16,359	--
Other Industrial/ Commercial							
Cement Plants	--	--	--	--	--	--	--
Commercial Cooking	2,692	--	--	--	--	2,692	--
ODS Substitutes/Nat. Gas Distrib./Other	--	128	--	40	--	74,966	--
Reciprocating Engines	17,563	106	--	--	--	19,801	15,424
Turbines	6	--	--	--	--	6	--
Natural Gas- Major Combustion Sources	32,505	1	--	--	--	32,534	--
Natural Gas- Minor Combustion Sources	94,281	2	2	--	--	94,854	--
Coke Coal	--	--	--	--	--	--	--
Other Fuels Combustion	460	2	--	--	--	511	1,106
Subtotal	147,506	953	45	40	--	253,621	32,580
RESIDENTIAL FUEL USAGE							
Natural Gas	121,376	2	2	--	--	122,115	--
LPgas/Liquid Fuel	6,334	--	--	--	--	6,468	--
Solid Fuel	--	275	3	--	--	6,688	27,600
Subtotal	127,710	278	6	--	--	135,271	27,600
ELECTRICITY/ CO-GENERATION							
Co-Generation	12,847	1	--	--	--	12,874	--
Electricity Generation	7,917	1	--	--	0.02	8,474	--
Electricity Imports	213,069	2	1	--	--	213,410	--
Subtotal	233,833	3	1	--	0.02	234,758	--
OFF-ROAD EQUIPMENT							
Lawn and Garden Equipment	2,252	4	2	--	--	2,850	--
Construction Equipment	31,597	5	--	--	--	31,763	--
Industrial Equipment	10,994	6	--	--	--	11,267	--
Light Commercial Equipment	4,050	2	1	--	--	4,340	--
Subtotal	48,893	17	3	--	--	50,220	--
TRANSPORTATION							
Off-Road							
Locomotives	4,353	--	2	--	--	4,899	--
Ships	--	--	--	--	--	--	--
Boats	24,628	29	6	--	--	27,189	--
Commercial Aircraft	--	--	--	--	--	--	--
General Aviation	16,274	3	1	--	--	16,517	--
Military Aircraft	--	--	--	--	--	--	--
On-Road							
Passenger Cars/Trucks up to 10,000 lbs	747,955	89	50	--	--	765,209	--
Medium/Heavy Duty Trucks > 10,000 lbs	83,920	6	6	--	--	85,933	--
Urban,School and Other Buses	9,640	--	2	--	--	10,289	--
Motor-Homes and Motorcycles	6,650	4	2	--	--	7,213	--
Subtotal	893,420	131	68	--	--	917,248	--
AGRICULTURE/ FARMING							
Agricultural Equipment	32,463	6	--	--	--	32,716	--
Animal Waste	--	1,131	21	--	--	30,180	--
Soil Management	7,431	--	93	--	--	36,148	35
Biomass Burning	--	2	1	--	--	423	596
Subtotal	39,894	1,139	115	--	--	99,467	631
GRAND TOTAL EMISSIONS	**1,491,256**	**2,522**	**237**	**40**	**0.02**	**1,690,586**	**60,810**

Table P:

Annual GHG Emissions: SAN FRANCISCO Year 2007 (Metric Tons / Year)

SOURCE CATEGORY	**CO_2**	**CH_4**	**N_2O**	**PFC/HFC**	**SF_6**	**Total GHG CO_2-Equivalent**	**Biogenic CO_2**
INDUSTRIAL/ COMMERCIAL							
Oil Refineries							
Refining Processes	--	--	--	--	--	--	--
Refinery Make Gas Combustion	--	--	--	--	--	--	--
Natural Gas and Other Gases Combustion	--	--	--	--	--	--	--
Liquid Fuel Combustion	--	--	--	--	--	--	--
Solid Fuel Combustion	--	--	--	--	--	--	--
Waste Management							
Landfill Combustion Sources	--	--	--	--	--	--	--
Landfill Fugitive Sources	--	--	--	--	--	--	--
Composting/POTWs	--	8	74	--	--	23,029	--
Other Industrial/ Commercial							
Cement Plants	--	--	--	--	--	--	--
Commercial Cooking	25,576	--	--	--	--	25,576	--
ODS Substitutes/Nat. Gas Distrib./Other	--	1,595	--	279	--	534,013	1
Reciprocating Engines	68,970	56	--	--	--	71,300	--
Turbines	45,071	1	--	--	--	45,197	--
Natural Gas- Major Combustion Sources	674,021	5	1	--	--	674,276	--
Natural Gas- Minor Combustion Sources	470,505	9	9	--	--	473,368	--
Coke Coal	--	--	--	--	--	--	--
Other Fuels Combustion	11,686	43	--	--	--	12,653	16,498
Subtotal	1,295,829	1,716	84	279	--	1,859,413	16,499
RESIDENTIAL FUEL USAGE							
Natural Gas	838,011	16	15	--	--	843,111	--
LPgas/Liquid Fuel	23,226	--	1	--	--	23,647	--
Solid Fuel	--	104	1	--	--	2,525	10,988
Subtotal	861,237	120	18	--	--	869,283	10,988
ELECTRICITY/ CO-GENERATION							
Co-Generation	178,970	89	--	--	--	180,863	100
Electricity Generation	7,504	--	--	--	0.14	10,753	--
Electricity Imports	1,136,370	9	5	--	--	1,138,188	--
Subtotal	1,322,843	99	5	--	0.14	1,329,804	100
OFF-ROAD EQUIPMENT							
Lawn and Garden Equipment	11,250	20	8	--	--	14,235	--
Construction Equipment	292,545	47	2	--	--	294,071	--
Industrial Equipment	60,802	38	4	--	--	62,697	--
Light Commercial Equipment	41,246	20	7	--	--	43,884	--
Subtotal	405,842	125	21	--	--	414,888	--
TRANSPORTATION							
Off-Road							
Locomotives	2,176	--	1	--	--	2,450	--
Ships	146,036	17	6	--	--	148,381	--
Boats	38,535	25	12	--	--	42,843	--
Commercial Aircraft	--	--	--	--	--	--	--
General Aviation	--	--	--	--	--	--	--
Military Aircraft	--	--	--	--	--	--	--
On-Road							
Passenger Cars/Trucks up to 10,000 lbs	2,059,302	221	106	--	--	2,096,833	--
Medium/Heavy Duty Trucks > 10,000 lbs	212,244	14	19	--	--	218,391	--
Urban,School and Other Buses	140,574	4	48	--	--	155,441	--
Motor-Homes and Motorcycles	7,248	12	3	--	--	8,344	--
Subtotal	2,606,117	292	195	--	--	2,672,683	--
AGRICULTURE/ FARMING							
Agricultural Equipment	110	--	--	--	--	111	--
Animal Waste	--	--	--	--	--	--	--
Soil Management	4	--	--	--	--	4	1,007
Biomass Burning	--	--	--	--	--	--	--
Subtotal	115	--	--	--	--	116	1,007
GRAND TOTAL EMISSIONS	**6,491,990**	**2,352**	**322**	**279**	**0.14**	**7,146,187**	**28,594**

Table Q:

Annual GHG Emissions: SAN MATEO Year 2007 (Metric Tons / Year)

SOURCE CATEGORY	CO_2	CH_4	N_2O	PFC/HFC	SF_6	Total GHG CO_2-Equivalent	Biogenic CO_2
INDUSTRIAL/ COMMERCIAL							
Oil Refineries							
Refining Processes	--	--	--	--	--	--	--
Refinery Make Gas Combustion	--	--	--	--	--	--	--
Natural Gas and Other Gases Combustion	--	--	--	--	--	--	--
Liquid Fuel Combustion	--	--	--	--	--	--	--
Solid Fuel Combustion	--	--	--	--	--	--	--
Waste Management							
Landfill Combustion Sources	--	245	--	--	--	5,234	115,658
Landfill Fugitive Sources	--	8,537	--	--	--	179,414	22,593
Composting/POTWs	--	60	86	--	--	27,932	--
Other Industrial/ Commercial							
Cement Plants	--	--	--	--	--	--	--
Commercial Cooking	13,461	--	--	--	--	13,461	--
ODS Substitutes/Nat. Gas Distrib./Other	--	835	--	236	--	442,682	20
Reciprocating Engines	46,803	112	--	--	--	49,172	18,613
Turbines	18	--	--	--	--	18	--
Natural Gas- Major Combustion Sources	76,685	1	--	--	--	76,754	--
Natural Gas- Minor Combustion Sources	747,061	14	14	--	--	751,608	--
Coke Coal	--	--	--	--	--	--	--
Other Fuels Combustion	5,274	16	--	--	--	5,632	4,455
Subtotal	889,303	9,821	101	236	--	1,551,908	161,338
RESIDENTIAL FUEL USAGE							
Natural Gas	719,218	14	13	--	--	723,595	--
LPgas/Liquid Fuel	19,086	--	1	--	--	19,481	--
Solid Fuel	--	379	4	--	--	9,203	38,326
Subtotal	738,303	393	18	--	--	752,278	38,326
ELECTRICITY/ CO-GENERATION							
Co-Generation	36,874	24	--	--	--	37,399	1,344
Electricity Generation	--	--	--	--	0.12	2,939	--
Electricity Imports	994,323	8	5	--	--	995,915	--
Subtotal	1,031,197	32	5	--	0.12	1,036,254	1,344
OFF-ROAD EQUIPMENT							
Lawn and Garden Equipment	11,208	20	8	--	--	14,182	--
Construction Equipment	173,531	28	1	--	--	174,445	--
Industrial Equipment	52,204	32	3	--	--	53,797	--
Light Commercial Equipment	25,539	12	5	--	--	27,226	--
Subtotal	262,482	93	17	--	--	269,650	--
TRANSPORTATION							
Off-Road							
Locomotives	4,353	--	2	--	--	4,899	--
Ships	347,697	41	16	--	--	353,471	--
Boats	17,157	19	6	--	--	19,293	--
Commercial Aircraft	1,076,869	61	39	--	--	1,090,389	--
General Aviation	25,284	4	1	--	--	25,664	--
Military Aircraft	4,449	--	--	--	--	4,470	--
On-Road							
Passenger Cars/Trucks up to 10,000 lbs	2,972,646	281	154	--	--	3,026,154	--
Medium/Heavy Duty Trucks > 10,000 lbs	224,315	14	21	--	--	230,958	--
Urban,School and Other Buses	74,368	2	19	--	--	80,264	--
Motor-Homes and Motorcycles	12,494	16	4	--	--	14,098	--
Subtotal	4,759,634	437	261	--	--	4,849,664	--
AGRICULTURE/ FARMING							
Agricultural Equipment	9,233	2	--	--	--	9,305	--
Animal Waste	--	257	9	--	--	8,101	--
Soil Management	348	--	30	--	--	9,520	139
Biomass Burning	--	3	--	--	--	136	98
Subtotal	9,581	262	39	--	--	27,062	237
GRAND TOTAL EMISSIONS	**7,690,500**	**11,037**	**440**	**236**	**0.12**	**8,486,808**	**201,245**

Table R:
Annual GHG Emissions: SANTA CLARA Year 2007 (Metric Tons / Year)

SOURCE CATEGORY	CO_2	CH_4	N_2O	PFC/HFC	SF_6	Total GHG CO_2-Equivalent	Biogenic CO_2
INDUSTRIAL/ COMMERCIAL							
Oil Refineries							
Refining Processes	--	--	--	--	--	--	--
Refinery Make Gas Combustion	--	--	--	--	--	--	--
Natural Gas and Other Gases Combustion	--	--	--	--	--	--	--
Liquid Fuel Combustion	--	--	--	--	--	--	--
Solid Fuel Combustion	--	--	--	--	--	--	--
Waste Management							
Landfill Combustion Sources	--	440	--	--	--	9,374	207,134
Landfill Fugitive Sources	--	12,444	1	--	--	261,566	35,205
Composting/POTWs	--	1,382	309	--	--	124,918	--
Other Industrial/ Commercial							
Cement Plants	841,350	13	3	--	--	842,475	--
Commercial Cooking	30,961	--	--	--	--	30,961	--
ODS Substitutes/Nat. Gas Distrib./Other	--	2,426	--	582	--	1,167,459	--
Reciprocating Engines	193,233	742	1	--	--	209,049	104,164
Turbines	349	--	--	--	--	350	--
Natural Gas- Major Combustion Sources	361,211	5	1	--	--	361,532	--
Natural Gas- Minor Combustion Sources	1,691,662	32	31	--	--	1,701,957	--
Coke Coal	--	--	--	--	--	--	--
Other Fuels Combustion	15,307	5	--	--	--	15,554	15,266
Subtotal	3,134,075	17,489	346	582	--	4,725,195	361,770
RESIDENTIAL FUEL USAGE							
Natural Gas	1,555,938	30	29	--	--	1,565,407	--
LPgas/Liquid Fuel	40,631	1	3	--	--	41,464	--
Solid Fuel	--	1,238	13	--	--	30,092	124,813
Subtotal	1,596,569	1,268	44	--	--	1,636,963	124,813
ELECTRICITY/ CO-GENERATION							
Co-Generation	322,599	242	--	--	--	327,843	24,023
Electricity Generation	1,207,022	88	--	--	0.30	1,216,062	1,510
Electricity Imports	2,059,670	17	9	--	--	2,062,967	--
Subtotal	3,589,291	347	10	--	0.30	3,606,872	25,533
OFF-ROAD EQUIPMENT							
Lawn and Garden Equipment	26,010	47	19	--	--	32,912	--
Construction Equipment	370,445	60	2	--	--	372,396	--
Industrial Equipment	316,775	216	22	--	--	328,058	--
Light Commercial Equipment	52,797	26	10	--	--	56,451	--
Subtotal	766,027	349	53	--	--	789,817	--
TRANSPORTATION							
Off-Road							
Locomotives	16,064	1	6	--	--	18,081	--
Ships	--	--	--	--	--	--	--
Boats	17,199	10	4	--	--	18,721	--
Commercial Aircraft	318,300	9	12	--	--	322,137	--
General Aviation	63,542	10	2	--	--	64,493	--
Military Aircraft	46,921	9	2	--	--	47,627	--
On-Road							
Passenger Cars/Trucks up to 10,000 lbs	6,428,736	704	363	--	--	6,555,920	--
Medium/Heavy Duty Trucks > 10,000 lbs	678,747	40	45	--	--	693,658	--
Urban,School and Other Buses	88,596	3	22	--	--	95,407	--
Motor-Homes and Motorcycles	39,091	32	10	--	--	42,785	--
Subtotal	7,697,197	818	466	--	--	7,858,834	--
AGRICULTURE/ FARMING							
Agricultural Equipment	35,719	7	--	--	--	35,997	--
Animal Waste	--	2,095	44	--	--	57,557	--
Soil Management	1,034	--	225	--	--	70,707	318
Biomass Burning	--	2	--	--	--	139	141
Subtotal	36,753	2,104	269	--	--	164,401	459
GRAND TOTAL EMISSIONS	**16,819,916**	**22,376**	**1,189**	**582**	**0.30**	**18,782,077**	**512,575**

Table S:

Annual GHG Emissions: SOLANO* Year 2007 (Metric Tons / Year)

SOURCE CATEGORY	CO_2	CH_4	N_2O	PFC/HFC	SF_6	Total GHG CO_2-Equivalent	Biogenic CO_2
INDUSTRIAL/ COMMERCIAL							
Oil Refineries							
Refining Processes	534,305	9	--	--	--	534,495	--
Refinery Make Gas Combustion	1,317,135	27	2	--	--	1,318,241	--
Natural Gas and Other Gases Combustion	86,885	9	1	--	--	87,249	--
Liquid Fuel Combustion	--	--	--	--	--	--	--
Solid Fuel Combustion	--	--	--	--	--	--	--
Waste Management							
Landfill Combustion Sources	--	8	--	--	--	172	3,803
Landfill Fugitive Sources	--	1,550	--	--	--	32,564	4,120
Composting/POTWs	--	218	44	--	--	18,110	--
Other Industrial/ Commercial							
Cement Plants	--	--	--	--	--	--	--
Commercial Cooking	8,077	--	--	--	--	8,077	--
ODS Substitutes/Nat. Gas Distrib./Other	--	498	--	84	--	160,727	--
Reciprocating Engines	27,294	71	--	--	--	28,792	1,775
Turbines	273,754	6	--	--	--	273,970	--
Natural Gas- Major Combustion Sources	76,427	14	--	--	--	76,769	--
Natural Gas- Minor Combustion Sources	348,389	7	6	--	--	350,509	--
Coke Coal	--	--	--	--	--	--	--
Other Fuels Combustion	3,506	6	--	--	--	3,635	2,344
Subtotal	2,675,772	2,421	53	84	--	2,893,310	12,042
RESIDENTIAL FUEL USAGE							
Natural Gas	245,980	5	5	--	--	247,477	--
LPgas/Liquid Fuel	3,763	--	--	--	--	3,843	--
Solid Fuel	--	264	3	--	--	6,426	26,568
Subtotal	249,744	269	8	--	--	257,747	26,568
ELECTRICITY/ CO-GENERATION							
Co-Generation	375,712	12	--	--	--	375,968	--
Electricity Generation	32,350	1	--	--	0.05	33,595	--
Electricity Imports	--	--	--	--	--	--	--
Subtotal	408,063	12	--	--	0.05	409,563	--
OFF-ROAD EQUIPMENT							
Lawn and Garden Equipment	3,045	6	2	--	--	3,853	--
Construction Equipment	68,732	11	--	--	--	69,093	--
Industrial Equipment	66,709	12	1	--	--	67,152	--
Light Commercial Equipment	6,391	3	1	--	--	6,848	--
Subtotal	144,876	32	5	--	--	146,946	--
TRANSPORTATION							
Off-Road							
Locomotives	5,804	--	2	--	--	6,532	--
Ships	12,274	2	1	--	--	12,478	--
Boats	11,276	25	3	--	--	12,732	--
Commercial Aircraft	--	--	--	--	--	--	--
General Aviation	--	--	--	--	--	--	--
Military Aircraft	423,131	21	13	--	--	427,663	--
On-Road							
Passenger Cars/Trucks up to 10,000 lbs	1,139,899	114	63	--	--	1,161,801	--
Medium/Heavy Duty Trucks > 10,000 lbs	168,039	9	9	--	--	170,918	--
Urban,School and Other Buses	30,727	--	5	--	--	32,268	--
Motor-Homes and Motorcycles	9,193	8	2	--	--	10,048	--
Subtotal	1,800,342	178	98	--	--	1,834,440	--
AGRICULTURE/ FARMING							
Agricultural Equipment	32,041	6	--	--	--	32,291	--
Animal Waste	--	1,037	3	--	--	22,727	--
Soil Management	871	--	197	--	--	62,013	39,600
Biomass Burning	--	2	--	--	--	95	76
Subtotal	32,912	1,045	201	--	--	117,126	39,676
GRAND TOTAL EMISSIONS	**5,311,708**	**3,957**	**364**	**84**	**0.05**	**5,659,130**	**78,287**

*** BAAQMD Jurisdiction only**

Table T:

Annual GHG Emissions: SONOMA* Year 2007 (Metric Tons / Year)

SOURCE CATEGORY	CO_2	CH_4	N_2O	PFC/HFC	SF_6	Total GHG CO_2-Equivalent	Biogenic CO_2
INDUSTRIAL/ COMMERCIAL							
Oil Refineries							
Refining Processes	--	--	--	--	--	--	--
Refinery Make Gas Combustion	--	--	--	--	--	--	--
Natural Gas and Other Gases Combustion	--	--	--	--	--	--	--
Liquid Fuel Combustion	--	--	--	--	--	--	--
Solid Fuel Combustion	--	--	--	--	--	--	--
Waste Management							
Landfill Combustion Sources	--	--	--	--	--	1	13
Landfill Fugitive Sources	--	4,416	--	--	--	92,809	11,669
Composting/POTWs	--	573	89	--	--	39,574	--
Other Industrial/ Commercial							
Cement Plants	--	--	--	--	--	--	--
Commercial Cooking	9,423	--	--	--	--	9,423	--
ODS Substitutes/Nat. Gas Distrib./Other	--	337	--	117	--	217,310	--
Reciprocating Engines	24,824	379	--	--	--	32,820	58,586
Turbines	7	--	--	--	--	7	--
Natural Gas- Major Combustion Sources	25,653	--	--	--	--	25,676	--
Natural Gas- Minor Combustion Sources	215,498	4	4	--	--	216,810	--
Coke Coal	--	--	--	--	--	--	--
Other Fuels Combustion	6,357	3	--	--	--	6,433	415
Subtotal	281,761	5,713	93	117	--	640,862	70,682
RESIDENTIAL FUEL USAGE							
Natural Gas	362,837	7	7	--	--	365,045	--
LPgas/Liquid Fuel	16,199	--	1	--	--	16,546	--
Solid Fuel	--	1,020	11	--	--	24,786	102,209
Subtotal	379,036	1,027	19	--	--	406,377	102,209
ELECTRICITY/ CO-GENERATION							
Co-Generation	6,478	96	--	--	--	8,506	6,294
Electricity Generation	--	--	--	--	0.08	1,940	--
Electricity Imports	568,185	5	3	--	--	569,094	--
Subtotal	574,663	101	3	--	0.08	579,541	6,294
OFF-ROAD EQUIPMENT							
Lawn and Garden Equipment	6,143	11	5	--	--	7,773	--
Construction Equipment	118,009	19	1	--	--	118,631	--
Industrial Equipment	34,416	20	2	--	--	35,374	--
Light Commercial Equipment	12,601	6	2	--	--	13,506	--
Subtotal	171,169	56	9	--	--	175,285	--
TRANSPORTATION							
Off-Road							
Locomotives	5,078	--	2	--	--	5,716	--
Ships	--	--	--	--	--	--	--
Boats	17,376	19	5	--	--	19,213	--
Commercial Aircraft	--	--	--	--	--	--	--
General Aviation	24,133	4	1	--	--	24,492	--
Military Aircraft	--	--	--	--	--	--	--
On-Road							
Passenger Cars/Trucks up to 10,000 lbs	1,790,567	211	115	--	--	1,830,792	--
Medium/Heavy Duty Trucks > 10,000 lbs	174,035	12	14	--	--	178,612	--
Urban,School and Other Buses	25,221	1	5	--	--	26,677	--
Motor-Homes and Motorcycles	16,221	13	4	--	--	17,730	--
Subtotal	2,052,632	260	146	--	--	2,103,232	--
AGRICULTURE/ FARMING							
Agricultural Equipment	32,040	6	--	--	--	32,290	--
Animal Waste	--	6,735	37	--	--	152,783	--
Soil Management	3,703	--	78	--	--	27,848	277
Biomass Burning	--	16	1	--	--	750	657
Subtotal	35,744	6,757	116	--	--	213,671	934
GRAND TOTAL EMISSIONS	**3,495,004**	**13,914**	**386**	**117**	**0.08**	**4,118,968**	**180,119**

*** BAAQMD Jurisdiction only**

Table U: Bay Area Greenhouse Gas Emission Inventory Projections : 1990 - 2029

(Million Metric Tons CO_2- Equivalent)

SOURCE CATEGORY Year	1990	1993	1996	1999	2002	2005	2008	2011	2014	2017	2020	2023	2026	2029
INDUSTRIAL/ COMMERCIAL														
Oil Refineries														
Refining Processes	3.3	3.5	3.6	3.7	3.5	3.4	3.5	3.6	3.7	3.8	3.9	4.0	4.2	4.3
Refinery Make Gas Combustion	3.8	4.0	3.7	4.4	4.5	4.7	4.8	5.0	5.1	5.3	5.4	5.6	5.8	5.9
Natural Gas and Other Gases Combustion	4.5	4.3	4.5	4.5	4.6	4.8	4.9	5.1	5.2	5.4	5.5	5.7	5.9	6.1
Liquid Fuel Combustion	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Solid Fuel Combustion	0.8	0.9	0.9	0.9	1.0	1.0	1.0	1.0	1.1	1.1	1.1	1.2	1.2	1.2
Waste Management														
Landfill Combustion Sources	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Landfill Fugitive Sources	1.7	1.4	1.1	1.1	1.1	1.2	1.2	1.2	1.2	1.2	1.2	1.2	1.1	1.1
Composting/POTWs	0.2	0.3	0.3	0.4	0.3	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4
Other Industrial/ Commercial														
Cement Plants	0.9	0.9	0.9	1.0	0.8	0.9	0.9	0.9	0.9	1.0	1.0	1.0	1.1	1.1
Commercial Cooking	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.2	0.2	0.2
ODS Substitutes/Nat. Gas Distrib./Other	1.1	1.3	1.8	2.4	2.9	3.6	4.8	5.9	7.1	8.3	9.4	10.6	11.7	12.8
Reciprocating Engines	0.5	0.5	0.6	0.5	0.5	0.6	0.6	0.6	0.6	0.7	0.7	0.7	0.8	0.8
Turbines	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.5
Natural Gas- Major Combustion Sources	1.9	1.8	1.6	1.8	1.5	1.6	2.4	2.5	2.6	2.7	2.8	3.0	3.1	3.2
Natural Gas- Minor Combustion Sources	2.3	7.2	8.5	8.5	8.5	8.8	9.1	9.4	9.7	10.1	10.4	10.7	11.0	11.3
Coke Coal	1.7	1.5	1.5	1.0	1.0	1.0	1.0	1.0	1.1	1.1	1.2	1.2	1.2	1.3
Other Fuels Combustion	0.4	0.4	0.4	0.3	0.3	0.3	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.5
Subtotal	23.9	28.5	29.9	31.1	31.4	32.8	35.6	37.7	39.9	42.0	44.2	46.4	48.6	50.8
RESIDENTIAL FUEL USAGE														
Natural Gas	5.4	5.6	5.9	6.1	6.2	6.4	6.5	6.7	6.9	7.0	7.2	7.4	7.5	7.7
LPgas/Liquid Fuel	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Solid Fuel	0.1	0.2	0.2	0.1	0.1	0.1	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Subtotal	5.8	6.0	6.2	6.4	6.6	6.7	6.9	7.0	7.2	7.4	7.5	7.7	7.9	8.0
ELECTRICITY/ CO-GENERATION														
Co-Generation	8.6	7.2	6.7	5.2	5.4	5.5	5.4	5.6	5.9	6.1	6.4	6.7	6.9	7.2
Electricity Generation	7.6	8.5	4.0	4.6	4.7	2.8	3.0	3.1	3.3	3.4	3.5	3.7	3.8	4.0
Electricity Imports	8.9	8.8	10.3	10.1	6.9	6.8	7.2	7.5	7.8	8.1	8.3	8.6	8.9	9.1
Subtotal	25.1	24.6	20.9	19.8	17.0	15.1	15.6	16.3	16.9	17.6	18.3	18.9	19.6	20.4

Table U: Bay Area Greenhouse Gas Emission Inventory Projections : 1990 - 2029

(Million Metric Tons CO_2- Equivalent)

SOURCE CATEGORY **Year**	**1990**	**1993**	**1996**	**1999**	**2002**	**2005**	**2008**	**2011**	**2014**	**2017**	**2020**	**2023**	**2026**	**2029**
OFF-ROAD EQUIPMENT														
Lawn and Garden Equipment	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Construction Equipment	1.3	1.4	1.5	1.6	1.6	1.7	1.8	1.9	2.0	2.1	2.2	2.3	2.3	2.4
Industrial Equipment	0.6	0.6	0.6	0.7	0.7	0.7	0.8	0.8	0.9	0.9	1.0	1.1	1.1	1.2
Light Commercial Equipment	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Subtotal	2.2	2.3	2.5	2.6	2.7	2.8	3.0	3.1	3.3	3.4	3.6	3.8	3.9	4.1
TRANSPORTATION														
Off-Road														
Locomotives	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Ships	0.6	0.6	0.6	0.6	0.7	0.7	0.8	0.8	0.8	0.9	1.0	1.1	1.2	1.3
Boats	0.5	0.5	0.5	0.5	0.6	0.6	0.6	0.6	0.5	0.5	0.6	0.6	0.6	0.7
Commercial Aircraft	1.6	1.7	1.8	1.9	1.7	1.8	1.9	2.1	2.2	2.4	2.6	2.7	2.9	3.2
General Aviation	0.3	0.3	0.3	0.3	0.2	0.2	0.2	0.2	0.3	0.3	0.3	0.3	0.3	0.3
Military Aircraft	0.6	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
On-Road														
Passenger Cars/Trucks up to 10,000 lbs	22.3	22.6	23.1	24.3	26.4	26.6	26.9	27.6	28.6	29.9	30.9	31.9	33.5	34.7
Medium/Heavy Duty Trucks > 10,000 lbs	3.1	2.8	2.8	2.9	3.0	3.3	3.3	3.4	3.5	3.6	3.7	3.8	3.9	4.0
Urban,School and Other Buses	0.5	0.6	0.6	0.7	0.8	0.8	0.8	0.8	0.8	0.8	0.9	0.9	0.9	0.9
Motor-Homes and Motorcycles	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.3	0.3	0.3
Subtotal	29.8	29.8	30.4	32.0	34.1	34.8	35.3	36.3	37.6	39.3	40.7	42.2	44.2	46.0
AGRICULTURE/ FARMING														
Agricultural Equipment	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2
Animal Waste	0.5	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6
Soil Management	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Biomass Burning	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Subtotal	1.0	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1
GRAND TOTAL EMISSIONS	**87.7**	**92.2**	**91.0**	**93.0**	**92.8**	**93.4**	**97.4**	**101.5**	**106.0**	**110.8**	**115.4**	**120.2**	**125.3**	**130.4**

Table V: 2007 BAY AREA MAJOR (TOP 200) GHG EMITTING FACILITIES

						CO2 Equivalent Emissions (Metric Tons per year)		
No.	**Plant #**	**Plant Name**	**Plant Address**	**City**	**Zipcode**	**Biogenic**	**Non-Biogenic**	**Total**
1	11	Shell Martinez Refinery	3485 Pacheco Blvd	Martinez	94553	-	**4,976,544**	**4,976,544**
2	10	Chevron Products Company	841 Chevron Way	Richmond	94802	-	**4,303,800**	**4,303,800**
3	14628	Tesoro Refining and Marketing Company	150 Solano Way, Avon Refinery	Martinez	94553	-	**2,804,678**	**2,804,678**
4	12626	Valero Refining Company - California	3400 E 2nd Street	Benicia	94510	-	**2,568,988**	**2,568,988**
5	12095	Delta Energy Center	Arcy Lane	Pittsburg	94565	-	**1,895,320**	**1,895,320**
6	16	ConocoPhillips - San Francisco Refinery	1380 San Pablo Ave	Rodeo	94572	-	**1,577,872**	**1,577,872**
7	11866	Los Medanos Energy Center	750 E 3rd Street	Pittsburg	94565	-	**1,368,588**	**1,368,588**
8	12183	Metcalf Energy Center	One Blanchard Road	Coyote	95013	-	**1,120,115**	**1,120,115**
9	17	Lehigh Southwest Cement Company	24001 Stevens Creek Blvd	Cupertino	95014	-	**842,475**	**842,475**
10	26	Mirant Potrero, LLC	1201 Illinois Street	San Francisco	94107	-	**462,505**	**462,505**
11	8664	Crockett Cogeneration, A Cal Ltd Partnership	550 Loring Avenue	Crockett	94525	-	**427,300**	**427,300**
12	1820	Martinez Cogen Limited Partnership	550 Solano Way, Avon Refinery	Martinez	94553	-	**410,970**	**410,970**
13	10295	Air Products & Chemicals, Inc	Tesoro, Avon Refinery	Martinez	94553	-	**361,179**	**361,179**
14	2066	Waste Management of Alameda County	10840 Altamont Pass Rd	Livermore	94551	**121,019**	**143,881**	**264,900**
15	2266	Browning-Ferris Industries of CA, Inc	12310 San Mateo Road	Half Moon Bay	94019	**109,503**	**130,538**	**240,041**
16	3981	GWF Power Systems,LP (Site 4)	3400 Wilbur Avenue	Antioch	94509	-	**201,672**	**201,672**
17	3243	GWF Power Systems,LP (Site 1)	895 E 3rd Street	Pittsburg	94565	-	**200,700**	**200,700**
18	3244	GWF Power Systems,LP (Site 2)	1600 Loveridge Road	Pittsburg	94565	-	**196,800**	**196,800**
19	3245	GWF Power Systems,LP (Site 3)	1900 Wilbur Avenue	Antioch	94509	-	**184,660**	**184,660**
20	3246	GWF Power Systems,LP (Site 5)	555 Nichols Road	Pittsburg	94565	-	**183,237**	**183,237**
21	1812	Kirby Canyon Landfill	910 Coyote Creek Glf Dr	San Jose	95198	**120,641**	**50,361**	**171,002**
22	1179	Redwood Landfill Inc	8950 Redwood Hwy	Novato	94948	**72,532**	**93,942**	**166,474**
23	2254	Sonoma County Department of Public Works	500 Mecham Road	Petaluma	94952	**68,119**	**87,311**	**155,430**
24	732	Bluegrass Mills Holding Co	2600 De La Cruz Blvd	Santa Clara	95050	-	**128,963**	**128,963**
25	11326	PE Berkeley, Inc	Univ of Calif, Berkeley Campus	Berkeley	94720	-	**126,541**	**126,541**
26	51	United Airlines, SF Maintenance Center	SF Int'l Airport	San Francisco	94128	-	**123,861**	**123,861**
27	12	Mirant Delta, LLC	696 W 10th Street	Pittsburg	94565	-	**121,624**	**121,624**
28	2246	Tri-Cities Recycling	7010 Auto Mall Pkwy	Fremont	94538	**54,680**	**65,197**	**119,877**
29	11928	Calpine Pittsburg LLC	Loveridge Road	Pittsburg	94565	-	**116,440**	**116,440**

Table V: 2007 BAY AREA MAJOR (TOP 200) GHG EMITTING FACILITIES

						CO2 Equivalent Emissions (Metric Tons per year)		
No.	Plant #	Plant Name	Plant Address	City	Zipcode	Biogenic	Non-Biogenic	Total
30	5095	Republic Services Vasco Road, LLC	4001 N Vasco Road	Livermore	94550	48,063	57,316	105,378
31	6044	O L S Energy-Agnews	3530 Zanker Road	San Jose	95134	-	104,777	104,777
32	2740	City of Mountain View (Shoreline)	2600 Shoreline Boulevard	Mountain View	94043	53,132	48,687	101,819
33	11180	LLC	1400 Pacheco Pass Hwy	Gilroy	95020	-	94,712	94,712
34	10978	Seagate Technology	311 Turquoise Street	Milpitas	95035	-	86,229	86,229
35	151	Hexion Specialty Chemicals, Inc	41100 Boyce Road	Fremont	94538	-	80,865	80,865
36	18	Mirant Delta, LLC	3201 Wilbur Avenue	Antioch	94509	-	80,803	80,803
37	606	Anheuser-Busch, Inc	3101 Busch Drive	Fairfield	94533	38,630	37,409	76,039
38	778	San Jose/Santa Clara Water Pollution Control	700 Los Esteros Road	San Jose	95134	47,537	26,689	74,226
39	2371	USS-POSCO Industries	900 Loveridge Road	Pittsburg	94565	-	68,215	68,215
40	3921	Seagate Technology, LLC	47010 Kato Road	Fremont	94538	-	68,048	68,048
41	2478	UCSF/Parnassus	3rd Avenue & Parnassus	San Francisco	94122	-	64,882	64,882
42	16151	NRG Energy Center LLC	465 Stevenson Street	San Francisco	94103	-	64,559	64,559
43	1840	West Contra Costa County Landfill	Foot of Parr Blvd	Richmond	94801	28,773	34,212	62,984
44	907	Central Contra Costa Sanitary District	5019 Imhoff Place	Martinez	94553	33,215	29,239	62,454
45	4618	Keller Canyon Landfill Company	901 Bailey Road	Pittsburg	94565	26,956	32,134	59,089
46	30	Owens-Brockway Glass Container Inc	3600 Alameda Avenue	Oakland	94601	-	53,055	53,055
47	14483	California Water Service Company	1720 N 1st Street	San Jose	95112	-	50,164	50,164
48	591	East Bay Municipal Utility District	2020 Wake Avenue	Oakland	94607	42,742	6,212	48,954
49	14017	American Lithographers & Business Forms	21062 Forbes Street	Hayward	94545	-	45,790	45,790
50	621	City of Santa Clara, Silicon Valley Power	560 Robert Avenue	Santa Clara	95050	-	45,288	45,288
51	1364	Cypress Amloc Land Co , Inc	1 Sand Hill Road	Colma	94014	19,074	22,738	41,812
52	11670	Gas Recovery Systems, Inc	1804 Dixon Landing Rd	San Jose	95134	36,129	4,913	41,042
53	1438	New United Motor Manufacturing, Inc	45500 Fremont Blvd	Fremont	94538	-	39,344	39,344
54	2039	Potrero Hills Landfill, Inc	3675 Potrero Hills Lane	Suisun City	94585	7,064	29,268	36,332
55	13289	Los Esteros Critical Energy Facility	800 Thomas Foon Chew Way	San Jose	95134	-	35,555	35,555
56	83	United States Pipe & Foundry Company, LLC	1295 Whipple Road	Union City	94587	-	34,560	34,560
57	173	Georgia Pacific Gypsum LLC	801 Minaker Street	Antioch	94509	-	34,348	34,348
58	3464	City of Santa Clara	5401 Lafayette	Santa Clara	95050	15,098	17,998	33,096

Table V: 2007 BAY AREA MAJOR (TOP 200) GHG EMITTING FACILITIES

						CO2 Equivalent Emissions (Metric Tons per year)		
No.	**Plant #**	**Plant Name**	**Plant Address**	**City**	**Zipcode**	**Biogenic**	**Non-Biogenic**	**Total**
59	12728	Waste Management Inc	2615 Davis Street	San Leandro	94577	**14,847**	**17,734**	**32,581**
60	153	PABCO Gypsum	37851 Cherry Street	Newark	94560	**-**	**32,171**	**32,171**
61	1201	Rolls-Royce Engine Services - Oakland, Inc	6711 Lockheed Street	Oakland	94621	**-**	**31,795**	**31,795**
62	15128	Cardinal Cogen Inc	Campus & Jordan Way	Palo Alto	94305	**-**	**30,626**	**30,626**
63	7265	San Jose State University (Cogen Plant)	San Carlos Street	San Jose	95192	**-**	**30,390**	**30,390**
64	706	New NGC, Inc	1040 Canal Boulevard	Richmond	94804	**-**	**29,252**	**29,252**
65	3011	IPT SRI Cogeneration Inc	333 Ravenswood Drive	Menlo Park	94025	**-**	**28,540**	**28,540**
66	13566	Norcal Waste Systems Pacheco Pass Landfill, Inc	Bloomfield Rd & Highway 152	Gilroy	95021	**12,486**	**14,884**	**27,370**
67	85	Hitachi Global Storage Technologies Inc	5600 Cottle Road	San Jose	95193	**-**	**26,909**	**26,909**
68	1257	Genentech, Inc	460 Point San Bruno Boulevard	Francisco	94080	**-**	**26,324**	**26,324**
69	41	Owens Corning Insulating Systems, LLC	960 Central Expressway	Santa Clara	95050	**-**	**25,432**	**25,432**
70	13193	Valero Benicia Asphalt Plant	3001 Park Road	Benicia	94510	**-**	**24,851**	**24,851**
71	11669	Gas Recovery Systems, Inc	15999 Guadalupe Mines Rd	San Jose	95120	**21,411**	**2,912**	**24,323**
72	11327	ConAgra Foods, Gilroy Foods	1350 Pacheco Pass Hwy	Gilroy	95020	**-**	**23,928**	**23,928**
73	1464	Acme Fill Corporation	950 Waterbird Way	Martinez	94553	**2,953**	**20,968**	**23,920**
74	1784	San Francisco International Airport	SF Int'l Airport	San Francisco	94128	**321**	**23,490**	**23,811**
75	698	Georgia-Pacific Gypsum LLC	1988 Marina Boulevard	San Leandro	94577	**-**	**23,535**	**23,535**
76	31	Dow Chemical Company	901 Loveridge Road	Pittsburg	94565	**-**	**22,900**	**22,900**
77	11887	Dynegy Oakland LLC	50 Martin Luthr Kng, Jr Way	Oakland	94607	**-**	**22,764**	**22,764**
78	3294	Guadalupe Rubbish Disposal	15999 Guadalupe Mines Rd	San Jose	95120	**11,947**	**10,813**	**22,759**
79	568	San Francisco South East Treatment Plant	1700 Jerrold Avenue	San Francisco	94124	**13,360**	**9,176**	**22,535**
80	617	Palo Alto Regional Water Quality Control Plant	2501 Embarcadero Way	Palo Alto	94303	**15,163**	**6,407**	**21,570**
81	3974	San Francisco General Hospital	1118	San Francisco	94110	**-**	**21,370**	**21,370**
82	1190	Evergreen Oil, Inc	6880 Smith Avenue	Newark	94560	**-**	**21,055**	**21,055**
83	13160	University of California SF	600 16th Street	San Francisco	94107	**-**	**19,847**	**19,847**
84	55	Lockheed Martin Corporation	1111 Lockheed Martin Way	Sunnyvale	94089	**-**	**18,632**	**18,632**
85	1634	Napa State Hospital	2100 Napa Vallejo Hwy	Napa	94558	**-**	**18,048**	**18,048**
86	11247	Clover Flat Landfill Inc	4380 Silverado Trail	Calistoga	94515	**13,181**	**3,769**	**16,951**
87	22	Conoco Phillips Refining Company	2101 Franklin Canyon Rd	Rodeo	94572	**-**	**16,697**	**16,697**

Table V:

2007 BAY AREA MAJOR (TOP 200) GHG EMITTING FACILITIES

						CO2 Equivalent Emissions (Metric Tons per year)		
No.	Plant #	Plant Name	Plant Address	City	Zipcode	Biogenic	Non-Biogenic	Total
88	12071	Bayer Healthcare LLC	800 Dwight Way	Berkeley	94710	-	16,609	16,609
89	11668	Gas Recovery Systems, Inc	Marsh Road	Menlo Park	94025	14,574	1,982	16,556
90	3256	Turk Island Solid Waste Disposal Site	Union City Boulevard	Union City	94587	7,527	8,973	16,500
91	11671	Gas Recovery Systems, Inc	Landfill	Canyon	94558	14,231	1,935	16,167
92	94	Cargill Salt	7220 Central Ave	Newark	94560	-	16,147	16,147
93	13631	Morgan Advanced Ceramics	2425 Whipple Road	Hayward	94544	-	16,071	16,071
94	5905	City of Sunnyvale/Public Works Dept	301 Carl Road	Sunnyvale	94088	1,799	14,204	16,003
95	12967	TRC	James Donlon Blvd	Antioch	94509	7,188	8,569	15,758
96	733	City of Sunnyvale Water Pollution Control	1440 Borregas Avenue	Sunnyvale	94089	13,277	2,345	15,622
97	9029	Kie-Con	3551 Wilbur Avenue	Antioch	94509	-	15,494	15,494
98	4272	El Camino Hospital	2500 Grant Road	Mountain View	94040	-	15,403	15,403
99	2721	City of Palo Alto Landfill	Byxbee Park	Palo Alto	94301	4,722	10,665	15,387
100	9010	California Paperboard Corporation	525 Mathew Street	Santa Clara	95050	-	14,247	14,247
101	14512	Gilroy Energy Center,LLC for Riverview Energy Ctr	801 Minaker Road	Antioch	94509	-	14,035	14,035
102	1403	City of Santa Rosa Wastewater Treatment	4300 Llano Road	Santa Rosa	95407	6,302	7,534	13,836
103	3590	City of Berkeley/Engr Div/Public Works	Cesar Chavez Prk	Berkeley	94704	6,195	7,385	13,581
104	7264	California Pacific Medical Center	3700 California Street	San Francisco	94118	-	13,399	13,399
105	11661	Rhodia Inc	100 Mococo Road	Martinez	94553	-	13,294	13,294
106	227	Criterion Catalysts Company LP	2840 Willow Pass Road	Pittsburg	94565	-	12,953	12,953
107	79	Morton International Inc	7380 Morton Avenue	Newark	94560	-	12,135	12,135
108	1371	TP	7399 Johnson Drive	Pleasanton	94588	5,959.6	5,830.9	11,790.5
109	18198	New WinCup Holdings, Inc	195 Tamal Vista Boulevard	Corte Madera	94925	-	11,563	11,563
110	1579	Granite Rock Company	1321 Lowrie Avenue	Francisco	94080	-	11,397	11,397
111	5081	ALZA Corporation	1015 Joaquin Road	Mountain View	94043	9,680	1,549	11,229
112	475	Santa Clara Valley Health & Hospital System	751 So Bascom Avenue	San Jose	95128	-	10,969	10,969
113	11374	WD Media, Inc	1710 Automation Pkwy	San Jose	95131	-	10,965	10,965
114	1605	Services	2003 Diamond Blvd	Concord	94520	-	10,920	10,920
115	632	Intel Corporation	2150 Mission College Blvd	Santa Clara	95054	-	10,759	10,759
116	14511	Gilroy Energy Center, LLC (Wolfskill Energy Ctr)	2425 Cordelia Road	Fairfield	94534	-	10,660	10,660

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						CO2 Equivalent Emissions (Metric Tons per year)		
No.	**Plant #**	**Plant Name**	**Plant Address**	**City**	**Zipcode**	**Biogenic**	**Non-Biogenic**	**Total**
117	1004	SFSU Housing Facilities (Cogeneration Plant)	1600 Holloway Avenue	San Francisco	94132	-	10,192	10,192
118	2025	University of San Francisco	2130 Fulton Street	San Francisco	94117	-	10,070	10,070
119	450	Veterans Administration Medical Center	3801 Miranda Avenue	Palo Alto	94304	-	9,783	9,783
120	3312	Zanker Road Resource Management,Ltd	705 Los Esteros Road	San Jose	95134	1,250	8,243	9,493
121	550	NASA-AMES Research Center	Moffett Field NS	Mountain View	94035	-	9,347	9,347
122	3273	Pacific Union College	1 Angwin Avenue	Angwin	94508	-	9,217	9,217
123	1209	Union Sanitary District	5072 Benson Road	Union City	94587	8,069	889	8,958
124	10271	Darling International	429 Amador Street	San Francisco	94124	-	8,883.9	8,883.9
125	541	Pacific Gas & Electric Co	4690 Evora Road	Concord	94520	-	8,828	8,828
126	927	California Oils Corporation	1145 Harbour Way, South	Richmond	94804	-	8,797	8,797
127	10583	Monterey Mushrooms Inc	642 Hale Avenue	Morgan Hill	95038	-	8,482	8,482
128	705	Vulcan Materials Company Western Division	52 El Charro Road	Pleasanton	94566	-	8,215	8,215
129	12870	Shell Chemical LP	10 Mococo Road	Martinez	94553	-	8,092	8,092
130	9455	American Licorice Company	2477 Liston Way	Union City	94587	-	8,061	8,061
131	9573	Diageo North America, Inc	151 Commonwealth Drive	Menlo Park	94025	-	7,987	7,987
132	1995	Solano County Facilities Operations	501 Delaware Street	Fairfield	94533	-	7,864	7,864
133	459	Veterans Administration Medical Center	4150 Clement Street	San Francisco	94121	-	7,847	7,847
134	8025	Novartis Vaccines and Diagn	4560 Horton Street	Emeryville	94608	-	7,821	7,821
135	12557	AMCAN Beverages, Inc	1201 Commerce Boulevard	Canyon	94503	-	7,715	7,715
136	14415	Center	5975 Lambie Road	Suisun City	94585	-	7,695	7,695
137	10437	Byron Power Company,c/o Ridgewood Power Mgnt	4901 Bruns Road	Byron	94514	-	7,650	7,650
138	3194	City of Alameda, Maint Serv Center	Doolittle Drive	Alameda	94501	3,413	4,068	7,481
139	17559	Pacific Atlantic Terminals LLC	2801 Waterfront Road	Martinez	94553	-	7,467	7,467
140	10861	Northrop Grumman Systems Corporation	401 E Hendy Ave, 62/1	Sunnyvale	94088	-	7,351	7,351
141	14416	Goose Haven Energy Center	3853 Goose Haven Road	Suisun City	94585	-	7,146	7,146
142	13683	Dey L P	2751 Napa Valley Corp Dr	Napa	94558	-	7,002	7,002
143	4175	City of San Jose (Singleton Road Landfill)	885 Singleton Road	San Jose	95111	3,187	3,799	6,986
144	14414	Creed Energy Center LLC	6150 Creed Road	Suisun City	94585	-	6,867	6,867
145	17925	Transamerica Pyramid Properties, LLC	600 Montgomery Street	San Francisco	94111	-	6,860	6,860

Table V: 2007 BAY AREA MAJOR (TOP 200) GHG EMITTING FACILITIES

						CO2 Equivalent Emissions (Metric Tons per year)		
No.	**Plant #**	**Plant Name**	**Plant Address**	**City**	**Zipcode**	**Biogenic**	**Non-Biogenic**	**Total**
146	148	Ball Metal Beverage Container Corp	2400 Huntington Drive	Fairfield	94533	**-**	**6,795**	**6,795**
147	2531	Santa Rosa Memorial Hospital	1165 Montgomery Drive	Santa Rosa	95402	**-**	**6,772**	**6,772**
148	16855	Kaiser Antioch Deer Valley	4501 Sand Creek Rd	Antioch	94509	**-**	**6,594**	**6,594**
149	4446	Veterans' Home of California	100 California Dr, Administration	Yountville	94599	**-**	**6,483**	**6,483**
150	3024	Isola USA Corp	401 Whitney Place	Fremont	94539	**-**	**6,362**	**6,362**
151	556	Stanford Linear Accelerator	2575 Sand Hill Road	Menlo Park	94025	**-**	**6,274.2**	**6,274.2**
152	3613	St Mary's Medical Center	450 Stanyan Street	San Francisco	94117	**-**	**6,228**	**6,228**
153	255	Lawrence Livermore National Laboratory	7000 East Avenue	Livermore	94550	**-**	**6,218**	**6,218**
154	4116	San Francisco, City & County, PUC	3500 Great Highway	San Francisco	94132	**2,780**	**3,428**	**6,208**
155	849	Roche Palo Alto LLC	3431 Hillview Avenue	Palo Alto	94304	**-**	**5,885**	**5,885**
156	110	Burke Industries, Inc	2250 So 10th Street	San Jose	95112	**-**	**5,849**	**5,849**
157	12965	John Zink Company	2150 Kruse Drive	San Jose	95131	**-**	**5,777**	**5,777**
158	11924	California Pacific Medical Center	Castro & Duboce Street	San Francisco	94114	**-**	**5,767**	**5,767**
159	15816	Cal-Pox, Inc	103 Shoreline Parkway	San Rafael	94901	**786**	**4,926**	**5,713**
160	2035	PEPSICO Beverages and Foods	1175 57th Avenue	Oakland	94621	**-**	**5,700**	**5,700**
161	167	Maxwell House, Div of Kraft Foods	100 Halcyon Drive	San Leandro	94578	**-**	**5,609**	**5,609**
162	9339	San Jose State University	One Washington Square	San Jose	95192	**-**	**5,540**	**5,540**
163	1067	Oro Loma Sanitary District	2600 Grant Avenue	San Lorenzo	94580	**4,315**	**1,203**	**5,519**
164	16930	San Mateo County Youth Services Center	70 Loop Road	San Mateo	94402	**-**	**5,369**	**5,369**
165	8287	Coca-Cola	5800 3rd Street	San Francisco	94124	**-**	**5,263**	**5,263**
166	15117	Bay Sheets	6791 Alexander St	Gilroy	95020	**-**	**5,204**	**5,204**
167	1472	Unimin Corporation	Camino Diablo Rd	Byron	94514	**-**	**5,093**	**5,093**
168	16023	Georgia-Pacific Corrugated LLC	2800 Alvarado Street	San Leandro	94577	**-**	**5,014**	**5,014**
169	18447	PPF OFF One Market Plaza LLC	Tow	San Francisco	94105	**-**	**4,979**	**4,979**
170	2168	Jelly Belly Candy Company	One Jelly Belly Lane	Fairfield	94533	**-**	**4,858**	**4,858**
171	17315	C & H Sugar Company, Inc	830 Loring Avenue	Crockett	94525	**-**	**4,817**	**4,817**
172	10408	County Asphalt	5501 Imhoff Drive	Martinez	94553	**-**	**4,807**	**4,807**
173	2561	Shoreline Amphitheatre	One Amphitheatre Parkway	Mountain View	94043	**2,171**	**2,592**	**4,763**
174	1860	Laguna Honda Hospital	375 Laguna Honda Boulevard	San Francisco	94116	**-**	**4,686**	**4,686**

Table V: 2007 BAY AREA MAJOR (TOP 200) GHG EMITTING FACILITIES

						CO2 Equivalent Emissions (Metric Tons per year)		
No.	**Plant #**	**Plant Name**	**Plant Address**	**City**	**Zipcode**	**Biogenic**	**Non-Biogenic**	**Total**
175	14327	Silgan Containers Mfg Corp	2200 Wilbur Avenue	Antioch	94509	-	4,632	4,632
176	2451	Pleasanton Garbage Service, Inc	2512 Vineyard Avenue	Pleasanton	94566	1,301	3,298	4,599
177	17560	Mission Foods	23423 Cabot Blvd	Hayward	94545	-	4,594	4,594
178	1700	California Pacific Med Ctr, Pacific Campus	2333 Buchanan Street	San Francisco	94115	-	4,593	4,593
179	15501	Spansion LLC	915 De Guigne Drive	Sunnyvale	94086	-	4,557	4,557
180	3919	Kaiser Permanente San Jose Medical Center	250 Hospital Parkway	San Jose	95119	-	4,546	4,546
181	3885	Highland General Hospital	1411 E 31st Street	Oakland	94602	-	4,517	4,517
182	1258	Delta Diablo Sanitation District	2500 Pittsburg & Antioch Hwy	Antioch	94509	2,049	2,400	4,449
183	653	Central Marin Sanitation Agency	Andersen Drive, East end	San Rafael	94901	1,776	2,542	4,318
184	73	Gallagher & Burk, Inc	344 High Street	Oakland	94601	-	4,301.4	4,301.4
185	128	Syar Industries, Inc	Lake Herman Road	Vallejo	94591	-	4,182	4,182
186	1351	City of Burlingame, Waste Water Treatment Plant	1103 Airport Boulevard	Burlingame	94010	1,937	2,220	4,157
187	1009	Hayward Waste Water Treatment Plant	3700 Enterprise Ave	Hayward	94545	3,563	491	4,053
188	453	Good Samaritan Hospital	2425 Samaritan Drive	San Jose	95124	-	4,034	4,034
189	460	Alta Bates Hospital	2450 Ashby Avenue	Berkeley	94705	-	4,004	4,004
190	12001	Quikrete Northern California	6950 Stevenson Blvd	Fremont	94538	-	3,866	3,866
191	16888	Ingersoll-Rand Energy Systems	5858 Horton Street	Emeryville	94608	-	3,864	3,864
192	92	Antioch Building Materials Company	1375 California Ave	Pittsburg	94565	-	3,839	3,839
193	1404	Fairfield-Suisun Sewer District	1010 Chadbourne Road	Fairfield	94534	1,775	2,061	3,836
194	13617	Delta Air Lines Inc	SJ Int'l Airport	San Jose	95120	-	3,805.7	3,805.7
195	5691	Sunquest Properties Inc	Landfill, Brisbane	Brisbane	94005	1,711	2,040	3,750
196	2957	Super Store Industries/Fairfield Dairy Division	199 Red Top Road	Fairfield	94533	-	3,689	3,689
197	10162	EXAR Corporation	48760 Kato Road	Fremont	94538	-	3,688	3,688
198	1534	South Bayside System Authority	Radio Road, End of	Redwood City	94065	3,356	233	3,589
199	232	Onizuka Air Force Base	1080 Innovation Way	Sunnyvale	94089	-	3,568	3,568
200	3893	Sonoma Valley Hospital District	347 Andrieux Street	Sonoma	95476	-	3,550	3,550
Total (Metric Tons per year)						**1,183,471**	**28,052,724**	**29,236,195**

Attachment 3

Table 1

Bay Area Winter Emissions Inventory for Primary PM2.5 & PM Precursors: Year 2010

	Winter Emissions tons/day			
SOURCE CATEGORY	**PM2.5**	**ROG**	**NOx**	**SO2**
INDUSTRIAL/COMMERCIAL PROCESSES				
PETROLEUM REFINING FACILITIES				
Basic Refining Processes	0.1	0.2	--	0.2
Wastewater (Oil-Water) Separators	--	1.2	--	--
Wastewater Treatment Facilities	--	0.5	--	--
Cooling Towers	--	0.9	--	--
Flares & Blowdown Systems	--	0.8	1.2	0.6
Other Refining Processes	--	0.3	--	--
Fugitives	--	0.5	--	--
Subtotal	**0.2**	**4.3**	**1.2**	**0.9**
CHEMICAL MANUFACTURING FACILITIES				
Coating, Inks, Resins & Other Facilities	--	0.1	0.1	2.7
Pharmaceuticals & Cosmetics	0.4	0.8	1.4	3.2
Fugitives - Valves & Flanges	--	0.6	--	--
Subtotal	**0.4**	**1.5**	**1.5**	**5.9**
OTHER INDUSTRIAL/COMMERCIAL PROCESSES				
Bakeries	--	0.8	--	--
Cooking(Charbroiling)	1.5	0.9	--	--
Cooking(Deep Fat Frying)	0.1	0.1	--	--
Cooking(Griddles)	0.2	0.1	--	--
Wineries & Other Food & Agr. Processes	0.3	1.1	--	--
Metallurgical & Minerals Manufacturing	1.8	0.2	0.2	0.2
Waste Management	0.6	3.1	--	--
Semiconductor Manufacturing	--	0.2	--	--
Fiberglass Products Manufacturing	--	0.1	--	--
Rubber & Plastic Products Manufacturing	--	0.3	--	--
Contaminated Soil Aeration	--	0.1	--	--
Other Industrial Commercial	0.4	0.9	--	--
Subtotal	**4.9**	**7.9**	**0.3**	**0.3**
PETROLEUM PRODUCT/SOLVENT EVAPORATION				
PETROLEUM REFINERY EVAPORATION				
Storage Tanks	--	3.3	--	--
Loading Operations	0.1	0.1	--	--
Subtotal	**0.1**	**3.4**	**--**	**--**
FUELS DISTRIBUTION				
Natural Gas Distribution	--	0.7	--	--
Bulk Plants & Terminals	--	0.7	--	--
Trucking	--	2.6	--	--
Gasoline Filling Stations	--	5.2	--	--
Aircraft Fueling	--	1.1	--	--
Recreational Boat Fueling	--	0.9	--	--
Portable Fuel Container Spillage	--	3.8	--	--
Other Fueling	--	0.4	--	--
Subtotal	**--**	**15.3**	**--**	**--**
OTHER ORGANIC COMPOUNDS EVAPORATION				
Cold Cleaning	--	2.7	--	--

Table 1

Bay Area Winter Emissions Inventory for Primary PM2.5 & PM Precursors: Year 2010

	Winter Emissions tons/day			
SOURCE CATEGORY	**PM2.5**	**ROG**	**NOx**	**SO2**
Vapor Degreasing	--	--	--	--
Handwiping	--	2.3	--	--
Dry Cleaners	--	0.6	--	--
Printing	--	4.1	--	--
Adhesives & Sealants	--	9.7	--	--
Structures Coating	--	21.2	--	--
Industrial/Commercial Coating	--	8.1	--	--
Storage Tanks	--	0.8	--	--
Lightering & Ballsting	--	--	--	--
Other Organics Evaporation	--	1.5	--	--
Subtotal	**--**	**51.0**	**--**	**--**
COMBUSTION - STATIONARY SOURCES				
FUELS COMBUSTION				
Natural Gas(Space/Water Heating/Cooking)	1.7	1.0	18.3	0.1
LPG and Liquid Fuel	--	--	0.5	0.3
Solid Fuel (Woodstoves)	3.7	2.8	0.5	0.1
Solid Fuel (Fireplaces)	13.1	5.3	1.1	0.2
Cogeneration	0.7	1.9	3.9	0.3
Power Plants	0.3	0.1	1.0	0.1
Oil Refineries External Combustion	2.4	1.0	10.5	14.0
Glass Melting Furnaces - Natural Gas	--	--	0.9	--
Reciprocating Engines	0.2	0.5	2.9	0.1
Turbines	0.1	--	0.8	--
Landfill/Cement Plant Combustion	1.6	1.4	15.4	3.0
Subtotal	**23.8**	**14.0**	**55.6**	**18.2**
BURNING OF WASTE MATERIAL				
Incineration	0.1	0.1	0.2	--
Planned Fires	0.4	0.4	0.2	--
Subtotal	**0.5**	**0.4**	**0.4**	**--**
Banked Emissions	--	--	--	--
Alternative Compliance Allowance	--	--	--	--
Subtotal (All stationary source categories)	**29.8**	**97.9**	**59.0**	**25.3**
COMBUSTION - MOBILE SOURCES				
ON-ROAD MOTOR VEHICLES				
Passenger Cars	2.3	42.1	35.5	0.4
Light Duty Trucks<6000lbs	1.0	23.4	27.1	0.2
Medium Duty Trucks 6001-8500 lbs	0.5	10.1	17.9	0.2
Light Heavy Duty Trucks 8501-14000lbs	0.4	6.2	23.1	0.1
Medium Heavy Duty Trucks 14001-33000lbs	0.8	2.4	21.6	--
Heavy Heavy Duty Trucks>33000 lbs	1.6	2.8	51.7	0.1
School/Urban Buses	0.6	1.4	16.9	--
Motor-Homes	--	1.4	1.4	--
Motorcycles	--	17.5	2.3	--
Subtotal	**7.3**	**107.4**	**197.6**	**0.9**
OFF-HIGHWAY MOBILE SOURCES				
Lawn and Garden Equipment	0.2	13.6	2.1	--

Table 1

Bay Area Winter Emissions Inventory for Primary PM2.5 & PM Precursors: Year 2010

	Winter Emissions tons/day			
SOURCE CATEGORY	**PM2.5**	**ROG**	**NOx**	**SO2**
Transportation Refrigeration Units	0.6	1.9	7.7	--
Agricultural Equipment	0.3	1.0	5.9	--
Construction and Mining Equipment	0.6	1.6	11.7	--
Industrial Equipment	0.2	1.1	6.5	--
Light Duty Commercial Equipment	0.6	4.5	7.4	--
Trains	0.2	0.9	10.7	--
Off Road Recreational Vehicles	--	1.3	--	--
Ships	0.2	0.4	8.3	1.8
Commercial Harborcraft	0.6	1.2	16.1	--
Recreational Boats	0.5	8.5	2.3	--
Subtotal	**4.0**	**35.9**	**78.7**	**1.9**
AIRCRAFT				
Commercial Aircraft	0.1	2.4	9.1	0.8
General Aviation	0.1	0.5	0.1	--
Military Aircraft	0.1	0.7	1.7	0.1
Airport Ground Support Equipment	--	0.1	0.8	--
Subtotal	**0.3**	**3.7**	**11.7**	**0.9**
Subtotal (All mobile source categories)	**11.6**	**147.0**	**288.0**	**3.7**
MISCELLANEOUS OTHER SOURCES				
Construction Operations	1.0	--	--	--
Farming Operations	0.2	--	--	--
Entrained Road Dust-Paved Roads	3.5	--	--	--
Entrained Road Dust-Unpaved Roads	0.4	--	--	--
Wind Blown Dust	0.7	--	--	--
Animal Waste	0.9	3.9	--	--
Agricultural Pesticides	--	0.6	--	--
Non-Agricultural Pesticides	--	0.3	--	--
Consumer Products(Excluding Pesticides)	--	44.1	--	--
Accidental Fires and Cigarette Smoking	1.2	0.4	0.2	--
Subtotal	**7.9**	**49.2**	**0.2**	**--**
Subtotal: All Stationary Source Categories	**29.8**	**97.9**	**59.0**	**25.3**
Subtotal: All Mobile Source Categories	**11.6**	**147.0**	**288.0**	**3.7**
Subtotal: All Miscellaneous Other Sources	**7.9**	**49.2**	**0.2**	
GRAND TOTAL EMISSIONS	**49**	**294**	**347**	**29**

Attachment 4

UNDERSTANDING PARTICULATE MATTER:

PROTECTING PUBLIC HEALTH IN THE SAN FRANCISCO BAY AREA

NOVEMBER 2012

BAY AREA AIR QUALITY
MANAGEMENT DISTRICT

UNDERSTANDING PARTICULATE MATTER: PROTECTING PUBLIC HEALTH IN THE SAN FRANCISCO BAY AREA

The Air District has prepared this report for informational purposes only. This report builds upon the Bay Area 2010 Clean Air Plan, which defines the Air District's current particulate matter (PM) control strategy. This report will inform future PM planning in the Bay Area, but does not commit the Air District to any specific course of regulatory action.

BAY AREA AIR QUALITY
MANAGEMENT DISTRICT

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EXECUTIVE SUMMARY

The air we breathe is saturated with millions of microscopic particles. This particulate matter (PM), especially the finest particles, can evade the body's natural defenses and penetrate deep into the lungs, blood stream, brain and other vital organs, and individual cells. Health studies in recent decades have shown that exposure to PM, on both a short-term and a long-term basis, can have a wide range of negative health effects, including respiratory effects such as asthma, bronchitis, and impaired lung development in children, as well as serious cardiovascular effects such as hardening of the arteries, heart attacks and strokes. Health studies in cities throughout the world have found that mortality (death) rates increase with higher PM levels.

PM is a complex pollutant composed of a diverse assortment of extremely small airborne particles, including a mixture of solid particles and liquid droplets that vary greatly in terms of their size and mass, chemical composition, and toxicity. PM originates from a variety of man-made processes and sources such as fossil fuel combustion, residential wood burning, and cooking, as well as from natural sources such as wildfires, volcanoes, sea salt, and geological dust. PM is generated indoors as well as outdoors. Indoor sources include stoves, heaters, fireplaces, consumer products, and cigarettes. Although researchers have been making progress in analyzing PM, there are still major gaps in our understanding of PM and its effects on public health, climate change, and ecosystems.

The Bay Area Air Quality Management District (Air District) and the California Air Resources Board (ARB) have been working to reduce PM levels and human exposure. Thanks to these efforts, PM levels in the Bay Area have been significantly reduced in recent decades. The reduction in PM levels has provided significant public health benefits and has helped to increase average life expectancy in the region. But despite this progress, PM is still by far the air pollutant most harmful to public health in the Bay Area. Although monitoring data shows that the Bay Area currently meets all national (and most State) standards for PM, health studies show that there are negative health effects associated with exposure to PM even below the current air quality standards.

The purpose of this report is to synthesize the latest scientific information about PM and its impacts, describe the Air District's current efforts to analyze and reduce PM in the Bay Area, and lay the groundwork for the Air District's continuing efforts to further reduce PM and better protect public health. The report provides technical information about how PM is emitted and formed in the Bay Area; describes progress in reducing PM levels in the San Francisco Bay Area in recent years; summarizes current regulations and programs to reduce PM emissions and concentrations; identifies future technical work needed to improve the Air District's understanding of PM; and explains the importance of continuing and enhancing the Air District's efforts to reduce PM in order to protect public health, the climate, and the environment.

INTRODUCTION

The mission of the Bay Area Air Quality Management District (Air District or BAAQMD) is to protect and improve air quality, public health, and the global climate. Although we have made great progress in improving Bay Area air quality over the past 50 years, new challenges continue to emerge. Research in recent years indicates that particulate matter (PM) suspended in the air we breathe is the air pollutant that imposes the greatest health burden on Bay Area residents. The available evidence indicates that the most effective way that the Air District can fulfill its mission of protecting public health is to reduce the population exposure of Bay Area residents to PM.

Although PM has been regulated by U.S. EPA as one of the original “criteria air pollutants” since the early 1970’s, early efforts to improve air quality primarily focused on other pollutants such as ground-level ozone (smog), carbon monoxide, and toxic air contaminants. PM moved to the forefront of the air quality agenda only in recent years, beginning in the mid-1990’s, in response to a series of compelling health studies that linked population exposure to PM with a wide range of respiratory and cardiovascular health effects, including premature death. Indeed, the recognition that PM must be treated as an air pollutant of the highest priority represents perhaps the most important development in the air quality arena in recent years.

This report describes particulate matter and its impacts on public health, climate change, and ecosystems; summarizes technical information about PM, and how it is emitted and formed in the Bay Area; describes progress in recent years in reducing PM levels in the San Francisco Bay Area in relation to State and national PM standards; describes current regulations and programs to reduce PM emissions and concentrations; identifies future technical work needed to improve our understanding of PM; and provides a roadmap to focus Air District resources in the effort to reduce PM and protect public health in the Bay Area in the years to come.

Reducing Population Exposure to PM

Air quality planning to date, in the Bay Area and elsewhere, has generally focused on reducing emissions and ambient concentrations of air pollutants in order to attain State and national ambient air quality standards. This approach has enabled the Bay Area and other regions to make substantial progress in improving air quality, especially for pollutants that are regional in nature, such as ozone. However, in recent years there is a growing recognition that if we want to achieve the ultimate goal of protecting public health, then reducing emissions and concentrations of air pollutants at the regional scale may not suffice. We need to directly consider where, when, and how people are being exposed to air pollution. This is especially true in the case of PM, a complex pollutant whose concentrations in the air can vary substantially depending upon location and time. Therefore, the major objective of this report is to advance our understanding of how the Bay Area public is exposed to PM, which sources and types of PM are most harmful, and where we should focus efforts to reduce PM in order to better protect public health.

What is PM?

PM includes a wide range of particles that vary in size, chemical composition, and toxicity.

The term **particulate matter** (PM) describes a complex pollutant composed of a diverse assortment of extremely small airborne particles, including a mixture of solid particles and liquid droplets known as aerosols. Most air pollutants (such as ozone, carbon monoxide, and sulfur dioxide) consist of a single molecule or compound. This means that the pollutant will have the same physical properties and the same impacts on public health and the environment, regardless of the source, or combination of sources, from which it is emitted. PM, by contrast, includes a wide range of disparate particles that vary greatly in terms of their size and mass (ultrafine, fine, and coarse), physical state (solid or liquid), chemical composition, toxicity, and how they behave and transform in the atmosphere.

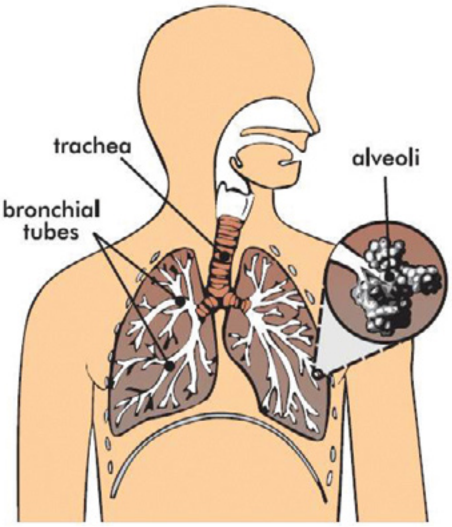
Particles originate from a variety of man-made processes and sources such as fossil fuel combustion, residential wood-burning, and cooking, as well as from natural sources such as wildfires, volcanoes, sea salt, and geological dust. PM is emitted directly from tailpipes, smokestacks, and fireplaces, and also formed indirectly by chemical reactions among precursor pollutants. Particulate matter is generated indoors as well as outdoors. Indoor sources include stoves, heaters, fireplaces, and consumer products, and cigarettes (if smoked inside). As described in Section 1-B, most people experience a significant percentage of their personal exposure to PM in the indoor environment where they are exposed to both ambient (outdoor) PM that penetrates inside as well as PM emissions produced by indoor sources.

Every breath we take contains millions of tiny airborne particles.

Public Health Impacts

PM is a stealthy pollutant - it is generally tasteless and odorless, and most particles are too small to be seen by the naked eye (though PM makes the air look hazy in the distance). But even on clear days when ambient PM concentrations are low and well within air quality standards, we inhale contains millions of tiny particles with each breath. Health studies show that these airborne particles cause and/or contribute to a wide range of respiratory and cardiovascular problems. In addition to effects such as asthma and bronchitis, exposure to PM can trigger major health impacts such as heart attacks or strokes. In fact, the evidence indicates that exposure to PM contributes to the death of more Bay Area residents each year than more visible causes such as auto accidents. Analysis shows that, even at the relatively moderate concentrations that prevail in the Bay Area, PM imposes economic and social costs to Bay Area residents and employers (in terms of sickness, lost productivity, and premature mortality) that run to billions of dollars per year, as discussed in Section 1-A.

How many particles do we breathe?



The number of airborne particles that we are exposed to on a daily basis is truly staggering. The air we breathe contains a very minute amount of PM in terms of its overall mass. However, even such a miniscule amount of mass contains enormous numbers of particles. For example, air with an ambient concentration of 10 micrograms of fine particulate matter (PM2.5, or particles less than 2.5 microns in diameter) per cubic meter - roughly the average concentration of Bay Area PM2.5 on an annual basis - may contain on the order of 100 million particles per cubic meter.

For this reason, we inhale huge numbers of particles. For purposes of illustration, consider the fact that urban air typically contains in the range of 5,000 to 30,000 particles per cubic centimeter, primarily in the ultrafine size range (particles less than 0.1 microns in diameter). The average adult inhales 450 cubic centimeters (roughly one pint) of air per breath. This means that in a typical urban environment we inhale from 1 million to 10 million minute particles with every breath we take. But that figure can spike to much higher levels in close proximity to high-volume roadways or other major outdoor emission sources, or indoor sources such as stoves and ovens. The bottom line is that, during the course of a single day, we inhale many trillions of fine and ultrafine particles, even when the air we are breathing meets air quality standards. Fortunately, our bodies have defenses in the nasal passages, throat, and lungs to filter out particles, so not all the pollutants that we inhale actually reach the air sacs (alveoli) where they can damage the lungs. But some of these tiny particles - which may be coated with acids, metals, and other toxic substances – are able to evade the body's defense mechanisms and penetrate deep into the lungs, bloodstream, cells, and vital organs where they can trigger various biological responses that harm the body.

PM Planning in the Bay Area

In response to concern about the health impacts of PM, in 2003 the State legislature enacted SB 656 (codified as Health & Safety Code Section 39614). This legislation required the Air Resources Board and local air districts to evaluate potential PM control measures and to develop a PM implementation schedule for appropriate PM-reduction measures. The Air District complied with this legislation; staff developed a Particulate Matter Implementation Schedule that was adopted by the Air District's Board of DIrectors in November 2005, and the Air District adopted the measures identified in the Implementation Schedule.

In fall 2010, the Air District adopted the Bay Area 2010 Clean Air Plan (2010 CAP) to update the region's plan to control ground-level ozone. In developing the 2010 CAP, the Air District was inspired by the recommendations issued by the National Research Council in 2004 which called for a new approach to air quality planning based on integrated multi-pollutant planning focused on achieving key outcomes such as protecting public health, the global climate, and ecosystems.

The Bay Area 2010 CAP identified two key goals: (1) protecting public health, and (2) protecting the climate. The 2010 CAP also pursued a multi-pollutant approach in developing an integrated control strategy to reduce four types of air pollutants: ground-level ozone; PM; toxic air contaminants (TACs); and greenhouse gases, such as carbon dioxide and methane, that contribute to climate change.

To inform the development of the 2010 CAP, Air District staff performed a "health burden" analysis, based upon the results of peer-reviewed health studies, in order to estimate:

- the public health effects of air pollution in the Bay Area, based upon key health endpoints such as chronic bronchitis, asthma emergency room visits, hospital admissions for respiratory or cardiovascular diseases, heart attacks, and premature mortality;
- the role of each air pollutant in causing or contributing to these health effects;
- the health benefits due to progress in improving Bay Area air quality in recent years; and
- the economic benefit of the improvement in public health.

Air quality issues are often presented in highly technical terms, using a specialized vocabulary and arcane units of measurement (such as parts per million or micro-grams per cubic meter) that mean little to the average person. The health burden analysis, by contrast, served to put a "human face" on the benefits of better air quality, by expressing its results in terms of tangible outcomes that make sense to policy-makers and the public.

From the policy perspective, the most important finding from the health burden analysis is that PM is the air pollutant that poses *by far* the greatest health risk to Bay Area residents, as shown in Figure 1-4. Although evidence of the public health risk related to PM has been mounting in recent years, it was only by performing this side-by-side health burden analysis for the 2010 CAP

that the predominant impact of PM on public health in comparison to other air pollutants was so starkly revealed.

Progress in Reducing PM Levels in the Bay Area

Although revelations about the health impacts of PM are cause for concern, the good news is that we have already achieved major progress in reducing PM levels in the Bay Area, as described in Section 3-C. Thanks to this progress, the Bay Area currently meets the national ambient air quality standards that apply to PM, and is making steady progress toward attaining more stringent, health-protective California PM standards. The reduction in PM levels in recent years translates into improved public health and vitality, and longer average life span. These benefits are worth billions of dollars in cost savings to Bay Area residents and employers.

Why We Need to Reduce PM Further

The fact that the Bay Area has made substantial progress in reducing PM levels does not mean that we can rest easy, however. There are several reasons why it is important to continue to enhance our efforts to reduce PM emissions, concentrations, and population exposure.

- Researchers have not been able to establish a safe threshold for population exposure to PM. Epidemiological studies have shown that there are health effects from PM2.5 exposure even at concentrations below current standards.
- The U.S. Environmental Protection Agency (EPA) reviews the national PM2.5 standards on a regular basis and may issue more stringent standards in the future.
- Even at the current, relatively low concentrations, PM is the most hazardous air pollutant in the Bay Area in terms of health impacts, including premature mortality, heart attacks, chronic bronchitis and other key health endpoints.
- PM concentrations – and population exposure to PM – can vary significantly at the local scale, as discussed in Section 1-B). Even though the Bay Area currently meets national PM standards based on the measurements from the regional PM monitoring network, some communities and individuals are exposed to higher concentrations of PM. People who live or work near major roadways, ports, distribution centers, or other major emission sources, or in proximity to wood-burning activities, may be disproportionately exposed to certain types of PM (e.g. ultrafine particles), so it is important to implement effective measures to reduce their exposure and health risks.

Challenges

Because PM is a complex pollutant and has become the focus of intense research only in recent years, there are still major gaps in our understanding of PM and its effects on public health, climate change, and ecosystems. These gaps are especially profound in regard to ultrafine PM, the smallest particles. As discussed in Section 1-A, health researchers are working to better understand which sizes and types of PM are most damaging to public health, and to explain the precise biological mechanisms by which PM damages our health. Similarly, climate scientists are striving to better define the various mechanisms by which different types of particles act upon the climate, and the overall effect of ambient PM on the climate.

Although many health studies to date suggest that breathing PM of all types may have negative health effects, researchers suspect that certain types of particles may be especially harmful. And intuitively, it makes sense that the size and chemical composition of particles should make a difference in terms of their health effects. Because health studies have not yet clearly defined the specific particle types that are most harful, regulators currently treat all fine PM as equal in terms of its health impacts.[1] However, determining the types of particles which are the most harmful is vitally important in the quest to understand PM. The current across-the-board approach to reducing PM has clearly provided major benefits in terms of reducing PM concentrations and protecting public health. But if certain particle types can be identified as the key culprits, and if specific sources account for the bulk of their emissions, then we may be able to identify appropriate control measures with a higher degree of precision, rather than pursuing reductions in all types of PM across the board. The ability to target the particles with the most severe health impacts would enable us to better protect public health, and also to identify measures that would achieve the greatest benefit at the lowest cost.

Since it is clear that we need to enhance our efforts to reduce PM in order to protect public health, PM will continue to be a major focus of air quality planning, regulation, and public education in the Bay Area over the next decade. Despite the gaps in our understanding of PM, we must develop and implement policies to control PM in the near-term, while refining our policies and priorities to reflect new information as it becomes available. Because we still have a great deal to learn, this report cannot serve as the final word on how to address PM in the Bay Area. But it is meant to lay the groundwork to guide the Air District's efforts to reduce PM in the coming years.

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[1] Certain types of particles, such as metals, polyaromatic hydrocarbons (PAHs), and diesel PM, are classified as toxic air contaminants (TACs), and are thus subject to regulation as TACs.

SECTION 1 : PM IMPACTS

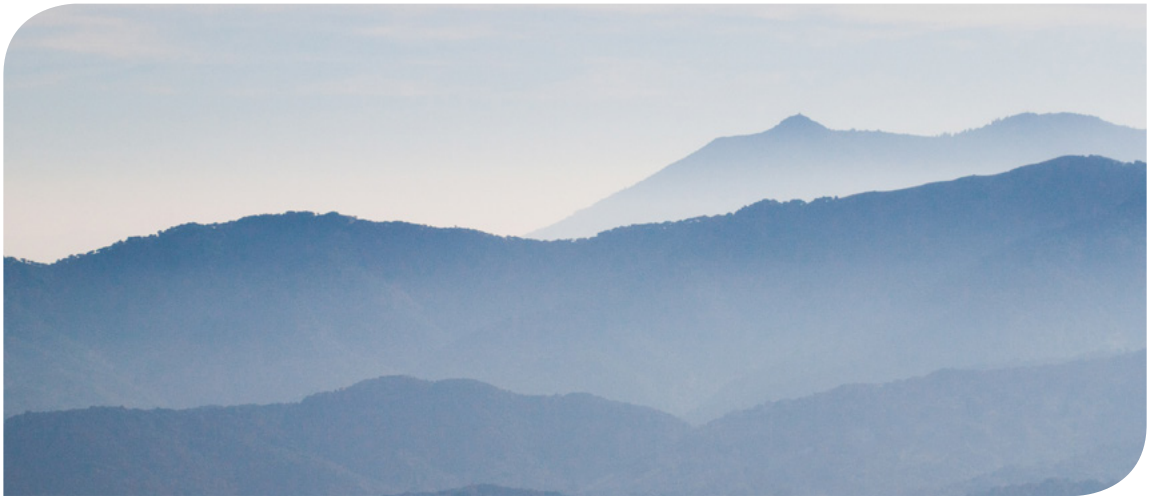
SECTION 1-A: THE PUBLIC HEALTH EFFECTS OF PM

This section summarizes methods used to study PM health effects, the current evidence regarding public health impacts related to exposure to PM; the biological pathways by which PM affects the body; which sizes and types of particles pose the greatest health risk; the estimated health burden from PM in the Bay Area; and why it is important to further reduce PM levels to protect the health of Bay Area residents.

This section will consider several key questions regarding the health effects of particulate matter:

- What types of negative health impacts are associated with exposure to PM?
- Does evidence show that exposure to PM is bad for public health?
- How does PM damage the body?
- Which types and sizes of particles are most harmful to health?
- Are there safe levels of PM?
- How does PM affect public health in the San Francisco Bay Area?

The discussion presented below attempts to synthesize information from the vast literature of studies that have analyzed the health effects of various particle sizes, including PM10 (particles less than 10 microns in diameter), PM2.5 (particles less than 2.5 microns in diameter), and ultrafine PM (particles less than 0.1 microns in diameter). An explanation of the various PM size categories is presented in Section 2 of this report.



A great deal of research has been performed in the past 25 years to identify and quantify the health risks of particulate matter. Health studies have linked exposure to PM with a wide range of negative health effects. The research provides evidence that exposure to PM, even at low and moderate levels, can cause or contribute to a wide range of respiratory and cardiovascular disease, including:

- irritation of the airways, coughing, or difficulty breathing
- decreased lung function
- aggravated asthma
- chronic bronchitis
- irregular heartbeat
- strokes
- heart attacks
- premature death in people with heart or lung disease

Challenges in Analyzing the Health Effects of PM

Determining the health effects of air pollutants is inherently difficult. But because PM is a heterogeneous pollutant comprised of particles that vary in size, mass, and chemical composition, this presents special challenges in determining its health effects. In the case of air pollutants composed of a single molecule, such as ozone (O3) or carbon monoxide (CO), the pollutant has exactly the same chemical composition – and thus the same potential health effects – regardless of the emissions source. However, in the case of PM, the composition of particles in a given air sample – and the corresponding health effects – will vary depending on the mix of emission sources.

Early Evidence of PM Health Effects

Several dramatic episodes in the first half of the 20th century demonstrated that very high levels of PM and other air pollutants can cause sickness and death. Early scientific research into the health effects of PM and air pollution was triggered by the December 1930 episode in the heavily populated and industrialized Meuse Valley of Belgium; this extreme air pollution episode killed more than 60 people over a three-day period. A similar tragedy occurred in Donora, Pennsylvania in October 1948 when an inversion layer trapped a lethal mix of PM, sulfuric acid, nitrogen dioxide, and other pollutants from local industrial plants for five days. Nearly half of the town's 14,000 residents became sick, twenty people perished, and 800 animals died.

Perhaps the most infamous air pollution episode occurred in London in December 1952, when a combination of coal combustion, cold weather and windless conditions trapped a thick layer of PM and other pollutants over the city, killing thousands. By comparing death rates during this event with normal conditions, London's Ministry of Health estimated at the time that nearly 4,000 deaths occurred as a result of the extreme air pollution. However, more recent research (Bell et al. 2004) suggests that the number of fatalities was considerably greater, on the order of 12,000. Death certificates and autopsies show that the main causes of death were respiratory and cardiovascular disease. These extreme episodes showed the need to study the effects of air pollution on public health.

Analyzing the Health Effects of PM

Researchers have developed various methods to analyze the health effects of PM and other air pollutants in terms of both morbidity (disease or illness) and mortality (premature death). Two of the most important techniques used to analyze the health effects of PM are epidemiological studies and clinical studies. Epidemiological studies analyze health data for a defined population group; the objective is to tease out the health effects of air pollution by looking for correlations between the amount of exposure to a pollutant and the observed incidence rate for various health endpoints (e.g., cases of respiratory or cardiovascular disease, hospital admissions, or premature mortality). If a correlation is observed, researchers must then try to determine whether the relationship may be inferred to be causal, meaning that exposure to the pollutant actually causes the observed health effect.

Establishing a causal relationship between PM and a given health effect is difficult because exposure to PM is only one factor among many that may cause, contribute to, or exacerbate a specific health effect. Other factors that affect our health include genetic and biological factors, environmental conditions (air quality, water quality, climate), and lifestyle (diet, exercise, drinking, and smoking), to name but a few. Therefore, in designing studies to analyze the health effects of air pollutants, epidemiologists attempt to isolate the effect of the air pollutants by controlling for (masking) the effect of these other socioeconomic (e.g. income and education), demographic (age, gender, etc.), environmental, and lifestyle factors that impact public health. One of the difficulties in air pollution epidemiology is that the health risks associated with current ambient levels of air pollution, while significant, are nonetheless extremely small when compared to other known risk factors, such as cigarette smoking, lack of physical activity, obesity, etc. Because these other risk factors have a powerful impact on health, it is difficult to distinguish the more subtle effects of air pollution from the health effects attributable to these other factors.

One of the key challenges in epidemiology is estimating how much the people in a study group have been exposed to the pollutant in question. Exposure estimates are generally based upon PM monitoring data and/or results of computer modeling to simulate ambient PM concentrations. Many studies rely on ambient air quality data from monitoring networks, but these data may not capture exposures in micro-environments. And even if accurate estimates of population exposure to ambient (outdoor) PM are available, epidemiological studies generally do not include indoor exposure to PM, which accounts for a significant portion of total exposure for many people, as discussed in Section 1-B. Improved methods to estimate population exposure to PM across the full range of indoor and outdoor environments would be valuable to enable epidemiologists to better analyze the health effects related to exposure to PM.

Analyzing health effects related to PM is also complicated by the fact that the various species of PM, as well as a variety of other pollutants and toxic air contaminants, are all mixed together in the air that we breathe. Therefore, isolating the health effects of PM from other types of pollutants, or distinguishing the effects of a certain size fraction of PM, or a certain chemical species of PM, from the overall mass of PM is a difficult task.

Despite these caveats, epidemiological studies are of great value in helping to illuminate the relationship between air pollutants and health effects. For example, exposure to PM is rarely, if ever, cited as the cause of death in a coroner's report when someone dies of a heart attack or stroke or lung disease. However, epidemiological studies indicate that exposure to PM is an important contributing factor in hundreds, perhaps thousands, of deaths in the Bay Area each year. Epidemiological studies are used to analyze:

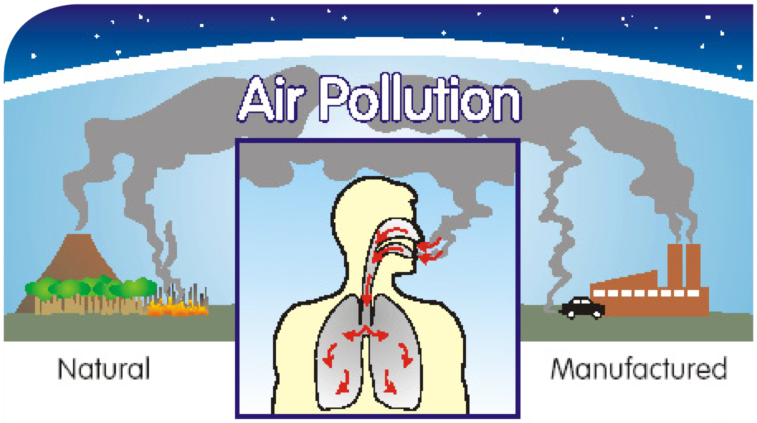
- Correlations between exposure to air pollutants and the incidence rate of both acute (short-term) and chronic (long-term) health effects among the general population or defined sub-groups;
- Concentration-response functions; i.e., how a change in the ambient concentration of a pollutant may affect the incidence rate of a specific health effect. Concentration-response functions can be used to estimate the reduction in health effects that can be expected from a given improvement in air quality, such as attaining an ambient air quality standard in the Bay Area or other metropolitan area.
- Safe thresholds, i.e., a "no-effects" threshold such as an ambient concentration below which no health effects can be observed. This is especially important for purposes of ensuring that ambient air quality standards are sufficiently health protective.

Epidemiological studies are an essential tool to discern correlations between exposures to air pollutants and health outcomes. However, these studies are based on statistical analysis of large population groups; they cannot definitively "prove" that air pollution causes a specific health effect either in an individual case or among a larger population group. Nor can they explain the precise biological mechanisms by which PM causes or contributes to the negative health effects observed. To investigate these issues, researchers perform clinical studies of small groups of people or animals in which they can carefully control the exposure and dosage and observe the impacts over a specific timeframe. These clinical studies are valuable in terms of confirming results of large-scale epidemiological studies at the individual level. Clinical studies also help to define biological mechanisms; that is, exactly how PM or other air pollutants act on and harm the body.

Recent Research on Health Effects of PM

By the 1970s, a link between exposure to PM and respiratory disease, such as triggering asthma episodes or other pulmonary disorders, had been well established, although there was uncertainty as to the level of PM exposure required to trigger significant public health impacts. Based on the evidence regarding the **respiratory effects** of particulate matter, such as asthma, bronchitis, and diminished lung function, PM was included among the six original “criteria pollutants” identified in the ground-breaking federal Clean Air Act of 1970. However, in the ensuing two or three decades after the Clean Air Act was enacted, other air pollutants such as ozone, carbon monoxide, and lead, were thought to pose a greater health risk than PM. As a result, air pollution control efforts focused primarily on reducing these other pollutants.

PM began to move to the forefront of concern about the health impacts of air pollution beginning in the 1990s in response to a new series of studies on PM health effects. Research in recent years provides evidence that, even at moderate or low levels, PM has a wide range of negative health impacts and can contribute to premature mortality. Whereas earlier research focused primarily on respiratory effects of PM, recent years have seen a great deal of research into the effects of PM on the **cardiovascular system**, the heart and blood system which takes oxygen from the lungs and distributes it throughout the body. Negative impacts of PM on the cardiovascular system include atherosclerosis (hardening of the arteries), ischemic strokes (caused by obstruction of the blood supply), and heart attacks. These new findings about the cardiovascular effects of exposure to PM, especially the increase in premature mortality in adults, have given greater urgency to the need to reduce PM.



PM Impact on Premature Mortality and Life Expectancy

Concern about PM health impacts was crystalized in the early to mid-1990s by a series of epidemiological studies that analyzed the correlation between PM and premature mortality (death).

- Studies in various cities with different climates, pollution mixes, and demographics consistently found a correlation between daily changes in PM levels and daily mortality.
- The two most important studies were the Harvard “Six Cities Study” (Dockery et al. 1993) which followed the health of over 8,000 people for a period of 14 to 16 years, and the March 1995 American Cancer Society study (Pope et al. 1995) which analyzed a study group of over half a million people in 151 cities. Both studies found that long-term exposure to PM is associated with cardiopulmonary mortality in adults. The Six Cities Study found that an increase of 10 $\mu g/m^3$ in ambient PM2.5 concentrations increases the risk of death from all cardiovascular causes by 19%. The American Cancer Society

study found that an increase of 10 µg/m^3 in ambient PM2.5 levels increases the risk of death from all cardiovascular causes by 13%.

It was already known, based on the severe pollution episodes in years past described above, that exposure to extremely high concentrations of PM and other air pollutants can kill people. But these new studies found that people may experience serious health effects, including premature mortality, from exposure to ambient PM at concentrations that most people would not even notice, including clear days when PM levels are below the current national ambient PM standards. For example, a recent study (Wellenius et al. 2012) on the link between PM and ischemic strokes in the Boston area (a region which attains the national 24-hour PM2.5 standard) found that the risk of stroke was 34% higher on days with "moderate" PM2.5 levels compared with days with "good" PM2.5 levels, as defined by the EPA Air Quality Index. The study also found that exposure to PM2.5 levels considered safe by the EPA increases the risk of ischemic stroke onset within hours of exposure and that the increase in risk was greatest within 12 to 14 hours after exposure to PM2.5.

People may experience negative health effects from exposure to PM even on clear days when PM levels are below the current national standards.

In assessing the linkage between PM and premature mortality, it is instructive to consider the leading causes of death in America. According to Center for Disease Control data for 2009, heart disease (25%), chronic lower respiratory disease (5%), and strokes (5%) are three of the four leading causes of death in the U.S., and collectively they account for 35% of all mortality. So if exposure to PM exacerbates cardiovascular and respiratory conditions even to a modest extent, this can be expected to exert a tangible impact in terms of increasing the overall mortality rate.

Since exposure to PM has been found to increase the incidence of premature mortality, it stands to reason that reducing PM levels should prevent premature death and thus help to extend average life expectancy. One recent study (Pope et al., 2009) analyzed the change in life expectancy as PM levels declined over the 20-year period from 1980 through 2000, based on data from 211 U.S. counties in 51 metropolitan areas. This study found that a 10 µg/m^3 decrease in PM2.5 levels was associated with a 7.3 (± 2.4) month increase in life expectancy. Analysis by Air District staff estimated that the improvement in air quality from 1990 to 2008 increased average Bay Area life expectancy by approximately six months per person during this period.[2] Since PM is estimated to be responsible for roughly 90% of the premature mortality related to air pollution in the Bay Area, most of this improvement in life expectancy due to improved air quality can be attributed to reduced PM concentrations.

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[2] See Appendix A in Bay Area 2010 Clean Air Plan: www.baaqmd.gov/Divisions/Planning-and-Research/Plans/Clean-Air-Plans.aspx

Reaction to Findings

The findings of these studies in the 1990s linking PM to premature mortality provoked controversy and skepticism. Researchers were surprised by results showing a broad range of health impacts from exposure to low ambient concentrations of PM. Health experts were perplexed by these findings, because at that time there were no known biological mechanisms to explain how exposure to relatively low concentrations of PM would produce the health effects observed in the epidemiological studies, especially in terms of cardiovascular disease and death.

In response to the controversy generated by these studies, researchers reexamined the results of the studies, and also embarked on a search for biological mechanisms to explain the health effects observed in these studies. To address concerns about methodological issues with previous studies, the Health Effects Institute funded the *National Morbidity, Mortality, and Air Pollution Study* (NMMAPS) (Samet et al. 2000). Over a five-year period through 2005, NMMAPS performed a time-series study using a consistent method to analyze health impacts of PM10 in the 90 largest American cities, cities that cover a wide geographic area and have varying levels of air pollutants. The NMMAPS largely confirmed the the findings in the original studies that, on average, for every 10 $\mu g/m^3$ increase in PM10, there was a 0.5% increase in overall mortality on the following day, as well as a 2% increase in hospital admissions for pneumonia and chronic obstructive pulmonary disease.

Summary of PM Health Effects

To inform its period review (once every five years) of PM air quality standards, U.S. EPA prepared a detailed synthesis of the vast body of literature on PM health effects and issued its December 2009 *Integrated Science Assessment for Particulate Matter*. Based on the cumulative weight of the studies in the literature, EPA's conclusions regarding the strength of the evidence to support a finding of causality between exposure to PM2.5 and key health effects are summarized in Table 1-1. EPA also reviewed the evidence as to whether exposure to coarse PM and to ultra-fine PM has been proven to cause negative health effects; they found that the evidence currently available was either **suggestive** of causality or **inadequate to establish causality** in the case of coarse PM and ultra-fine PM.

Table 1-1 US EPA Findings on Health Effects for Particulate Matter

Health Outcome	Causality Determination	Examples of Health Effects
Size Fraction: PM$_{10-2.5}$ Coarse PM		
Short -Term Exposure		
Cardiovascular Effects	Suggestive	Increase in hospital admissions and emergency room visits for cardiovascular causes Reduction in heart rate variability
Respiratory Effects	Suggestive	Increase in hospital admissions and emergency room visits for respiratory causes, particularly in children Pulmonary inflammation
Mortality	Suggestive	
Central Nervous System	Inadequate	
Long -Term Exposure		
Cardiovascular Effects	Inadequate	Increase in hospital admissions for ischemic heart disease Arrhythmia Reduction in heart rate variability
Respiratory Effects	Inadequate	
Mortality	Inadequate	
Reproductive & Developmental	Inadequate	Low birth weight
Cancer, Mutagenicity, Genotoxicity	Inadequate	
Size Fraction: PM$_{2.5}$ Fine PM		
Short -Term Exposure		
Cardiovascular Effects	Causal	Myocardial ischemia (reduced blood flow to the heart) Congestive heart failure Altered vasomotor function (stiffening and reduced flexibility of blood vessels)
Respiratory Effects	Likely to be causal	Alterations in lung function & respiratory symptoms in asthmatic children Chronic obstructive pulmonary disease Respiratory infections

Health Outcome	Causality Determination	Examples of Health Effects
Mortality	Causal	Cardiovascular- and respiratory-related mortality
Central Nervous System	Inadequate	Pro-inflammatory responses in the brain that may lead to neurodegenerative diseases
Long -Term Exposure		
Cardiovascular Effects	Causal	Higher blood pressure Increased blood coagulation Enhanced development of atherosclerosis (hardening of the arteries) Reduction in heart rate variability Increased risk of heart disease and stroke
Respiratory Effects	Likely to be causal	Impaired lung development Increased respiratory symptoms Asthma Altered pulmonary function Chronic bronchitis
Cancer, Mutagenicity, Genotoxicity	Suggestive	Lung cancer
Reproductive & Developmental	Suggestive	Low birth weight
Mortality	Causal	Cardiovascular mortality, lung cancer mortality, and Infant mortality due to respiratory causes
Size Fraction: Ultrafine PM		
Short -Term Exposure		
Cardiovascular Effects	Suggestive	Increased markers of oxidative stress Changes in vasomotor function Alterations in heart rate variability parameters
Respiratory Effects	Suggestive	Oxidative, inflammatory and allergic responses Decreases in pulmonary function

Health Outcome	Causality Determination	Examples of Health Effects
Mortality	Inadequate	
Central Nervous System	Inadequate	
Long -Term Exposure		
Cardiovascular Effects	Inadequate	
Respiratory Effects	Inadequate	Pulmonary inflammation
		Oxidative and allergic responses
Mortality	Inadequate	
Reproductive & Developmental	Inadequate	
Cancer, Mutagenicity, Genotoxicity	Inadequate	

Source: EPA Integrated Science Assessment, December 2009, Table 2-6

Table 1-2 presents the findings of a recent Harvard School of Public Health study (Kloog et al. 2012) that analyzed hospital admission rates throughout New England in terms of the correlation between PM2.5 exposure and hospital admission rates for respiratory disease, cardiovascular disease, strokes and diabetes. The effects from long-term exposure to PM2.5 are significantly higher than for short-term exposure for all four causes of admission.

Table 1-2: Estimated increase in hospital admissions rate for a 10 $\mu g/m^3$ increase for short-term and long-term exposure to PM2.5 by cause of admission

PM2.5 Exposure Type	All Respiratory	Cardiovascular Disease	Stroke	Diabetes
Short-term	0.70%	1.03%	0.24%	0.96%
Long-term	4.22%	3.12 %	3.49%	6.33%

Source: Kloog et al. Acute and Chronic Effects of Particles on Hospital Admissions in New England. Harvard School of Public Health, 2012. www.hsph.harvard.edu/clarc/sac2012/kloog-ne.pdf

No Safe Threshold Has Been Identified

The federal Clean Air Act requires US EPA to adopt ambient air quality standards for PM and other criteria pollutants at a level that provides an "adequate margin of safety ... requisite to protect the public health." EPA is charged with reviewing the standards every five years based on the latest scientific evidence on health effects. A key issue in setting standards is whether researchers can identify a safe threshold below which level no negative health effects are observed. To date, researchers have not been able to identify a "no-effects" threshold for PM. The evidence suggests that in terms of the effect of PM on premature mortality, the concentration-response function (i.e., how the incidence of a given health effect varies in response to a change in ambient concentration of the pollutant) is essentially linear (EPA Integrated Science Assessment for PM, 2009). These findings suggest that people exposed to PM at levels below the current EPA standards may still experience negative health effects. (PM air quality standards are discussed in Section 3.)

Recent Findings

Research on the health effects of PM is on-going. Our understanding of PM health impacts is gradually enhanced as new studies and journal articles appear at a steady rate. The new research reinforces earlier findings regarding negative impacts of PM on both respiratory and cardiovascular health, and increased rates of health impacts such as heart attacks, strokes, and premature death in response to PM exposure. However, in addition to confirming the results of earlier research, new research is also uncovering evidence of a wider range of potential health effects from exposure to PM, including, linkages to diabetes, reduced cognitive function in older adults, and oxidative damage to DNA.

New research is uncovering evidence of a wider range of potential health effects from exposure to PM, including linkages to diabetes, reduced cognitive function in older adults, and oxidative damage to DNA.

Diabetes: The incidence of Type 2 diabetes (sometimes referred to as "adult onset" diabetes) has increased rapidly in recent years in response to sedentary lifestyles, changes in diet, and higher rates of obesity. People are also contracting Type 2 diabetes at a younger age as well. Experts predict major impacts on public health and enormous costs to the health care system as a result of increasing diabetes rates. Although diet and lifestyle are key factors in diabetes incidence, a recent nationwide study (Pearson et al. 2010) found that air pollution may also be a risk factor for diabetes. The study concluded that diabetes prevalence increases with increasing PM2.5 concentrations, with a 1% increase in diabetes prevalence seen with a 10 $\mu g/m^3$ increase in PM2.5 exposure. These results suggest that PM2.5 may contribute to increased diabetes prevalence in the adult U.S. population and that air pollution is a risk factor for diabetes. There is also some evidence that people with diabetes may be more vulnerable to the negative health effects of PM. A recent study (O'Donnell et al. 2011) by the Harvard School of Public Health found that diabetics exposed to PM may be at higher risk for ischemic stroke compared to the background population.

Impacts on the Brain and Cognition: Although the lungs and the circulatory system are the primary path by which particles are transported throughout the body, studies (e.g., Oberdörster et al. 2010) have found that particles can also enter the central nervous system and then the brain via olfactory neurons in the nasal passages. When ultrafine particles travel via olfactory nerves to the brain, they are able to bypass the blood-brain barrier, the defensive shield that blocks unwanted chemicals from reaching sensitive brain cells. Studies which exposed mice to both fine and ultrafine particles showed inflammatory responses in the brain. Ultrafine particles can also damage brain cells in the basal ganglia, the region of the brain impacted by degenerative nerve diseases such as Parkinson's (Peters et al. 2006).

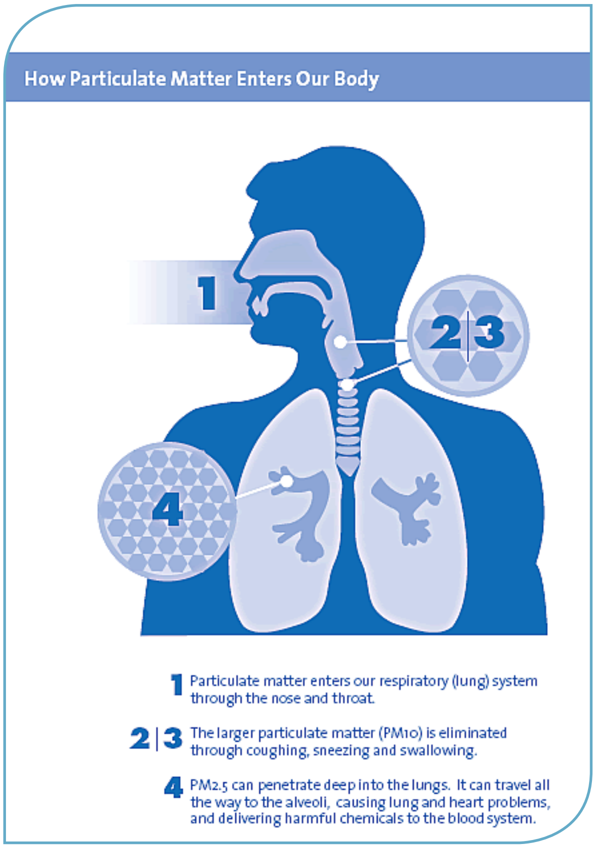
A recent study (Weuve et al. 2012) found an association between long-term exposure to both fine PM and coarse PM and cognitive capability in older women. This study, based on the longitudinal Nurses' Health Study Cognitive Cohort, examined the effects of PM exposure over 7-14 years for nearly 20,000 American women aged 70 to 81. The study found that women exposed to higher levels of PM experienced more rapid cognitive decline, and concluded that the effect of a 10 μg/m^3 increment increase in long-term PM exposure is equivalent to approximately two years of cognitive aging. The study noted that higher levels of exposure to ambient PM are associated with worse cognitive decline, and that the effects observed occurred at levels of exposure typical of many areas of the United States. The authors suggest that reducing particulate levels may help to reduce the future incidence of age-related cognitive decline and dementia. In a clinical postmortem study (Calderon-Garciduenas et al. 2004) that also points to a potential connection between PM and cognitive impairment, researchers found higher levels of amyloid-B42, a hallmark of Alzheimer's disease, in the brains of people who lived in cities with higher pollution levels. Clinical studies in animals have shown increased brain inflammation in response to PM exposures.

Biological Pathways: How PM Harms the Body

External exposure to PM can cause short-term impacts to external organs, such as irritation of the eyes. But the most damaging effects are caused when pollution enters the body via the respiratory system. The mechanisms by which PM and other air pollutants damage the lungs and the respiratory system are well understood. Our lungs serve as the entry point to the body for PM and other pollutants, so they are the organ most directly impacted by air pollution. In addition, the lungs are especially sensitive to air pollutants because they contain a large surface of exposed membrane to facilitate the delivery of oxygen to the blood system. Our respiratory system has defenses in the nasal passages, throat, and lungs that filter out particles, but the smallest particles are most likely to elude the body's filtration mechanisms. For example, as much as 50% of ultrafine particles with a diameter of 0.02 microns or smaller are estimated to be deposited in the alveolar region of the lung. Particles inhaled deep into the lungs can then be transported to cells and organs throughout the body. And once particles become deeply embedded in our body, they can remain there for weeks, months, or even years.

Figure 1-1 depicts how PM enters the body. The larger particles are typically filtered out; in contrast, particles less than 2.5 microns in diameter can penetrate deep into the lungs which are where most health problems begin.

Figure 1-1 How Particulate Matter Enters Our Body



Source: British Columbia Air Quality (www.bcairquality.ca/health/air-quality-and-health.html)

On the individual basis, the health impact of long-term exposure to PM is likely to be determined by the number of particles that are transported from the lungs into the body, the chemical composition of the particles, and how quickly the particles are cleared from the body.

Researchers have been making progress in recent years in explaining how PM damages the cardiovascular system and other organs and systems, but this is still an area of on-going research. Research to date indicates that inflammation and oxidative stress are two of the key ways that PM damages the body.

Inflammation: When foreign substances are deposited in the body, this irritates the impacted area and causes an inflammatory response. Studies (e.g., Araujo et al. 2010) have found that exposure to PM can lead to chronic, low-level inflammation. An inflammatory response to PM or other air pollutants can damage the body in many ways. In the vascular system, an inflammatory response to PM can stiffen blood vessels and reduce their flexibility, leading to higher blood pressure, increased blood coagulation, hardening of the arteries (atherosclerosis), altered cardiac autonomic function (the system that controls the heart), and reduction in heart rate variability (a risk factor for future cardiovascular problems). All these effects can increase long-term risk of heart disease or stroke. Based on high particle numbers, high lung deposition efficiency and surface chemistry, ultrafine PM may be especially dangerous in terms of its potential to induce inflammation.

Oxidative Damage to DNA: Studies (e.g., Risom et al. 2005) indicate that exposure to PM increases **oxidative stress**. This term describes the effect of oxidation in which an elevated level of reactive oxygen species, such as free radicals (e.g. hydroxyl, nitric acid, superoxides) or non-radicals (e.g. hydrogen peroxide, lipid peroxide) causes oxidative damage to specific molecules, thereby injuring cells or tissue. There is evidence that ultrafine PM may cause oxidative damage to DNA. For example, a Danish study (Vinzents et al. 2005) found that participants who rode bicycles in traffic in Copenhagen, and were thus subjected to increased exposure to ultra-fine PM, sustained oxidative damage to their DNA, thus demonstrating an association between DNA damage and ultrafine PM exposure in live subjects.

Cardiovascular effects: In terms of explaining how PM damages the cardiovascular system, more study is needed to determine how exposure to PM affects intermediate health outcomes such as heart rate variability and inflammation markers. In a paper (Pope & Dockery 2006) reviewing research on the health effects of PM, two of the leading researchers summarize their discussion of biological mechanisms as follows: “Various plausible pathways have been identified. However, none has been definitively demonstrated to be the pathway that directly links exposure to PM with cardiopulmonary morbidity and mortality. In fact, it is unlikely that any single pathway is responsible. There are almost certainly multiple pathways with complex interactions and interdependencies.”

Which sizes and types of particles are most dangerous?

Evidence suggests that PM health effects depend upon both particle size and particle composition.

Particle Size: Research indicates that the health effects of PM depend upon particle size. Smaller particles (in the fine and ultrafine size ranges) are generally more harmful than coarse particles. Smaller particles typically remain suspended in the air for longer periods; penetrate more readily and deeply into the lungs, bloodstream and organs; and present a large amount of reactive surface area relative to their mass.

Ultrafine PM: A growing body of evidence documents public health effects from ultrafine particles. Motor vehicles are a major source of ultrafine particle emissions, and these particles are highly reactive when emitted from internal combustion engines. Because ultrafine particles are so miniscule, they can travel deep into the lungs and organs and pass through cell membranes. These particles can also carry toxic compounds into the body. Based on high particle numbers, high lung deposition efficiency, and surface chemistry, ultrafine particles may have a greater potential than PM2.5 for inducing inflammation and oxidative stress, key mechanisms by which PM harms the body. In clinical studies, greater inflammatory and oxidative stress (cell, tissue or organ damage), resulting in damage to DNA, has been associated with exposure to ultrafine particles compared to the larger particles at comparable mass doses. In some cases, the substances absorbed on to the ultrafine particles may be responsible for some of the effects observed, including oxidative stress, rather than the particles themselves. A study (Oberdoster et al. 2010) that examined the effects of combustions fumes on laboratory rats found that, compared to larger particles, ultrafine particles cause a greater inflammatory response in the lungs of rats and increased antioxidant levels in their lung tissues.

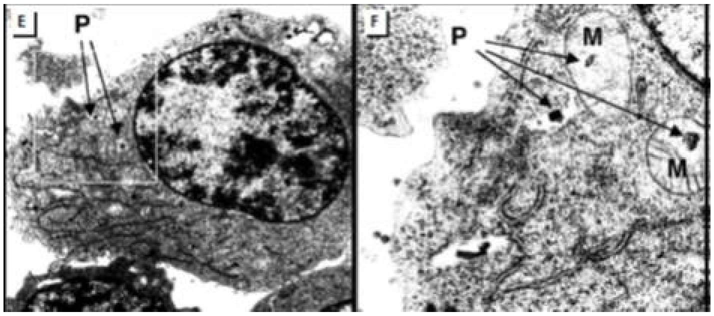
Numerous studies (e.g., Wichmann 2000) have found a correlation between exposure to ultrafine PM and increased incidence of health effects such as premature mortality, hospital admissions, lung cancer and other cancers, cardiovascular disease, adverse birth outcomes, effects on the immune system, and neurotoxicity. In cell cultures exposed to ambient particles, ultrafine particles were found in the mitochondria where they induced structural damage.

Research on health effects related to exposure to ultrafine PM is still very limited compared to the amount of research that has been performed into the health effects of PM2.5. For example, specific mechanisms of health effects from exposure to ultra-fine nitrate and sulfate particles are not well defined, nor are the health effects of semi-volatile organic compounds and trace metals found in ultrafine PM. However, existing studies suggest that ultrafine PM may have significant health effects, and that some of the health effects related to ultrafine particles may be independent of the effects from exposure to PM2.5 and/or PM10.

PM Damage to Cells

Figure 1-2 depicts an image of what happens when PM passes through the lungs and penetrates into cells. Photos E and F (magnified x 6,000 and 21,000, respectively) show a cell exposed for 16 hours to fine PM (PM2.5). The "P" indicates damage to **cristae**, the inner membrane of the mitochondria, which are studded with proteins and increase the surface area for chemical reactions, such as cellular respiration, to occur. The "M" points to the presence of particles inside the mitochondria as well as ultra-structural damage to the mitochondria. Mitochondria, sometimes described as "cellular power generators" supply cellular energy, and are involved in a number of processes such as signaling, cellular differentiation, cell death and the control of the cell cycle and cell growth. Mitochondria have a central place in cell metabolism and their damage plays an important role in a wide range of health effects.

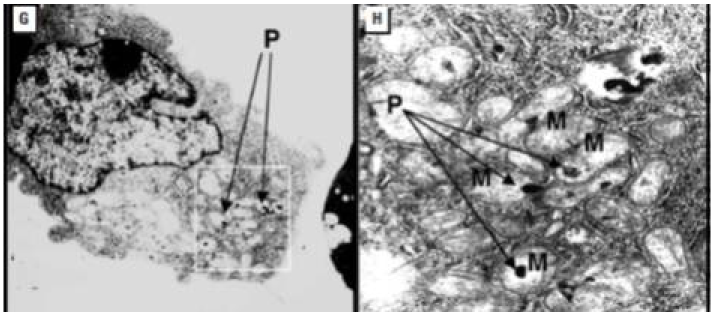
Figure 1-2 Cells Exposed to Fine PM (PM2.5) for 16 Hours



Source: Cho et al. *Ultrafine Particulate Pollutants Induce Oxidative Street and Mitochondrial Damage.* Environmental Health Perspectives. 2003 April; 111(4): 455–460.

Figure 1-3 shows images of a cell exposed to ultrafine PM for 16 hours. As in the figure above, the "P" points to damage to the cristae, and the "M "shows the presence of particles inside the mitochondria as well as structural damage. The degree of ultra-structural damage in this study was found to be greater in the cells exposed to ultrafine PM than the cells exposed to fine PM (and even more so than those exposed to coarse PM, which experienced little if any mitochondrial damage).

Figure 1-3 Cells Exposed to Ultrafine PM for 16 Hours



Source: Cho et al. 2003.

Relationship Between Particle Composition and Health Effects

The available evidence from epidemiological studies to date suggests that fine particles themselves are harmful, regardless of their emissions source or chemical composition. Isolating and pinpointing the health effects of a specific particle type through epidemiological studies is difficult, because many particle types and sizes are co-emitted by the same sources and processes, and the air we breathe always includes a diverse mix of particle sizes and types. Because risk estimates for any specific particle type are subject to confounding by co-pollutants, the evidence for differential health risk among PM2.5 components is not as robust as for PM2.5 as a whole. Therefore, when estimating the health impacts of PM, most researchers currently assume that all mixtures of PM2.5 are equally potent. And in the absence of information to clearly distinguish the relative risk of different particle types, US EPA and ARB currently treat all particles alike, regardless of their chemical make-up, for purposes of PM air quality standards. (It should be noted, however, that ARB does call out diesel PM for special attention as a toxic air contaminant.)

But even though it is difficult to determine the relative harmfulness of different particle types, variation in particle composition and behavior suggests that their health effects are likely to differ as well. There is some evidence that specific particle types, such as black carbon and diesel PM2.5, may be especially harmful. For example, fine and ultrafine particles produced by fuel combustion may be more toxic, because they are highly reactive and because they include sulfates, nitrates, acids, trace metals and other toxic contaminants. Researchers have also hypothesized that insoluble ultrafine particles with a solid core may be more harmful than soluble particles. Whereas soluble particles will dissolve as they interact with blood and body liquids, the insoluble particles persist in solid form and can thus penetrate through protective barriers to irritate and inflame deep within the body (Ostiguy et al., IRSST, 2006).

As the science advances, at some future date it may be possible to link the various health effects associated with PM to specific particle types, or combinations thereof. From the standpoint of protecting public health, determining which sizes or types of particles are most harmful is vitally important in the quest to understand PM. If new information becomes available to identify and help target the most harmful particle types, then we should be able to develop more effective control measures to maximize public health benefits in the most cost-effective manner.

From the standpoint of protecting public health, determining which sizes or types of particles are most harmful is vitally important in the quest to understand PM.

Motor Vehicle Emissions

From the standpoint of population exposure, PM from both gasoline and diesel powered vehicles is of special concern, because of the large number of people exposed to motor vehicles emissions in urban areas, including pedestrians, bicyclists, motorists, and people who live, work or go to school in proximity to busy roadways. Vehicle tailpipe emissions include primary ultrafine and fine PM from fuel combustion and lubricating oil, as well as gaseous PM precursors such as ROG, NOx, and ammonia. Vehicles also produce particles from brake and tire wear that include toxic chemicals such as copper and cadmium; these toxic substances pollute soil and water as well as the air. In addition, motor vehicles re-suspend dust (primarily coarse particles) that has been deposited on roadways from a variety of emission sources, so that these particles once again become available to be inhaled.

ARB's *Air Quality and Land Use Handbook* (April 2005) cites several key findings regarding health impacts from near-roadway exposures.

- Reduced lung function in children was associated with traffic density, especially trucks, within 1,000 feet; this association was strongest within 300 feet. (Brunekreef, 1997)
- Increased asthma hospitalizations were associated with living within 650 feet of heavy traffic and heavy truck volume. (Lin, 2000)
- Asthma symptoms increased with proximity to roadways; the risk was greatest within 300 feet. (Venn, 2001)
- Asthma and bronchitis symptoms in children were associated with proximity to high traffic in a San Francisco Bay Area community with good overall regional air quality. (Kim, 2004)
- A San Diego study found increased medical visits in children living within 550 feet of heavy traffic. (English, 1999)

The ARB *Handbook* notes that truck traffic densities and distance from the roadway are key factors affecting the strength of the association with adverse health effects. In the health studies cited in the ARB *Handbook*, adverse health effects diminished with distance; adverse health effects were seen within 1,000 feet of high-volume roadways, with the strongest effects within 300 feet.

Exposure to vehicle emissions has been linked to increased blood pressure and thickening of the arteries. A recent clinical study (Brook et al. 2009) that exposed subjects to PM levels typically found near highways showed an immediate increase in blood pressure. This study also found that exposure to PM induced inflammation; this effect typically manifested within roughly 24 hours after exposure. Another recent study (Kunzli et al. 2010) found that thickening of artery walls progressed more than twice as quickly among people living within 100 meters of a Los Angeles freeway (where ultrafine PM levels are typically elevated) compared to those who lived farther away.

The Brugge (2007) review of health studies concludes that there is strong evidence that exposure to high volume roadways is linked to higher asthma rates and to reduction of lung function in children. In addition, recent studies (Schwartz et al. 2005, and Adar et al. 2007) have found that heart rate variability, a risk factor for future cardiovascular problems, is altered by traffic-related pollutants, particularly in older people and people with heart disease.

Lubricating Oil

Recent research indicates that engine lubricating oil may contribute to PM emissions from motor vehicles. To investigate how lubricating oil (and fuels) contributes to the formation of PM and semi-volatile organic compounds (SVOC) in vehicle exhaust, the National Renewal Energy Lab, ARB, South Coast AQMD and other partners worked together on the Collaborative Lubricating Oil Study on Emissions (CLOSE). The objective was to evaluate how much PM emissions come from lubricating oil and explore ways to reformulate oil so as to reduce PM emissions.

The CLOSE project found that in the vehicles tested unburned lubricating oil makes up more than 70% of organic carbon (OC), which constitutes a large portion of UFPM. The results of the CLOSE project indicate that lubricating oil and fuels may lead to the formation of PM. However, further study is needed to test a wider range of vehicles, and to investigate the effects of oil type on PM and SVOC formation.

Diesel PM

Diesel PM is a subset of PM2.5 that is emitted by diesel engines. Although diesel PM accounts for a small portion (less than 10%) of the overall PM2.5 emission inventory, it has been called out for special attention by ARB because of its toxicity. In 1998, in response to a comprehensive health assessment of diesel exhaust, ARB formally identified diesel PM as a toxic air contaminant (TAC), a special class of air pollutants that can impair public health even at very low exposures or dosages. TACs can cause both acute and chronic effects, including cancer. Diesel exhaust also contains more than 40 other TACs, including carcinogens such as benzene, arsenic, nickel, and formaldehyde. The Air District performed an analysis of TACs for its Community Risk Evaluation (CARE) program and found that diesel PM accounts for approximately 85% of the total cancer risk from TACs in the Bay Area. As discussed in Section 4, diesel PM has been the focus of control efforts by both ARB and the Air District.

Wood Smoke

Although wood fires may have an aesthetic appeal and some people may perceive wood smoke as natural or even healthy, findings from health studies to date indicates that wood smoke particles cause the same types of negative health effects as other types of fine PM. Wood smoke accounts for a major portion (38%) of the Bay Area PM2.5 inventory during the winter season.

Wood smoke is produced by incomplete combustion from residential fireplaces and wood stoves. Whereas combustion in diesel and gasoline engines is carefully controlled to regulate oxygen supply and the fuel-to-air ratio in order to maximize the efficiency of the combustion process, the wood-burning combustion process in fireplaces is largely uncontrolled. This leads to inefficient combustion, resulting in a high rate of smoke and PM emissions compared to other types of combustion. In addition to PM, wood smoke contains thousands of chemicals, including criteria pollutants such as sulfur oxides (SOx), nitrogen oxides (NOx), and carbon monoxide (CO); as well as several dozen toxic air contaminants such as acrolein and acetaldehyde, and carcinogenic compounds such as polycyclic aromatic hydrocarbons (PAHs), benzene, formaldehyde and dioxins. More research is needed to evaluate how the various pollutants in wood smoke may interact in terms of health impacts.

In addition to PM, wood smoke contains various toxic and carcinogenic compounds.

A review of research on the health effects of residential wood smoke (Zelikoff et al. 2002) found that prolonged inhalation of wood smoke contributes to chronic bronchitis, chronic interstitial lung disease, pulmonary arterial hypertension, and altered pulmonary immune defense mechanisms in adults. Studies (e.g., Larsen & Koenig, 1994) found that young children living in homes heated by a wood-burning stove had a greater occurrence of moderate and severe chronic respiratory symptoms than children who did not live in homes heated with a wood-burning stove. Effects on preschool children living in homes heated with wood burning stoves or in houses with open fireplaces include decreased lung function in young asthmatics, increased incidence of acute bronchitis, and increased incidence and duration of acute respiratory infections. One study (Danielsen et al. 2011) found that wood smoke PM has small particle size and a high level of polycyclic aromatic hydrocarbons (PAH). This study found that, in terms of health effects, exposure to wood smoke produces high levels of free radicals, may trigger inflammatory and oxidative stress in cultured human cells, and may damage DNA.

Studies of wood smoke health effects to date have focused primarily on respiratory impacts. Additional research is needed to evaluate potential cardiovascular effects from exposure to wood smoke.

Estimating PM Health Impacts in the Bay Area

The Air District has performed two recent analyses to estimate the public health impacts (morbidity and premature mortality) of PM and other air pollutants in the Bay Area.

- *Bay Area Air Pollution Burden: Past and Present* (see Appendix A in the Bay Area 2010 Clean Air Plan, issued September 2010).
- *Health Impact Analysis of Fine Particulate Matter in the San Francisco Bay Area,* September 2011.

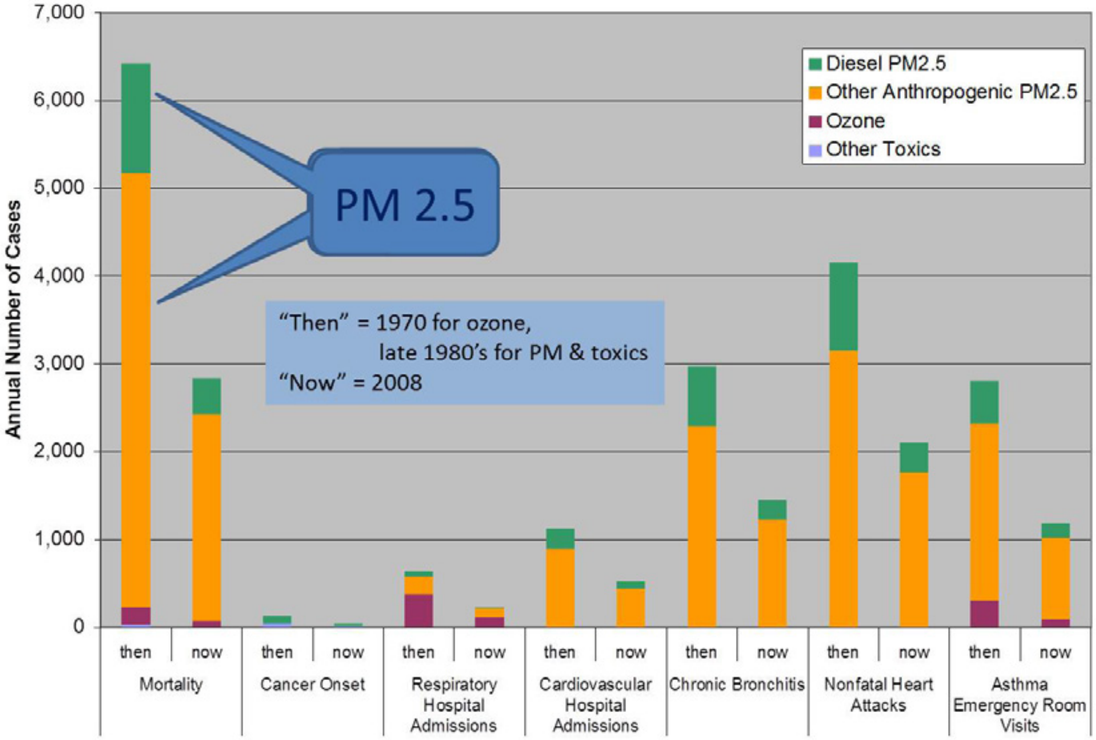
These analyses were based on methodologies employed by US EPA and ARB to estimate the health impacts and the monetary costs of air pollution. They rely on the results of peer-reviewed epidemiological studies and US EPA's BenMAP program, in combination with air quality modeling results performed in-house by Air District staff. Key findings from these studies include the following:

- Improvements in Bay Area air quality in recent decades have greatly reduced the health burden from air pollution. Most of the public health benefit is due to the substantial reduction in PM levels in recent years, as discussed in Section 3-C.
- The reduction in health impacts has provided cost savings (in terms of reduced treatment costs, increased productivity, and longer life span) to the region valued at multiple billions of dollars per year. Most of this economic benefit is due to progress in reducing PM levels.
- Average life expectancy in the Bay Area increased from 75.7 years in 1990 to 80.5 years in 2006. Improved air quality – primarily the reduction in PM levels – in the Bay Area during this period accounts for roughly 6 months of the overall increase in average Bay Area life expectancy.
- Despite progress in reducing PM levels and related health impacts, exposure to fine PM (PM2.5) remains the leading public health risk, and contributor to premature death, from air pollution in the Bay Area. The vast majority of premature deaths associated with air pollution - more than 90% - are related to exposure to fine particulate matter (PM2.5). Most of the deaths associated with PM2.5 are related to cardiovascular problems. Based on current PM levels, exposure to PM is estimated to contribute to approximately 1,700 premature deaths per year in the Bay Area; this represents roughly 3-5% of total annual deaths in the region. The economic cost of these negative health impacts is estimated at multiple billions of dollars per year.
- PM emitted by diesel engines is believed to be the leading toxic air contaminant (TAC) in the Bay Area. However, the current evidence suggests that only 10-20% of PM-related deaths in the Bay Area are linked to diesel exhaust. PM from other (non-diesel) sources (such as wood smoke, cooking, combustion of other (non-diesel) fossil fuels, and secondary PM formed by precursors such as NOx, SO2, and ammonia) appears to be responsible for most of the PM and the PM-related deaths in the Bay Area.
- The Air Resources Board has identified diesel PM as a carcinogenic pollutant that may cause lung cancer. Although lung cancer is clearly a major public health issue, it should be noted that exposure to diesel PM may cause a wide range of respiratory and cardiovascular effects in addition to lung cancer. In fact, to the extent that diesel PM contributes to premature mortality, analysis suggests that this is primarily due to its role as a component of PM2.5, in which it contributes to mortality related to heart attacks, emphysema, strokes, etc.

PM is the air pollutant that causes by far the greatest harm to public health in the Bay Area.

Figure 1-4 depicts findings from the health burden analysis performed for the Bay Area 2010 Clean Air Plan. The graph shows the number of cases of seven key health efects that are related to population exposure to current Bay Area air pollution levels (2008, labeled "now) compared to the estimated number of cases that would have occurred if the quantifiable air quality improvements had not been made (labeled "then"). The "then" data is based on the earliest data available - 1970 for ozone, and the late 1980's for toxics and PM. Figure 1-4 shows that the annual cases of health effects associated with exposure to air pollutants in the Bay Area has dropped dramatically, by more than half. Of particular interest, premature mortality related to air pollution has decreased from an estimated 6,400 per year to an estimated 2,800 per year. Despite this substantial progress, the health impacts from air pollution are still significant. And as the graph shows, PM2.5 accounts for the vast majority of the health effects in comparison to ozone or other (non-diesel) toxic air contaminants.

Figure 1-4 Bay Area Air Pollution Health Burden: Past & Present



Estimated contribution of PM, ozone, and key air toxics to health endpoints among Bay Area residents, based on Bay Area air pollution data for year 2008.

Summary of Key Points on the Health Effects of PM

- A robust body of epidemiological studies has established that there are a wide range of negative health impacts from both short-term and long-term exposure to PM2.5. These impacts include both respiratory and cardiovascular effects, including key health endpoints such as heart attacks and strokes.
- Although researchers are still working to define which specific particle types are more harmful to human health, the available evidence indicates that smaller particles are most dangerous because they can most easily penetrate into the lungs, bloodstream, organs, and cells of the body.
- There are documented health effects from exposure to PM2.5 even below current PM air quality standards, and researchers have not been able to establish a safe threshold below which there are no health risks.
- Even although the Bay Area either attains or is close to attaining State and national air quality standards for PM, analysis by Air District staff indicates that PM is the air pollutant that imposes by far the greatest harm to public health in the Bay Area.
- Even in regions with relatively low PM concentrations, such as the Bay Area, efforts to further reduce PM levels will results in public health improvements and longer average life expectancy, thus providing significant social and economic benefits.

Areas that Require Additional Research

This chapter has attempted to summarize current information regarding the health effects related to PM.

Although there is robust evidence that exposure to PM can cause a wide range of respiratory and cardiovascular effects, many fundamental questions have not yet been fully answered. Researchers are actively working to provide better answers to key questions, including the following:

- Which particle types are most dangerous?
- What biological mechanisms cause the observed health effects?

- Is there a safe level (no-effects threshold) for short-term or long-term exposure to PM, below which no health effects occur?
- Do ultrafine particles cause health effects independent of the health effects caused by exposure to PM2.5?
- If ultrafine particles do cause health effects independent of the effects caused by exposure to larger particles, are these effects related to short-term peak exposure, chronic exposure to lower levels of UFPM, or a combination of both acute and chronic exposure?
- How does exposure to PM affect intermediate health outcomes such as heart rate variability and inflammation markers?
- What is the relationship between exposure to PM emissions from roadways and health effects such as cardiovascular disease, lung cancer, and premature mortality?

SECTION 1-B: POPULATION EXPOSURE TO PM

This section summarizes key information regarding population exposure to PM.

In order to protect Bay Area residents from the negative health effects related to PM described above, we need to better understand population exposure to PM. The key questions are: Who is exposed to PM? And when, where, and how much PM are people exposed to?

Reducing personal (individual) exposure and population exposure (the aggregate exposure experienced by all Bay Area residents combined) to PM can provide important public health benefits. Key questions considered in this section include:

- Who is at greatest risk from exposure to PM?
- How do PM concentrations vary at the local scale?
- Which types of environments pose the greatest risk?
- How much exposure to PM occurs in the indoor environment compared to outdoors?
- How do PM concentrations and population exposure to PM change based upon distance from emission sources?

Linking PM to Health Effects

The connection between PM and negative health effects can be explained by a pathway that includes the following links:

1. **Emissions:** A wide range of sources release primary PM and PM precursors into the air.
2. **Ambient PM concentrations:** The combination of emissions and meteorological conditions determines the ambient concentration of PM, that is, the level of PM in the air.
3. **Exposure:** Exposure occurs when people actually inhale PM into their lungs. The level of exposure to PM is closely linked to the ambient PM concentration.
4. **Health effects:** Health effects may occur as a result of exposure to PM, depending upon the intensity, duration and frequency of population exposure, as well as the size and physical condition of the receptor population.

The Air District has detailed information about PM emissions and ambient concentrations of PM[3] at the regional scale, and there is a wealth of epidemiological studies analyzing the relationship

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[3] The emissions inventory described in Section 2 provides information on emissions by source. Information regarding ambient concentrations of PM is available from monitoring data and photochemical modeling.

between population exposure and health effects. However, as discussed below, estimating population exposure to PM at the regional scale is difficult. In analyzing how PM effects public health via the steps described above, the greatest uncertainty relates to estimating population exposure.

Measuring population exposure to PM, and the related health risks, is challenging for several reasons:

- PM concentrations vary both temporally and spatially
- The composition of PM varies depending upon the mix of emission sources and meteorology
- Personal activity patterns are complex
- Indoor exposure may account for a significant share of total exposure

Emissions: Many different sources, both stationary (factories, refineries, etc.) and mobile (cars, trucks, locomotives, marine vessels, and farm and construction equipment) emit direct emissions of PM and/or PM precursors such as NOx and SOx. Identifying the key emission sources and developing strategies to reduce emissions from these sources is the first and most fundamental step to improve air quality.

Ambient Concentrations: This term refers to the level of pollutants that are measured in the air. PM air quality standards are expressed in terms of ambient concentrations, as discussed in Section 3. The relationship between emissions and ambient concentrations is complex and depends upon many factors, including meteorological conditions (temperature, humidity, wind speed and direction, vertical mixing, etc.) the ratio of precursor pollutants, and regional topography. Ambient concentrations of PM can vary greatly at the local scale.

Population Exposure:

From the standpoint of protecting public health, the key objective is to reduce population exposure to PM and other air pollutants. The issue is not simply how much pollution may be degrading the quality of the air that we breathe, but rather how much people are exposed to the pollution and how much each individual actually inhales and absorbs (dosage). Key factors in determining population exposure include how much pollution is in the air at the time and location that exposure occurs, the number of people exposed, the duration

of exposure, and the frequency of exposure. People are exposed to air pollutants in both the outdoor and indoor environments. In the case of PM, the evidence suggests that most people experience a major portion of their total PM exposure when they are indoors, as discussed later in this Section.

Population exposure to PM can also be analyzed in terms of **intake fraction**, i.e., the fraction of an emitted pollutant that is actually inhaled by the population. High PM emissions in a sparsely-populated area may cause little damage to public health, because the intake fraction and population exposure are low. Conversely, moderate levels of emissions in proximity to a densely populated environment may result in a higher intake fraction, more population exposure, and greater public health impacts.

Factors that determine individual exposure to PM and other air pollutants include:

- The pollution profile: the level of pollution, mix of co-pollutants, the specific mix of particle sizes and their chemical composition, all of which vary depending on geographical location and emissions sources, season of the year, and weather conditions.
- Where people live, work, and play.
- Activity patterns and lifestyle choices such as how much time people spend outside, or how much time they spend driving on busy roadways.

Personal exposure to PM for a given individual on a specific day can be calculated by multiplying the time spent in each activity by the PM concentration in each location, and then summing the exposure for each activity. The degree of exposure may vary greatly depending on the type and location of the activity. Therefore, an activity that involves high exposure for a relatively short period of time, such as driving on a freeway for 30 minutes, may account for a major portion of total daily exposure.

Estimating total population exposure to PM is challenging because people are mobile and PM levels may vary substantially from place to place. Estimating population exposure to PM requires three steps:

1. Documenting activity patterns for a specific individual: i.e., where, when, and for how long he or she performs various activities.
2. Estimating the ambient concentration of PM that the individual is exposed to at each time and location. These estimates are derived from air quality monitoring data and/or computer modeling results.
3. Aggregating each individual's personal exposure across an entire defined population.

Dosage: The amount of an air pollutant that an individual actually inhales is called **dosage**. And the amount that is absorbed by the body and becomes available for interaction with biologically significant organs and tissues is called the **internal dose**. Once absorbed, the chemical can undergo metabolism, storage, excretion, or transport within the body. The amount transported to a specific organ or system is termed the **delivered dose**.

The air we breathe serves as the carrier medium by which harmful pollutants can enter the body. The average adult takes about 25,000 breaths and inhales a total volume of approximately 14,000 liters of air every day. So even though we may only be exposed to a tiny amount of pollution per breath, on a cumulative basis this can result in a substantial amount of pollution inhaled over time.

The pollutant dosage depends upon the amount of pollution in the air and an individual's inhalation rate relative to their body weight. Inhalation rate varies depending upon age, metabolic rate, body weight, and type of activity. When people are exercising, they inhale more frequently and more deeply, resulting in a higher dosage. So walkers, runners, and bicyclists experience greater dosage from a given exposure; this may be a concern if people engage in these activities in close proximity to busy roadways or other major emission sources.

Children may experience higher dosage than adults, because they are more physically active, have a higher metabolic rate and inhale more air on a per-pound basis than adults. Children also tend to breathe more through their mouths more than adults; as a result, more PM may reach their lungs because the mouth is less effective than the nose at filtering out PM. For these reasons, children typically experience a greater dosage into their bodies from a given exposure to pollution.

Sensitive Populations

Just as individual exposure differs, so does the ability of our bodies to tolerate exposure to pollutants. People vary in their susceptibility to health effects from air pollution depending upon factors such as genetic features, gender, age, lifestyle (e.g., smoking status and nutrition), and their health status. To protect public health, we need to focus on reducing exposure among the most sensitive populations and the most heavily impacted communities.

The key aspect to protecting public health is to reduce personal exposure for the people who are most susceptible to air pollution; these "sensitive populations" include children, pregnant women, seniors, and people burdened with existing cardiovascular or respiratory conditions. Children are especially vulnerable to air pollution due to their higher inhalation rates, narrower airways, less mature immune systems, and the fact that their lungs and other key organs are still developing. In addition, children with allergies may have an enhanced allergic response when exposed to pollutants such as diesel exhaust. Children also tend to spend more time outside than adults, so they may be more exposed to pollutants in the ambient air. Seniors and people with existing cardiovascular or respiratory conditions are more vulnerable to the effects of air pollution than healthy adults because their lungs, hearts, and immune systems may already be weakened or compromised.

Some epidemiological studies suggest that PM health effects may be influenced by gender. For example, a 2007 study by the Women's Health Initiative (Miller et al. 2007), which examined the impacts of PM exposure on post-menopausal women, found that a relatively modest increase in PM2.5 levels (an increase of 10 µg/m^3 or micrograms per cubic meter) was associated with a 76% increase in the risk of death from cardiovascular disease. This is much higher than the increase in risk associated with an increase of 10 µg/m^3 that was observed for the general population in the Harvard Six Cities study (19%) and the American Cancer Society study (13%).

Although members of sensitive populations live in communities throughout the Bay Area, we know that certain communities experience higher-than-average levels of air pollution. And in many cases, people who live in communities that are disproportionately impacted by air pollution may be especially vulnerable to the negative effects of air pollution because of their demographic and socioeconomic status. Studies have shown that socioeconomic factors such as income, race, access to health care, and level of educational attainment, can profoundly affect health status and life expectancy.

But although a correlation between socioeconomic factors and health outcomes has been well documented, determining the precise correlation between a particular socioeconomic factor and a specific health outcome is complicated because the various factors are closely intertwined in most communities. Identifying the specific mechanism(s) by which socioeconomic factors affect health outcomes is also difficult. But researchers have found that factors such as poverty and race contribute to stress, thus increasing susceptibility to the health effects of air pollution, since stress weakens the immune system and may be a factor in initiating some types of disease.

Recognizing that certain neighborhoods and communities in the Bay Area are disproportionately impacted by local air pollutants, the Air District launched the Community Air Risk Evaluation (CARE) program in 2004. Air District staff performed technical analysis which identified six impacted communities most affected by toxic air contaminants. The Air District has been making a concerted effort to reduce emissions and population exposure to PM and toxic air contaminants in these communities, as described in Section 4.

To sum up, many factors go into determining population exposure to air pollutants and the degree to which an individual's health may be affected by that exposure. The amount of pollution in the air and the frequency and duration of the exposure are key factors. But individual attributes such as age and gender; personal activity patterns; lifestyle (active or sedentary); socioeconomic status; and health status all play a role as well in determining how much pollution people are exposed to and how their bodies respond to that exposure.

Which Environments Pose the Greatest Risk?

In the course of their daily activities and routines, Bay Area residents are exposed to PM and other air pollutants in a variety of locations and environments. Several of these environments may expose people to elevated levels of PM and account for a significant portion of total exposure to PM.

Population Exposure to Motor Vehicle Emissions

Most Bay Area residents are exposed to motor vehicle emissions on a daily basis. Motor vehicles emit the entire spectrum of ultrafine, fine, and coarse PM by means of several mechanisms. Gasoline and diesel vehicles produce tailpipe emissions of both primary PM in the fine and ultrafine size ranges, as well precursor pollutants that form secondary PM, such as ROG, NOx, and ammonia. Lubricating oil has also been identified as a potential important source of ultrafine PM. In addition to these combustion-related emissions, motor vehicles generate PM from brake wear and tire wear, and they cause dust that has settled on roads to become re-suspended in the air. Studies have found a wide range of negative health effects among people who are exposed to PM emissions from motor vehicles, as discussed in Section 1-A.

The amount of primary PM and PM precursors emitted by vehicles on a given roadway, and the chemical composition of those emissions, depend upon many factors, including:

- the volume of traffic and the level of traffic congestion;
- the composition of the vehicle fleet, and whether there are any restrictions for certain types of vehicles, such as trucks;
- the age mix of the vehicles, the effectiveness of their emissions control devices, and the fraction of vehicles that are high-emitters;
- the mix of fuel types: gasoline, diesel, or alternative fuel vehicles;
- season of the year and weather conditions; and
- vehicle speed and operating mode; vehicles emit more particles when accelerating and when traveling at high speed (Hall & Dickens, 1999).

Near-Roadway Concentrations and Population Exposure

Although the emissions inventory (see Section 2) indicates that on-road vehicles account for a relatively modest share of overall primary PM2.5 emissions in the Bay Area, anyone who drives in traffic, walks or cycles on urban streets, or lives in close proximity to a busy roadway incurs significant exposure to PM. Exposure to roadway emissions has emerged as an important social equity issue because major roadways, especially those that carry a high volume of heavy-duty diesel-powered trucks, often run through or in close proximity to low-income and minority communities.

Studies show that ambient concentrations of ultrafine and fine PM are generally much higher than average near major roadways, especially in the downwind direction. Numerous studies have found increased incidence of respiratory and cardiovascular disease among people who live in close proximity to heavily-traveled roadways. The good news is that PM concentrations, exposure, and health effects all tend to decrease rapidly with greater distance from the roadway.

Population exposure to roadway emissions depends upon the amount and the chemical composition of emissions generated by the vehicles on a given roadway; meteorological conditions, such as wind speed and prevailing wind direction; the number of people in proximity to the roadway, their distance from the roadway, and their demographic characteristics; the type of land uses and buildings near the roadway; and the presence of buildings, trees, sound walls or other barriers that affect air pollution dispersion patterns.

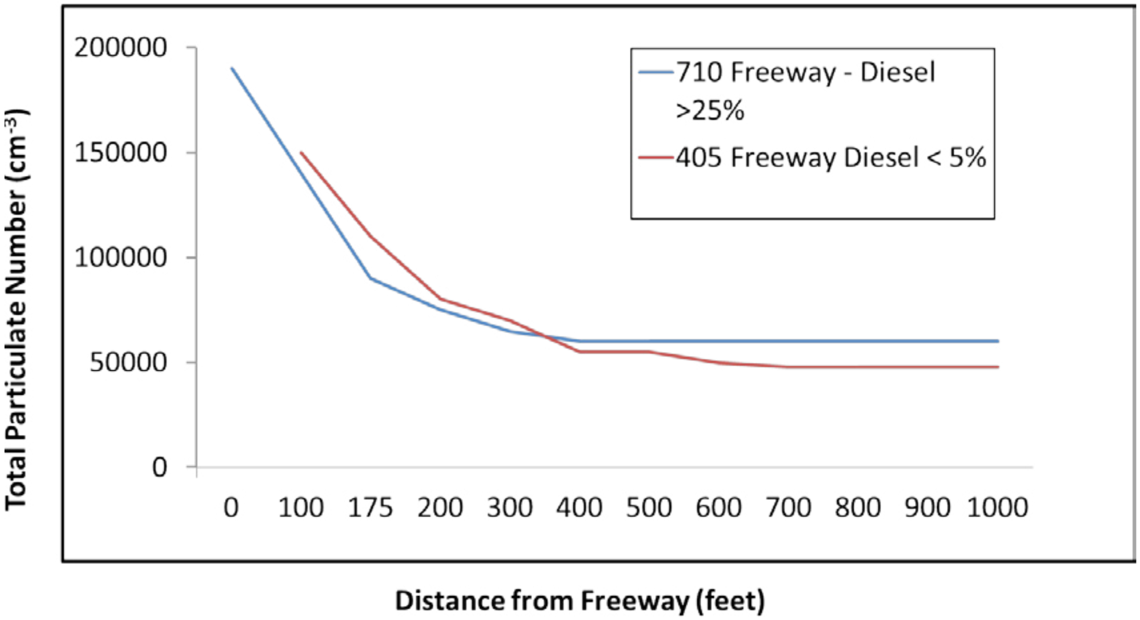
Exposure to PM from Busy Roadways

Key findings from studies on population exposure to PM emissions from roadways are summarized here:

- Numerous studies have found increased rates of respiratory and cardiovascular disease among people who live in close proximity to busy roadways.
- In close proximity to high-volume roadways, ambient concentrations of PM may be greatly elevated compared to background levels, especially in the downwind direction.
- When vehicle emissions are trapped in enclosed areas, such as urban street canyons and tunnels, this can lead to much higher pollution concentrations and population exposure on a local basis.
- In-vehicle exposure may be a leading source of exposure to PM and other air pollutants for people who drive on freeways or major arterials on a regular basis. Exposure rates may be 5 to 10 times higher than average when driving on busy roadways. Driving on a freeway or busy arterial road for even a modest time or distance can account for a significant portion of total daily exposure to ultrafine particles.
- Both PM2.5 mass and ultrafine particle number decrease as distance from the roadway increases. The concentration of ultrafine particles drops off rapidly within the first 50 to 100 meters from the source, and generally reverts to background levels 100-300 meters of the roadway.
- How quickly the particle concentration declines as a function of distance from the roadway depends upon the specific mix of particles emitted by the vehicles, as well as the atmospheric conditions and wind speed and direction in the area of the roadway.

A number of studies have analyzed how PM concentrations change in relation to distance from roadways. For example, a study in Southern California (Zhu et al., 2002) found that the concentration of ultrafine particles decreased rapidly within approximately 300 feet (~ 100 meters) of the I-710 and I-405 freeways, as shown in Figure 1-5. Although the 710 freeway had much more diesel traffic (more than 25% of the vehicles) than the 405 freeway (less than 5% of the vehicles), the results were similar for both freeways.

Figure 1-5 Number of Particles Versus Distance from Roadway: Two Freeways in Southern California (Zhu, 2002)



The chemical composition and size distribution of freshly-emitted ultrafine and fine PM are transformed during transport to downwind locations by the processes of condensation, evaporation, and dilution. Although PM2.5 mass and ultrafine particle number both decrease with distance from a roadway, the number of ultrafine particles generally drops off much more rapidly than PM2.5 mass. This is due to the fact that ultrafine particles coagulate very rapidly to form larger particles upon exposure to ambient air; coagulation reduces particle number, but does not significantly affect overall PM2.5 particle mass.

Another way to analyze how roadway emissions affect population exposure to PM is to consider **intake fraction**; i.e., the percentage of emitted PM that is actually inhaled by a human receptor. A study by the EPA-funded Harvard Center for Ambient Particle Health Effects performed dispersion modeling of primary PM2.5 emissions for 23,000 road segments in the Boston area. For each segment modeled, the study estimated how much of the intake fraction occurs among people in close proximity to the roadway. The study found that although the intake fraction varied considerably among the different segments, on average 46% of the total population exposure occurs within 200 meters of the road segment. These findings support the notion that in a dense urban environment, such as Boston or many parts of the San Francisco Bay Area, a considerable portion of the overall population exposure to roadway emissions occurs in close proximity to the roadway. However, the findings also suggest that even though emissions from a roadway become diluted and disperse rapidly within a few hundred meters of the road, a significant share of the total population exposure to primary PM2.5 emissions from a roadway still occurs at a distance greater than 200 meters from the roadway.

The spatial distribution and extent of roadway emissions may vary based upon temporal factors, such as time of day and season of the year. A study (Zhu et al. 2006) which compared ultrafine particle numbers for daytime and nighttime conditions near a major freeway (I-405) in Los Angeles found that the rate of decrease in ultrafine particles downwind of the freeway was much less at night than during the day. Although traffic volume on I-405 at night was only 25% of the daytime volume, the particle count 30 meters downwind of the freeway was about 80% of the daytime value. The authors attribute the higher ratio of particles to traffic volume at night to a combination of lower wind speed and weaker atmospheric dilution, as well as cooler temperatures which cause increased particle formation in the vehicle exhaust. The study also found that particle counts near the freeway were higher in winter than in summer, for similar reasons to the factors that lead to higher particle counts at night.

Dispersion is key to reducing ambient concentrations and exposure to PM. However, it is important to note that some urban environments, such as tunnels and “urban street canyons”, are not conducive to dispersion of air pollutants. When emissions are trapped in enclosed areas, this can lead to much higher local concentrations, and thus much higher population exposure. One study (Morwaska et al. 2008) found that ultrafine particle numbers in the near-roadway environment were roughly 18 times higher than in a non-urban background environment, while measured concentrations in street canyons and tunnels were 27 and 64 times higher, respectively, than background. Another study (Zhou et al. 2008) found that, due to high population density, combined with the lack of dispersion, the intake fraction of emissions in urban street canyons is very high, similar in magnitude to the intake fraction associated with indoor tobacco smoke.

In-Vehicle Exposure

Concerns about elevated exposure to PM near major roadways also apply to drivers and passengers traveling in vehicles on high-volume roads. In fact, the evidence suggests that in-vehicle exposure may be a leading source of exposure to PM and other air pollutants for people who drive on freeways or major arterials on a regular basis. In-vehicle exposure depends on the volume and mix of vehicles on a given road, as well as the type of ventilation system used in the vehicle. Moving vehicles typically have high air exchange rates, allowing emissions from the stream of traffic to penetrate into vehicles. One study (Fruin et al. 2008) found that 36% of total daily exposure to ultrafine particles occurred during a daily commute of 1.5 hours round trip (6% of the day) in Los Angeles, and that 22% of total exposure occurred during 0.5 hours (just 2% of the day) that was spend on freeways. This indicates that exposure rates may be 5 to 10 times higher than average when driving on busy roadways. Thus, even limited time on a freeway can account for a significant portion of total daily exposure to ultrafine particles.

Freeways are also where people are most likely to experience higher exposure to diesel PM, which has been classified by the Air Resources Board as a toxic air contaminant. The 2008 Fruin study found that on freeways in Los Angeles, concentrations of ultrafine PM, black carbon, nitric oxide, and polycyclic aromatic hydrocarbons (PAH) bound to small particles are generated primarily by diesel-powered vehicles, even though diesel vehicles account for only a small fraction (6%) of the traffic on LA freeways. This study also found, however, that on arterial roads concentrations of ultrafine particles appear to be emitted primarily by gasoline-powered vehicle undergoing hard accelerations. Concentrations of ultrafine particles on arterials were roughly one-third those of freeways.

“Self-pollution”, which occurs when the exhaust from a vehicle infiltrates its own passenger cabin, may also contribute to in-vehicle exposure. This has raised concern about risks to children who ride to school on diesel-powered buses. One study (Adar et al. 2008) found that PM2.5 on school buses was double the on-road levels, and that 35% of PM2.5 measured in school buses came from self-pollution. (See description of the Lower-Emission School Bus Program in Section 4 regarding actions to address this issue.)

Aircraft and Airports

Studies conducted by the South Coast AQMD suggest that jet aircraft may be major emitters of ultrafine particles. Typical ultrafine particle concentrations are on the order of 50,000-200,000 particles per cm^3 near freeways; by contrast, ultrafine particle concentrations near jet exhaust can reach 6,000,000 particles per cm^3. As shown in Table 1-3.

Table 1-3 Comparison of Ultrafine Particle Concentrations[4]

Environment	Ultrafine Particle Concentration
Clean background	500 – 2,000 particles per cubic centimeter
Typical urban air	5,000 – 30,000 particles per cc
Freeway	50,000 – 200,000 particles per cc
Jet exhaust	Up to 6,000,000 particles per cc

A study (Hu et al. 2009) that measured ultrafine particles near the Santa Monica Airport, at the residence closest to the airport, and at a nearby school showed correlations of ultrafine particle concentrations from jet exhaust at all three locations. Aircraft operations resulted in average ultrafine particle concentrations elevated by a factor of 10 at 100 meters downwind and by a factor of 2.5 at 660 meters downwind. In fact, the area impacted by elevated UFPM concentrations was found to extend beyond 660m downwind and 250m perpendicular to the wind on the downwind side of the

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[4] Presentation by Dr. Philip Fine of South Coast AQMD to BAAQMD Advisory Council Meeting: Ultrafine Particles 2012 Atmospheric Monitoring of Ultrafine Particles, February 2012.

Santa Monica Airport. This study demonstrated that there may be health implications for residences living in proximity to jetports, especially in the downwind direction.

A study by Carnegie Mellon University researchers (Miracolo et al. 2011) evaluated the effects of photo-oxidation on ultrafine PM emissions from a gas turbine engine designed to mimic a jet aircraft engine. The study found that photo-oxidation created substantial secondary PM, suggesting that it is also important to consider potential secondary PM formation when assessing the influence of jet aircraft emissions.

Back-up Generators

Back-up generators (BUGs), also known as stationary engines and emergency generators, are used frequently by hospitals, office buildings, schools, grocery stories, and government facilities to supply power to a building during a power failure. While power failures are generally rare, BUGs are operated several times a year for testing. Diesel BUGs emit diesel particulate matter and other toxic air contaminants and may contribute significantly to people's exposure to toxics and health risks. In addition, BUGs tend to be concentrated in populated areas, where high numbers of people may be already exposed to high levels of pollution.

A new BUG installed today in the Bay Area poses little health risk during its operating testing hours due to the Air District's and ARB's regulations. However, old BUGs that were installed prior to regulations and continue to be in use today generate high levels of toxics and pose a serious health risk challenge. Even though these BUGs may be used as little as 100-50 hours a year, they can emit enormous amounts of diesel PM since their engines do not comply with any emission standards or contain retrofit technologies. In the Air District's general screening of health risks for BUGs in the Bay Area, the cancer risk for grandfathered BUGs ranges from 20 to 200 in a million in some cases. There are close to 3,000 BUGs in the Bay Area, approximately 1,500 of which may have cancer risks over 10 in a million. The majority of these BUGs are located in Bay Area urban centers. These BUGs contribute heavily to health risks already experienced by people living near roadways and other mobile emissions of diesel PM. The Air District's general health risk screening for stationary sources indicates that addressing emissions from grandfathered back-up generators could significantly reduce exposure to diesel PM, especially in urban areas with already high exposure rates.

Indoor Exposure to PM

Studies have found that most people experience a major portion of their total PM exposure when they are indoors. This is not surprising, since people spend the majority of their time indoors, in the home, office, school, stores, restaurants, etc. According to one study (Qing Yu Meng et al. 2005), adults typically spent 87% of their time indoors, 7% in vehicles, and just 6% outside. The PM that we breathe indoors is a combination of ambient (outdoor) PM that penetrates to the indoor environment, as well as PM emissions produced by indoor sources.

Most people experience a major portion of their exposure to PM when they are indoors.

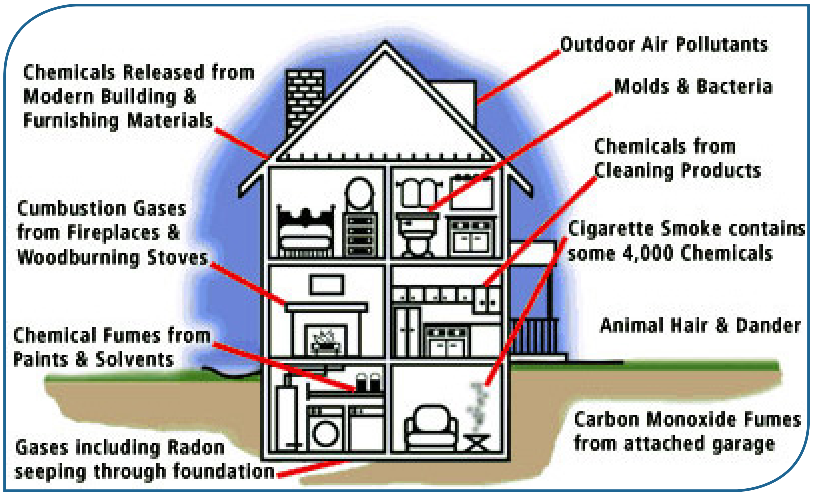
Studies to date to measure indoor PM levels and population exposure have generally been limited to small numbers of sites, because gaining access to suitable sites (private homes, schools, etc.), installing monitors, and analyzing data requires substantial time and resources. Analyzing indoor concentrations and exposures in multi-unit buildings, such as apartment buildings, is complicated by the fact that PM created indoors can move between units, as well as the fact that heating and ventilation systems, if not properly designed and maintained, can transfer pollutants between units. Nonetheless, the findings of existing studies suggest that indoor exposure to PM is a serious issue that merits more attention.

Factors that determine indoor exposure to PM include (1) the ambient (outdoor) PM concentration in the vicinity of the building, 2) the **infiltration rate**: i.e., how much of the ambient outdoor PM penetrates indoors, 3) the **air exchange rate**: how quickly indoor air is replaced by outdoor air, and (4) the amount of primary PM emissions and PM precursors produced in the indoor environment from sources such as cooking, wood-burning, and cigarette smoking. These factors can vary considerably depending upon building type and location, the type of heating and ventilation system, and meteorological conditions.

The infiltration rate of ambient (outdoor) PM to the indoor environment depends upon building materials, characteristics, and design, such as the type of ventilation system, the location of air intake units, whether windows are open or closed, and whether a building has air conditioning or an air filtration system. The PM infiltration rate also varies upon the size and composition of the particles present in the ambient PM. Because different sizes and types of particles have different infiltration rates, the composition of PM in the indoor environment generally differs from the ambient outdoor PM. Ammonium nitrate levels, for example, are generally higher outside than indoors. Ammonium nitrate can exist in either particle or gaseous form in the atmosphere, depending upon temperature. In colder weather, ammonium nitrate particles account for a sizable portion of total ambient PM2.5 in the Bay Area. However, when they encounter warmer air in the indoor environment, ammonium nitrate particles generally volatize (convert to the gaseous form), such that they no longer exist in particle form.

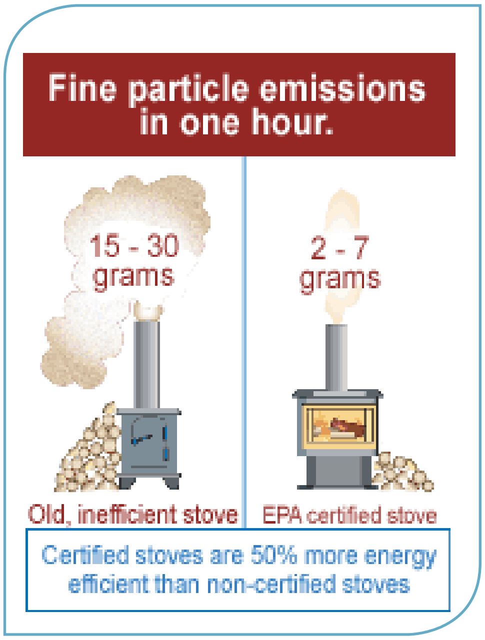
Ultrafine particles are less likely to penetrate through a building envelope because they deposit more rapidly on building surfaces due to Brownian motion at the molecular level. Whereas typical infiltration factors for PM10 and PM2.5 are in the range of 50%, (Ott et al. 2000), infiltration factors for ultrafine particles are on the order of 30% (Wallace & Howard-Reed, 2002). Since ultrafine particles do not easily penetrate to the indoors, this suggests that indoor sources of ultrafine particles play an important role in determining total personal exposure to UFPM.

Sources of PM in the Home Environment



Although PM in outdoor air does penetrate to the indoor environment, particles generated within the home often account for a substantial share of indoor PM levels and exposure. Indoor sources of PM include fireplaces and wood stoves, cooking, gas stoves, cleaning products, cigarette smoking, candles, and incense, laser printers, as well as human activities that may re-suspend PM2.5. Indoor PM may also include a mixture of dander from pets, other types of allergens, chemical substances, mineral particulate, mold spores, viruses and bacteria. The RIOPA study (Polidari, A. et al. 2006) found that fine organic particles dominates indoor-generated PM2.5 in the homes that were studied. Indoor sources of PM can cause PM levels to spike, especially because the emissions are often retained within a confined area. Several of the key sources of PM generated in the home environment are briefly described below.

Cooking: Studies have found that cooking is a leading source of ultrafine particles in many homes. Indoor monitors show that ultrafine particle counts spike whenever cooking occurs. Studies suggest that emissions of UFPM are higher from natural gas stoves than from electric stoves, but the particle emission rates are high in both cases. Ultrafine particle levels tend to be significantly higher in homes with gas stoves that use a pilot light (compared to pilot-less stoves). Emission rates when the oven is in use may be greater than for stove-top cooking. One study found that the indoor concentration of ultrafine particles jumped from 5,000 particles per cubic centimeter to 1 million particles – a 200-fold increase – within a few minutes after the oven in a residential kitchen was turned on. [5]



Wood-burning devices: People are exposed to wood smoke in both indoor and outdoor environments. In addition to its negative impact on outdoor air quality, residential wood-burning can be a major source of indoor PM, especially if the chimney or stovepipe does not vent smoke to the outdoors effectively. This problem occurs most commonly when a fire is first ignited and the fireplace flue is not warmed up, thus failing to draw smoke efficiently. One

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[5] Presentation by Susanne Hering, Ph. D., of Aerosol Dynamics to BAAQMD Advisory Council on March 9, 2011.

study (Pierson et al. 1989) found that 70% of smoke from chimneys can reenter the home where it originated and/or neighboring dwellings.

Appliances: Common household appliances, such as clothes dryers, toaster ovens, irons, and laser printers can also produce ultrafine particles, especially appliances that operate by heating metal surfaces.

Cleaning products: Household cleaning products can also produce ultrafine and fine particles in the indoor environment. Scented cleaning products contain terpenes such as pinene (pine scent) and limonene (citrus scent); these terpenes can react with ozone to form ultrafine particles.

Contribution of Indoor Exposure to Total PM Exposure

Lance Wallace and Wayne Ott have done pioneering work using portable particle counters to measure personal exposure to ultrafine particles. In one of their recent studies (Wallace & Ott, 2010) using personal monitors to measure exposures in environments such as homes, cars, and restaurants, they estimated that, on average, 47% of daily personal exposure to ultrafine particles for the participants in the study can be attributed to indoor sources, 36% to outdoor sources, and 17% to in-vehicle exposure. Consistent with the SHEDS-PM estimates for PM2.5 described below, cooking and cigarette smoking were the dominant sources of indoor emission of UFPM. In households with one or more smokers, the cigarette smoke more than doubled the exposure from all other sources. By measuring the particle count per cubic centimeter (cm^3) and multiplying this by the size of the impacted indoor area, this study estimates that smoking a single cigarette emits approximately 2 trillion (2×10^{12}) ultra-fine particles.

Lynn Hildebrand at Stanford University and William Nazaroff at UC Berkeley have also done important research to advance our understanding of exposure to PM in various micro-environments. A recent study directed by Professor Nazaroff (Bhangar at al. 2011) monitored ultrafine particle concentrations and exposures in seven residences (with non-smoking inhabitants) in urban and suburban Alameda County. This study provides several findings of interest:

- Ultrafine particle concentrations in the home environment are heavily impacted by episodic **indoor source events** that cause sharp spikes in particle counts. These events are triggered by activities such as cooking on the stove; uses of appliances such as toaster ovens, steam irons, or clothes dryers; burning candles; and use of the furnace.
- Frequency of use of the cooking range (either gas or electric) is the single most important determinant of exposure from episodic indoor sources.
- Gas stoves with pilot lights are a key source of indoor emissions and exposures to ultrafine particles.
- Indoor particle counts are much higher when occupants are at home and active (thus generating particles via indoor source events), compared to when they are away from home, or at home but asleep.

- Emissions from indoor sources of ultrafine particles accounted for roughly 60% of the indoor particles; the remaining 40% represent particles that infiltrated from outdoor air.
- Active particle removal systems can reduce indoor particle levels (of both particles generated indoors, as well as particles that infiltrate from outdoors) by a factor of 2 to 4.

The papers cited above analyzed personal exposure to PM at the individual level. Efforts have also been made to estimate the major sources of aggregate population exposure to PM in various urban areas. Many of these studies have employed the *Stochastic Human Exposure & Dosage Simulation for PM* (SHEDS-PM) model developed by the US EPA National Exposure Research Laboratory. Synthesizing data from many sources, including personal activity logs, ambient PM2.5 concentrations for outdoor air, and results from studies of indoor PM, the SHEDS-PM model has been used to estimate the contribution of outdoor exposure and indoor exposure to total population exposure, and to examine the role of key indoor sources of PM2.5 such as cigarette smoking and cooking.

An analysis (Burke et al. 2001) using SHEDS-PM for Philadelphia found that, on average, ambient (outdoor) PM2.5 accounted for only 37.5% of total exposure; however, this percentage varied greatly within the population. The study found relatively low variation in personal exposure to ambient (outdoor) PM2.5; however, exposure to PM in the indoor environment varied greatly, with high levels of indoor exposure caused primarily by emissions from cigarette smoking and/or cooking. Another study (Cao & Frey, 2011) had similar findings, using SHEDS-PM to analyze and compare PM exposures in three different areas and climate zones (New York City; Harris County, Texas; and six counties along the I-40 corridor in North Carolina). This study found that ambient exposure accounted for approximately 40% of the estimated total daily average PM2.5 exposure in each of the three areas. As in the case of the Burke study of Philadelphia, the Cao study also found that some individuals have extremely high PM exposures, primarily due to indoor emissions from cigarette smoking and/or cooking.

The *Relationship of Indoor, Outdoor and Personal Air* (RIOPA) study (Polidari et al. 2006) investigated residential indoor, outdoor and personal exposures to PM2.5 in three cities with different climates: Houston, TX; Los Angeles, CA; and Elizabeth, NJ. The study found that the median contribution of ambient (outdoor) sources to indoor PM2.5 concentrations was 56% for all study homes (63%, 52% and 33% for California, New Jersey and Texas study homes, respectively).

Exposure to PM in Schools

Another recent study directed by Professor Nazaroff (Mullen et al. 2011) measured PM concentrations in six elementary school classrooms in Alameda County; measurements were performed for a total of 18 days (from 2-4 days in each classroom). None of the schools was in close proximity to a major freeway; distance from the nearest freeway was 0.5 km or greater in all cases. Two of the classrooms were equipped with mechanical ventilation systems; the other four used natural ventilation (windows and doors that open). The study found that (1) indoor particle counts were typically about half of the outdoor concentrations, and (2) roughly 90% of the ultrafine particles measured in the classrooms originated outdoors. The authors compared exposure in the classrooms to exposure in the homes (per Bhangar 2011), noting that the results suggest that elementary school students are subject to much greater overall exposure to ultrafine particles in the home environment, because in-home particle counts are higher and because the students spend more time at home than at school. The authors attribute the difference in concentrations to the fact that fewer ultrafine particles are emitted in classrooms than in homes. In particular, indoor source events, such as cooking, that lead to sharp spikes in UF particle levels, are common in the home, but much less prevalent in the school setting.

Summary of Indoor Population Exposure to PM

Key findings regarding indoor exposure to PM can be summarized as follows:

- Ambient contribution to indoor PM exposure depends on outdoor concentrations in combination with the infiltration rate.
- When indoor sources are present, indoor PM concentrations can be substantially higher than outdoor PM concentrations.
- Indoor PM emissions are generated primarily by specific activities and sources: cooking, cleaning, ironing clothes, burning candles, use of forced-air furnaces, fireplaces, etc.
- PM levels in the home are characterized by sharp spikes triggered by the types of activities mentioned above.
- Ventilation to control PM spikes can greatly reduce indoor concentrations and population exposure.
- PM concentrations in the home are generally much lower at night (when people are sleeping, and PM-generating activities are not occurring) than when people are at home and active.

Occupational Exposure

Exposure to PM and other pollutants on the job is regulated by the Occupational Safety & Health Administration (OSHA). Occupational exposure to PM may differ from ambient exposure in terms of particle type and composition, as well as the intensity, frequency, and duration of exposure. Certain job types may expose workers to significant occupational exposures. For example, truck drivers and other people who drive a lot on the job may be exposed to higher levels of PM from both diesel and gasoline vehicles. Restaurant workers may be exposed to PM from cooking and wood smoke from charbroilers. Construction workers and quarry workers may be exposed to diesel PM, as well as to geologic dust particles from mechanical processes. Firefighters, especially those who combat wildfires, may be subject to extremely high acute exposures to PM. Janitorial workers may be exposed to high levels of PM in the indoor environment when they use cleaning products that contain chemicals which react with ambient ozone to form PM. Researchers (Morawska et al. 2007) have founds that people who work in office buildings may be exposed to PM (as well as VOCs) from printers.

Brigham and Woman's Health Hospital conducted a study (Laden et al. 2007) of mortality patterns associated with job-specific exposure to fine particulate and especially particulate matter from vehicle exhaust. They examined rates of cause-specific mortality and compared this to the general population. This study concluded that in the U.S. trucking industry there was an excess of mortality due to lung cancer and heart disease particularly among drivers.

Summary

Population exposure to PM is heavily dependent on individual activity patterns and the types of PM emissions sources that people are exposed to in the course of their day-to-day activities. PM levels, and population exposure to PM, may be greatly elevated in certain micro-environments, such as in-vehicle, near-roadway, and in the home.

The key to avoiding negative health impacts from PM is to reduce population exposure to PM among Bay Area residents. Recognizing the importance of reducing population exposure to air pollutants, the Air District has been working to identify areas that are disproportionately impacted and implementing policies and programs to protect these communities, as described in Section 4.

But to better protect public health, we need to improve our understanding of population exposure to PM in the Bay Area. Future steps to enhance our understanding of population exposure to PM are discussed in Section 5.

Simple steps that Bay Area residents can take to reduce their exposure to PM in the course of their day-to-day activities are also described in Section 5.

SECTION 1-C: PM AND CLIMATE CHANGE

This section describes the complex interplay between particulate matter and climate change, including how PM affects climate, as well as how higher temperatures due to climate change may impact local PM levels.

Although more work is needed to fully discern the connections, research reveals a two-way relationship in which air pollutants impact the climate at both the local and global scale, while changes in climate impact air quality. Most discussion has focused on the need to reduce emissions of carbon dioxide and other greenhouse gases, but researchers have found that particulate matter also affects the climate, especially the type of PM known as black carbon.

How PM Affects Climate Change

The thin atmosphere that surrounds the Earth enables our planet to support life and the complex ecosystems that sustain us. There is irrefutable scientific evidence that the Earth's atmosphere is getting hotter, and that a wide range of human activities, such as combustion of fossil fuels, emit carbon dioxide (CO2) and other **greenhouse gases** (GHG) that are building up in the atmosphere and changing the climate at the global scale. The effects of this man-made global heating are already being experienced in California and on a global basis in terms of temperature trends, extreme weather events (e.g., drought, frequency and intensity of hurricanes and cyclones), sea-level rise, changes in precipitation patterns, the frequency and intensity of wildfires, changes in habitat for flora and fauna, etc.

Certain types of PM, especially black carbon, can have a potent effect in heating the climate.

Efforts to date to protect the climate have focused primarily on reducing man-made emissions of GHGs that trap solar radiation (heat) that would normally escape back into space. Reducing emissions of CO2 has been the main focus of climate protection efforts to date, because on a mass basis emissions of CO2 dwarf the other GHGs, and because CO2 remains in the atmosphere for a very long time.

However, in recent years researchers have discovered that other short-lived air pollutants, including particulate matter and tropospheric ozone, also affect the climate. Although the effects are complex, there is evidence that certain types of particulate matter, especially **black carbon**, can have a potent effect in heating the climate at both the local scale (in the area where PM is emitted) and the global scale. In response to this research, there is a growing recognition that we need to incorporate strategies to reduce emissions of black carbon into climate protections efforts. Reducing black carbon can help to slow the rate of atmospheric heating in the near-term, while also protecting air quality and public health. Emission control opportunities that provide co-benefits in terms of protecting both air quality and the climate are highly desirable from the policy perspective.

Climate Forcing

Climate change is primarily caused by man-made activities that impact the Earth's energy balance (Denman et al. IPCC, 2007). Energy constantly flows to the Earth in the form of sunlight and other forms of solar radiation. Some of this solar energy is reflected back into space, and the rest is absorbed by the planet and stored in the atmosphere, as well as in oceans, forests, etc. Factors external to the natural energy system – so-called **external forcings** - can disturb the Earth's energy balance. These external forcings can be positive or negative. Positive forcers, such as carbon dioxide, methane, and other greenhouse gases, cause more of the sun's energy to be retained by the planet. In contrast, negative forcers, such as volcanic dust that reflects sunlight back into space, cause less of the sun's energy to be retained by the planet. The overall impact of human activities on the climate depends upon the net sum of positive and negative forcings caused by a wide spectrum of man-made activities, including emissions of GHGs and other air pollutants, agriculture and forestry practices, land development and road-paving that affect the reflectivity (albedo) of the Earth's surface.

Climate Forcing Effects of Particulate Matter (PM)

Particulate matter is composed of solid or liquid particles that are suspended in the air; these particles are sometimes referred to as **atmospheric aerosols.** Fine particles affect the climate by means of several direct and indirect processes, some of which heat, and others of which cool, the climate. All PM in the atmosphere can affect the Earth's climate either by absorbing light or by scattering light. Particles that absorb sunlight add energy to the earth's system; they act as positive forcers that lead to climate heating. Particles that scatter light increase the reflection of incoming sunlight back to space; they serve as negative forcers that cool the climate. In addition to the direct effect caused by absorbing or scattering incoming sunlight, fine particles may also have indirect effects on the climate by altering the properties of clouds in various ways. [6] More analysis is needed to fully define the impacts of particles on clouds, but researchers have noted various different processes by which aerosols can affect the reflectivity and lifespan of clouds, in ways that can have both heating and cooling effects, as further describe below. (The 2007 IPCC report discusses five processes; Jacobson 2002 lists 12 processes.)

For purposes of analyzing the impacts of PM on climate, scientists have identified several types of carbon: black carbon, brown carbon, and organic carbon. The effect of primary (directly-emitted) PM on sunlight spans a continuum from light-absorbing to light-scattering, with **black carbon** at the light-absorbing end of the spectrum, most **organic carbon** at the opposite, light-scattering end of the spectrum, and **brown carbon** (a subset of organic carbon) somewhere in the middle. The ratio of black carbon, brown carbon and organic carbon produced by fuel combustion depends upon the specific fuel being burned and the type of combustion conditions. PM emitted by diesel engines is primarily black carbon, whereas the PM emitted by gasoline engines is mostly organic carbon.

Table 1-4 lists the most significant types of anthropogenic (man-made) aerosol particles in terms of impact on the climate, and their most common sources. At the global scale, the dominant negative

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[6] "Atmospheric Aerosol Properties and Climate Impacts" U.S. Climate Change Science Program Synthesis and Assessment Product 2.3; January 2009.

forcing agent is sulfate,[7] whereas the dominant particle as a positive forcing agent is black carbon. But organic carbon, brown carbon, and ammonium nitrate also affect the climate in various ways that can have both heating and cooling effects. In the Bay Area, ammonium nitrate levels are greater than sulfates.

Table 1-4 Climate-Forcing Properties of PM Components

Negative Forcer (Cooling Agent)	Positive Forcer (Heating Agent)	Direct Effect	Indirect Effect	Source
Sulfates				
X		Reflects sunlight	Increases reflectivity of clouds	Secondary PM formed by SO2 emissions from fossil fuel-burning
Ammonium nitrate				
X		Reflects sunlight	Increase reflectivity of clouds	Secondary PM formed by combination of NOx and ammonia emissions.
Black carbon				
	X	Absorbs sunlight	1) Reduces reflectivity of clouds; impacts cloud formation. 2) Heats snow & ice by reducing their reflectivity in polar regions.	Incomplete combustion of fossil fuels, biofuels, and biomass (wood-burning)
Brown carbon				
	X	Absorbs some wavelengths of sunlight		Incomplete combustion of fossil fuels, biofuels, and biomass (wood-burning)
Organic carbon				
?		Mildly absorbs sunlight		Incomplete combustion of fossil fuels, biofuels, and biomass (wood-burning)

The various particle types are never emitted into the atmosphere in isolation. The emissions produced by a given combustion process or event contain a mixture of black carbon, brown carbon,

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[7] Text from NASA Fact Sheet: "While a large fraction of human-made aerosols come in the form of smoke from burning tropical forests, the major component comes in the form of sulfate aerosols created by the burning of coal and oil. The concentration of human-made sulfate aerosols in the atmosphere has grown rapidly since the start of the industrial revolution. At current production levels, human-made sulfate aerosols are thought to outweigh the naturally produced sulfate aerosols."
http://www.nasa.gov/centers/langley/news/factsheets/Aerosols.html

and organic carbon, as well as other co-pollutants including nitrates and sulfates, and various air toxics. The climate effects from a given emissions source will depend on the mix and ratio of the PM components and other co-pollutants, and the way these pollutants interact in the atmosphere. Therefore, unlike greenhouse gases, the climate effects from aerosol particles can vary regionally.

PM as Negative Forcer (Cooling Agent): Sulfates & Ammonium Nitrate

As discussed in Section 2, sulfates and ammonium nitrate are the two leading forms of secondary PM, formed by interaction of precursor chemicals such as SOx, NOx, and ammonia (NH3). Sulfate particles reflect sunlight, thereby acting as a negative forcer (cooling agent) on the climate. Sulfate aerosols also have an indirect cooling effect on the climate by increasing the reflectivity of clouds. Sulfate particles serve as nuclei for the condensation of water vapor; higher rates of condensation increase the brightness of clouds and thus their reflectivity. Sulfates and other aerosols may also lengthen cloud lifetimes. The overall impact of sulfate and ammonium nitrate particles is to increase the Earth's atmospheric **albedo**, or reflectivity. This causes less sunlight to reach the Earth and thus has a cooling effect on the climate. The cooling effects of sulfates are somewhat regional in nature, that is, the cooling effects seem to be concentrated near areas where the emissions occur, such as areas of industrial activity.

Due to the atmospheric presence of sulfate and nitrate particles which function as cooling agents, the evidence suggests that the climate has experienced less heating in recent decades than would have otherwise occurred. However, since these particles are harmful to human health, we need to further reduce sulfate and nitrate levels. As we reduce sulfate and nitrate levels to protect public health, this will lend increasing urgency to the need to reduce emissions of the greenhouse gases, as well as particles such as black carbon that contribute to climate heating.

PM as Positive Forcer (Heating Agent): Black Carbon & Brown Carbon

Several types of particles act as heating agents. Black carbon, often referred to as "soot", is a solid particle formed of mostly pure carbon that absorbs solar radiation (light) at all wavelengths. Black carbon has been identified as a potent climate heating agent. In fact, black carbon can absorb a million times more energy than carbon dioxide per unit of mass. Black carbon is black in color because it is highly efficient in absorbing all the wavelengths of light in the visible spectrum. The vast majority of black carbon is man-made. Black carbon is produced by incomplete combustion of fossil fuels, biofuels and biomass (wood-burning).

Combustion of fossil fuels (gasoline, diesel, coal, and natural gas) and biomass (wood and vegetation) are the major sources of black carbon on a global basis. In general, fossil fuel combustion from diesel engines, energy production and industrial processes accounts for most black carbon in developed countries, with the major contribution coming from diesel engines. Approximately 75% of the PM in diesel exhaust is black carbon. By contrast, biomass burning produces most of the black carbon in developing countries. In developing countries, burning of biomass occurs both to clear land and for domestic uses such as cooking and home-heating.

A recent analysis by Air District staff, based on the Bay Area emissions inventory and chemical mass balance (CMB) analysis of PM captured on filters, indicates that fossil fuel combustion accounts for approximately 65% of black carbon emissions (diesel engines accounts for 50%; other fossil fuels 15%) in the Bay Area. Wood smoke from all sources accounts for 35% of black carbon emissions in the Bay Area, most of which (25%) is produced by residential wood-burning.

CO2 and other GHGs heat the atmosphere primarily by retaining heat from the outgoing infrared radiation (produced when sunlight is reflected off the Earth's surface) that would otherwise escape the atmosphere. By contrast, the impact of black carbon on the climate is more complex. It acts on the climate through multiple mechanisms while suspended as a particle in the atmosphere, and also when deposited on snow and ice. The impact of the black carbon heating mechanisms described below can be greater if other pollutants in the air adhere to the black carbon, thus making the particles bigger and their heat absorption greater.

Direct effect: Black carbon absorbs both incoming and outgoing radiation of all wavelengths (whereas GHGs only absorb outgoing radiation in the infrared range). When sunlight hits black carbon in the atmosphere, the carbon particle absorbs that solar radiation and heats the atmosphere. Black carbon also has a heating impact when it absorbs solar radiation reflected by the Earth and clouds, thus reducing the amount of heat that would otherwise radiate back into space.

Snow/ice albedo effect: When black carbon falls on to snow or ice, it darkens their surface. This decreases the reflectivity (albedo) of the snow or ice, so that more sunlight is absorbed, thus accelerating the melting of ice caps and glaciers. On a global basis, the albedo effect of black carbon on ice and snow accounts for about 25% of the total heating effect of black carbon (Hansen & Nazarenko, 2004). Nonetheless, the melting of ice caps and glaciers, which reduces and alters habitat for key arctic species such as polar bears, is one of the most dramatic manifestations of climate change. In addition, the impact of black carbon in polar regions is of special concern, because it may lead to abrupt transitions or "tipping points".

Two examples of potential tipping point phenomena include the melting of ice in the Arctic Sea, as well as the release from thawing permafrost of vast quantities of CO2 and methane which could further accelerate the process of global heating. While the most dramatic manifestations of the snow/ice albedo effects may occur in polar regions, it has implications for California and the western U.S. as well. For example, one study (Hadley et al. 2010) has found deposition of black carbon on snowpack in the Sierra Nevada and Rocky mountains contributes to faster melting of the snowpack earlier in the spring, thus reducing the amount of snowmelt that would normally occur later in the spring and summer. This may have impacts on water supply in the western U.S. by reducing the supply that can be captured for human use. Given the importance of the Sierra snowpack to water supplies in California, this could be one of the most important effects of black carbon emissions within the state.


Grinnell Glacier 2005.jpg courtesy of the U.S. Geological Survey

Indirect and semi-direct effects: Black carbon also alters the properties of clouds, affecting cloud reflectivity, precipitation, and the surface dimming caused by cloud cover. Clouds permeated with black carbon reflect less sunlight back into space, thus causing more heat to be absorbed in the atmosphere. Because these effects are so complex, estimating the indirect heating effects of black carbon and other particles in terms of cloud formation and albedo and precipitation patterns is one of the greatest challenges for modelers who study climate change.

Net effect of black carbon: Although black carbon has a complex mix of both cooling and heating effects, the evidence suggests that its net climate impact is positive forcing with significant climate heating potential. In fact, recent studies indicate that black carbon may be the second biggest contributor to global heating, after carbon dioxide (Jacobson 2010). However, because of the complexities related to analyzing the climate impacts of black carbon, most of the global-scale climate change models currently in use only consider black carbon in a simplified way by addressing a subset of its various forcing mechanisms.

Short-Term vs. Long-Term Impacts

Black carbon typically remains suspended in the atmosphere for a relatively short time, on the order of 10-12 days. This is a very brief timespan compared with greenhouse gases such as methane, which typically remains in the atmosphere for approximately 10-15 years, or carbon dioxide (CO2), which stays in the atmosphere for decades or even up to hundreds of years. Because its atmospheric timespan is brief, this means that reducing emissions of black carbon in the near-term will provide immediate climate cooling benefits. This could mitigate, in the short-term, the heating that continues to occur in the absence of effective national and international policies to reduce the on-going increase in emissions of longer-lived GHG emissions such as CO2 and methane. In other words, efforts to reduce black carbon can yield immediate cooling benefits, thus buying us time to address the longer-term solutions needed to reduce longer-lived GHG emissions.

Geographic Scale of Impacts

Whereas the climate impacts of the traditional greenhouse gases are global in scale, the evidence suggests that the climate impact of black carbon is more localized in nature. Certain regions of the world are more likely to be impacted by black carbon heating effects, either due to transport and deposition, such as polar regions, or to high levels of PM emissions in the region, such as Asia. This has several implications:

- Efforts to reduce black carbon can be targeted on the sources and locations where the heating effect of black carbon is most damaging. One of the great unknowns in terms of global heating is where and when climate change may

> go beyond irreversible tipping points that could trigger disastrous impacts, such as greatly accelerated melting of the Greenland ice sheet or thawing permafrost. Well-targeted efforts to reduce black carbon emissions in the most sensitive regions may provide a means to avert or delay such tipping point scenarios.

Reductions in black carbon at the local level will provide direct local benefits, both in terms of reducing heating in the specific region where the black carbon reductions occur, as well as reducing the serious health impacts related to exposure to PM and black carbon.

Brown Carbon

The term "brown carbon" refers to organic carbon compounds that absorb visible and/or ultraviolet light, and thus heat the atmosphere. Like black carbon, it is a product of incomplete combustion. Brown carbon compounds are chemically diverse, so that the wavelengths of light they absorb, and thus their color, vary. The mixture of colors of brown carbon compounds appears brown to the human eye. The total quantity of solar energy absorbed by a brown carbon mixture depends upon the molecular structure of the compounds and the total mass of material. The net contribution of brown carbon to climate change is presently uncertain; this represents a key gap in our understanding of the net impact of PM on climate.

Net impact of PM on the Climate

Assessing the net impact of PM on the climate system is challenging. It requires analyzing the mix of the various particle types in the atmosphere in a given region, and then evaluating the different heating and/or cooling properties (both direct effects and the diverse indirect effects) for each particle type. And because fine particles are generally short-lived in the atmosphere, it is difficult to measure them on a global scale.

Despite the uncertainties, the available evidence suggests that, even though black carbon and brown carbon have a heating effect, fine particles as a whole currently have a net cooling effect on the climate.[8] In fact, a recent study (Sriver 2011) led by the National Oceanic and Atmospheric Administration found that a rapid build-up of aerosols (fine particles) in the stratosphere over the past decade has offset about one-third of the climate heating influence of CO2 during this period. The NOAA study concludes that the amount of aerosols in the stratosphere will play an important part in determining the overall change in climate in coming decades. As noted above, it is important to reduce emissions of fine particles to protect public health, but doing so may exacerbate the challenge we face in attempting to control the climate heating impacts of greenhouse gases and black carbon.

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[8] Stratospheric Pollution Helps Slow Global Warming," David Biello, Scientific American, July 22, 2011; www.scientificamerican.com/article.cfm?id=stratospheric-pollution-helps-slow-global-warming.
Quote from this article: "By analyzing satellite data and other measures, Daniel and his colleagues found that such aerosols have been on the rise in Earth's atmosphere in the past decade, nearly doubling in concentration. That concentration has reflected roughly 0.1 watts per meter squared of sunlight away from the planet, enough to offset roughly one-third of the 0.28 watts per meter squared of extra heat trapped by rising atmospheric concentrations of greenhouse gases such as carbon dioxide. The researchers calculate that the aerosols prevented 0.07 degrees Celsius of warming in average temperatures since 2000."

Air District staff has performed a preliminary analysis to examine how decreasing PM levels in the Bay Area may be affecting the local climate. Staff looked at pyranometer[9] readings for three sites with data stretching back to 1990: Bethel Island, Santa Rosa and San Martin. The data suggest that insolation (the amount of solar radiation hitting the earth's surface) for these sites has increased on the order of 2 Watts/m^2 per decade, or about 1% per decade. This finding, though preliminary, is consistent with the results from one paper (Wild et al. 2008) which found that surface net radiation over land rapidly increased by about 2 W/m2 per decade on a global basis for the 15-year period 1986–2000.

An April 2012 study (Leibensperger et al.) by the Harvard School of Engineering & Applied Sciences also sheds light on the cooling effect of sulfate particles and other aerosols. The Harvard study found that in the later part of the 20th century particulate pollution created a "cold patch" over the eastern United States where the effects of global warming were temporarily obscured. In addition to directly scattering incoming sunlight, the particles also helped form clouds that further reflected sunlight, thus indirectly leading to greater cooling at the earth's surface. The study found that as a result of efforts to reduce sulfates and other particles in recent years to protect public health, this "cold patch" effect has now been largely removed. In the words of the authors, "What we've shown is that particulate pollution over the eastern United States has delayed the warming that we would expect to see from increasing greenhouse gases. For the sake of protecting human health and reducing acid rain, we've now cut the emissions that lead to particulate pollution, but these cuts have caused the greenhouse warming in this region to ramp up to match the global trend. No one is suggesting that we should stop improving air quality, but it's important to understand the consequences. Clearing the air could lead to regional warming."

In analyzing the effects of PM on climate, one of the key technical issues is how to compare the climate forcing effects of fine particles with the effects of carbon dioxide and other greenhouse gases. A metric called **Global Warming Potential** (GWP) is generally used to compare the heating potential of various greenhouse gases in comparison to carbon dioxide. Although a range of values has been published in the literature, there is as yet no consensus as to the appropriate GWP value for black carbon or for fine PM as a whole; this is due to the complex combination of heating and cooling effects described above, as well as the fact that the impacts of PM on climate may vary from region to region depending upon the specific sources and composition of PM in a given area.

In comparing the impact of different climate forcing agents, it is important to consider the different atmospheric lifetimes of the various pollutants that impact the climate. For example, CO2 and methane remain in the atmosphere for many years; therefore, the benefit of reducing these gases in cooling the atmosphere will be spread over many years. By contrast, black carbon and other

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[9] A pyranometer measures solar irradiance, that is, the amount of solar energy hitting a flat horizontal surface. It works by using a black-coated flat disk called a thermopile: The black surface absorbs the solar energy and converts it to heat. The thermopile converts this thermal energy to electrical energy.

fine particles remain in the atmosphere for just a few days or weeks; therefore, the cooling effect from reducing black carbon is experienced immediately. So for purposes of policy decisions with a short-term time horizon, it may be appropriate to give greater weight to black carbon, since reducing black carbon (1) provides immediate climate protection benefits by helping to offset the rise in temperatures caused by the continued build-up of greenhouse gases in the atmosphere, and (2) provides localized health benefits by reducing PM concentrations and population exposure to PM.

Impacts of PM on Precipitation Patterns and Storms

In addition to the impacts of fine particles on climate summarized above, recent studies also describe potential impacts of PM on cloud formation and precipitation patterns. A study (Zhanqing Li et al. 2011) that analyzed rainfall patterns and aerosol level over a 10-year period in the southern Great Plains of the U.S. found that aerosol pollution will suppress cloud formation and reduce precipitation in relatively dry environments. Conversely, aerosols are likely to increase cloud formation and rainfall in the summertime in areas with an existing moist environment, thus worsen flooding. The authors conclude that "These findings have important implications for the redistribution, availability, and usability of water resources in different regions of the world."

Impacts of Climate Change on PM Levels

Meteorology plays a critical role in determining air pollution levels. Climate change may impact future PM levels by affecting key meteorological variables such as surface temperature, relative humidity, precipitation rate and patterns, wind speed, and mixing height (vertical mixing). Atmospheric mixing is important to disperse PM and prevent it from building up in the air, so any climate effects that would reduce horizontal or vertical mixing could lead to higher ambient concentrations of PM. In considering potential impacts of climate change on future PM levels, one of the main concerns is to determine whether there will be a "climate penalty" – i.e., whether climate change will increase ambient PM levels. Any such climate penalty would either lead to higher PM levels, or require additional controls, beyond those already enacted or anticipated, in order to achieve PM standards.

Researchers are still attempting to evaluate the potential impacts of climate change on future PM levels. There are many factors that introduce uncertainty into this exercise, including uncertainty about the degree of future change in the climate and future emissions of PM and its precursors. Key findings from a June 2010 report to ARB prepared by University of California scientists entitled *Climate Change Impact on Air Quality in California* include the following:

- The impact of climate heating on PM levels is difficult to pin down because some of the likely effects act in opposite directions. For example, higher temperatures discourage the formation of ammonium nitrate, a component of secondary PM that constitutes a sizable fraction of Bay Area PM2.5. However, as an offsetting effect, higher temperatures will also lead to increased background concentrations of ozone, which encourages the formation of ammonium nitrate.

- Impacts of climate change on PM levels may vary by region, but, overall, climate change is expected to have only a small effect on PM levels and population exposure to PM in California's major air basins on an annual average basis.
- Climate change is likely to increase average wind speeds in coastal regions of California; this may lead to lower concentrations of primary PM, especially in coastal regions, such as the Bay Area.
- However, climate change may cause PM levels to be higher during extreme pollution events in the future, because future stagnation events which trap pollutants close to the emissions source will increase in strength.

Increased Wildfires

Climate change may cause an increase in the frequency and severity of wildfires by altering snowmelt and precipitation patterns. At least one study (Westerling, et al. 2006) has found an association between climate change and increased wildfires in forests in the western US. Wildfires can emit huge quantities of fine particles such as black carbon, as well as other air pollutants, such as carbon monoxide, NOx, and air toxics. Most of the particles from wildfires are in the very fine size range, the types of particles that can most effectively penetrate deep into the lungs. Wildfires can cause dramatic short-term spikes in pollution levels, and greatly increase population exposure to PM and other harmful pollutants. The outbreak of wildfires that swept across California in late June 2008 caused ambient concentrations of ozone and PM to soar to unprecedented levels.[10] A recent study (Wegesser et al. 2009) found that the PM concentrations not only reached high levels, but that the PM released by these June 2008 fires was much more toxic than the PM more typically present in the California atmosphere. Smoke from wildfires can cause a variety of acute health effects, including irritation of the eyes and the respiratory tract, reduced lung function, bronchitis, exacerbation of asthma, and premature death.

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[10] During the final week of June 2008, PM2.5 levels increased five or ten-fold compared to normal readings at several Bay Area monitoring stations.

In addition to these health effects, wildfires also release immense quantities of carbon dioxide stored in trees and vegetation into the atmosphere. Therefore, to the extent that climate change increases wildfires, this will increase emissions and atmospheric concentrations of black carbon and GHGs that contribute to climate change, in an unwelcome feedback loop.

Policy Considerations

The evidence suggests that there is a compelling rationale to reduce black carbon emissions for several reasons.

- Reducing black carbon will provide immediate climate protection benefits by (1) helping to offset temperature increases related to greenhouse gases, and (2) potentially averting or delaying the onset of "tipping point" scenarios, such as collapse of arctic ice cap or the sudden release of carbon stored in permafrost, which could have profound and irreversible impacts at the global level. Since emissions of greenhouse gases, especially CO2, continue to increase at the national and global scale, and efforts to reverse this trend have made little headway to date, reducing black carbon may buy society time to implement the more fundamental changes required to stem the rise of greenhouse gas emissions.
- In addition to protecting the climate, reducing black carbon will provide important public health benefits by reducing population exposure to PM2.5 and diesel PM, both of which have been shown to cause a range of negative health effects.
- Reducing black carbon will provide these health and climate benefits at the local scale; i.e. in the region where the emissions reductions occur.

The good news is that, thanks to efforts over the past 10-15 years to reduce emissions of fine PM and diesel PM in California and the Bay Area (described in Section 4) in order to protect public health, we have already made major progress in reducing ambient levels of fine PM and black carbon. Analysis of elemental carbon, which is closely associated with black carbon, indicate that Bay Area levels decreased 73% from 1989-91 through 2008-10. [11] Reductions in ambient concentrations of black carbon in California and the Bay Area have presumably helped to reduce the amount of climate heating caused by man-made emissions of GHGs and other climate forcers at both the local and global scale. Looking forward, black carbon emissions will be further reduced in response to adopted regulations, such as ARB's heavy-duty diesel engine regulations, that will be implemented over the next 10-15 years. The other side of the coin, however, is that because we have a "head start" on reducing diesel PM and black carbon in California, much of the "low-hanging fruit" has already been picked. Therefore, measures to reduce the remaining increment of fine PM and black carbon may entail higher cost or effort per unit of emissions reduced. However, because reducing black carbon provides benefits in protecting both public health and the climate,

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[11] See pages 36-37 in *Trends in Bay Area Ambient Particulates*, BAAQMD, November 2011. www.baaqmd.gov/Divisions/Planning-and-Research/Research-and-Modeling/Publications/Reports.aspx

it should still be possible to craft additional black carbon control measures or incentive programs that reduce black carbon at a favorable benefit/cost ratio.

Trade-offs

Because PM includes a variety of particle types with differing effects on climate, it is important to identify and mitigate potential trade-offs in developing control measures to reduce PM. As noted above, reducing emissions of particles that act as cooling agents (e.g., sulfates and ammonium nitrate) should protect public health, but may entail a trade-off in terms of protecting the climate. In another example of trade-offs, a recent study (Anenberg et al. 2011) that analyzed the global-scale health benefits of reducing black carbon noted that reducing black carbon could lead to a modest increase in sulfate (SO4) concentrations. Because sulfate particles scatter incoming sunlight, an increase in sulfates could provide additional benefit in terms of cooling the atmosphere. However, an increase in sulfates could have a negative impact on public health, depending on how this would affect PM2.5 levels and local population exposure to PM. As this example suggests, identifying and evaluating potential trade-offs, and determining the net benefit of potential control strategies, can be a complex exercise. The Air District will closely track ongoing research on these issues to inform policy decisions.

Next Steps

Although the impact of PM and aerosols on the global climate is complex, some type of particles such as black carbon contribute to climate heating. This suggests that efforts to reduce PM should place high priority on reducing emissions of those particles, such as black carbon, that both damage public health and contribute to climate heating. Potential policies and actions to address the climate impacts of PM are discussed in Section 5.

SECTION 1-D: PM IMPACTS ON ECOSYSTEMS & VISIBILITY

This section describes impacts of PM on ecosystems, visibility, and the built environment.

In addition to directly impacting public health and the climate, particulate matter also can have negative effects on water quality and on the ecosystems and environment that sustain us. These effects, as summarized in Table 1-5, include acid rain which leads to acidification of lakes and streams, changes in the nutrient balance of coastal waters and river basins, leaching of nutrients from soil and reduced nutrient uptake in plants, damage to forests and crops, reduced diversity and productivity of ecosystems, damage to stone and man-made materials, and reduced visibility and aesthetic values. According to the US EPA, the scientific evidence is sufficient to conclude that a causal relationship is likely to exist between deposition of PM and a variety of effects on individual ecosystems.

It is important to note that the varying chemical compositions found in PM concentrations affect different aspects of the ecosystem. As discussed in Section 2, PM is a complex pollutant made up of a number of compounds and originates from a variety of sources and processes. As such, specific chemicals and components of PM are linked to specific ecosystem affects. For example, elemental carbon and some crustal minerals are the most commonly occurring airborne particle components that absorb light. Reduced visibility is caused by light absorption (as well as light scattering). In another example, toxic responses in plants and foliage have been documented when exposed to PM concentrations containing acids, trace metal content, or saline compounds. Also, different chemical compounds can negatively affect segments of the environment to varying degrees. One significant trace metal component of PM is mercury, which is toxic and can move readily through ecosystems and food chains. Mercury emitted from a smoke stack of a factory, for instance, may settle into soil, then be transported from soils into a nearby water body, accumulate in the bodies of plankton and other smaller fish who are then consumed by larger predators such as animals and humans, thereby transferring the mercury content all the way up the food chain.

In summary, the overall impact of PM on the environment depends upon many factors, including the chemical composition of the particles, meteorology, season of the year, and the characteristics and viability of the impacted ecosystem.

Table 1-5: Summary of Negative Impacts of PM on Ecosystems, Visibility, and Materials

Effects of PM on:	Effects:	Economic/Social Ramifications:
Vegetation	Damaged leaf tissue; stunted growth; reduced starch storage capacity.	Reduced crop yields; less robust growth of vegetation that humans, animals, and insects depend upon.
Soils	Altered soil chemistry.	Plants absorb metals and other toxic elements, which are then transferred to animals & people that eat the plants. Can lead to an increase in invasive plant species. Polluted soil can run off into stream/water bodies.
Water & Aquatic Systems	Altered chemistry; increased acidity; increased mortality in fish, decreased stamina in fish.	Decline in fish populations; bio-accumulation effect which impacts plants, fish, and mammals from the bottom of the food chain to the top
Wildlife	Lung damage; neurological damage; chromosome damage.	Stress on wildlife populations. Bio-accumulation of pollutants up the entire food chain.
Visibility	Reduced visibility; aesthetic degradation.	Reduction in tourism & economic benefits from tourism. Reduced ability to enjoy scenic and/or historical vistas.
Materials	Damage and degradation of building materials and finishes.	Increased maintenance costs; degradation of historic structures.
Property Values	Reduction in property values.	Economic disbenefit to homeowners & landlords. Reduction in local government tax revenue.

Effects of PM on Vegetation and Soils

Airborne particles are deposited to a variety of surfaces, including soil and open ground; forests, crops, and other vegetation; water bodies such as lakes, streams, and oceans; and man-made surfaces such as buildings, roads, and parking lots. Particles are deposited via two processes: **wet deposition** (rain or snow) and **dry deposition**. Coarse particles typically are deposited by means of dry deposition, and generally settle close to the location where they were emitted. Conversely, fine particles are typically deposited via wet deposition and may travel farther from the point of origin.

PM can affect plant life through direct deposition on surfaces, or indirectly through altered soil chemistry. When directly deposited onto vegetation, PM can affect the metabolism and photosynthesis of plants by blocking light, obstructing stomata apertures, increasing their temperature, and altering pigment and mineral content. In general, the toxic responses documented in plants after exposure to high levels of PM are typically associated with acidity, trace metal content, or salinity of the deposited particles. Fine PM has been shown to enter the leaf through the stomata, penetrate the structure of the leaf, and alter its chemistry. Coarse PM can form a "crust" on the leaf, which reduces photosynthesis, damages the leaf tissues, inhibits new growth of the tissue, and reduces starch storage.

The soil environment is one of the most dynamic sites of biological interaction in nature. Deposition of PM on soil can have a negative effect not just on the chemical composition of the soil itself, but also upon the plants which grow in it, the animals and people who eat those plants, and even nearby groundwater systems which run through the impacted soils. This phenomenon, whereby a substance (such as trace metal) moves up the food chain and becomes more concentrated during ascension, is also known as biomagnification. In general, plant growth is negatively impacted by the presence of trace elements and heavy metals in soils which can then enter the plant tissue. As the plants absorb heavy metals and other pollutants via PM deposition into the soil, this can have a biomagnification effect and negatively impact the health of the people or animals that eat them.

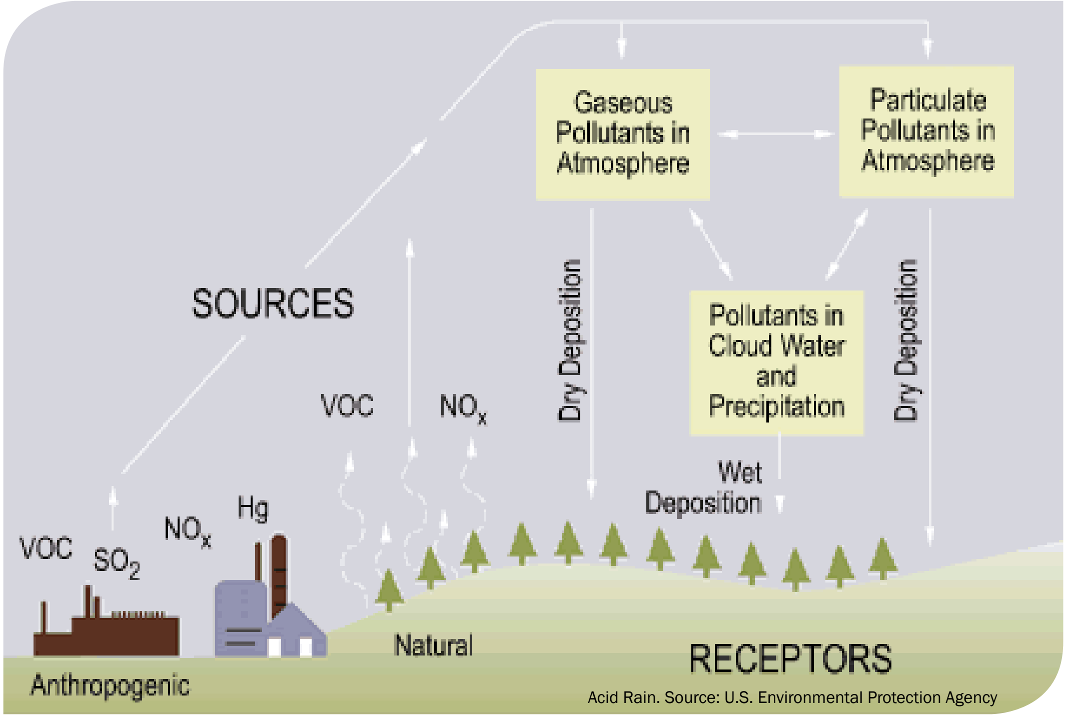
PM may have another potential impact on soil, flora, and fauna, to the extent that the ammonium nitrate component of PM acts as a source of reactive nitrogen. Deposition of reactive nitrogen on land acts as an unintended fertilizer which can have impacts on terrestrial flora and fauna. A growing body of literature documents the impacts of nitrogen deposition on ecosystems in western states. Studies demonstrate that increased nitrogen deposition is negatively affecting native plant communities which are adapted to live in low-nitrogen environments. These changes have enhanced invasion of exotic plant species such as annual grasses. Of the 225 plant species in California listed as threatened or endangered by the state or federal government, 101 are exposed to levels of nitrogen suspected of causing ecological disruption (Weiss, CEC 2006). In areas where reactive nitrogen is deposited on nutrient-poor soil, this can facilitate the expansion of invasive, non-native species that choke out native plants. As the flora changes, animal species that depend on the native vegetation may be adversely impacted.

The case of the Bay Checkerspot Butterfly, which has been on the federal endangered species list since 1987, provides an example of the impact of reactive nitrogen on diversity of native flora and fauna. The Checkerspot depends on native grasses that grow on nutrient-poor serpentine soils. The serpentine ecosystem provides food for both the larval and adult stages of the butterfly. Edgewood Natural Preserve in San Mateo County historically supported a healthy population of Checkerspots. However, nitrogen deposition from vehicles on Interstate 280, which is adjacent to the Preserve, has allowed aggressive, non-native grasses, such as Italian rye grass, to crowd out native grass species in recent years (Weiss 2002). As a result of habitat reduction, the Checkerspot population at Edgewood is in jeopardy.

PM and Acid Rain

Acid deposition, more commonly known as acid rain, is a widespread problem which effects water quality and ecosystems, and its effects have been well studied. Acid rain is a broad term which refers to a mix of wet and dry deposited particles from the atmosphere which contain high proportions of nitric and sulfuric acids. The precursors of such acids result from both natural and manmade sources. Natural sources include volcanoes and decaying vegetation, and manmade sources include emissions of SO2 and NOx from fossil fuel combustion. Figure 1-6 shows the process whereby natural and man-made emissions combine in the atmosphere to produce acid rain.

Figure 1-6 Formation and Deposition of Acid Rain



Acid Rain. Source: U.S. Environmental Protection Agency

Acid rain has negative effects on soil, water (freshwater and saltwater), aquatic ecosystems, and building materials. Regions where a high percent of ambient PM2.5 is composed of secondary particles such ammonium sulfate and ammonium nitrate (e.g., the eastern US) are more likely to experience greater negative impacts of acid rain. While a majority of the PM2.5 in the Bay Area is attributable to primary PM from wood smoke and fossil fuel combustion, during winter months a large portion of PM2.5, on the order of 35%, is composed of ammonium nitrate. Accordingly, while acid rain is not a serious problem in the western United States, due to the levels of ammonium nitrate in overall PM concentrations in the Bay Area, it is worthwhile to take precautions.

On land, acid rain can damage trees, especially at higher elevations, where exposure to acid-heavy clouds and mist is greater. The ability of a forest to cope with acid rain depends on the buffering capacity of its soil. Acid dissolves and removes the nutrients in forest soils before trees and other plants can use them to grow. At the same time, acid rain causes the release of substances that are toxic to trees and plants, such as aluminum, into the soil.

While acid rain is not a serious problem for water bodies in the Bay Area, because primary PM and PM precursors can travel a considerable distance in the atmosphere before depositing elsewhere, pollution emitted in the Bay Area may impact ecosystems in downwind areas including the Sierra Nevada. According to a National Parks Service report,[12] acid rain and snow is not as serious a problem in the Sierra Nevada as in the eastern U.S. or the Colorado Rockies. However, many high-elevation Sierra lakes have low buffering capacity (ability to cope with acid), so it is important to minimize any future acid deposition.

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[12] See http://www.nature.nps.gov/air/Pubs/pdf/techInfoEpaDeposition.pdf

According to the National Acid Precipitation Assessment Program Report to Congress (2011) and the U.S. Environmental Protection Agency, numerous negative ecosystem effects are attributed to increased acid deposition, including:

- Impaired visibility;
- Acidification of lakes and streams, which has a cascading effect onto fish in terms of reductions in total population, hardiness of the fish, age distribution, and size;
- Reduction in plankton biodiversity (specific to the western US);
- Reduction in acid neutralizing capacity;
- Decrease in pH (increase in acidity level) which can affect the ability of certain plant, insect and aquatic species to survive;
- High levels of nitrates in water which are toxic to aquatic life; and
- Depletion in oxygen levels of the water from accelerated plant life/death.
- Slower growth, injury or death of forests and plant species from altered soil chemistry, and/or damage to leaves or plant organs;
- Increases in atmospheric nitrogen deposition which tends to decrease species diversity (particularly in alpine plant communities); and
- Degrading effect on built structures and monuments, particularly those made of limestone, marble, lime mortars and carbonate-cemented sandstone.

Studies to date have found that the rise in CO2 concentrations in oceans via absorption, which causes decreases in ocean pH and alkalinity, is the major issue regarding ocean acidification. However, acid rain also contributes to ocean acidification. Approximately one-third of all nitrogen oxide emissions end up in the oceans. The contribution of acid rain to ocean acidification is likely greater in coastal regions such as the Bay Area, where the acidifying effect of nitrogen oxides can be as high as 10 to 50 percent of the impact of carbon dioxide (Doney, 2007). Studies have shown that increased acidity interferes with the formation of the shells and skeletons in coral, crabs, marine snails, and clams (World Wildlife Fund 2011).

Effects of PM on Water, Aquatic Systems, and Wildlife

As previously discussed, some components of anthropogenic (manmade) PM such as trace metals have a particularly damaging effect on ecosystems, including mercury, a significant trace metal component of PM that moves readily through ecosystems; as well as pesticides and polyaromatic hydrocarbons (PAHs). Once deposited, these pollutants may travel through the snow pack and feed into the water system. Deposition of PM containing these compounds has been found in the Sierra Nevada mountains in California, the major source of the state’s water supply.

Some environments produce PM concentrations more toxic than others. In urban areas, motor vehicles emit toxic metals and other particles which are deposited on roads and parking lots, from where they are washed into the streams and bays, thus degrading nearby water quality. For example, particles from tire wear are a significant source of zinc, and brake pad wear is a significant source of copper (Stolzenbach 2006). Copper from brake pads is toxic to aquatic organisms such as phytoplankton that serve as the foundation of the food chain, thus affecting the health of entire ecosystems. Elevated copper levels may also be one of the factors contributing to the decline of salmon populations.[13]

Physiological responses of fish to higher pollutant levels include increased mortality rates, chromosomal damage, retarded growth and development, and disruption of normal biological functions, including reduced stamina for swimming and maintaining positions in streams. An increase in concentrations of certain heavy metals such as aluminum, nickel, cadmium, copper, and mercury can poison fish and shellfish, and those who prey upon fish/shellfish.

Deposition of PM on land and water can have a range of negative impacts on ecosystems and wildlife from the bottom of the food chain to the top, due to the process of bio-magnification or bioaccumulation. For example, in addition to impairing the health of fish populations, air pollutants deposited in the aquatic environment can damage the broader ecosystem. To the extent that PM and related air pollutants are deposited in water and then absorbed by fish, frogs, snails and other marine life, these then travel up the food chain, increasing in concentration with each step up the ladder, to fish-eating predators including bald eagles, osprey, otters, pelicans, and grizzly bears.

PM and Visibility (Haze)

Particulate matter is a major cause of reduced visibility, or haze, in both urban and rural areas. Haze is one of the most visible manifestations of air pollution. Reduced visibility is of special concern in areas of great natural beauty such as national and state parks and wilderness areas. In addition to detracting from the aesthetic enjoyment of vistas and landscapes, haze can have negative economic impacts in areas that depend on tourism. The emissions that create haze in parks and wilderness areas often originate elsewhere, sometimes from distant urban areas. For example, 33% of the haze found in the Grand Canyon is attributed to sources of particulate pollution in California.

To address regional haze problems, the US EPA created a Regional Haze Program and issued regulations to improve visibility, particularly in national parks and recreation areas. The original regulations, adopted in 1999, required states to develop plans to address the emissions that contribute to regional haze. In addition, all 50 states were required to submit a regional haze state

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[13] For discussion regarding the impact of copper from brake pads on water bodies, see http://www.suscon.org/bpp/#

implementation plan by December 2007 to demonstrate each state's long-term strategy for making reasonable progress towards achieving natural visibility conditions. In June 2012, US EPA proposed to issue a new urban visibility standard to provide increased protection from particle-induced haze in urban areas. As discussed in Section 3-B, the proposed standard would measure visibility on the basis of light extinction as expressed in units called **deciviews**.

Haze is caused when fine particulates in the air scatter and absorb sunlight. Some light is absorbed by particles, while other light is scattered away before it reaches the observer. More particles lead to greater absorption and scattering of light, reducing visual clarity and color. Some types of particles, such as sulfates, scatter more light. Haze-causing particles come from a variety of both manmade and natural sources, including windblown dust, wildfires, motor vehicles, electric utility and industrial fuel burning, and so on. Some particles which cause haze are produced primarily, while others are produced secondarily.

Visibility is closely tied to wind and weather conditions. Wind affects how pollutants are mixed and dispersed. On very windy days, the air is normally clear because particles are well dispersed. On days when surface winds are present but weaker, particles usually form a plume which causes reduced visibility. When no surface wind is present, haze typically forms near the ground and continues to build as long as the stagnant condition persists. These conditions are most conducive to reduced visibility.

Key sources contributing to the formation of haze include combustion of fossil fuels or biomass burning in electric utilities, manufacturing processes, and transportation. Natural sources of haze include wildfires, volcanoes and wind-blown dust. As a result of regulations to reduce emissions of PM2.5 and PM10, visibility has improved in many US cities and national scenic areas in recent years.

Reduced visibility may also impose monetary costs in terms of reduced property values, and negative impacts on tourism. An analysis performed by the South Coast AQMD for its 2007 Air Quality Management Plan (AQMP) found that visibility improvements related to achieving compliance with federal PM2.5 standards would provide an estimated $3.6 billion benefit per year in terms of increase property values in the South Coast air basin. South Coast staff analyzed the sales price of owner-occupied single family homes between 1980 and 1995 and found that visibility has an impact on property values. Their analysis demonstrated a willingness on the part of home-buyers to pay a premium for visibility. (Final Socioeconomic Report for the 2007 AQMP, June 2007, SCAQMD)

Effects of PM on Materials and Property Values

In addition to negative impacts on soil and water quality and ecosystems, PM and other air pollutants also damage the man-made built environment. Based on the available evidence, US EPA has determined that a causal relationship exists between PM and damage to building materials and other surfaces. Exposure to air pollutants can accelerate the natural wear and tear on buildings from wind, rain, moisture, and temperature changes, further damaging these surfaces. For example, PM deposition on buildings affects the durability of paint finishes and promotes discoloration, chalking, loss of gloss, erosion, and causes blistering and peeling of surface material. This requires costly cleaning or washing, and potentially re-painting, depending upon the soiled surface. The effect of PM deposition on national monuments and other cultural treasures and historical structures is of particular concern. Other effects on PM deposition on buildings and surfaces include:

- Enhanced weathering process on stone in combination with exposure to PM. Black crusts commonly develop from airborne particles deposited on stone surfaces;
- Corrosion of metals and masonry;
- Soiling of motor vehicles and damage to their finish;
- Increased building maintenance and repair costs; and
- Reduced property values.

Additionally, PM contributes to the formation of acid rain, which has a serious effect on structures and monuments, particularly those made of limestone, marble, lime mortars and carbonate-cemented sandstone.

The 2007 AQMP prepared by the South Coast AQMD also quantified the damage to wood and stucco surfaces of residential properties, as well as the cost of household cleaning, from PM2.5 emissions at eight locations in southern California. The total benefit of the decrease in costs for repainting stucco and wood surfaces, cleaning, and replacing damaged materials is projected to be $204 million, on average, every year between 2007 and 2025. Further, this figure is likely to be understated, because it takes into account only residential buildings. As previously discussed, damage to historic buildings or monuments, such as statues, cemetery gravestones, and the like, as well as non-residential buildings, occurs from exposure to PM2.5. Taking into account other building types, especially special and historic ones, is likely to greatly increase the benefit from reduced PM2.5 emissions. (Final Socioeconomic Report for the 2007 AQMP, June 2007, SCAQMD)

SECTION 2: PM TECHNICAL INFORMATION

This chapter provides technical information about particulate matter, including PM size ranges, PM formation and dynamics, the results of PM air quality modeling, PM speciation data as to the contribution of key emission sources to ambient concentrations of PM, and the Bay Area PM emission inventory.

PM Characteristics

The term **particulate matter** (PM) encompasses a diverse assortment of microscopic airborne particles. Many air pollutants, such as ozone or carbon monoxide, consist of a single molecule or compound. PM, by contrast, includes a potpourri of disparate particles that vary greatly in terms of their size and mass, physical state, chemical composition, toxicity, and how they behave and transform in the atmosphere.

A variety of chemical & physical processes are involved in PM formation and transformation. Because PM is so heterogeneous and dynamic, this presents technical challenges in terms of measuring emissions and ambient concentrations, estimating population exposure, determining PM health impacts, assessing PM impacts on ecosystems and climate change, and devising appropriate control strategies.

PM typically consists of a mixture of solid particles as well as liquid droplets known as aerosols. The components of PM include elemental carbon, organic carbon, and trace metals; compounds such as nitrates, organics, and sulfates; and complex mixtures such as diesel exhaust, wood smoke, and geologic dust. Types of particles include:

- **Dusts and fibers** generated by handling, grinding, abrasion or cutting of bulk materials.
- **Mists** composed of liquid droplets generated by condensation from a gaseous state or breaking up of bulk liquid.
- **Smoke** produced by incomplete combustion of carbonaceous materials.
- **Fumes** composed of solid particle generated by condensation of vapors or gases from high temperature processes.
- **Bio-aerosols** composed of solid or liquid particles from biological sources.

Particles emanate from a variety of man-made processes and sources, such as fuel combustion, as well as from natural sources, such as wildfires, volcanos, and sea salt. Particulate matter is generated both indoors and outdoors. Emission sources that affect ambient (outdoor) air are described in the

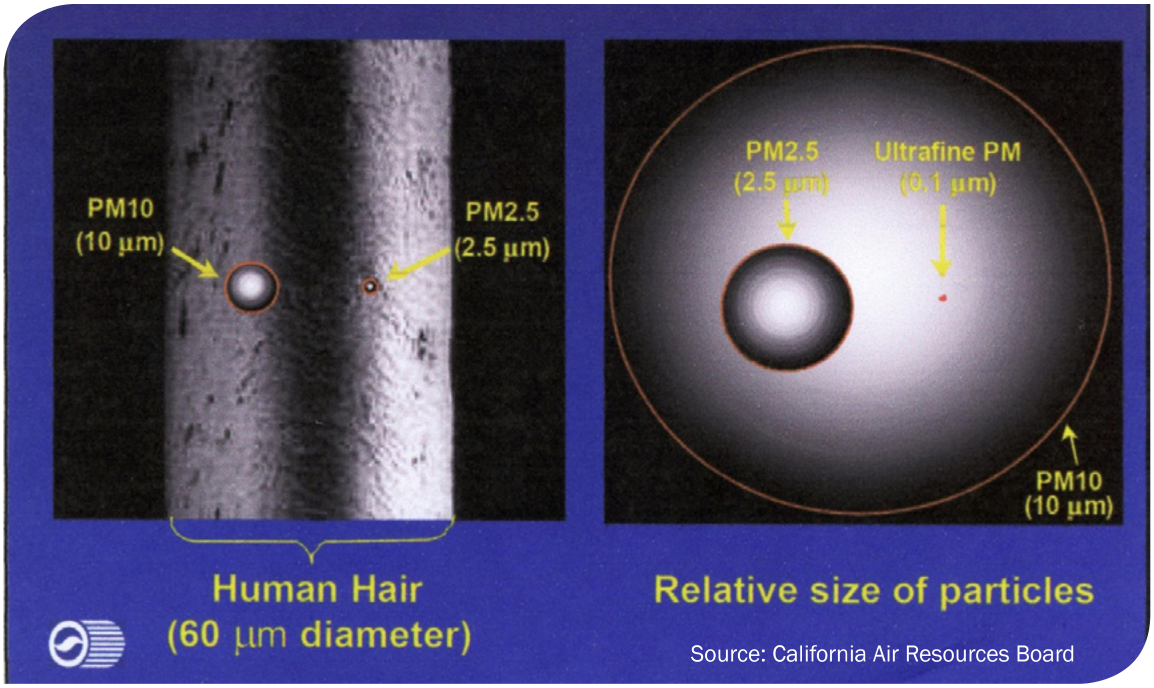
Emissions Inventory section below. Indoor sources of PM emissions, which include cooking, heating, fireplaces, appliances, smoking, and consumer products, are discussed in Section 1-B.

Particle Size

PM is commonly characterized on the basis of particle size. Figure 2-1 shows the various particle sizes in comparison to the diameter of a human hair.

- **Ultrafine PM** (PM0.1) includes the very smallest particles. The term generally refers to particles less than 0.1 micron in diameter (one micron equals one-millionth of a meter, or m x 10-6). Because ultrafine particles are so small, their size is often described in nanometers, or billionths of a meter (m x 10-9). By definition, the largest ultrafine particles measure 0.1 microns or 100 nanometers in diameter, but many ultrafine particles are as small as 3 nanometers to 20 nanometers at the time they are emitted.
- **Fine PM** or PM2.5 consists of particles 2.5 microns or less in diameter (including ultrafine PM).
- **Coarse PM** refers to particles between 2.5 microns and 10 microns in diameter. The term “coarse” particles may be misleading; it should be emphasized that even “coarse” particles are still very tiny, many times smaller than the diameter of a human hair.
- **PM10** consists of particles 10 microns or less in diameter (including ultrafine, fine and coarse PM).
- **Total suspended particles** (TSP) includes particles of all sizes, including particles larger than 10 micron in diameter.

Figure 2-1 Comparison of PM10, PM2.5, and Ultrafine PM



Source: California Air Resources Board

These distinctions based on particle size are important, because the different size ranges vary in terms of how the particles within each size range are formed and emitted; how long they remain suspended in the atmosphere and how far they travel; how easily they can evade the body's defenses and how deeply they can penetrate into the lungs and key organs; and the mechanisms by which the particles are removed from the air. However, although particle size is a useful way to categorize PM, it should be emphasized that the particles within each size range are by no means homogeneous. The diameter and mass of the particles within each of these ranges varies considerably, and there is great variation in terms of the chemical composition of the particles.

In terms of composition, coarse PM is generally dominated by geologic particles of dust and soil (from farms, quarries, mines, volcanos) and other particles of natural origin (sea salts, pollen, mold, spores, etc.). By contrast, fine and ultrafine PM are primarily the product of combustion and therefore contain compounds such as black carbon, sulfates, nitrates, acids, and metals which are more harmful to health.

Table 2-1 provides a summary of PM characteristics by particle size range.

Table 2-1 Characteristics of Particulate Matter by Size Range

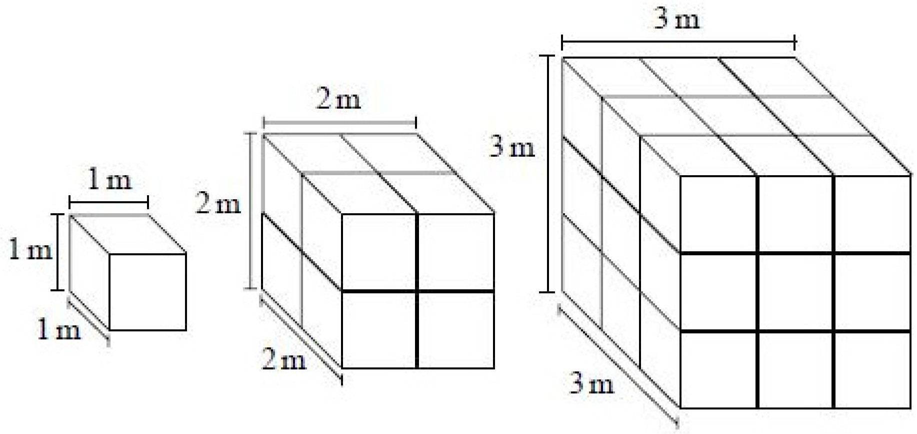
Particle Size	Characteristics		
	Key Attributes	How Measured?	Key Emission Sources
Ultrafine PM: Particles less than 0.1 micron in diameter	Mostly primary PM. Short-lived in atmosphere. Local impacts. Produced by fossil fuel combustion. Particles can be inhaled deep into the body.	Particle count	Motor vehicles, diesel engines.
Fine PM: Particles less than 2.5 microns in diameter	Combination of primary & secondary formation. Produced by fossil fuel combustion & wood-burning. Particles can be inhaled deep into the body.	Mass; expressed in µg/m^3 (micro-grams per cubic meter)	Wood-burning, motor vehicles, off-road engines & equipment, industrial processes & combustion.
Coarse PM: Particles between 2.5 and 10 microns in diameter	Mostly primary PM. Relatively few particles on a number basis, but they account for about half of PM10 on a mass basis.	Mass; expressed in µg/m^3 (micro-grams per cubic meter)	Geologic dust, brake and tire wear, residential wood-burning, motor vehicles.

Relationship between Particle Count, Particle Mass, and Surface Area

The particles that comprise PM vary in both size (diameter) and mass (weight). Larger particles are much heavier than small ones; a single coarse particle may weigh more than thousands of ultrafine particles. Although larger particles account for most of PM on a mass basis, they represent only a small percentage of the total number of particles. Conversely, smaller particles contribute less PM mass, but dominate in terms of the number of particles. Ultrafine particles account for the greatest **number** of particles in PM, but only a small proportion of the total **mass** of PM. Ultrafine particles account for roughly 90% of the total number of particles (Wu et al. 2008). There tends to be little correlation between the number of particles (most of which are in the ultrafine and fine size range) in a given air sample and the mass of PM2.5 or PM10 in that sample. So measuring the mass of PM inthe air generally reveals little as to the number of particles that make up that mass.

As shown in Figure 2-2, as particle size increases, particle mass (volume) increases much faster than the surface area. So a given mass of ultrafine PM will have a much greater particle number and total surface compared to an equal mass of fine PM or coarse PM. The huge number of fine and ultrafine particles suspended in the air collectively presents a great deal of surface area relative to their small mass. Surface area is a concern for two reasons. First, greater surface area means more surface to which microscopic airborne toxics can adhere. Fine and ultrafine particles coated with toxics can penetrate deep into the lungs when they are inhaled. Second, because of their relatively large surface area, once these fine and ultrafine particles enter the respiratory system, they interact with a large area of lung tissue; this means that they can do greater damage to the lungs (or other organs with which they come into contact).

Figure 2-2 **Ratio of surface area to volume**



surface area	$6m^2$	$24m^2$	$54m^2$
volume	$1m^3$	$8m^3$	$27m^3$
surface area: volume	6:1	3:1	2:1

Source: http://cikgurozaini.blogspot.com/2010/09/gas-exchange-in-animals.html

Primary PM versus Secondary PM

In addition to size ranges, PM is also categorized on the basis of how the particles are formed and emitted. **Primary PM** refers to particles that are directly emitted in solid or aerosol form. **Secondary PM** refers to particles that are formed in the atmosphere through chemical reactions among different pollutants.

Primary PM includes black carbon (soot) and fugitive dust from a wide variety of sources, including cars, trucks, buses, industrial facilities, cooking, power plants, construction sites, tilled fields, paved and unpaved roads, rock quarries, and burning wood. Some primary particles are emitted directly from a tailpipe or smokestack in particle form. However, primary PM also includes condensable PM (discussed below) which is formed when organic compounds that are emitted as hot gases condense into particles upon exposure to cooler ambient air. Organic carbon is the largest directly-emitted constituent of Bay Area PM2.5; its main sources are wood-burning, fossil fuel combustion, and cooking.

Secondary PM describes particles formed indirectly via chemical processes when precursor pollutants, such as sulfur oxides (SOx), nitrogen oxides (NOx), volatile organic compounds (VOC), and ammonia (NH_3), react in the presence of sunlight and water vapor. These precursor pollutants are emitted from fuel combustion, industrial processes, household activities, agriculture, natural vegetation, and other sources. Combustion of fossil fuels produces NOx, which converts to nitric acid (NO_3) and combines with ammonia (NH_3) in the atmosphere to form ammonium nitrate, as well as sulfur dioxide (SO_2), which converts to sulfuric acid (H_2SO_4) and combines with ammonia to form ammonium sulfate. In determining whether ammonia is a significant contributor to PM formation, the key question is which pollutant – ammonia, or NOx (in the form of nitric acid) - is the limiting factor in ammonium nitrate formation.

These secondary compounds account for roughly one-third of Bay Area PM2.5 on an annual-average basis and approximately 40-45% during winter peak periods. Ammonium nitrate, which is stable in solid form only during the cooler winter months, contributes an average of about 40% of total PM2.5 under peak PM conditions. The contribution of ammonium sulfate to Bay Area PM2.5 is relatively low, accounting for approximately 10% of total PM2.5 on an annual-average basis.

The distinction between primary PM and secondary PM is important for understanding and analyzing how the various emissions sources contribute to ambient PM concentrations. However, in ambient air where particles are constantly interacting and transforming, most individual particles are actually composed of a mix of primary and secondary PM. An individual primary particle typically has a core of carbonaceous material, often containing trace metals and other toxic materials. Layers of organic and inorganic compounds are then deposited onto the core particle. Depending on the composition of the material deposited on the core particle, the particle may become more toxic as it grows in size.

Physical Processes that Affect PM Formation

In addition to chemical processes in which precursor compounds react to form secondary PM, several physical processes also play an important role in determining how particles interact and transform while suspended in the air. The processes that affect PM vary depending on particle size.

Key physical processes that affect the formation of fine and ultrafine particles at the micro scale include **condensation, nucleation, and coagulation.** These processes occur very rapidly, especially in the initial seconds after a plume of emissions is released by a combustion process. During "plume processing", the hot particles and gases produced by combustion interact vigorously upon exposure to cooler ambient air. As a result of these processes, particle count and particle size distribution can change very rapidly. These physical processes are especially potent among ultrafine particles; because ultrafine particles are so numerous and have a great deal of surface area, they interact and agglomerate more rapidly than other types of PM. Therefore, ultrafine PM is very dynamic and short-lived in the atmosphere.

Condensation and nucleation are related processes. Combustion processes emit a great variety of organic compounds in gaseous form. Upon exposure to cooler ambient air, these hot gases seek to condense. When the hot gases condense by adhering to existing particles, this is called condensation. When the hot gases condense by forming new particles, this is called nucleation. There is competition between condensation and nucleation.[14] If the ambient air already contains an abundance of fine particles, then the hot gases will generally condense on to the existing particles. However, if the supply of existing particles is limited, then in the absence of existing particles on which to condense, the gaseous emissions will nucleate to form new particles, primarily in the ultrafine size range. The number of particles produced as hot gases condense thus depends in large part upon the supply of pre-existing particles in the air.

Because the presence of existing particles promotes condensation (instead of nucleation which forms new particles), this means that not only is PM2.5 mass concentration a poor surrogate for ultrafine particle count, but that PM2.5 mass and ultrafine particle count may actually be negatively correlated. The number of new ultrafine particles produced as a by-product of combustion will generally be low when the existing PM2.5 mass concentration is high, and vice versa. This helps to explain measurements showing that when particulate filters are installed on diesel engines, the mass of PM2.5 emitted by diesel combustion is greatly reduced, but the number of ultrafine particles in the diesel exhaust may actually increase (Van Setten et al. 2001).

Coagulation occurs when two or more existing particles join to form a larger particle. Coagulation is very prevalent among ultrafine particles, but tends to decrease as particles grow to a larger, more stable size. Ultrafine particles produced by combustion coagulate very rapidly to form larger particles upon exposure to ambient air. Coagulation reduces particle number and increases particle size, but does not affect overall PM2.5 particle mass. Due to the processes of coagulation and condensation, the number of ultrafine particles tends to drop off rapidly as distance from the emission source increases, whereas PM2.5 mass is more stable.

PM is removed from the air through processes such as diffusion, coagulation, and deposition. Because ultrafine particles are so small that they are only weakly affected by the force of gravity, they are removed mainly by diffusion, in which their random thermal motion (known as "Brownian motion") causes the particles either to adhere to man-made or natural surfaces or to adhere to other

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[14] For purposes of developing PM emissions inventories, the particles formed by both the condensation and the nucleation processes are referred to as "condensable" emissions.

particles (coagulation). As particles grow larger and heavier via coagulation, they are eventually deposited to the earth's surface by means of gravity through the processes of dry deposition or wet deposition (rain and snow). Fine and ultrafine particles often provide a nucleus that facilitates the condensation of water vapor in the atmosphere, thus forming water droplets; so these particles can also be removed from the air via wet deposition. Although beneficial for purposes of clearing the air, PM deposition may have negative impacts on soil, water, flora and fauna, as discussed in Section 1-D.

Spatial Variation in PM Concentrations (Concentration Relative to Distance from Emission Source)

The ambient concentration of a directly-emitted air pollutant, such as primary PM, generally decreases rapidly via dispersion as distance from the emission source increases. This means that concentrations of primary pollutants will vary considerably on a spatial basis. By contrast, ambient concentrations of pollutants that are formed by means of chemical processes in the atmosphere, such as ozone and secondary PM, are not so directly related to distance from the emission source; these secondary pollutants tend to be more broadly and evenly distributed on a spatial basis.

Since ultrafine PM is composed mainly of primary PM, the number of ultrafine particles typically decreases rapidly as distance from the emission source increases. Fine PM (PM2.5) and coarse PM, on the other hand, include a mix of both primary and secondary particles. So concentrations of primary PM from emission sources such as engine combustion and wood-burning can vary greatly at the local scale, whereas the distribution of secondary PM such as ammonium nitrate and ammonium sulfate tends to be more uniform across a region.

Relationship between PM and Toxic Air Contaminants

Air pollutants are generally regulated either as **criteria air pollutants** or as **toxic air contaminants** (TACs). Criteria pollutants are generally controlled on a regional scale in an effort to attain air quality standards which are based on ambient concentrations in the atmosphere. TACs are generally present in the atmosphere only in very low concentrations. But because of their high toxicity, TACs are regulated at the emissions source so as to limit individual exposure on the basis of risk-based standards; for example, a maximum cancer risk no greater than 10 in one million. Although PM is categorized and regulated as a criteria air pollutant, PM displays some characteristics of a TAC to the extent that it acts as a local air pollutant. Areas of overlap between fine PM and toxic air contaminants (TACs) include the following:

- In the case of both fine PM and TACs, exposure to even small amounts of the pollutant can cause negative health effects;
- PM and TACs share common emissions sources, such as combustion of fossil fuels and biomass;
- Diesel PM has been identified as a TAC by the California Air Resources Board;

- Some air toxics may be emitted in particle form, such as cadmium emitted from combustion of fossil fuels;
- TACs and PM are fellow travelers; air toxics frequently adhere to fine particles and then enter the lungs when these particles are inhaled.

PM Formation & Dynamics in the Bay Area

The basic chemical and physical processes described above that govern PM formation and transformation at the micro level hold true for the Bay Area. However, local meteorology and climate, the specific mix of PM sources and their geographical distribution within the region, and air exchange with neighboring air basins all influence PM formation and dynamics in the Bay Area.

Temporal & Seasonal Variation in Bay Area PM Levels

Ambient PM in the Bay Area varies considerably both in composition and spatial distribution on a day-to-day basis and on a seasonal basis, due to changes in emissions and weather. Changes in meteorological conditions are the most important factor in explaining the day-to-day and seasonal variation in PM concentrations. The Bay Area experiences its highest PM concentrations in the winter; exceedances of the 24-hour national PM2.5 standard almost always occur from November through February. High PM2.5 episodes are typically regional in scale, impacting multiple Bay Area locations. During other seasons, by contrast, Bay Area PM2.5 levels tend to be relatively low, due largely to the region’s natural ventilation system. Thus, on an annual-average basis, PM2.5 levels in the Bay Area are among the lowest measured in major U.S. metropolitan areas.

Meteorological factors are the main reason that the Bay Area experiences its highest PM levels in winter months. However, it is important to note that winter is also the season when the most residential wood-burning occurs; in some parts of the Bay Area, wood smoke accounts for the majority of airborne PM2.5 during high PM episodes. In addition to higher wood smoke emissions, secondary PM2.5 levels are also elevated during the winter months. Cool weather is conducive to the formation of ammonium nitrate which contributes an average of about 40% of total PM2.5 under peak PM conditions.

Studies in Southern California have found that seasonal variation in meteorological conditions also affect emissions and concentrations of ultrafine particle. Similar to PM2.5, ultrafine particle numbers are higher in winter compared to spring and summer. This is likely due to fact that lower temperatures promote particulate formation; condensable organics emitted as hot vapors from tailpipes and other combustion sources quickly cool and condense to form particles. One study (Zhang et al. 2005) found that the ultrafine particle formation rate from vehicle exhaust is higher in winter than in summer because average particle size is smaller in winter (~ 10 nanometers) than in summer (~ 60 nanometers).

While PM emissions and concentrations tend to be highest in winter for the reasons described above, meteorological conditions primarily determine whether the concentrations will build up to levels that exceed the national 24-hour PM2.5 standard. Horizontal mixing (i.e., surface winds) and vertical mixing (which occurs when air temperatures increase with height) are the key to dispersing particulates in the atmosphere, and thus keeping ambient concentrations below the PM standards. Winter is associated with decreased atmospheric mixing height and more stagnant, less windy weather.

Weather & Wind Patterns Conducive to High PM Concentrations

The Central Valley (comprised of the Sacramento and San Joaquin valleys) borders the Bay Area to the east, and there is considerable air exchange between them. In summer, the typical pattern is westerly winds blowing from the Bay Area into the Central Valley. In the winter, during periods of stagnation when PM concentrations are high, the pattern tends to reverse with easterly flow draining from the Central Valley through the Bay Area and into the Pacific Ocean.

Using a technique called **cluster analysis** to identify correlations between weather conditions and PM concentrations in historical data, Air District staff collaborated with UC Davis staff to analyze how meteorological conditions impact Bay Area PM levels during winter months. Weather systems conducive to high PM2.5 levels have shallow flows through the complex terrain that form stagnating cold pools in valleys that trap pollutants. Consecutive stagnant and rainless winter days are typically prerequisites for development of elevated PM2.5 episodes.

A single weather pattern was found to account for approximately 80% of all Bay Area PM2.5 exceedances. This pattern is characterized by a ridge of high pressure moving over the Bay Area during a period of multiple days. This system leads to calm conditions within the Central Valley, coupled with persistent easterly winds from the Central Valley into the Bay Area. The lower levels of solar radiation (sunlight) in the winter lead to stronger temperature inversions; these inversions prevent vertical mixing and are therefore conducive to the buildup of PM in ambient air near ground level.

During this weather pattern, PM levels in the Central Valley can be about 2-3 times higher than in the Bay Area. This is largely because meteorological conditions are more conducive to the transformation of NOx to nitric acid (needed for the formation of ammonium nitrate) in the Central Valley than for coastal locations. Conditions that enhance daytime and nighttime conversion of NOx to nitric acid include low wind speeds combined with abundant sunlight during the day and high humidity at night. Nitric acid then rapidly reacts with ammonia emissions, mostly from dairy activities, which are especially concentrated in the northern San Joaquin Valley, to form ammonium nitrate. The easterly winds that prevail during this pattern transport both primary and secondary PM from the Central Valley into the Bay Area, as further discussed in the modeling section below.

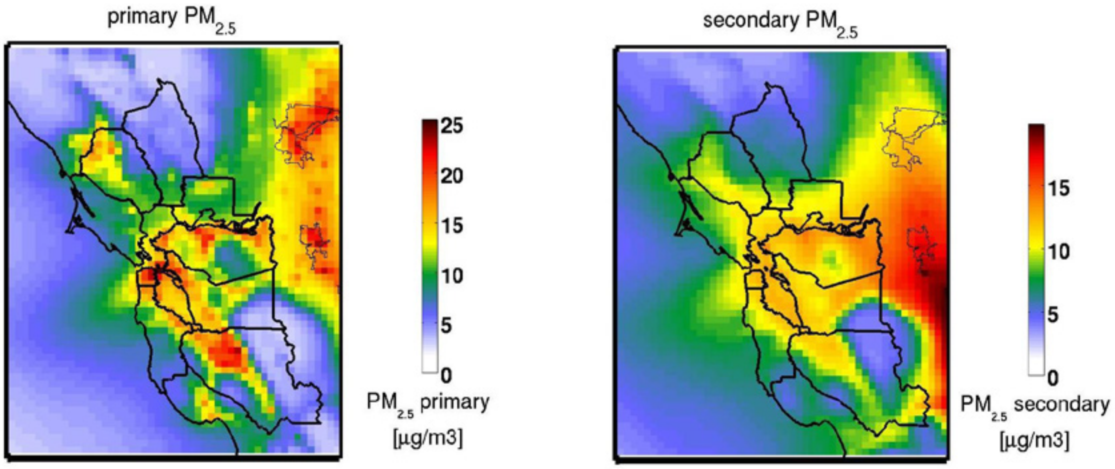
Although the weather pattern described above accounted for over 80 percent of all Bay Area exceedances, it should be noted that only around one in three days belonging to this pattern resulted in an exceedance. Therefore, this weather pattern constitutes a necessary, but not sufficient, condition for an exceedance to occur. Days with this pattern were further analyzed to distinguish the characteristics of days that result in exceedances of the 24-hour PM2.5 standard. Exceedance days could be defined in terms of a number of simultaneous meteorological characteristics: a ridge of high pressure over the Bay Area from strongly stable conditions aloft providing a weak surface pressure gradient over Central California; persistent shallow flows from the east through the Bay Area; winds channeled by terrain; enhanced nocturnal cooling under clear-sky conditions leading to enhanced overnight drainage flows off the Central California slopes; and at least two consecutive days of these listed conditions.

PM Photochemical Modeling Results

The Air District performs photochemical modeling to better understand the complex relationship between emissions, ambient concentrations, and population exposure to air pollutants. Air District staff has employed EPA's Community Multiscale Air Quality (CMAQ) model to simulate PM2.5 formation and dynamics in the Bay Area. PM2.5 simulations were performed with the CMAQ model for four months (December-January, 2000-01 and 2006-07). The modeling domain included the Bay Area and the entire Central Valley to account for the impact of inter-basin transport. The model was applied on 4-km horizontal grids. The results of the PM modeling have been summarized in the October 2009 report entitled *Fine Particulate Matter Data Analysis and Modeling in the Bay Area.*

Figure 2-3 shows the spatial distribution of simulated primary and secondary PM2.5 concentrations around the Bay Area. These results were averaged across the 52 simulated days for which measured Bay Area 24-hour PM2.5 levels exceeded 35 $\mu g/m^3$. For most of these episodic days, light winds flowed through the Bay Area from the east, and Central Valley conditions were near calm. Primary PM2.5 levels were elevated mainly in and around major Bay Area cities, including Oakland, San Francisco and San Jose; near industrial facilities and highways along the Carquinez Strait; at Travis AFB; and Santa Rosa. Secondary PM2.5, present mostly as ammonium nitrate, was not localized near the sources of its precursor emissions, NO_x and ammonia. Rather, secondary PM2.5 was regionally elevated. A sharp gradient existed, with very high secondary PM2.5 levels in the Central Valley decreasing westward through the Bay Area. Around San Francisco and San Jose, PM2.5 levels were dominated by primary (directly-emitted) PM. For other areas affected by PM episodes, such as the eastern, northern, and southern Bay Area and also the Delta, primary and secondary PM2.5 levels were comparable. Both primary and secondary build-up were required for exceedances to occur in these locations.

Figure 2-3 Spatial distribution of simulated 24-hr primary and secondary PM2.5 levels averaged across the 52 simulated days when measured Bay Area 24-hr PM2.5 level exceeded 35 µg/m³



(Bay Area counties and the California coastline are drawn using thick black lines.
City limits for Sacramento and Stockton are drawn using thin black lines.)

As noted above, analysis of meteorological patterns found that more than 80% of Bay Area PM exceedances occur when easterly winds blow into the Bay Area from the Central Valley. Therefore, Air District staff also performed photochemical modeling to estimate the contribution of PM transport from the Central Valley during elevated PM episodes in the Bay Area. Transport impacts were evaluated for 55 days (from 2000-01 and 2006-07) having simulated base-case PM2.5 concentrations of 35 µg/m^3 or higher. Anthropogenic Bay Area emissions were eliminated to estimate the cumulative transport impacts from all sources outside of the Bay Area. These simulations found that significant amounts of both primary and secondary PM2.5 in the form of ammonium nitrate were transported into the Bay Area. On days when the Bay Area exceeded the 24-hour PM2.5 standard, modeling indicated that transported primary PM2.5 levels averaged as high as 8 µg/m^3 and transported secondary PM2.5 levels averaged as high as 13 µg/m^3. The largest transport impacts for both primary and secondary PM2.5 occurred along the eastern boundary of the Bay Area.

The modeling also examined the sensitivity of ambient PM concentrations in response to hypothetical reductions in Bay Area emissions of primary PM2.5, as well as reductions in precursor pollutants (ROG, NOx, SOx, and NH3) individually and in combination. Reducing Bay Area primary (directly-emitted) PM2.5 emissions provided far greater reductions in ambient Bay Area PM2.5 levels than reducing Bay Area secondary PM2.5 precursor emissions. Of the precursor emissions reductions simulated, Bay Area ammonia reductions were most effective. Reducing ammonia emissions by 20% was found to decrease PM2.5 concentrations by approximately 0.5 to 1.0 µg/m^3. The ammonia emissions reductions lowered the ammonium nitrate component of PM2.5 only for relatively cold winter days favoring ammonium nitrate buildup. (Ammonium nitrate PM2.5 tends to evaporate faster than it forms at temperatures above around 60 degrees Fahrenheit.) Combined

NOx and ROG emissions reductions for the Bay Area were relatively ineffective. NOx emissions reductions were relatively ineffective because ammonium nitrate PM2.5 formation involves the relatively slow and incomplete conversion of NOx to nitric acid. Reducing Bay Area sulfur-containing PM precursor emissions typically had a small impact on Bay Area ambient PM2.5 levels. Under certain conditions, however, reducing Bay Area sulfur-containing emissions did decrease ambient Bay Area PM2.5 levels by approximately 1 µg/m^3.

The Air District also modeled wood smoke to estimate the impact of efforts to reduce residential wood-burning during the winter season. The modeling period included 8 of the 11 Spare the Air periods during the winter of 2008-09. Bay Area wood-smoke levels were simulated with and without wood-burning restrictions during these periods. Without burning restrictions during these Spare the Air periods, the simulations indicated that peak wood-smoke levels of up to 10-20 µg/m^3 would have occurred over the areas that generally have high wood-burning emissions. For many of the remaining populated locations within the Bay Area, wood-smoke levels would have been approximately 5 µg/m^3. Peak benefits of the wood-smoke rule were about 10 mg/m3 of reduced wood smoke. Maximum simulated benefits of the wood-smoke rule occurred for areas that generally have the highest wood-smoke levels. Often, the areas most heavily impacted by wood smoke are away from the Air District's PM monitoring locations. Therefore, reductions of population exposure to wood smoke resulting from the rule may be significantly greater than indicated by the monitoring data.

Source Contributions to Ambient PM Concentrations

Determining the relative contributions of various sources of direct PM2.5 emissions and PM2.5 precursors to total PM concentrations is complex. The Air District collects PM on filters at four monitoring sites (Livermore, West Oakland, San Jose, and Vallejo) for PM speciation purposes. The filters are then analyzed to estimate the contribution of various emission sources to the total ambient concentration of PM2.5 using a technique known as **chemical mass balance** (CMB).[15] To estimate the overall contribution of various sources, the results of the source apportionment analysis are combined with emissions data from the 2010 Emissions Inventory. The CMB analysis presented here updates a previous CMB analysis prepared in 2008[16] and includes data from July 2009 through December 2011. The goal was to represent the mix of PM2.5 sources as of 2010, including the reduced contributions from ships following the effective date of the ARB rule to require ocean-going ships to use low-sulfur fuel within 24 miles of the coast.

Figure 2-4 shows the estimated contributions to Bay Area annual PM2.5 concentrations using the average of the results from the four sites. Note that this includes contributions to secondary PM2.5; i.e., ammonium nitrate formed from NOx and ammonia, and ammonium sulfate formed from SO2 and ammonia. The source apportionment analysis shows that combustion of both fossil fuels and biomass are major PM2.5 contributors for all seasons. The largest contributor to annual PM2.5 is wood-burning,

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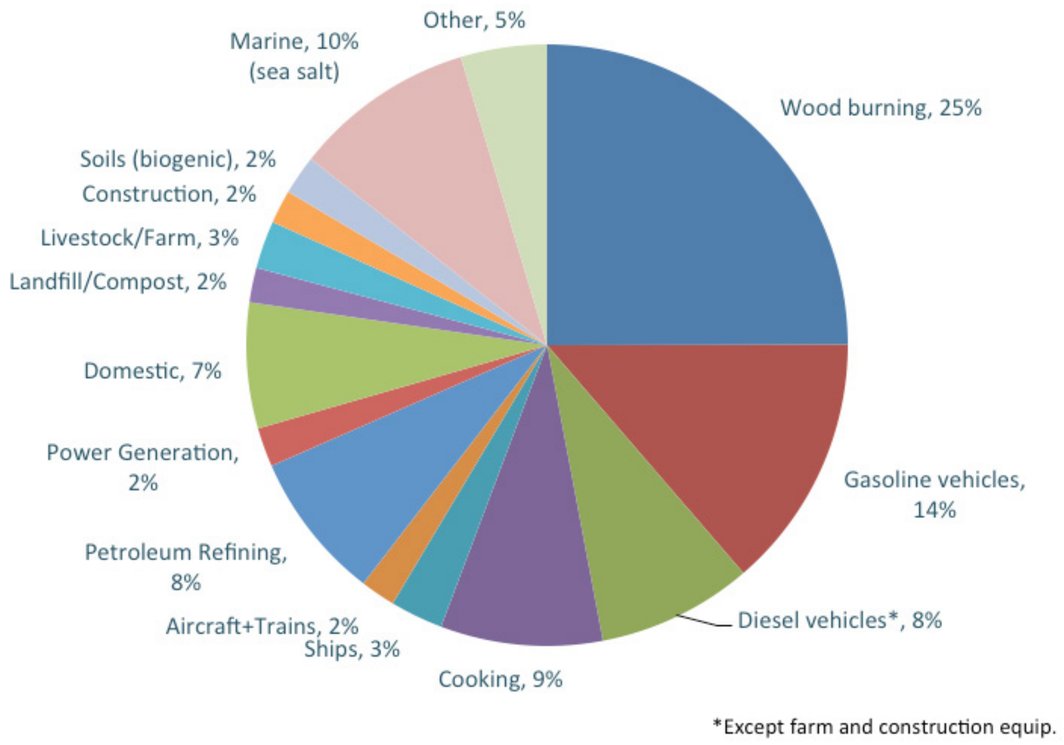
[15] Chemical mass balance (CMB) analysis is a methodology in which a computer model is used to apportion ambient PM2.5 collected on filters over 24-hour periods at monitoring sites around the Bay Area to a set of source categories. Each filter was analyzed for a range of chemical species. The same species were measured in special studies of emissions from various sources, such as motor vehicles and wood burning. The CMB model finds the mix of these source measurements that best matches the ambient sample, chemical species by chemical species.

[16] See report entitled Sources of Bay Area Fine Particles, April 2008 at: www.baaqmd.gov/Divisions/Planning-and-Research/Research-and-Modeling/Publications/Reports.aspx

contributing about a quarter of the total. This is mainly from residential wood burning, but also includes wood smoke from controlled burns and wildfires. Nearly another quarter comes from on-road motor vehicles, 14% from gasoline and 8% from diesel.

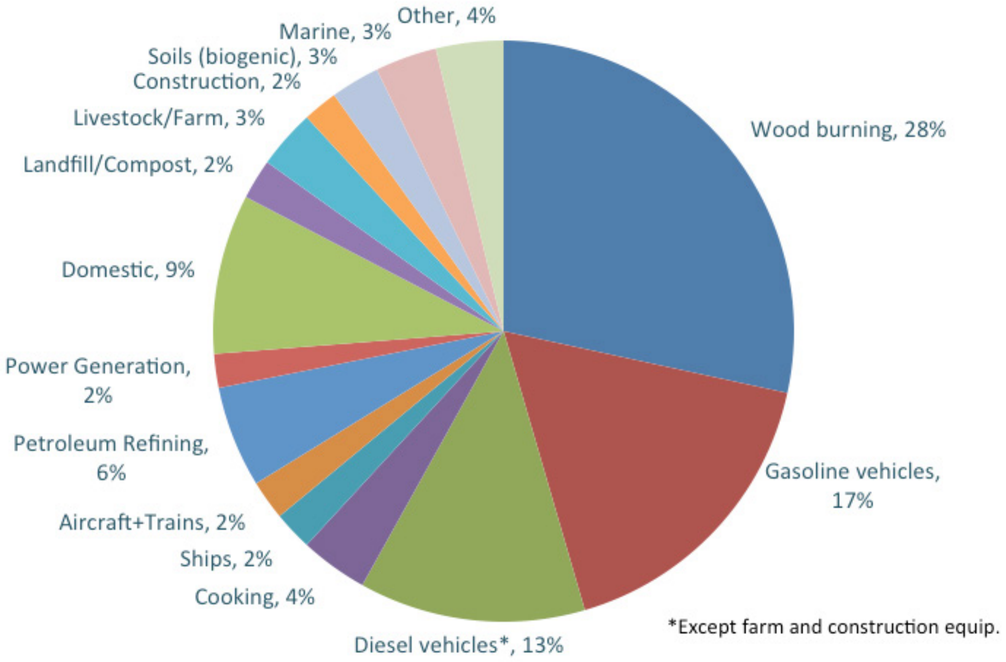
Figure 2-4 Estimated Source Contributions to Annual PM2.5 Concentrations



Cooking, petroleum refining, and domestic sources are also sizeable contributors, along with the naturally occurring sea salt from marine air. Domestic sources, for purposes of Figures 2-4 and 2-5, include emissions from use of natural gas for residential heating and cooking, as well as the contribution to formation of secondary PM2.5 as a result of ammonia emissions from pet waste and human perspiration and respiration Contributions from ships have been cut by two-thirds or more, thanks to the ARB rule mandating low sulfur fuel near ports. The estimate for construction equipment has also dropped substantially because of a revision in ARB's estimation method. Note that several sources – soils, livestock, and landfill – appear because of their ammonia emissions, which contribute to formation of ammonium nitrate and ammonium sulfate.

Figure 2.5 shows the sources contributing to peak PM2.5 concentrations, specifically, the 10 samples at each monitoring site with the highest PM2.5 concentrations. Almost all the highest Bay Area PM2.5 concentrations occur in winter, so the wood-burning contribution is higher than the annual, representing almost 30% of the peak total. The biomass combustion contribution to peak PM2.5 levels is about 3-4 times higher in winter than the other seasons, as confirmed by isotopic carbon (^{14}C) analysis, due to increased levels of wood burning during the winter season. Motor vehicles are also a larger contributor during winter months because their NOx emissions contribute most to the formation of secondary PM in the form of ammonium nitrate. Similarly, domestic sources are another large source of secondary PM because of their ammonia emissions. Marine air is a smaller contributor in the winter than on an annual-average basis, because winter winds are frequently offshore, unlike the typical on-shore pattern the rest of the year.

Figure 2-5 **Estimated Source Contributions to Peak PM2.5 Concentrations**



PM Emissions Inventory

An emissions inventory is an essential tool for air quality planning. The inventory contains a detailed breakdown of the estimated emissions from each source category, thus providing information as to the source, magnitude, and location of emissions. Emissions inventories are used to perform air quality modeling, to identify source categories where there may be opportunities for additional emission reductions, and to predict trends in terms of future air quality.

This section presents the Air District's latest Bay Area inventory for primary emissions (directly-emitted particles) of both PM2.5 and PM10, as well as precursors that combine via chemical reactions to form secondary PM. These precursors include reactive organic gases (ROG), oxides of nitrogen (NOx), sulfur oxides (SOx), and ammonia (NH3). The Air District released it's first-ever emissions inventory for ultrafine PM in August 2012[17].

The inventory includes annual-average emissions, as well as emissions during the winter season when the Bay Area normally experiences its highest PM concentrations. In addition to the inventory for base year 2010, projected emissions for future years out to 2030 are also provided. Tables in Appendix A provide the detailed inventory showing estimated emissions by source category.

This inventory does not include "condensable" emissions that also form PM. Condensables are emissions which are emitted in a gaseous phase, but then condense to form particles upon exposure to cooler ambient air, as discussed in the section above entitled **Physical Processes that Affect PM Formation**. Depending on the emission source and combustion conditions, this condensable component may account for a significant share of overall PM emissions for certain emission sources. Recent US EPA guidelines call on air quality agencies to consider the condensable component of PM in developing PM emission inventories and control strategies. EPA has published a source test method to be used for source testing stationary sources (with a few minor exceptions). However, test methods and methodologies to estimate condensable emissions are not available at the current time for certain emission source categories. Therefore, in the interest of methodological consistency, the Air District has chosen to exclude condensable emissions in the current inventory. The Air District will work with ARB and other partners to develop estimates of condensable emissions for future PM inventories.

Methodologies Used to Estimate Emissions

The estimated emissions provided in the inventory are based upon data from source tests, published emission factors, and engineering calculations. Emission inventories are revised and improved on a regular basis. The PM inventory provided in this report differs significantly from the inventory provided in the Bay Area 2010 Clean Air Plan (2010 CAP). This is due to the fact that, since the release of the 2010 CAP, ARB and Air District staff reviewed and improved the methodologies used to estimate emissions from several of the most important PM source categories, as explained below.

Stationary (Point) Sources

Actual 2010 reported emissions from permitted facilities are included in this inventory. Examples of stationary sources that emit primary PM and/or PM precursors including oil refineries, metal smelters, charbroilers, and back-up generators used to supply emergency power at many facilities.

Residential Fuel Combustion

This category includes residential combustion of both wood and natural gas for space-heating, water-heating, and cooking. Most homes rely primarily (or completely) on natural gas (or electricity) for these purposes. Nonetheless, although only a minority of Bay Area households burn wood, PM from wood smoke is the largest single source of winter-time PM emissions in the Bay Area, and greatly exceeds the PM emissions from (the much more common) residential natural gas

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[17] See the report entitled, *Ultrafine Emissions Inventory for the San Francisco Bay Area*, BAAQMD August 2012

combustion. Previous estimates of the amount and location of residential wood burning were developed based on a Bay Area 2005-2006 telephone survey on wood burning. In 2008 the Air District adopted Regulation 6, Rule 3 to limit emissions of particulate matter (PM) and visible emissions from wood-burning devices, as described in Section 4. The District also implemented a robust outreach effort to inform Bay Area residents about "no-burn days" and the detrimental health effects of wood smoke through its winter Spare the Air program. Annual surveys of Bay Area households, as well as chemical mass balance analysis of PM captured on filters, both indicate that wood-burning has decreased approximately 40% since 2008 when the Air District's wood burning rule went into effect. Emissions from residential fuel combustion in this inventory have been revised to reflect this decrease in wood-burning.

Commercial Cooking

Previous estimates for this source category included condensable PM. The reason for this was that source testing conducted as part of the regulatory process to control emissions from this source category (via Regulation 6, Rule 2) included condensable PM. However, because methods to estimate condensable emissions for other source categories are not yet available, as discussed above, the condensable emissions for the Commercial Cooking source category have been omitted here for purposes of methodological consistency.

On-Road Motor Vehicles

Estimates for on-road motor vehicle emissions are based on ARB's latest emissions factor model, EMFAC2011. In preparing EMFAC2011, ARB staff conducted major research to determine the population and compositions (e.g. construction trucks, port trucks, in-state trucks, etc.) of medium and heavy duty diesel truck fleets (>14,000 pounds Gross Vehicle Weight), as well as age distribution and vehicle miles traveled for these vehicles. Also, emission factors for these vehicles have been updated to reflect the major benefits of recent ARB regulations to reduce emissions from diesel trucks and buses. The EMFAC2011 model also includes the impact of the recent economic recession on both diesel and gasoline vehicle activity.

The on-road category includes PM emissions from both diesel and gasoline engines. PM emissions from late-model light-duty gasoline vehicles are extremely low on a grams-per-mile basis. However, emissions from gasoline vehicles are significant on an aggregate basis because gasoline vehicles account for approximately 95% of the 175 million miles driven on an average day in the Bay Area.

Motor vehicle emissions that contribute to PM include tailpipe emissions of both primary PM and PM precursors from fuel combustion, lubricating oil, and particles generated by brake and tire wear. PM from tire wear and brake wear are included for on-road motor vehicles. There is insufficient information to produce estimates for tire-wear and brake-wear for off-road equipment; however, these emissions are expected to be very low as they are related to vehicle miles traveled (VMT) and most of the off-road equipment with tires have very low VMT.

A breakdown of annual average motor vehicle emissions in the current inventory is shown in Table 2-2.

Table 2-2 Bay Area Annual Average Primary PM2.5 Emissions – Year 2010: On-Road and Off-Road Motor Vehicles

On-Road Motor Vehicle	tons/day	%
Gasoline Exhaust	0.8	10%
Diesel Exhaust	2.7	37%
Tire/Brake-Wear (Gasoline Vehicles)	3.2	44%
Tire/Brake-Wear (Diesel Vehicles)	0.6	8%
Total On-Road Motor Vehicles	**7.3**	**100%**
Off-Road Motor Vehicles	**tons/day**	**%**
Gasoline Exhaust	1.7	37%
Diesel Exhaust	2.9	63%
Total Off-Road Motor Vehicles	**4.6**	**100%**
On-Road and Off-Road Motor Vehicles Combined		
All Motor Vehicles	**tons/day**	**%**
Total Gasoline Exhaust	2.5	21%
Total Diesel Exhaust	5.6	47%
Tire/Brake Wear	3.8	32%
Grand Total	**12.0**	**100%**

The new emission factors show a reduction in tailpipe emissions from both gasoline and diesel engines. However, the decrease in tailpipe exhaust has been offset to a considerable extent by a major increase in estimated PM emissions from brake wear. Brake-wear emissions of PM2.5 in

EMFAC2011 are three times the estimates produced by EMFAC2007. (It should be noted, however, that chemical mass balance analysis performed by Air District staff shows better agreement with the EMFAC2007 factors for brake and tire wear. Therefore, Air District staff will work with ARB staff to continue to investigate and improve PM emission factors for brake wear.)

Whereas brake wear and tire wear made up a relatively small portion of PM2.5 from on-road vehicles using the previous EMFAC2007 emission factors, they now account for half of total PM2.5 emissions from on-road vehicles, according to the EMFAC2011 model. Although PM2.5 exhaust (tailpipe) emissions from diesel vehicles are much greater than from gasoline vehicles, when the new brake wear factors are included, gasoline vehicles account for 54% of total PM2.5 from on-road vehicles, compared to 45% for diesel vehicles.

It is important to note that, in addition to their direct emissions of primary PM, motor vehicles are the major source of precursor pollutants that combine to form secondary PM. For example, on-road vehicles account for 37% of ROG, 57% of NOx, and 18% of ammonia emissions in the Bay Area inventory for year 2010, as shown in Figures 2-10, 2-11, and 2-12, respectively.

Although the Air District does not yet have an ultrafine PM inventory, emissions testing indicates that both gasoline and diesel engines are a major source of ultrafine particles. Tests indicate that gasoline vehicles emit especially large amounts of ultrafine particles when in a hard acceleration mode. Research also suggests that engine lubricating oil may be an important source of ultrafine particles. It is likely that when an ultrafine PM inventory for the Bay Area becomes available, it will show that combustion from both diesel and gasoline vehicles account for significant shares of UFPM emissions in the region.

Construction, Industrial and Airport Ground Support Equipment

Emissions estimates for off-road mobile sources (such as construction, cargo handling at ports, and airport ground support equipment) are taken from ARB's latest emissions factor model, OFFROAD2011. Emissions for this category have decreased significantly compared to the estimates in the previous inventory derived from ARB's OFFROAD2007 model. Research on fuel sales for off-road equipment showed that fuel usage, and hence emissions from these vehicles, had been substantially over-estimated in the OFFROAD2007 model.

In response, ARB staff improved the inventory for this category by revising equipment population estimates based on historical equipment sales data, and incorporating data from industry regarding

hours of equipment operation and load factor, a measure of how intensively the equipment is being used. When coupled with the impact of the 2007 recession, the new OFFROAD2011 model estimated PM2.5 emissions from this category to be just one-fifth of the estimate produced by EMFAC2007 that was used in the 2010 CAP.

Ships and Commercial Boats

ARB recently revised the methodology to calculate emissions from ships and commercial boats. For ships, the new inventory increases the specificity of the earlier inventory by including vessel-specific characteristics and activity for each port. Emissions were calculated by estimating ship emissions on a ship-by-ship and a port-by-port basis, using actual ship engine power, speeds, and berthing times where possible. Projected emissions for future years were estimated using a set of growth factors specific to each port and each ship type. For commercial boats, emissions were calculated based on data collected from ARB's 2004 Statewide Commercial Harbor Craft Survey. This survey collected information from boat owners as to vessel type, home port, engine characteristics, hours of operation, annual fuel usage, etc. This information along with other studies, such as emission inventories developed for the Ports of Los Angeles and Long Beach, were used to estimate emissions for boats. Cruise ship berthing emissions were taken from the 2005 Bay Area Seaports Air Emissions Inventory Report.

Another major difference in the current estimate of ship emissions, compared to the estimate provided in the 2010 CAP, is that ship emissions reported in this inventory are based on ship activity within three nautical miles of the coastline, whereas the 2010 CAP reported emissions for ship activity up to 100 nautical miles. Based on direction from ARB, using a limit of three nautical miles is consistent with the limit being used by other air districts that are in the process of preparing PM2.5 SIP submittals.

Aircraft

Aircraft emissions are based on actual 2010 activity data (landing and take-off data, taxi times between the runway and the terminal, etc.) at Bay Area airports, based on the current aircraft fleet mix and the latest emissions factors for PM, ROG, NOx, and SO2. Aircraft emissions are decreasing on a per-passenger mile basis, due to the shift toward larger planes and the development of more fuel efficient engines. As a result of new aircraft engine technological changes that rely on higher combustion temperatures to improve fuel economy, ROG emissions are decreasing, but NOx emissions are increasing per landing and take-off.

Paved and Unpaved Road Dust (Re-entrained Dust)

Previous paved road dust emissions, including those reported in the 2010 CAP, were believed to be over-estimated when compared to observed ambient concentrations and source apportionment (chemical mass balance) analysis. A new methodology from US EPA was used to estimate PM emissions from vehicular travel on paved roads.[18] This methodology results in significantly lower estimates of PM10 and PM2.5 emissions compared with previous estimates. The revised estimates of road dust emissions from paved roads for PM10 and PM2.5 are only about one-third of the estimated emissions reported in the 2010 CAP.

Unpaved road dust emissions were also updated to reflect the latest information from Caltrans on Bay Area unpaved road miles and vehicle miles traveled on these roads. As with the paved road dust estimates, the revised estimates of road dust emissions from unpaved roads for PM10 and PM2.5 are only about one-third of the estimated emissions reported in the 2010 CAP.

2010 Annual Average Primary PM2.5 Emissions

Estimated annual average emissions of primary PM2.5 have decreased significantly in the current inventory compared to the estimates provided in the 2010 CAP. Whereas the 2010 CAP reported 47 tons per day of primary PM2.5 for year 2009, the current inventory shows an estimated 87 tons per day of primary PM2.5 for year 2010, a decline of 46%. The revised inventory includes significant reductions in several major source categories, such as residential wood-burning, commercial cooking, off-road vehicles (“Other Mobile Sources”), and road dust (included in “Geological dust”). This decrease in the inventory is based in part on real emission reductions due to factors such as turnover in the vehicle fleet and the impact of new regulations that had not been accounted for in previous emission factors. However, changes in methodologies used to estimate emissions, as discussed above, also account for a significant portion of the decrease in the inventory. Figure 2-6 shows the annual average emissions of PM2.5 for year 2010 broken down by major source categories.

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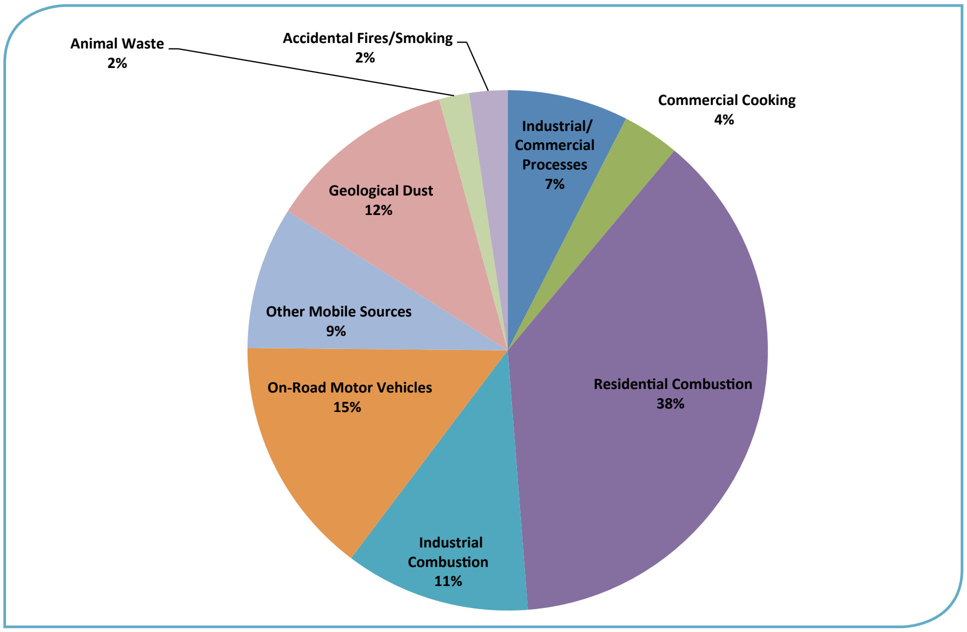
[18] The new methodology for road dust emissions is set forth in two documents:
1) US EPA guidance, January 2011: www.epa.gov/ttn/chief/ap42/ch13/final/c13s0201.pdf and
2) Air Resources Board guidance re: Draft Entrained Paved Road Travel Paved Road Dust, (Section 7.9) 6/15/11

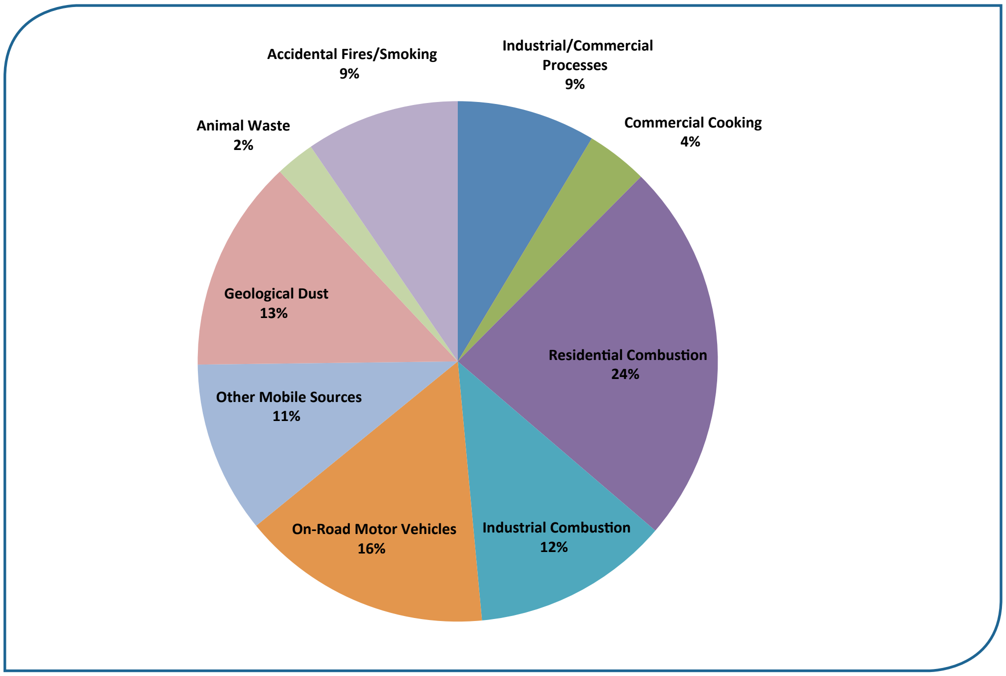
Figure 2-6 2010 Annual Primary Emissions of PM2.5, 47 tons/day



2010 Winter PM2.5 Emissions

The breakdown of primary PM2.5 emissions by source category differs for the winter PM2.5 inventory compared to the annual inventory. The key difference is that PM2.5 emissions from residential fuel combustion account for 38% of the winter inventory versus 24% in the case of the annual inventory. PM2.5 emissions from residential fuel combustion, which is dominated by wood-burning in fireplaces and wood stoves, are 65% higher (an additional 7 tons per day) in the winter months than the annual average emissions for this category. However, emissions from several source categories are lower in winter. For example, winter emissions from off-road equipment, as well as from the industrial sector, are lower than annual average emissions due to reduced activity during the winter months. Accidental fires and geological dust emissions are also lower in the winter than the annual average. Overall, however, winter-time PM2.5 emissions are 4% higher (an additional 2 tons per day) than annual-average emissions. Figure 2-7 shows the winter emissions of PM2.5 for year 2010 broken down by major source categories.

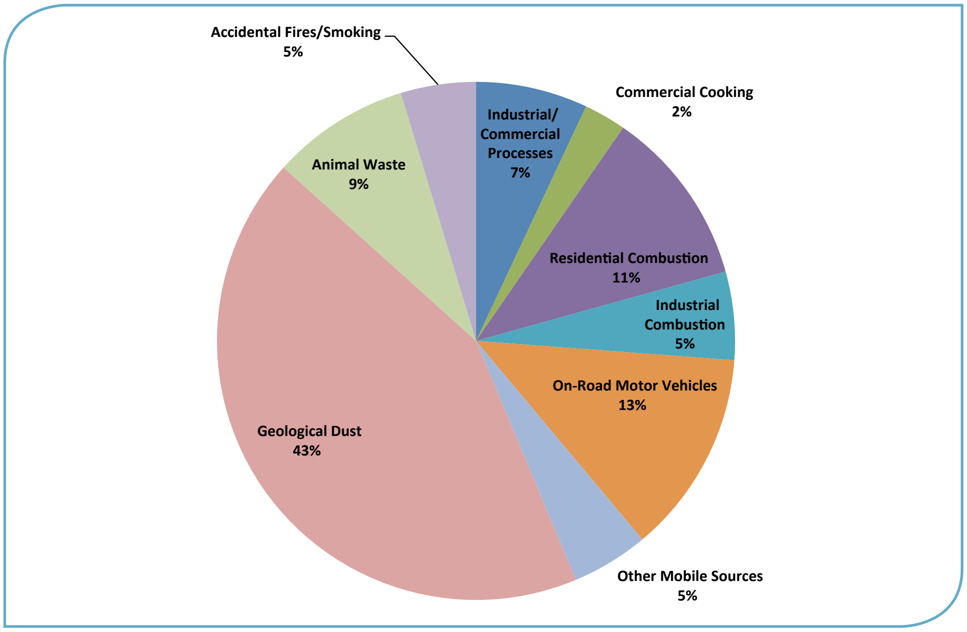
Figure 2-7 2010 Winter Primary Emissions of PM2.5, 49 tons/day



2010 Annual Average Primary PM10 Emissions

PM2.5 emissions are a sub-set of PM10 emissions. For some sources, such as the various types of engine combustion, virtually all PM10 actually consists of fine particles less than 2.5 microns in diameter. For example, 97% of diesel PM10 is PM2.5, meaning that diesel particulate is essentially all PM2.5. But for other sources, only a fraction of PM10 is made up of particles less than 2.5 microns in diameter, and the rest consists of coarse particles between 2.5 and 10 microns in diameter. For example, only 15% of PM10 from road dust is PM2.5; the remaining 85% is coarse particles. For the emissions inventory as a whole, on a mass basis, roughly half of PM10 is composed of fine particles less than 2.5 microns in diameter and half is composed of coarse particles between 2.5 and 10 microns in diameter. In Figures 2-8 and 2-9 showing annual and winter emissions of PM10, source categories that emit coarse particles become more prominent, thus contributing more to PM10 emissions compared to Figures 2-6 and 2-7 for PM2.5. In particular, geological dust, which includes dust from construction and farming operations, re-entrained road dust from paved and unpaved roads, and wind-blown dust, accounts for a much greater percentage of PM10 (43% on an annual average basis) than PM2.5 (13% on an annual average basis).

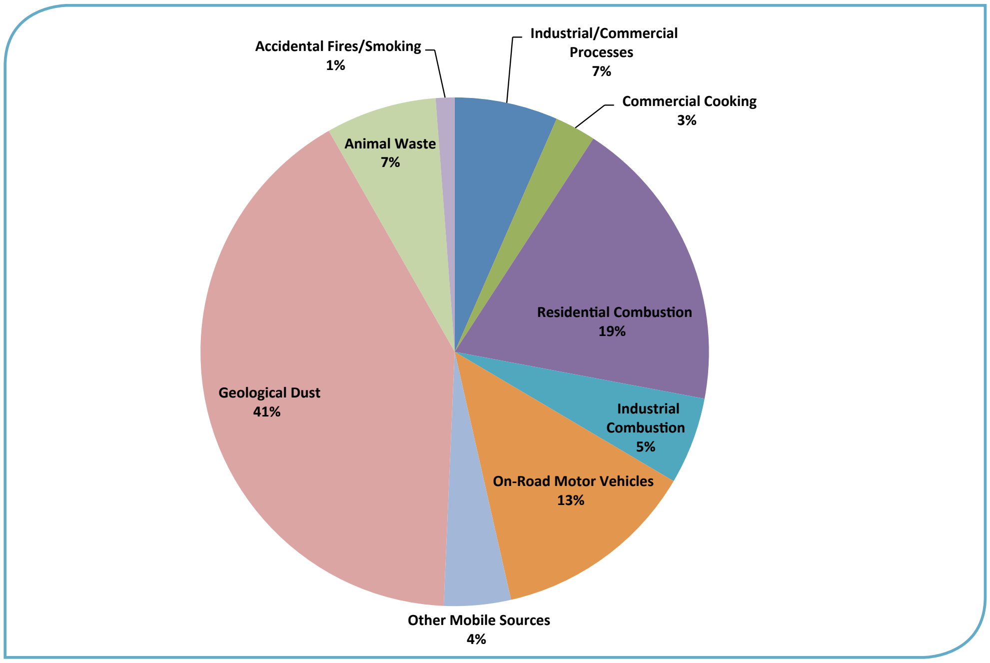
Figure 2-8 **2010 Annual Average Primary Emissions of PM10, 106 tons/day**



2010 Winter Primary PM10 Emissions

The relationship between annual average PM10 and winter PM10 (i.e., the relative contributions from various source categories) is similar to that for PM2.5. The main difference is that emissions for residential fuel combustion (primarily wood-burning) increase from 11% of the annual PM10 inventory to 18% of the winter PM10 inventory, while emissions from accidental fires (primarily wildfires) decrease from 5% of the annual PM10 inventory to 1% of the winter PM10 inventory. Figure 2-9 shows the winter emissions of PM10 for year 2010 broken down by major source categories.

Figure 2-9 **2010 Winter Primary Emissions of PM10, 104 tons/day**



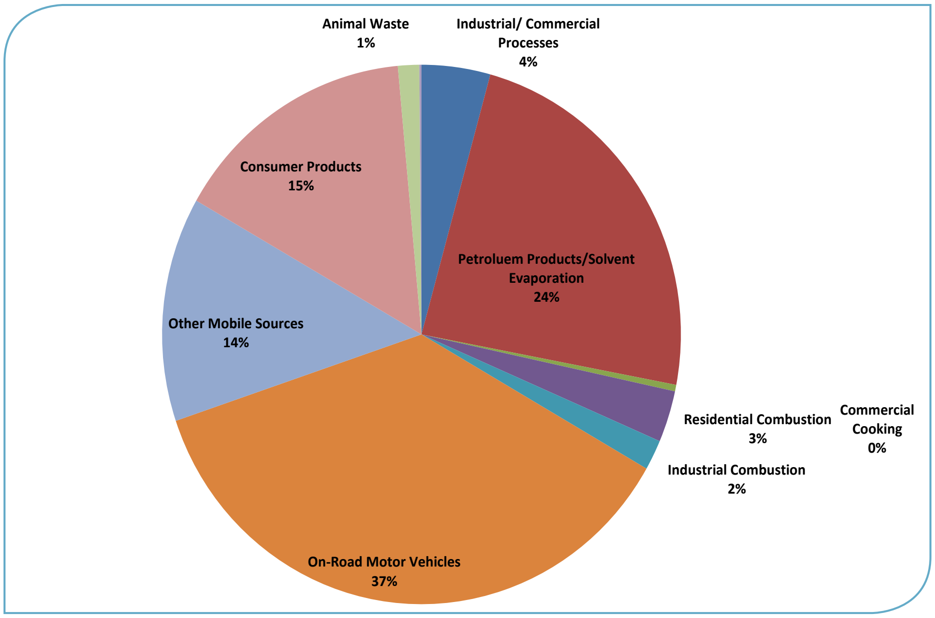
Sources of Precursors Pollutants That Form Secondary PM

Precursor pollutants that combine via chemical processes to form secondary PM include reactive organic gases (ROG), oxides of nitrogen (NOx), sulfur dioxide (SO2), and ammonia (NH3). The sources that produce these pollutants are described below, with pie charts depicting emissions during the winter season when secondary PM formation is greatest.

Reactive Organic Gases (ROG)

As shown in Figure 2-10, on-road motor vehicles (37%) and off-road vehicles (14%) together produce approximately 50% of the winter-time ROG emissions. Evaporation from petroleum products (including those from refineries and fuels distribution) and solvents (such as those from structures coating, adhesives, and sealants) are the second largest contributors to ROG emissions, accounting for approximately 24% of winter emissions. Emissions from consumer products contribute another 15% of the winter-time ROG emissions.

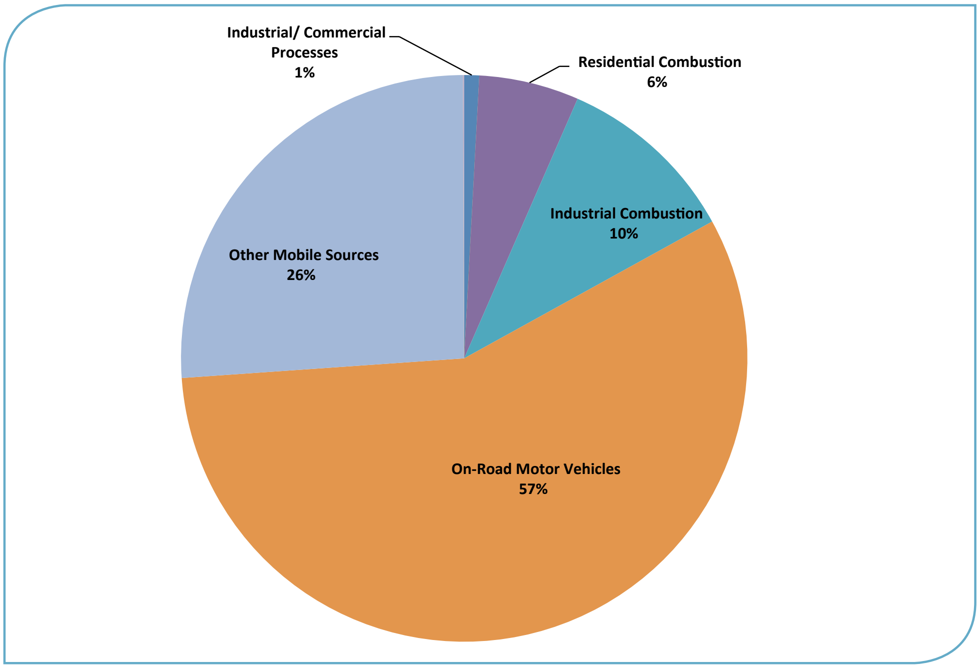
Figure 2-10 2010 Winter Emissions Reactive Organic Gases (ROG) 294 tons/day



Oxides of Nitrogen (NOx)

As shown in Figure 2-11, on-road motor vehicles are the single largest source of NOx emissions in the Bay Area. Together with off-road mobile sources, they produce over 80% of the winter-time NOx emissions. Industrial combustion and residential fuel combustion (including wood-burning) produce 10% and 6% of the winter-time NOx inventory, respectively.

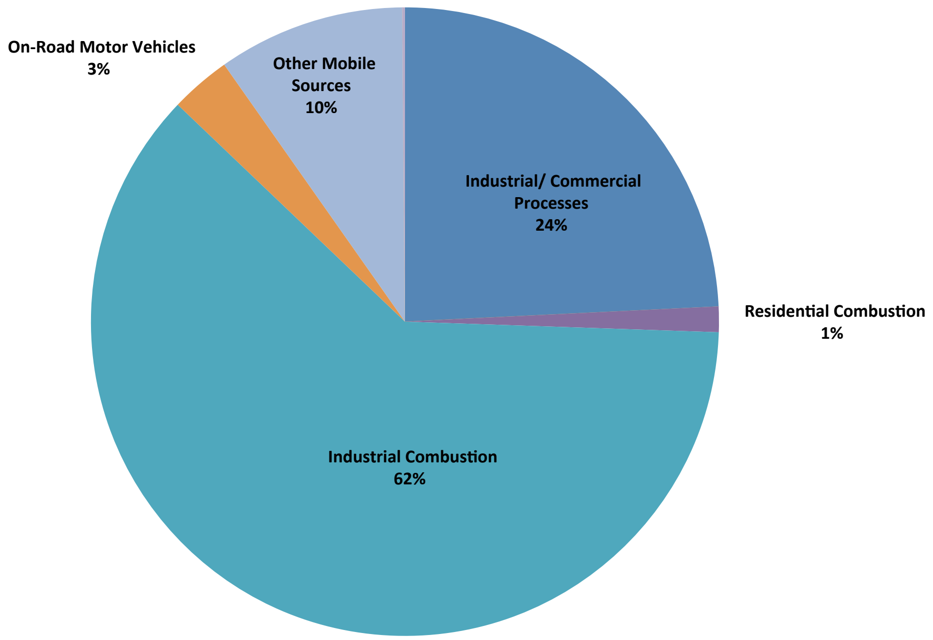
Figure 2-11 **2010 Winter Emissions Oxides of Nitrogen (NOx), 347 tons/day**



Sulfur Dioxide (SO2)

Figure 2-12 provides a breakdown of winter SO2 emissions for year 2010. The majority of SO2 emissions in the Bay Area are from combustion at industrial facilities; the Industrial Combustion wedge (62%) in Figure 2-12 includes SO2 emissions from refineries (50%) and other industrial facilities (12%). Industrial and commercial processes employed in the production of pharmaceuticals, cosmetics, inks, and resins, as well as those used at refineries, create another 24% of SO2 emissions in winter. Off-road and on-road motor vehicles produce 13% of the Bay Area winter emissions. Although most of the SO2 in the Bay Area (over 50%) is emitted during refinery operations, it is worth noting that the refinery products are used to fuel motor vehicles in the Bay Area and throughout California. This suggests that SO2 levels can be reduced both by further controlling emissions at the refineries, as well as by reducing motor vehicle use.

Figure 2-12 2010 Winter Emissions Sulfur Dioxides (SO_2), 29 tons/day



Predicting Future Emissions Trends

Predicting future trends in emissions is challenging, since many factors come into play, such as changes in control technologies; emission standards and fuel formulations for mobile sources; population growth and household formation; economic growth rates; land use decisions; and changes in transportation infrastructure and travel mode choice which affect motor vehicle travel. The inventory projections presented here include anticipated changes in population and economic activity, and well as emission reductions from ARB and Air District regulations that have already been adopted and turnover in the motor vehicle fleet (whereby older, dirtier vehicles are replaced by newer, cleaner vehicles)ARB. Implementation of ARB regulations on mobile sources is typically phased in, so the entire benefit of these adopted regulations will not be realized until they have been fully implemented over the next 10-15 years. At that point, emissions are projected to slowly increase in response to population and economic growth, if no additional regulations are adopted.

The projected emissions are based on a conservative "business as usual" assumption that no additional regulation or polices will be adopted to reduce emissions in the future. However, past experience suggests that it is likely that future measures will in fact be adopted and implemented to provide additional emission reductions. Past experience also suggests that the projected inventory may underestimate the future reduction in emissions. For example, whereas previous PM emissions inventories for the Bay Area predicted that overall emissions would increase over the past 10-15 years, monitoring data and CMB analysis shows that PM emissions and ambient concentrations actually declined substantially during that period. Given the fact that previous inventories under-predicted the emission reductions that were actually achieved, it is possible that this could occur again over the coming years.

Assumptions and methods used to estimate future emissions for key source categories are briefly discussed below.

Industrial and Commercial Processes and Combustions

PM emissions from industrial and commercial sources are projected to increase at a rate of around 1% per year based on previous observed growth and regulations adopted to date.

Commercial Cooking

PM emissions from commercial cooking are also projected to increase at a rate of around 1% per year based on previous observed growth and regulations adopted to date.

Residential Fuel Combustion

As noted above, estimated PM emissions from residential wood-burning were reduced for the 2010 inventory to reflect recent progress in response to the Air District's wood-burning regulation and its winter Spare the Air program. The projected inventory assumes that residential wood-burning rates will hold steady for the foreseeable future, with a slight increase based upon growth in population and households. Although it is possible that future emissions may decrease if the District is successful in increasing compliance with the wood-burning regulation and further educating Bay Area residents as to the health risks from wood smoke, Air District staff has opted to hold the wood-burning emissions steady, pending development of better data to use in estimating future wood-burning trends, so as to provide a conservative estimate. Modest increases of emissions are expected in other residential combustion categories, mainly for space heating. This is assumed to grow in-line with population growth resulting in modest increases in overall residential combustion emissions.

On-Road and Off-Road Motor Vehicles

Emissions from on-road and off-road motor vehicles are expected to decline until 2020 due to aggressive regulations on diesel engines; retirement of older, dirtier vehicles; and penetration of cleaner gasoline vehicles into the fleet. After 2020, vehicle emissions are expected to increase by less than 1% per year until 2030. The projected increase in vehicle emissions rests upon two assumptions:

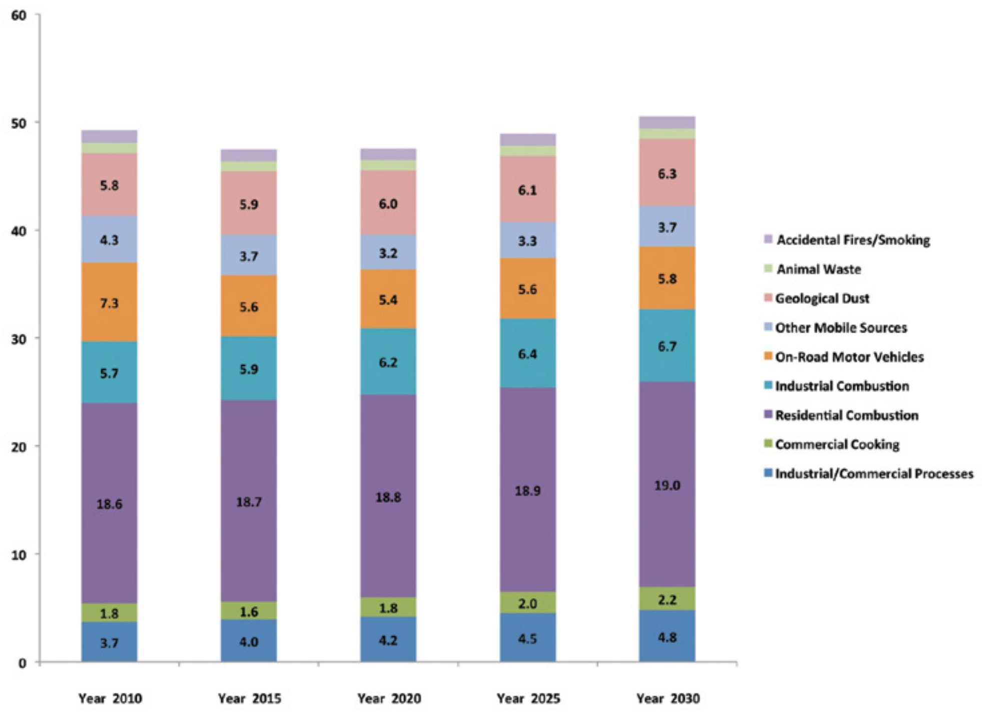
1. An increase in vehicular activity following recovery from the economic recession. ARB staff examined various economic recovery scenarios; the projected rate of growth in vehicle activity is based on an average between a slow-recovery scenario and a fast-recovery scenario.
2. No additional regulation apart from those already adopted by ARB. (The assumption that no future regulations will be adopted is unlikely, however. Although it will be technically challenging to further reduce motor vehicle emissions rates below the stringent standards already adopted in California, experience to date suggests motor vehicle emission standards will be further tightened in coming years as needed to continue progress toward attainment of air quality standards in the major urban areas of the state.)

It is important to note that the projected trends for diesel and gasoline vehicles differ. Whereas diesel PM emissions are projected to sharply decrease over the next decade in response ARB regulations, PM emissions from gasoline engines are expected to hold steady. For year 2010, diesel vehicles account for about half of the primary PM emissions from on-road vehicles, including both tailpipe exhaust and brake and tire wear. However, by 2030, this figure is projected to decrease to about 27%. It should be noted, however, that the EMFAC2011 model does not include the potential reduction in PM from light- and medium-duty vehicles that may occur in response to ARB's LEV III program, as described in Section 4. Efforts to reduce PM from mobile sources in recent years have focused on heavy-duty, diesel-powered vehicles. However, the fact that gasoline vehicles are projected to account for an increasing share of the PM from motor vehicles in coming years suggests that future efforts to reduce PM emissions from on-road vehicles will need to focus on reducing tailpipe emissions and emissions due to brake and tire wear from light-duty vehicles.

Projected Trends for Primary PM and Precursors To Secondary PM

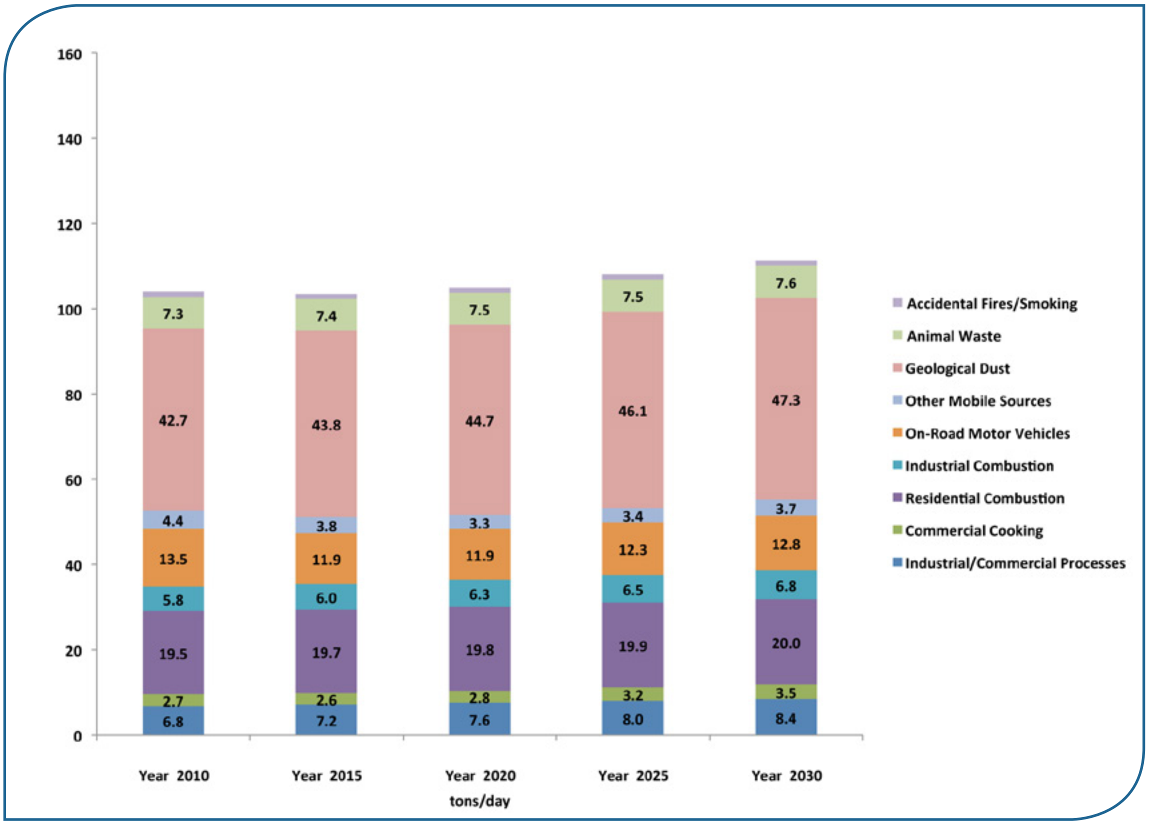
Projected trends for emissions of primary PM and PM precursors are presented in summary form in the bar charts below. A more detailed breakdown of projected future emissions by source category is provided in the tables in Appendix A. As shown in Figures 2-13 and 2-14, the overall inventory for emissions of primary PM2.5 and PM10 is projected to decrease thru 2020, then to begin to rise slowly through 2030 in tandem with population and economic growth. The same trend is projected for emissions from on-road motor vehicles, as well as other mobile sources.

Figure 2-13 Bay Area Winter Primary PM2.5 Emissions Trends



The trend for winter emissions of primary PM10 is similar to that for PM2.5, as shown in Figure 2-14.

Figure 2-14 Bay Area Winter Primary PM10 Emissions Trends



Emissions projections for key precursors to formation of secondary PM are mixed. Similar to primary PM, ROG emissions are projected to decrease through 2020 in response to already-adopted control measures, then to begin to increase slowly through 2030, as shown in Figure 2-15. NOx emissions are projected to decrease steadily and substantially through 2030 in response to already-adopted control measures, as shown in Figure 2-16. SO2 emissions, by contrast, are projected to increase slowly but steadily through 2030 in the absence of additional regulations or controls, as shown in Figure 2-17.

Reactive Organic Gases (ROG)

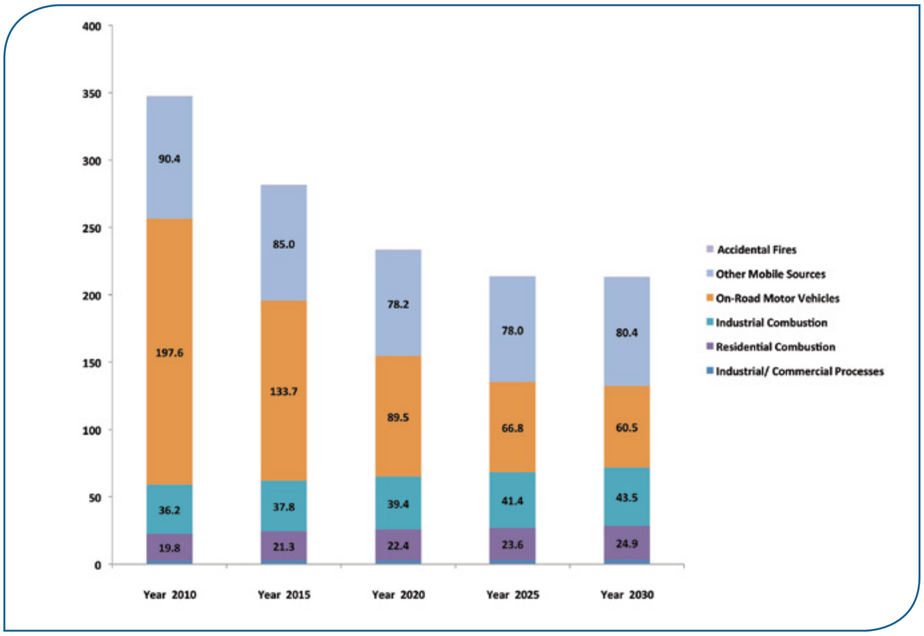
As shown in Figure 2-15, overall ROG emissions are expected to decline until 2025. ROG emissions from on-road motor vehicles are expected to decline due to fleet turnover, despite increases in vehicle population and VMT. Emissions from off-road mobile sources will continue to decline until 2020 due to implementation of already-adopted regulations. After 2020, a projected increase in off-road vehicular activity is expected to lead to increases in ROG emissions from off-road mobile sources. For the inventory as a whole, ROG emissions are projected to increase slightly after 2025 due to increased population and economic activity, in the absence of future regulatory measures.

Figure 2-15 Bay Area Winter ROG Emissions Trends

Oxides of Nitrogen (NOx)

NOx emissions from on-road motor vehicles are expected to decline due to fleet turnover despite increases in vehicle population and VMT, as shown in Figure 2-16. Emissions from other mobile sources will continue to decline until 2025 due to aggressive regulations on diesel vehicles. After 2025, projected increase in off-road vehicular activity is expected to lead to increases in NOx emissions from off-road mobile sources. Other major contributors to NOx emissions are expected to increase due to population increase unless new regulations are introduced. Overall NOx emissions are expected to decline until 2025 and then increase slightly. However, it is expected that with introduction of new rules on major sources of NOx emissions in the future, further reduction in NOx emissions is likely to occur.

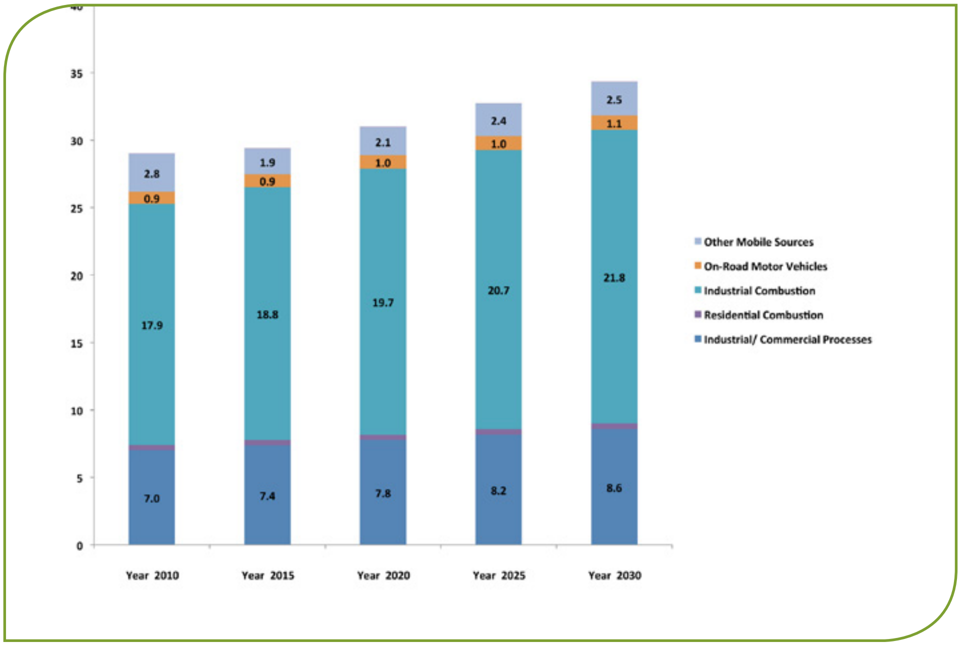
Figure 2-16 Bay Area Winter NOx Emission Trends



Sulfur Dioxide (SO2)

After decreasing substantially in the past few years in response to regulations on sulfur content used in ships and commercial boats, SO2 emissions are projected to increase slowly in future years, as shown in Figure 2-17, primarily due to projected expansion in industrial activity. Overall SO2 emissions are expected to increase in line with SO2 emissions increases mentioned above. The rate of increase is less than 1% per year until 2030.

Figure 2-17 Bay Area Winter SO2 Emission Trends



SECTION 3: PM AIR QUALITY STANDARDS AND TRENDS

SECTION 3-A: PM MONITORING PROGRAM

Regional PM Monitoring Network

The Air District operates a network of monitoring stations to measure ambient concentrations of particulate matter in the Bay Area. The Air District's PM monitoring network has evolved in tandem with the evolution of the PM air quality standards. Because the original PM standards issued in response to the Clean Air Act of 1970 were based on total suspended particles (TSP), the initial PM monitoring stations measured TSP. When US EPA adopted standards for PM10 in 1987, the District established monitoring sites to measure ambient PM10 concentrations. Likewise, when US EPA issued standards for PM2.5 in 1997, the District established a network of monitoring sites to measure ambient PM2.5 concentrations. Therefore, we can track progress in reducing TSP in the Bay Area back to the 1970's, PM10 to the late 1980's, and PM2.5 to the late 1990's.

Until recently, all PM measurements were performed by collecting particles on filters, and PM concentrations were estimated by weighing the filters before and after collecting the particles. A filter is pre-weighed, and then placed in a sampler that draws air through the filter, typically for 24 hours. The PM concentration is estimated by comparing the before and after weight difference of the filter, divided by the total air flow, yielding a measurement of ambient PM expressed in micrograms per cubic meter ($\mu g/m^3$). Filter-based methods that meet specific operational requirements are called Federal Reference Method (FRM) and have been used for determining compliance with the national air quality standards. Because this process is labor-intensive, measurements have not been made every day at every station. At most stations, they were made either on a 1-in-3 or 1-in-6 day schedule. More recently, instruments have been developed to measure PM continuously; the Air District employs a continuous method known as a Beta-Attenuation Monitor (BAM).

The Air District began measuring PM10 concentrations at a number of sites in 1989 on a 1-in-6 day schedule. In addition to total PM10 concentrations, a set of ions has been measured: nitrate, sulfate, ammonium, and chloride. Potassium was added in 1995, and elemental and organic carbon were added in 2004. The District began measuring PM2.5 in 1999. In recent years, the number of sites measuring PM10 has been reduced, as US EPA guidelines have placed greater emphasis on the need to monitor PM2.5.The Bay Area PM monitoring network meets and exceeds both state and US EPA requirements. The network provides data to measure regional PM levels relative to state and national standards. The network includes 8 sites which measure PM10 and 13 sites which measure PM2.5. There are three categories of PM monitors:

- Filter-based Federal Reference Method (FRM);
- Real-time Beta-Attenuation Monitor (BAM) monitors designated as Federal Equivalent Method (hereafter referred to as FEM/BAM); and
- Ordinary Beta-Attenuation Monitors that are not designated FEM (hereafter referred to as BAM).

Ten PM2.5 monitoring sites provide data to determine whether the Bay Area meets national PM2.5 standards. This includes eight FEM/BAM sites that monitor PM2.5 on a continuous basis, as well as two FRM (Federal Reference Method) sites that use filters to measure PM2.5 on a schedule based on the location's PM2.5 level relative to the national standard.[1] The PM2.5 network includes four additional sites with BAM monitors that are used (in conjunction with the FEM and FRM sites) in determining whether the Bay Area attains State PM2.5 standards. (These four BAM sites are not included for purposes of determining compliance with federal PM2.5 standards.)

In addition to the sites used to determine compliance with State and federal PM2.5 standards, the Air District also operates SASS (Speciation Air Sampler System) instruments at four sites (San Jose, Vallejo, Livermore, and West Oakland) that provide PM2.5 speciation data; these speciation data are used to analyze PM by chemical type and emissions source category in order to refine the Air District's PM emissions inventory and to help identify emission source categories that may warrant additional control measures. The speciation data provided by these four monitors are the source for the information provided in Figures 2-4 and 2-5 in Section 2. The Air District also operates one mobile sampling van, as well as one re-locatable trailer-mounted unit that can be deployed to monitor local concentrations for special purpose monitoring studies lasting a minimum of one year. For example, the mobile van provided measurements used in the 2010 *West Oakland Monitoring Study* described below.

The Air District's current PM monitoring sites are shown in Table 3-1.

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[1] The schedule can be every day, once every three days, or once every six days depending on the site and season.

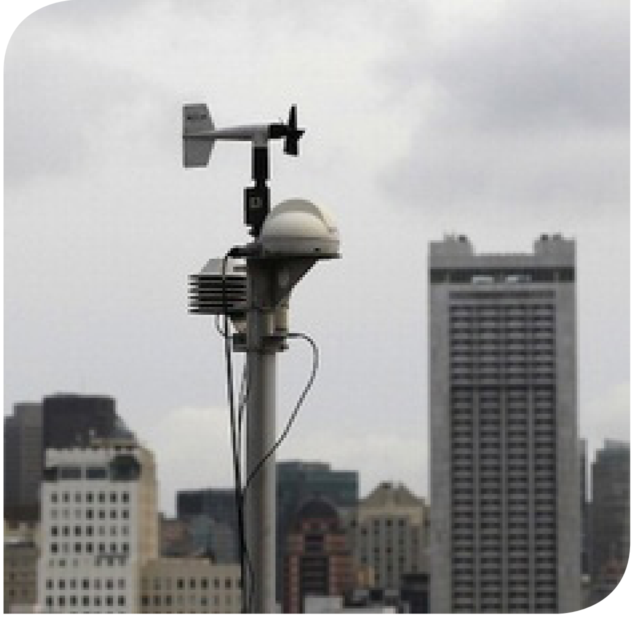
Table 3-1 **Bay Area PM Monitoring Sites**

Location	PM10	PM2.5	Type of Monitor
Livermore		√	FEM/BAM & Speciation Sampler
Oakland		√	FEM/BAM
Concord	√	√	FRM
Bethel Island	√		
San Rafael	√	√	FEM/BAM
San Francisco	√	√	FEM/BAM
Redwood City		√	FEM/BAM
Gilroy		√	FEM/BAM
San Jose	√	√	FRM, BAM & Speciation Sampler
San Pablo	√		
Vallejo		√	FEM/BAM & Speciation Sampler
Santa Rosa		√	FEM/BAM
Cupertino	√	√	BAM
Napa	√	√	BAM
West Oakland		√	BAM & Speciation Sampler

Measuring PM Concentrations at the Local Scale

The primary objective of the Bay Area air quality monitoring network for PM is to measure ambient PM levels at the regional scale in comparison to State and federal PM standards. The network is not intended to measure ambient concentrations of PM and other air pollutants at a fine-grain local scale. Also, while the network can measure PM impacts from larger-scale incidents such as wildfires, it is not designed to measure localized PM impacts from short-term incidents and episodes at a specific facility or source. However, concentrations of pollutants such as PM, carbon monoxide, and air toxics can vary greatly at the local scale. Among its limitations, for example, the existing regional monitoring network cannot accurately measure the local impacts of residential wood-burning on ambient PM concentrations due to the highly localized and variable nature of this activity.

The Air District recognizes that information about local PM levels is needed in order to identify impacted areas and develop strategies to reduce PM concentrations in such areas. Since it is neither technically nor financially feasible to install and operate PM monitors in every neighborhood, the Air District has been working to estimate PM concentrations and population exposure at the local scale by means of computerized photochemical modeling. This approach has been used to develop local estimates of PM concentrations and population exposure in support of the District's Community Air Risk Evaluation (CARE) program, as well as two pilot Community Risk Reductions Plans (CRRPs) that are under development in partnership with the cities of San Francisco and San Jose.



The Air District has also performed special monitoring to develop better measurements of PM and other pollutants in various locations with specific air quality issues or concerns. Monitoring was performed in Berkeley from December 2007 through December 2009 in proximity to Pacific Steel Casting; in Benicia from April 2007 through December 2008 in proximity to the Valero oil refinery; and in Cupertino beginning in September 2010 (scheduled to run through December 2012) in proximity to the Lehigh cement plant, and in areas, such as West Oakland, using its mobile van. Several examples of localized studies of PM that were extremely helpful in quantifying PM levelsin communities believed to be significantly impacted by PM are briefly described below.

The Air District sponsored Desert Research Institute to assist with the West Oakland Monitoring Study (WOMS) [1] developing and analyzing air quality monitoring data for the West Oakland area during two seasonal periods of four weeks in summer 2009 and winter 2009/10. The WOMS data were used by the District to evaluate local-scale dispersion modeling of diesel emissions and other toxic air contaminants for the area within and around the Port of Oakland. The monitoring data showed spatial patterns of higher pollutant concentrations that were generally consistent with proximity to vehicle traffic. Concentrations of directly-emitted pollutants were highest on heavily traveled roads with consistently lower concentrations away from the roadways.

The Air District also measured PM2.5 and PM10 and analyzed concentrations of certain metals and other chemical species in the ambient air of West Oakland from August 2009 through July 2010. The goals were to look for how these concentrations vary spatially within West Oakland and specifically in the neighborhood of Custom Alloy Scrap Sales (CASS), and to look for evidence of elevated metals concentrations in the West Oakland area. PM2.5 filters were collected at 7 sites as part of the West Oakland Monitoring Study for a month in the summer of 2009 and the winter of 2009-10. PM10 filters were collected at four sites near CASS from August 2009 through July 2010. The monitoring did not reveal a clear signature from CASS. The estimated cancer risk from measured metals concentrations

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[1] West Oakland Monitoring Study, Desert Research Institute, 2010.
See: www.baaqmd.gov/Divisions/Planning-and-Research/CARE-Program/CARE-Documents.aspx

was less than 10 in a million, considerably less than from diesel exhaust. The concentrations are all within the corresponding reference exposure levels (RELs) for morbidity effects.

Air District staff is currently working to identify several sites to monitor near-roadway levels of nitrogen dioxide (NO2), as required by EPA regulations. NO2 monitors are expected to be installed in near-roadway environments in San Francisco, San Jose, and the I-80 or I-880 corridor in Alameda County. The Air District may install PM2.5 and/or ultrafine PM monitors at one or more of these sites.

To expand the Bay Area monitoring network, one possibility may be to deploy smaller and less expensive monitoring units to supplement the PM monitors that comprise the Air District's official PM2.5 monitors, provided that equipment and personnel funds can be secured to purchase and operate such units.

Measuring Ultrafine PM

As noted in Section 1-A, evidence suggests that ultrafine particles may be especially harmful to public health. However, measuring ultrafine particles (UFPM) presents unique challenges. Due to their extremely small size, conventional technologies are not well-suited to measuring ultrafine particles. PM2.5 and PM10 monitors measure the mass of particles in a given volume of air; however, UFPM is negligible on a mass basis. Therefore, UFPM measurements usually count the **number** of particles rather than the particle **mass**. Measuring UFPM is especially difficult because many of the particles are actually smaller than the wave length of light. Only in recent years has measurement technology progressed such that the size distribution of nanometer-size particles can be measured in the atmosphere. Current methods to measure UFPM typically expose the particles to water vapor to make them grow large enough that they can be counted. Although several UFPM monitoring devices are currently available, technologies are still evolving, equipment and maintenance costs are relatively high, and accuracy and dependability of the devices can be an issue.

There are currently no State or national requirements for monitoring ambient concentrations of ultrafine PM. Most of the ultrafine particle UFPM monitoring performed to date has occurred in the Los Angeles area; UFPM monitoring in the LA area has focused on measuring ultrafine particles in close proximity to major roadways. (See discussion of near-roadway measurements in Section 1-B.)

In spring 2012 the Air District purchased and installed UFPM particle counters (TSI EPC 3783) at three sites in Santa Rosa, Redwood City and Livermore. An additional UFPM counter on loan to the District has been in operation in San Pablo; the District is in the process of purchasing this monitor, so that it will continue to provide monitoring data for the San Pablo area. The Air District also plans to install an ultrafine particle counter in conjunction with the near-roadway NO2 monitor described above, once the location for that monitor has been finalized. These UFPM monitors will provide data on ambient concentrations of UFPM at the regional scale and on a near-roadway basis. The data from these monitors will be used to track progress in reducing ultrafine particle concentrations in the Bay Area and to inform the District's future UFPM computer modeling efforts.

In addition, measurements of ambient levels of UFPM levels for the Bay Area (and many other urban areas) is needed to provide data that epidemiologists can use to study the health effects of exposure to UFPM.

Although the monitors described above should be useful for purposes of determining baseline concentrations of ambient UFPM in the Bay Area, using traditional air quality monitoring networks to measure UFPM may be of limited value, given the great spatial and temporal variability exhibited by UFPM. To adequately measure the great variability in UFPM levels, air quality agencies may need to explore the use of smaller, cheaper devices (perhaps including personal monitors) that can be deployed more densely on a neighborhood scale.

Challenges with PM Monitoring: Air quality monitoring stations are expensive to build and maintain. Due to budgetary constraints in recent years, the Air District is currently hard-pressed to operate its existing air quality monitoring network. Expanding the monitoring network would entail securing funds for the initial capital cost to purchase monitoring equipment, finding good locations that meet applicable criteria, and deploying human resources to operate and maintain the sites on an on-going basis. As the Air District continues its technical work to develop a better understanding of the dynamics and distribution of PM in the atmosphere by means of computer modeling and special studies, this should enable the Air District to deploy its limited monitoring resources so as to measure ambient concentrations and population exposure to PM in the most cost-effective manner.

As mentioned above, the PM monitoring network is not designed to measure localized PM impacts from short-term incidents and episodes at a specific facility or source. Therefore, the Air District is investigating the possibility of augmenting the current network with incident response capabilities that would allow for accurate, real-time, mobile measurement of localized PM impacts from short-term episodes.

See Section 5 for additional discussion regarding future directions in monitoring ambient PM concentrations.

SECTION 3-B: PM STANDARDS AND PM PLANNING REQUIREMENTS

To protect public health and welfare, US EPA and the California Air Resources Board have both adopted **ambient air quality standards** for particulate matter. The federal Clean Air Act requires the US EPA Administrator to adopt standards for six “criteria pollutants”, including PM, with an “adequate margin of safety to protect public health.” EPA is charged with reviewing the standards every five years based on the latest scientific research on health and welfare effects, and considering recommendations provided by an expert panel called the Clean Air Scientific Advisory Committee (CASAC). PM standards have evolved and become more stringent over the past several decades in response to better understanding of the negative effects of PM on public health. In addition to **primary standards** which are designed to protect public health, U.S. EPA also issues **secondary standards** for PM to protect “public welfare”, including visibility (clarity of the air), flora, fauna, and the built environment. The national secondary standards for PM are currently set at the same level as the primary standards.

PM Standards

Ambient air quality standards are based on three key elements: the averaging time period (e.g., 24-hour or annual); the form of the standard; and the level of the standard.

Annual average standards are intended to protect public health from chronic (long-term) health impacts related to PM. EPA adopted an annual average PM2.5 standard of 15 µg/m^3 in 1997. In 2002, the State of California adopted a (more protective) annual average standard of 12 µg/m^3 in 2002. In June 2102, EPA proposed to lower the national annual standard to a value in the range of 12 to 13 µg/m^3, as discussed below.

24-hour standards are intended to protect public health from acute (short-term) health impacts related to PM. In 2006, EPA significantly tightened the 24-hour PM2.5 NAAQS from 65 µg/m^3 to 35 µg/m^3. The State of California has not yet adopted a short-term 24-hour PM standard.

For criteria pollutants, the level of the standard is generally defined in terms of the **ambient concentration** of a pollutant in outdoor air, as expressed in terms of either a **parts per million** ratio (e.g., the state 8-hour ozone standard is 0.070 parts per million) or a **mass per volume** basis. For example, the national 24-hour PM2.5 standard is 35 mg/m^3, or micrograms per cubic meter (one microgram equals one-millionth of a gram). State and national PM standards for PM2.5 and PM10 are based on the mass (i.e., the total weight), rather than the number, of particles suspended in the air.

Design Value: Determining whether an air basin attains a given air quality standard requires comparing ambient pollutant levels with the standard to calculate the region's **design value**.[2] (For purposes of State standards, the term **designation value** is used.) The design value is calculated for each station in the official monitoring network (See Section 3-A for a description of the Bay Area PM monitoring network). A region meets the standard only if the design value for each and every official monitoring site does not exceed the standard. The stringency of an air quality standard depends upon (1) the numerical threshold and (2) the **form of the standard** which specifies the method and statistical protocol used to calculate the design value. The form of a standard may allow each region to exceed the standard on a limited number of occasions over a given time period. For example, the design value for the national 24-hour PM2.5 standard for any site is defined as the site's annual 98th percentile PM measurements averaged over a three-year period; thus, a site may exceed the standard on a limited number of days and still attain the standard. The basic steps to calculate the design value for the national PM2.5 standards are shown in Table 3-2.

The PM standards established by the State of California are more difficult to attain than the national standards; not only are the State standards set at lower numerical thresholds, but also they have a more stringent form of the standard. The State 24-hour PM10 standard allows fewer exceedances in order to remain in attainment. The State annual standard for PM2.5 is more stringent because it is based on the **maximum** of three annual averages, rather than the **average** of three annual averages). The design values that determine whether the Bay Area attains the various PM standards are calculated using measurements of ambient PM concentrations from the regional monitoring network described in Section 3-A.

Table 3-2 Basic Design Value Calculation Method for National PM2.5 Standards

Averaging Period	Ambient Concentration	Calculation Method – performed for each official monitoring station
24-hour	35 µg/m³	**Step 1:** Determine the 98th percentile value for each year over a consecutive three year period. (In practice this means that the seven highest values per year are excluded.) **Step 2:** Average the three 98th percentile values. **Step 3:** Round the resulting value to the nearest 1.0 µg/m³. **Step 4:** Compare the result to the standard.
Annual	15.0 µg/m³	**Step 1:** Calculate the average of each quarter of each year over a three year period. **Step 2:** Average the four quarters in a calendar year to determine the average for each year. **Step 3:** Average the three annual values. **Step 4:** Round the resulting value to the nearest 0.1 µg/m³. **Step 5:** Compare the result to the standard.

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[2] Details on how design values are calculated are provided in 40 Code of Federal Regulations (CFR) Part 50 Appendix N, and the April 1999 EPA document Guideline on Data Handling Conventions for the PM NAAQS. See http://epa.gov/ttncaaa1/t1/memoranda/pmfinal.pdf.

Bay Area Attainment Status for Current PM Standards

The current State and national standards for PM2.5 and PM10, the Bay Area's attainment status relative to those standards, and the region's design value for each standard, are summarized in Table 3-3. PM standards recommended by the World Health Organization (WHO) in 2005 are also shown for purposes of comparison; the national PM standards issued by US EPA are less stringent than the recommended WHO guidelines. Despite increasing concern about the health impacts of ultra-fine particles, as yet there are no State or national ambient air quality standards for ultra-fine PM.

The Bay Area attainment status shown in Table 3-3 is based on the current formal designation by US EPA or ARB. However, although the Bay Area is formally designated as non-attainment for these standards, monitoring data shows that the region currently meets the national 24-hour PM2.5 standard, the State annual PM2.5 standard, and the State annual PM10 standard, as indicated by the fact that the design value is less than or equal to the standard in each case. For purposes of attainment status, although monitoring data for an air basin may show that it meets a standard, once a region has been designated as non-attainment, it is still formally designated as non-attainment until such time as the region submits a redesignation request and maintenance plan which is approved by EPA.

National standards: The Bay Area attains the national 24-hour PM10 standard and the national annual PM2.5 standard. The region's design value for both these standards is well below the threshold. The national 24hour PM2.5 standard was tightened to 35 mg/m^3 in 2006. The Bay Area's attainment status for this standard is explained below in Federal PM Planning Requirements for the Bay Area.

State standards: The California Air Resources Board has adopted PM standards that are more stringent (health-protective) that the national standards. The most recent monitoring data (through 2011) demonstrates that the Bay Area continues to meet the State annual average PM2.5 standard. Recent data also shows that the Bay Area has attained the State annual PM10 standard for the first time ever, based on data for the 2009-2011 period. The Bay Area does not attain the State 24-hour PM10 standard; however, the region's design value for this standard has been decreasing in recent years, a sign that we are making progress toward attaining this standard.

Table 3-3 PM Standards, Bay Area Attainment Status, and Design Values

Averaging Time	National / California	Standard	Bay Area Design Value *	Attainment Status	World Health Organization PM Guidelines
Pollutant: PM2.5					
24-hour	National	35 µg/m³	30 µg/m³	Non-attainment	25 µg/m³
Annual	National	15 µg/m³	10.3 µg/m³	Attainment	10 µg/m³
	California	12 µg/m³	10.4 µg/m³	Non-attainment	
Pollutant: PM10					
24-hour	National	150 µg/m³	72 µg/m³	Unclassified	50 µg/m³
	California	50 µg/m³	70 µg/m³	Non-attainment	
Annual	California	20 µg/m³	20 µg/m³	Non-attainment	20 µg/m³

* Design values are calculated based on PM monitoring data thru year 2011.

Federal PM Planning Requirements for the Bay Area

Any state or region that fails to attain the national standard for any criteria pollutant is required to submit a State Implementation Plan (SIP) to U.S EPA to demonstrate how it will reduce ambient concentrations in order to attain the national standard. U.S. EPA designated the Bay Area as "non-attainment" for the revised 24-hour PM2.5 national standard in December 2009, based on air quality monitoring data for the three-year period 2006-2008. Areas designated as non-attainment for the revised standard, including the Bay Area, are required to submit a PM2.5 SIP to U.S. EPA by December 2012 to show how they will attain the standard by December 2014.

Although the Bay Area was designated as non-attainment for the national 24-hour PM2.5 standard based on monitoring data for the 2006-2008 period, the region exceeded the standard by only a slight margin. Since then, Bay Area PM2.5 levels have continued to decline. Air quality data from the regional monitoring network shows that the Bay Area met the national 24-hour PM2.5 standard during the three-year period from 2008 through 2010, as well as the three-year period from 2009 through 2011.

Under US EPA guidelines, a region with monitoring data showing that it currently attains an air quality standard can submit a "redesignation request" and a "maintenance plan" in lieu of a SIP attainment plan. However, the Air District believes that it would be premature to submit a PM2.5 redesignation request for the Bay Area at this time. Instead, the Air District is pursuing another option provided by US EPA guidelines for areas with monitoring data showing that they currently meet the PM2.5 standard. In December 2011, the Air Resources Board submitted a "clean data finding" request on behalf of the Bay Area. This request is currently under review by EPA. If EPA verifies that monitoring data shows that the Bay Area currently meets the standard (i.e., has “clean data”), then EPA will suspend the SIP provisions that apply to preparing an attainment plan to demonstrate how the region will attain the standard by the specified target date. These SIP provisions will remain suspended as long as Bay Area monitoring data continues to show compliance with the standard. Although the SIP requirements related to demonstrating attainment would be suspended, the region will still be required to submit a “clean data” SIP consisting of the following elements:

- Amendments to the Air District’s New Source Review (NSR) regulations to address PM2.5; and
- An emissions inventory for PM2.5 for the attainment year: i.e. the year in which monitoring data shows that the Bay Area first achieved attainment. For the Bay Area, the attainment year is 2010. The inventory must include both primary PM2.5 emissions by source category, as well as precursors to secondary PM formation.

The Air District is preparing these required SIP elements for submittal to US EPA in fall 2012.

State PM Planning Requirements

The California Clean Air Act of 1988, the primary legislation that defines State air quality planning requirements, is focused primarily on reducing ground-level ozone. The California Clean Air Act does not require that local air districts prepare plans to reduce PM. In response to concern about the health impacts of PM, in 2003 the State legislature enacted SB 656 (codified as Health & Safety Code Section 39614); this legislation required ARB and local air districts to evaluate potential PM control measures and to develop a PM implementation schedule for appropriate PM-reduction measures. The Air District complied with this legislation; staff developed a Particulate Matter Implementation Schedule that was adopted by the Air District’s Board of Directors in November 2005. The three measures identified in the PM Implementation Schedule have been adopted and implemented: stationary internal combustion engines (Regulation 9-8); commercial charbroiling operations (Regulation 6-2); and the residential wood-burning rule (Regulation 6-3) which is further described in Section 4.

The SB 656 legislation sunset on January 1, 2011 and is therefore no longer in effect. Thus, despite the fact that State PM standards are more stringent than the national standards, formal PM planning efforts in California are governed primarily by the national standards and the SIP process describe above.

Proposed Revisions to National PM Standards

As noted above, EPA is required to review the national standards for PM and other criteria pollutants every five years based on the latest scientific research on health and welfare effects. After reviewing the evidence, EPA issued a draft proposal on the national PM standards for public comment on June 14, 2012. EPA proposes to:

- Strengthen the annual health standard for fine particles (PM2.5) by setting the standard at a level within the range of 12 µg/m^3 to 13 µg/m^3.
- Retain the existing 24–hour PM2.5 standard of 35 µg/m^3.
- Set a separate PM standard to improve visibility in urban areas, as discussed below.
- Retain existing secondary standards for PM2.5 and PM10 identical to primary standards to provide protection against other effects, such as ecological effects, effects on materials, and climate impacts.
- Retain the existing 24-hour standard for PM10 of 150 µg/m^3; this standard has been in place since 1987.

The proposed urban visibility standard would provide increased protection from particle-induced haze. The standard would measure visibility on the basis of light extinction as expressed in units called **deciviews**. Each deciview represents a constant change in visual air quality, with zero deciviews representing the most pristine conditions. EPA is proposing a 24-hour averaging time, a 90th percentile form averaged over 3 years, and a visibility level set at either 28 or 30 deciviews. Because monitors to directly measure visibility are not currently available, EPA proposes to use data on speciated PM2.5 mass concentrations as well as relative humidity, in conjunction with an algorithm, to calculate PM2.5 light extinction.

The Air District provided comments at a public hearing on the proposed standards in Sacramento on July 19, 2012, as well as written comments, urging EPA to adopt the most health-protective PM standards.

After reviewing public comments on the proposed revisions to the PM standards summarized above, EPA will issue final standards by December 14, 2012. Revisions to the current standards will trigger a process to evaluate monitoring data and issue new attainment designations for air basins throughout the nation. EPA expects to make attainment designations based on the revised standards by December 2014. A preliminary, unofficial review of Bay Area monitoring data for years 2008 through 2011 indicates that the region is likely to attain a more stringent annual standard set at either the 12 µg/m^3 or the 13 µg/m^3 level, provided that recent ambient PM2.5 concentrations prevail in future years.

SECTION 3-C: TRENDS IN AMBIENT PM CONCENTRATIONS

Trends in ambient PM concentrations can be analyzed in terms of **design value** (as explained in Section 3-B), as well as the number of days the region exceeds the PM standard. The Bay Area has been making progress in reducing PM levels as measured by both of these metrics. The graphs in Figures 3-1, 3-2, and 3-3 below show trends for ambient concentrations of PM2.5 and PM10. Trends in ambient PM10 concentrations are shown since the late 1980's, and trends in PM2.5 are shown since 1999, because these are the years for which monitoring data are available. The information presented here is derived from a more detailed report entitled *Trends in Bay Area Ambient Particulates* (BAAQMD, November 2011). The November 2011 *Trends* report also provides analysis showing trends in reducing several specific PM components, including nitrate, sulfate, potassium, elemental carbon, and organic carbon.

PM2.5 Trends

Figure 3-1 shows the trend in the District's design value relative to the national annual PM2.5 standard of 15 μg/m^3. The annual design value is the maximum of design values from individual PM2.5 sites. The annual design value for a particular site is the 3-year average of its quarterly averaged annual mean PM2.5 concentrations. The design values are marked at the third year of three year averages. The District's annual design value decreased from 14 μg/m^3 for 1999-2001 to 10 μg/m^3 for 2009-2011, a 28% reduction. As can be seen, the Bay Area met the national annual PM2.5 standard during the entire period.

Figure 3-1 Bay Area PM2.5 Annual Design Value 1999-2001 through 2009-2011

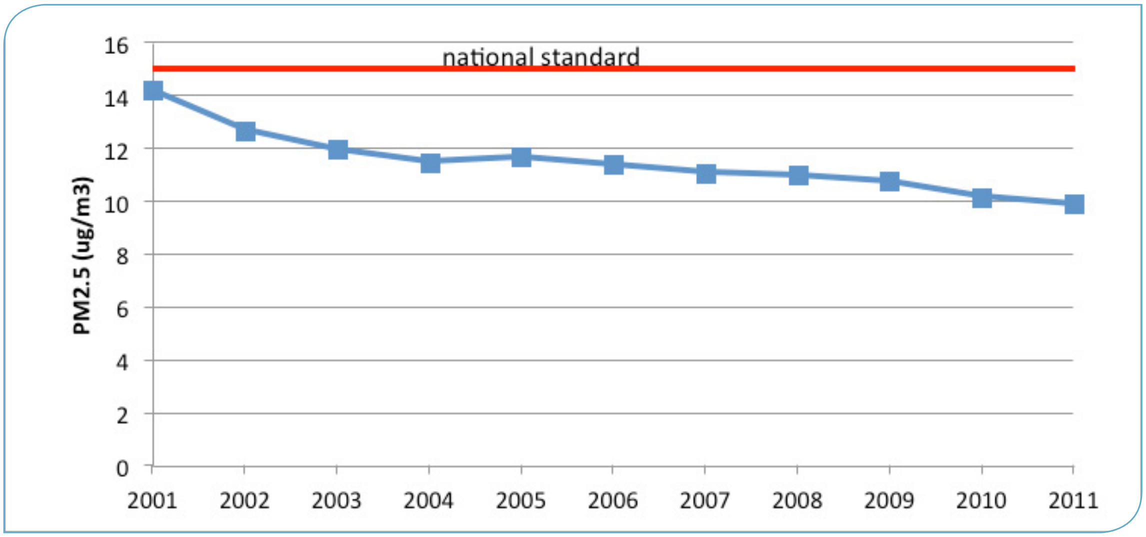


Figure 3-2 shows the District's design values relative to the national 24-hour PM2.5 standard. For the earliest period available, 1999-2001, the design value was 57 μg/m^3. By 2009-2011, it had declined to 30 µg/m³, well below the 35 µg/m³ standard. The reduction from 1999-2001 to 2009-2011 was approximately 46%.

Figure 3-2 Bay Area Design Values for the 24-Hour Standard 1999-2001 through 2009-2011

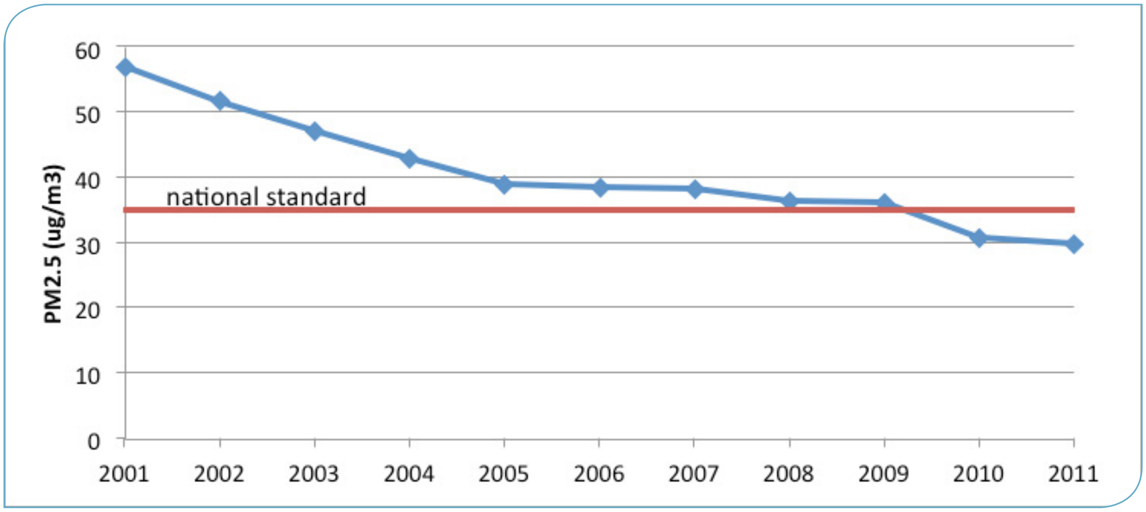
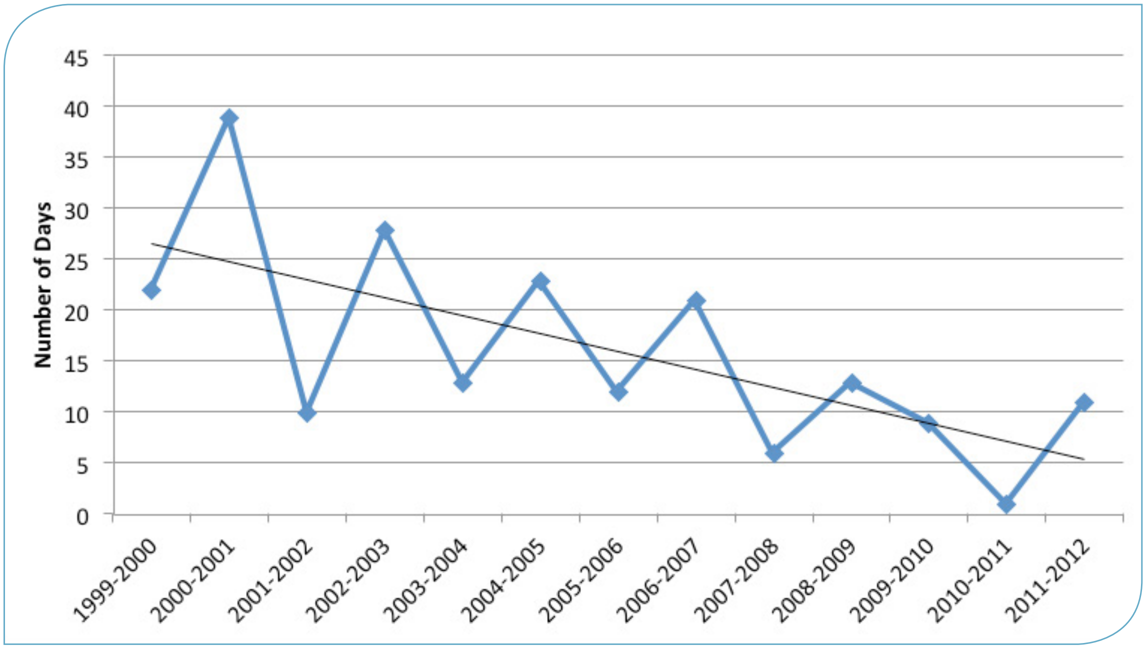


Figure 3-3 shows an overall downward trend in the number of days that Bay Area PM2.5 levels exceeded the 35 µg/m³ standard for each winter from 1999-2000 through 2011-2012. (Although the 35 µg/m³ standard did not take effect until 2006, the number of exceedance days per year is shown as if the 35 µg/m³ standard had been in effect for the entire period shown.) The overall downward trend reflects the reduction in PM emissions in response to ARB and Air District control measures, whereas the sawtooth pattern in the number of exceedances is primarily due to year-to-year variation in meteorology, rather than short-term changes in emissions.

Figure 3-3 Bay Area PM2.5 Exceedances by Winter Number of days exceeding the 24-hour NAAQS, November 15 – February 15



Not only is the Bay Area experiencing fewer exceedance days per year, but when exceedances do occur, they are generally less severe. Table 3-4 shows the number of exceedances per year relative to the national 24-hour PM2.5 standard adopted in 2006. Both the number of exceedance days per year has declined, as well as the average value of the exceedances that occur in a given year. The average PM2.5 exceedance value has decreased from over 50 µg/m^3 in the 1999-2002 period to approximately 40 µg/m^3 in the 2009-2011 period. The combination of fewer exceedance days and lower peak values on the days when an exceedance of the standard does occur translates into reduced population exposure to unhealthy PM2.5 levels for Bay Area residents.

Table 3-4 Exceedances of 24-Hour PM2.5 (35 µg/m^3) [3]

Year	Number of Exceedances	Mean Exceedance Value (µg/m3)
1999	29	52.2
2000	28	50.8
2001	17	61.1
2002	28	51.5
2003	16	42.0
2004	20	43.0
2005	21	41.9
2006	10	46.8
2007	14	44.1
2008	12	42.4
2009	11	38.2
2010	6	40.3
2011	8	40.5

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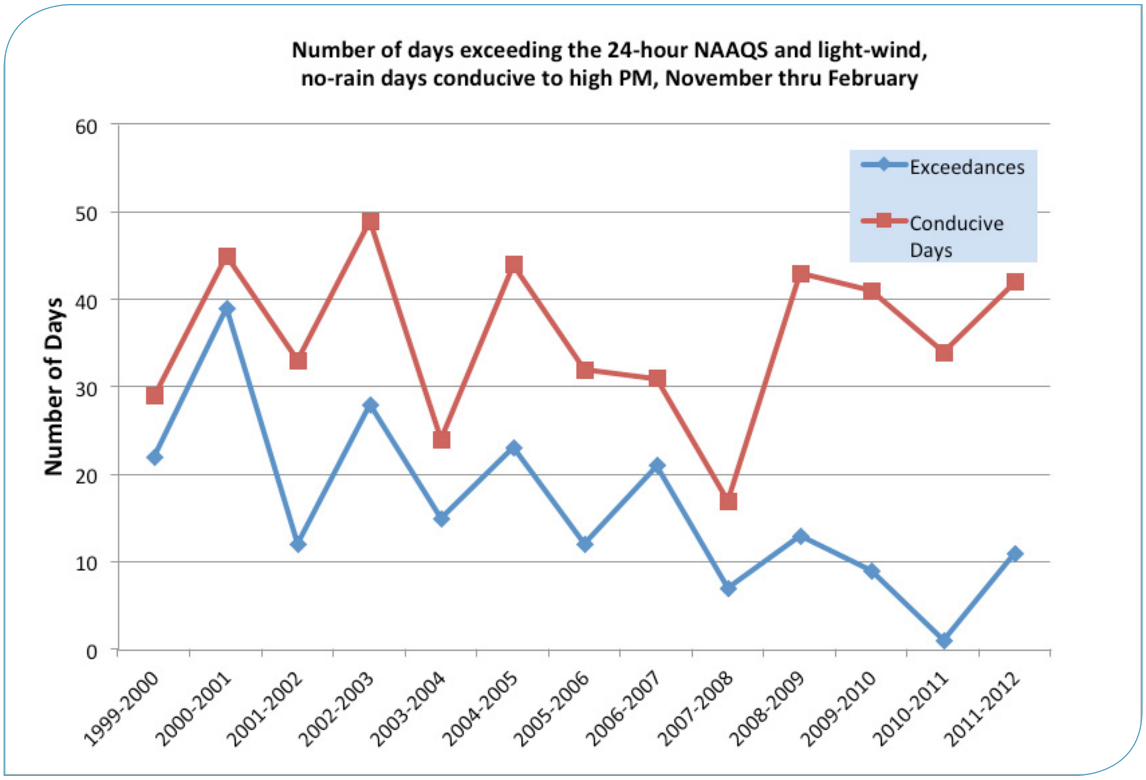
[3] Please note that the data shown in Table 3-4 is reported on a calendar year basis, whereas the data presented in Figures 3-3 and 3-4 is based on the winter season.

Relationship Between Meteorology and Exceedances of the 24-Hour PM2.5 Standard

Many factors affect PM ambient concentrations. Although emissions of primary PM and the precursor pollutants that contribute to secondary PM formation appear to be declining, meteorological factors (temperature, humidity, wind speed and direction) that affect PM build-up and transport have a strong impact on PM levels on a day-to-day basis. For example, analysis shows that winter periods of three or more days with light winds and no rain are conducive to build-up of PM, as discussed more fully in Section 2. Figure 3-4 shows that there is a clear correlation between the number of "PM-conducive" days (defined here as winter days with light wind and no rain) and the number of days that the Bay Area exceeds the national 24-hour PM2.5 standard in a given year.

Figure 3-4 Bay Area PM2.5 Exceedances and Conducive Days by Winter



Comparison with PM2.5 Trends in the Central Valley

Since there is considerable air flow between the Bay Area and the Central Valley – and vice versa – it is instructive to compare the current PM levels and design value trends among the three major central California districts: that is, Bay Area, Sacramento, and San Joaquin Valley. Figure 3-5 shows the trend in Bay Area design values for the national annual PM2.5 standard of 15 µg/m^3 compared with trends in the Central Valley. The trends in the Bay Area and Sacramento are similar, with an average reduction of 2.8% per year for the Bay Area and 1.9% per year for Sacramento. There is no clear trend for the San Joaquin Valley.

Figure 3-5 Design Values for Annual PM2.5 Standard for 3 Central California Air Basins

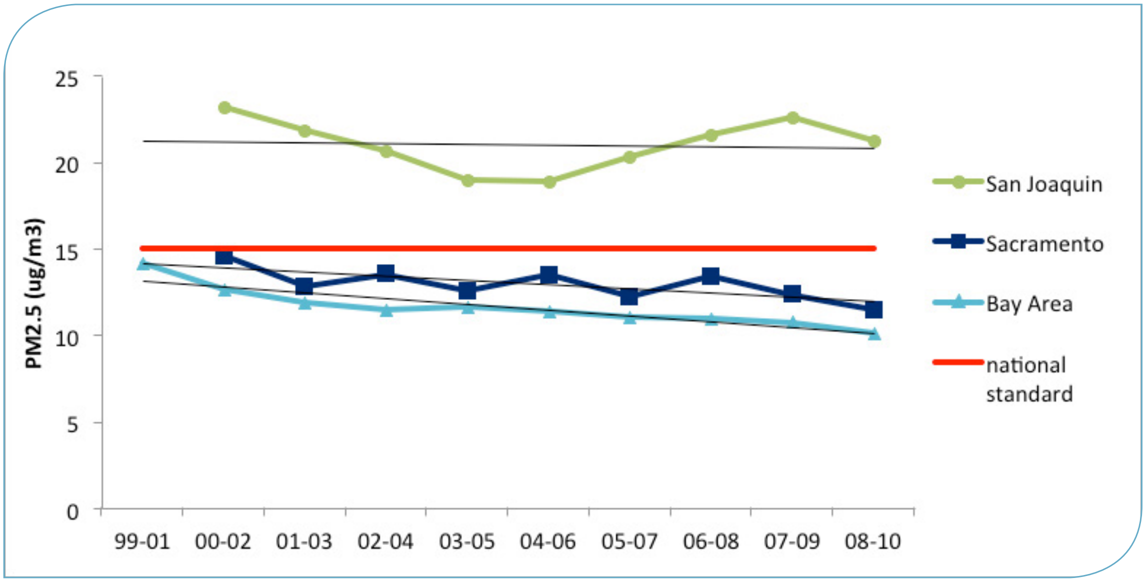
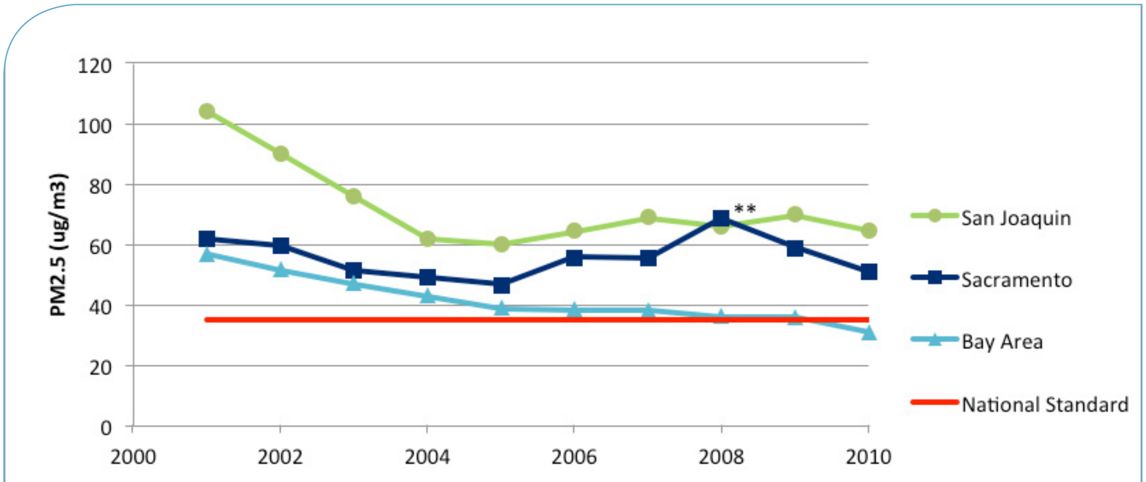


Figure 3-6 shows the design values for the national 24-hour PM2.5 standard of 35 µg/m^3 for the three air basins. The Bay Area's design value has decreased 5.8% per year from 2000 to 2010, and met the standard for 2008-2010. The design value in the San Joaquin Valley shows a decrease of 3.8% per year. Sacramento's design value shows no decrease, partly because of the 2008 wildfires. Excluding the wildfire months of June and July 2008, the decrease is 3.3% per year.

Figure 3-6 Design Values* for 24-Hour PM2.5 Standard for 3 Central California Air Basins



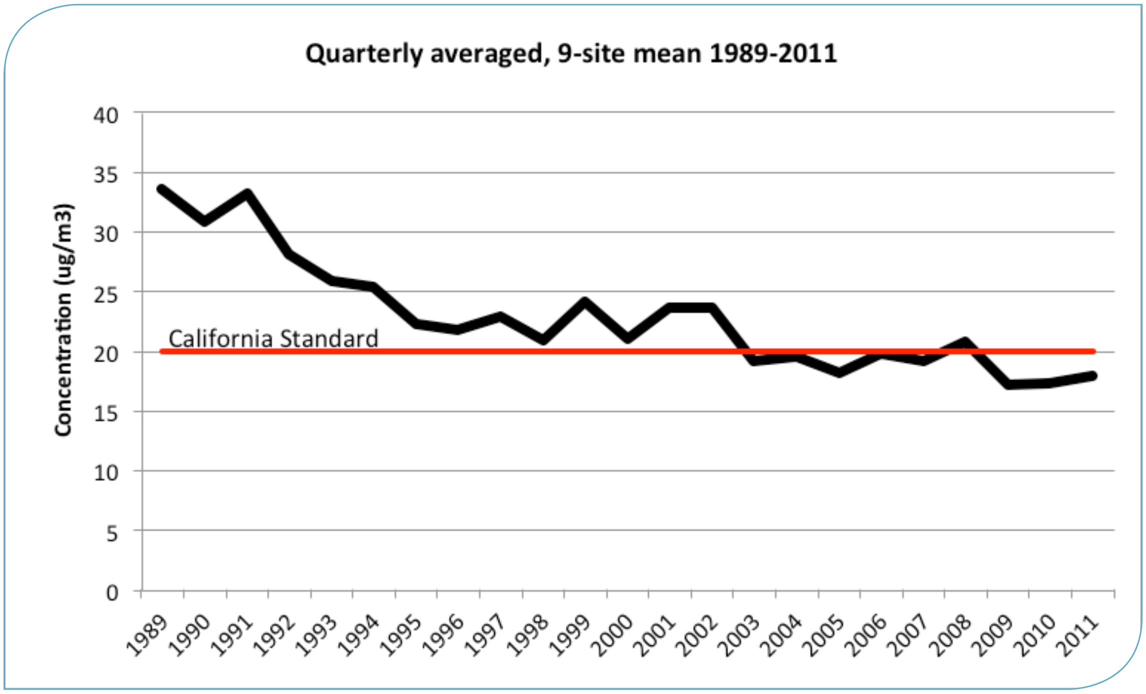
* Design values are 3-year averages of 98th percentiles, plotted vs. most recent year.
**Values for 2008-2010 influenced by wildfires in summer 2008.

PM10 Trend

Bay Area PM10 levels have decreased significantly since 1990; peak concentrations have declined by approximately half and annual average values have declined by about one-third. Figure 3-7 shows quarterly-averaged annual PM10 concentrations from 1989 through 2011 relative to the State annual standard. (There is no national annual PM10 standard.) The solid line shows the average, based upon data from nine PM10 monitoring sites. The reductions were approximately 3%

per year in the 1990s and 2% per year from 2000 through 2011. The average decreased from 33 µg/m3 in 1989-1991 to 17 µg/m3 in 2009-2011, a reduction of 46%.

Figure 3-7 Bay Area Annual Mean PM10



In summary, we have made substantial progress in reducing PM levels in the Bay Area, but further reductions in PM would provide additional benefit by reducing the negative health impacts of PM described in Section 1-A.

SECTION 4: SUMMARY OF PM CONTROL PROGRAM

This chapter summarizes the existing regulations, policies, and programs that the Air District, the California Air Resources Board, and the US Environmental Protection Agency are implementing to reduce emissions of primary PM and PM precursors, and to reduce population exposure to PM.

Because primary PM and PM precursors are emitted by a wide range of stationary and mobile sources, a comprehensive and multi-faceted effort is needed to reduce ambient PM levels. As described in Chapter Section 1-A, there are negative health effects caused by both acute (short-term) and chronic (long-term) exposure to PM. Therefore, PM control programs aim to reduce both short-term (peak) and long-term (annual average) PM concentrations in order to protect public health.

As discussed in Sections 2 and 3, emissions inventory data and air quality monitoring data demonstrate that PM emissions and ambient PM concentrations have been greatly reduced in the Bay Area in recent years. So even though the Air District has never been required to prepare a formal PM SIP attainment plan to date, this data offers tangible evidence that the PM control efforts implemented by the Air District and other agencies summarized in this chapter have been effective in reducing PM and related public health effects in the Bay Area.

Although the Bay Area has made substantial progress toward meeting State and national PM standards, the Air District recognizes that some communities are exposed to above average levels of PM and that some individuals are especially vulnerable to the negative health effects of PM. Therefore, in addition to reducing emissions of PM and its precursors at the regional scale, the PM control effort also focuses on the need to reduce population exposure to PM, especially in the most heavily impacted communities and among the most sensitive populations.

ARB PM Reduction Program

The California Air Resources Board (ARB) has played a vital role in reducing PM by regulating emissions of primary PM and PM precursors from most mobile sources, such as on-road cars and trucks as well as off-road equipment. ARB has pursued an aggressive program to reduce PM emissions from mobile sources throughout California over the past 15 years. ARB classified diesel PM as a toxic air contaminant (TAC) in 1998. In September 2000, ARB adopted a Diesel Risk Reduction Plan with a goal of 75 percent PM reduction by 2010 and 85 percent by 2020. ARB has adopted a comprehensive set of regulations to implement the Diesel Risk Reduction Plan (DRRP) and reduce population exposure to diesel PM. The ARB program to reduce emissions of diesel PM includes four key components:

- Regulations to reduce tailpipe emissions of primary PM and PM precursors from on-road and off-road mobile sources;
- Cleaner fuel, especially the requirement for ultra-low sulfur diesel;
- Restrictions on vehicle use, such as idling restrictions on diesel engines, and;
- Grants and incentives to encourage emission reductions over and above the regulatory requirements.

As summarized in Table 4-1, the heart of the DRRP is a set of regulations called **Air Toxics Control Measures** (ATCMs) to reduce diesel PM emissions from on-road trucks and buses; off-road equipment, including construction, farm and port equipment; harbor craft, and ocean-going ships. ARB has phased in the effective dates of these regulations in order to provide time for fleet owners to prepare and comply. These regulations will provide increasing benefit as they come into full effectiveness over the next 5-10 years. In addition to reducing emissions of primary PM, ARB regulations will reduce emissions of precursors that contribute to formation of secondary PM, such as ROG and NOx.

Table 4-1 ARB Diesel Air Toxic Control Measures for Heavy-Duty Vehicles, Equipment and Ships

Trucks and Buses	Since **2008**, idling limited to 5 minutes
	By **2016**, all trucks meet equivalent of 2007/2010 PM standard
	By **2023**, all trucks meet equivalent of 2010 NOx standard
Drayage Trucks	By **2010**, pre-MY 1994 trucks banned
	By **2010**, MY 1994-2003 trucks meet 2007/2010 PM standard
	By **2014**, all trucks meet 2007/2010 PM and 2007 NOx standards
	By **2023**, all trucks meet 2010 NOx standard
Public Fleet Vehicles	By **2012**, all vehicles meet equivalent of 2007/2010 PM standard
Garbage Trucks	By **2011**, all vehicles have installed Best Available Control Technology (BACT)
Transit Buses	By **2003**, met a NOx fleet average of 4.8 g/bhp-hr
	By **2007**, PM emissions reduced by 85% from 2002 baseline
	For fleets in the Bay Area with 200+ buses, 15% of new buses purchased from **2011-2026** must be zero emissions. (May be amended in 2012.)
Truck Refrigeration Units	By **2020**, engines must meet Ultra-Low Emission standard

Locomotives	In **2007**, begin using 15 ppm Sulfur fuel in California-based locomotives
	By **2008**, conduct health risk assessments for major rail yards
	By **2009**, install idling reduction devices on California based locomotives
Construction Equipment	Since June **2008**, idling limited to 5 minutes
	Between **2014 and 2023**, fleets with more than 5,000 total hp must meet fleet average NOx targets or turnover/replace 4.6-10% of fleet hp
	Between **2017 and 2023**, fleets with 2,501 to 5,000 total hp must meet fleet average NOx targets or turnover/replace 4.6-10% of fleet hp
	Between **2019 and 2029**, fleets with less than 2,501 total hp must meet fleet average NOx targets or turnover/replace 4.6-10% of fleet hp
Cargo Handling Equipment	By **2007**, new equipment meets equivalent of Tier 4 off-road engine standards or 2007 PM/NOx on-road engine standards
	By **2015**, pre-2007 yard trucks meet equivalent of Tier 4 off-road or 2007 PM/NOx on-road standards
	By **2017**, all other pre-2007 equipment must meet equivalent of Tier 4 off-road or 2007 PM/NOx on-road standards
Harbor Craft	Beginning **2009**, engines for new vessels or repowers meet Tier 2 or Tier 3 off-road standards; new ferries must be 85% below Tier 2 standards
	By **2016**, pre-2000 engines meet Tier 2,3 or 4 off-road standards
	By **2022**, all marine engines must meet Tier 2,3 or 4 off-road standards
Ships	In **2009**, ships began using Marine Diesel Oil (MDO) with 0.5% sulfur or Marine Gas Oil (MGO) with 1.5% sulfur. By August 2014, ships begin using MDO or MGO with 0.1% sulfur.
	By **2014**, 50% reduction in auxiliary engine use during 50% of visits by cruise and container ships (shore power)
	By **2017**, 70% reduction in auxiliary engine use during 70% of visits by cruise and container ships (shore power)
	By **2020**, 80% reduction in auxiliary engine use during 80% of visits by cruise and container ships (shore power)
Back-Up Generators (BUGs)	By **2008**, PM emissions for BUG’s reduced by 85% in new engines

Light-Duty Vehicle Emission & Fuel Economy Standards

Although ARB's program to reduce PM emissions from diesel engines may have stolen the spotlight, its efforts to control emission from light-duty vehicles (LDVs) and medium-duty vehicles (e.g. vans and pick-up trucks) have also provided important PM reduction benefits. These vehicles account for the vast majority of the 175 million miles that Bay Area residents drive every day. So even though PM emissions from gasoline-powered LDVs are very low on a per-mile basis, the combined emissions of primary PM from light-duty vehicles are significant. In addition, LDVs account for a major portion of ROG and NOx emissions, which are important precursors to the formation of secondary PM.

ARB's Low Emission Vehicle (LEV) program is the backbone of its effort to reduce emissions from light-duty vehicles. The LEV program has greatly reduced emissions of ROG, NOx, and PM from LDVs throughout the state. In its initial phase, LEV I regulations reduced emissions in model year 1994-2003 vehicles. The more stringent LEV II program, which took effect in model year 2004, continues to provide major air quality benefits. As discussed in Section 5, ARB is in the process of adopting a new iteration of the LEV program – LEV III – to further reduce pollution from LDVs.

ARB is in the process of finalizing proposed amendments to California's Low Emissions Vehicle (LEV) regulations to strengthen the LEV program. One element of the LEV III proposal is more stringent PM standards for passenger cars and light-duty trucks. Although PM emissions from new light-duty vehicles are already very low, ARB staff is aware that California and federal emission requirements to reduce greenhouse gas emissions have fostered development of advanced internal combustion technology such as gasoline direct injection engines (GDI). To encourage the continued development of GDI engines that emit PM at the same low levels as port fuel injection engines, the LEV III standards would reduce the PM standard from 0.010 grams per mile for passenger cars and light-duty trucks. These standards would be phased in from 2017 through 2021. The LEV III amendments propose to further reduce the PM standard to 0.001 grams per mile (one milligram per mile) to be phased in during the 2025 and 2028 period. At this 1 mg/mile emission rate, a car would emit a total of 150g or 1/3 of a pound of particulate matter over a typical lifetime of 150,000 miles. The LEV III provisions should help offset the potential increase in PM emissions from light-duty vehicles that would occur if vehicle miles of travel (VMT) in the Bay Area rise in response to population growth or other factors.

In addition to the LEV tailpipe emission standards which reduce emissions on a per-mile basis, ARB has also adopted fuel economy standards to implement the Pavley legislation[4]. Although the impetus for improved fuel economy is primarily to reduce carbon dioxide emissions that contribute to climate change, by reducing fossil fuel combustion these standards also provide important benefits in reducing criteria air pollutants and air toxics.

The ARB regulations summarized above are primarily responsible for the statewide reductions in PM emissions from mobile sources to date. These regulations are expected to provide continued reductions in emissions of primary PM and PM precursors from mobile sources over the next two decades.

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[4] State legislation enacted in 2002 (Assembly Bill 1493, Pavley) directed the ARB to adopt regulations to reduce greenhouse gases from passenger vehicles. For additional information, see www.arb.ca.gov/cc/ccms/ccms.htm

EPA Actions to Reduce PM from "Federal Sources"

In addition to establishing ambient air quality standards for PM, US EPA is responsible for establishing emission standards for aircraft engines, new locomotive engines and new non-road engines less than 175 horsepower used in construction or farm equipment. US EPA regulations help to reduce PM emissions from trucks, locomotives, and marine engines that operate across state boundaries when these vehicles or engines operate within California. These EPA actions complement the ARB mobile source regulations described above. A summary of US EPA programs and actions to control PM and PM precursors is available at US EPA at www.epa.gov/pm/links.html.

In 1998, US EPA adopted more stringent "Tier 2" and "Tier 3" emission standards for ROG, NOx, and PM from new non-road diesel engines. This program established the first emission standards for non-road diesel engines less than 50 horse-power (hp), including marine engines in this size range. The Tier 2 standards were phased in for all engine sizes from 2001 to 2006. More stringent Tier 3 standards for engines between 50 and 750 hp were phased in from 2006 to 2008. The Clean Air Non-Road Diesel: Tier 4 rule was adopted to provide a comprehensive program to reduce emissions from future non-road diesel engines. The Tier 4 standards require engine manufacturers to produce new engines with advanced emission control technologies similar to those already mandated for on-road trucks and buses. Emissions from these engines are expected to decrease by more than 90 percent as a result of this rule. In addition to requiring new locomotives to meet stringent standards, US EPA regulations also mandate that old locomotives must be rebuilt to comply with cleaner standards.

Pursuant to Annex VI to the International Convention on the Prevention of Pollution from Ships (MARPOL), the US and Canada have collaborated to establish a North American Emissions Control Area (ECA) effective August 1, 2012. The ECA will require the use of low-sulfur fuel in ships operating within 200 miles of the coast, effective in 2015. The reduction in sulfur will reduce emissions of SO2 which combine with ammonia to form ammonium sulfate, a type of secondary PM.

BAAQMD PM Reduction Program

The Air District has developed a comprehensive program to reduce PM in the Bay Area. This includes measures to reduce emissions and ambient concentrations of PM, as well as population exposure to PM. The Air District implements a number of regulations and programs to reduce PM emissions. These include rules limiting primary PM emissions from open burning of agricultural and non-agricultural waste; limiting emissions from combustion sources such as boilers, cement kilns and furnaces; controlling dust from earth-moving and construction/demolition operations; regulating residential wood-burning during the winter season; and reducing PM from activities that generate dust or smoke.

The Air District's Community Air Risk Evaluation (CARE) program has identified communities in the Bay Area that are disproportionately impacted by local emission sources. The CARE program, which is further discussed later in this chapter, serves as the foundation for the District's efforts to reduce population exposure to toxic air contaminants (TACs), including diesel PM.

PM Reductions from Control Strategy in the Bay Area 2010 Clean Air Plan

In fall 2010, the Air District adopted the Bay Area 2010 Clean Air Plan (2010 CAP). The legal impetus for the 2010 CAP was to update the region's plan to control ground-level ozone as required by the State Health & Safety Code. However, the Air District took the initiative to expand the scope of this plan by developing a multi-pollutant air quality plan. The 2010 CAP laid out an integrated control strategy to reduce four types of air pollutants: ground-level ozone; primary PM as well as PM precursors; toxic air contaminants (TACs); and greenhouse gases, such as carbon dioxide and methane, that contribute to climate change. The 2010 CAP control strategy included a total of 55 control measures in five categories, including:

- **Stationary Source Measures (SSMs):** The control strategy includes 18 measures to reduce emissions from stationary and area sources, as further described below.
- **Mobile Source Measures (MSMs):** The control strategy includes 10 measures reduce emissions by promoting the use of advanced-technology vehicles and cleaner fuels that reduce emissions of criteria pollutants and/or greenhouse gases, as well as accelerating the replacement or repair of older vehicles with high emission rates.
- **Transportation Control Measures (TCMs):** The control strategy includes 17 measures to reduce motor vehicle emissions by decreasing vehicle use, vehicle idling, or traffic congestion by improving transit service; encouraging walking, bicycling, and transit use; improving the efficiency of the regional transit and roadway systems; supporting focused growth; and developing and implementing transportation pricing strategies.
- **Land Use and Local Impact Measures (LUMs):** The control strategy includes six measures to promote mixed-use, compact development to reduce motor vehicle travel and emissions, and to ensure that we plan for focused growth in a way that protects people from exposure to air pollution from stationary and mobile sources of emissions.
- **Energy and Climate Measures (ECMs):** The control strategy includes four measures designed to protect air quality and the climate by promoting energy conservation and energy efficiency; promoting renewable forms of energy production; reducing "urban heat island" effects; and promoting the planting of shade trees in order to lower air temperatures, provide shading to reduce energy use, and absorb CO2 and other air pollutants.

> **The control strategy defined in the 2010 Clean Air Plan is the backbone of the Air District's current PM control program.**

In developing the 2010 CAP control strategy, the Air District sought to maximize reductions of primary PM as well as PM precursors, and to prioritize measures to reduce PM in the implementation schedule for the control strategy. The control strategy defined in the 2010 CAP is the backbone of the Air District's current PM control program. Emissions of primary PM and PM precursors will be reduced as the Air District adopts and implements the measures in the 2010 CAP.

Reducing PM from Stationary Sources

Controlling emissions from stationary sources (factories, refineries, gas stations, etc.) is the Air District's core regulatory function. The Air District has a long history of controlling PM emissions by means of (1) regulations that apply to certain categories of facilities or sources, and (2) permit conditions imposed on individual facilities. Permit conditions vary depending upon the size of the facility and/or magnitude of emissions that it generates and the type of permit required.

In addition to controlling emissions of primary PM from stationary sources, the Air Districts also adopts and enforces regulations to reduce emissions of PM precursors such as NOx and SO2 from power plants, industrial facilities, and other combustion sources, as well as reactive organic gases (ROG) from oil refineries, coatings and solvents, fuel storage, transfer and dispensing activities, and many other industrial and commercial facilities and processes.

The Air District already controls PM10 emissions from facilities subject to its New Source Review (NSR) program, and is in the process of amending the NSR requirements to include PM2.5 as well. Major PM emission sources are required to implement Best Available Control Technology (BACT) for PM in permit conditions when new sources are constructed or existing sources are modified. Three types of control equipment are commonly used to abate particulate emissions from industrial facilities:

- Wet mechanical scrubbers and/or cyclones
- Baghouses
- Electrostatic precipitators

The Air District has adopted five regulations that directly address primary PM:

- **Regulation 5:** Open Burning: Generally prohibits open burning, but also allows for exemptions such as agricultural burning, disposal of hazardous materials, fire training, and range, forest, and wildlife management.
- **Regulation 6:** Particulate Matter, Rule 1: General Requirements: Limits PM emissions from stationary sources by controlling emission rates, concentration, visible emissions and opacity.
- **Regulation 6:** Particulate Matter, Rule 2: Commercial Cooking Equipment: Regulates emissions from commercial charbroilers in restaurants.
- **Regulation 6:** Particulate Matter, Rule 3: Wood Burning Devices: Regulates emissions from residential wood-burning devices (fireplaces and woodstoves)
- **Regulation 12:** Miscellaneous Standards of Performance, Rule 4: Sand Blasting

The estimated reductions in emissions of primary PM and PM precursors (NOx and SO2) from Air District regulations are shown in Table 4-2.

Table 4-2 Estimated Reductions in Primary PM & PM Precursors from BAAQMD Regulations (tons per day)

Category	Rule(s)	Description	Dates of Rule-Making	Primary PM Reduced tons/day	NOx Reduced tons/day
Wood-Burning & Cooking	6-2	Commercial Cooking (Charbroiling)	2007	0.6	0
	6-3	Wood-burning Devices (annual)	2008	0.7	0
	6-3	Wood-burning Devices (during peak season)	2008	6.0	0
Refinery & Chemical Plant Processing	12-11, 12-12	Flare Monitoring and Minimization (SO2 reduction)	2003 2005 2006		(SO2) 6.3
Combustion of Fuels (Nitrogen Oxides controls)	9-7, 10	Boilers, Steam Generators & Process Heaters	1992, 1994, 2008, 2010	0	41.3
	9-8, 11-17	Internal Combustion Engines	1993, 2007, 2011	0	27.6
	9-11	Electric Power Generating Boilers	1994, 2000	0	17.5
	9-9	Stationary Gas Turbines	1993, 2006	0	7.4
	9-6	Gas-Fired Water Heaters	2007	0	2.5
	--	All Others	n/a	0	5.7
			Total	**7.3**	**NOx: 102.0** **SO2: 6.3**

In developing the control strategy for the Bay Area 2010 Clean Air Plan, the Air District performed a thorough review of its stationary source regulations, as well as regulations from other regions throughout the United States, and identified several new or amended rules to further reduce emissions of primary PM as well as precursors to secondary PM. Stationary Source Measures in the 2010 CAP to reduce emissions of primary PM and PM precursors are shown in Table 4-3.

Table 4-3 Stationary Source Measures to Reduce PM & PM Precursors in Bay Area 2010 Clean Air Plan

Stationary Source Measure (SSM)	Description	Status	Completion Date
SSM #1 - Metal Melting Facilities	Limit emissions of organic compounds, fine particulates, toxic compounds, and odors from foundry operations and metal melting facilities.	Initial workshops, July 2011. Second workshops July 2012.	Fall 2012
SSM #2 Digital Printing	Establish VOC limits or control requirements for inkjet, electro-photographic and other digital printing technologies.	Not yet initiated.	TBD*
SSM #3 - Livestock Waste	Establish management practices to reduce ROG, ammonia, PM, GHG.	Not yet initiated.	TBD
SSM #4 - Natural Gas Production and Processing	Reduce emissions of VOCs and methane from natural gas production facilities.	Not yet initiated.	TBD
SSM #5 - Vacuum Trucks	Require carbon or other control technology on vacuum trucks to reduce emissions of VOCs.	Adopted April 18, 2012.	April 2012
SSM #6 - General Particulate Matter Emission Limitation	Reduce particulate weight limitation as a function of exhaust gas volume and/or as a function of process weight rate.	Rulemaking initiated May, 2011.	TBD
SSM #7 - Opening Burning	Further limit agricultural burning of some crops to be burned on a given day to reduce VOCs, NOx, and PM.	Rulemaking not yet initiated.	TBD
SSM #8 - Sulfur Dioxide from Petroleum Code Calcining	Reduce SOx emissions from coke calcining.	Initiated April 2012.	TBD
SSM#9 – Cement Kilns	Further limit NOx and PM from cement production and reduce toxic emissions.	Workshop was held in December 2011. Public hearing expected Sept, 2012.	Expected Sept 2012
SSM #10 - Refinery Boilers and Heaters	Further reduce NOx emissions from refinery boilers, heaters, and steam generators.	Adopted December 15, 2010.	December 2010
SSM #11 - Residential Fan Type Furnaces	Reduce allowable NOx limits for residential furnaces.	Not yet initiated.	TBD

Stationary Source Measure (SSM)	Description	Status	Completion Date
SSM #12 - Large Residential and Commercial Space Heating	Establish NOx limits for industrial and commercial space heating.	Not yet initiated.	TBD
SSM #13 - Dryers, Ovens, and Kilns	Establish NOx limits for industrial dryers, ovens, and kilns.	Not yet initiated.	TBD
SSM #14 - Glass Furnaces	Reduce NOx limits for glass furnaces.	Not yet initiated.	TBD
SSM #16 - New Source Review Addressing PM 2.5	Amend Reg. 2, Rule 2 to address the District's anticipated non-attainment status of the 24-hour PM2.5 National Ambient Air Quality Standard.	Workshop March, 2012. Board hearing anticipated Sept/Oct 2012.	Fall 2012
SSM #17 - New Source Review for Toxic Contaminants	Implement more health-protective permitting requirements in Regulation 2, Rule 5, New Source Review of Toxic Air Contaminants based on revisions to OEHHA risk factors and method.	Adopted January 6, 2010.	January, 2010
SSM #18 – Revisions to Air Toxic Hotspots Program	Revise the District's Air Toxics Hot Spots program to incorporate more stringent risk reduction requirements from existing sources.	Awaiting OEHHA revisions to exposure assessment guidelines expected 2012.	TBD
*To be determined			

Pursuant to SSM #6 in the 2010 CAP, Air District staff has embarked upon a thorough review of the general PM rule (Regulation 6, Rule 1) with the objective of imposing more stringent emissions limits based upon the latest control technologies, as discussed in Section 5.

Reducing PM from Mobile Sources

The California Air Resources Board has primary legal authority to regulate emissions from mobile sources, as described above. However, recognizing that on-road and off-road vehicles are major sources of primary PM emissions and PM precursors in the Bay Area, the Air District also works to reduce emissions from mobile sources. The Air District seeks to reduce PM emissions from mobile sources by means of grants and incentives, targeted enforcement of ARB regulations, partnerships, and public education.

In the case of heavy-duty vehicles, the Air District focuses its efforts on ensuring compliance with ARB's diesel regulations in the Bay Area; incentivizing early compliance with ARB regulations; and promoting the use of alternative fuels and technologies. The Air District's efforts to reduce emissions from passenger vehicles include accelerating the repair or replacement of old vehicles with high emission rates; promoting the use of alternative fuels and technologies, such as zero emissions vehicles; and reducing motor vehicle use by promoting transit use, ridesharing, bicycling, walking, and telecommuting.

Reducing Emissions from Seaports and Goods Movement

Movement of goods and freight is a major source of particulate matter emissions and other air pollutants in major freeway corridors, in ports and rail yards, and in the disproportionately-impacted communities identified by the Air District's Community Air Risk Evaluation (CARE) program. Therefore, reducing emissions from seaports and the goods movement sector has been a major focus of Air District efforts in recent years. To provide a technical foundation, the Air District has developed detailed emissions inventories for each of the five Bay Area seaports (Oakland, Richmond, Redwood City, Benicia, and San Francisco). Much of the emission reduction effort has been directed at the Port of Oakland, since this port handles by far the greatest volume of goods and is located in the impacted western Alameda County area identified by the CARE program.

To develop a comprehensive approach to reducing emissions from port operations, the Port of Oakland, in partnership with the Air District and other stakeholders, developed the Maritime Air Quality Improvement Plan (MAQIP) in 2009, with the overall goal of protecting the local residents and workers by reducing their exposure to diesel PM. The Air District and the Port of Oakland have developed a joint work program that includes outreach to the regulated community to ensure compliance with state and federal regulations; and cooperating to identify and implement projects to reduce emissions, such as replacement and retrofit of drayage trucks; shore power (dockside electrification) for ships; vessel speed reduction; and development of a "marine highway" between the ports of Oakland, West Sacramento, and Stockton to help reduce on-road truck traffic between these ports.

Significant emission reductions at the Port of Oakland have already been achieved through a combination of grants and regulations. For example, over the past several years, ships have been required to switch to low-sulfur fuel. In addition, significant grant funding has been provided to equip drayage trucks that serve the Port with diesel particulate filters or with new cleaner engines, as discussed further below. Additional benefits will be achieved by 2015 as engines in cargo-handling equipment and harbor craft are either replaced or retrofitted, and ships begin using shore power while berthed.

In addition to these efforts to reduce emissions, the Air District also collaborated with ARB, the Port of Oakland, and Union Pacific Railroad in performing the 2008 West Oakland Health Risk Assessment (HRA). The HRA was performed to help understand the potential public health impacts from diesel PM emissions on the West Oakland community. The study addressed the health impacts from maritime activities at the Port, as well as locomotives, non-Port marine vessels and trucks and other significant sources of diesel PM emissions in and around the West Oakland community.

Mobile Source Compliance Plan

As noted above, ARB has adopted a comprehensive set of regulations to reduce emissions of PM from diesel engines. These regulations provide an option for local air districts to play a role in enforcing these regulations within their boundaries. Recognizing that effective enforcement of the diesel regulations is essential to protect the health of Bay Area residents, the Air District executed a Memorandum of Understanding (MOU) with ARB in fall 2009 and established a Mobile Source Compliance Plan (MSCP) which sets forth a comprehensive strategy to enforce specified ARB regulations, including regulations that apply to drayage trucks, commercial vehicle idling, transport refrigeration units, and off-road vehicles.

The goal of the MSCP is to reduce diesel PM health risk in CARE impacted communities through a robust enforcement and compliance assistance program. The initial focus of the MSCP was to provide a strong presence at the Port of Oakland to enforce ARB's 2010 Drayage Truck Rule. As of the first quarter of 2012, MSCP resources have been focused on preventing illegal "drayoffs" whereby drayage trucks switch loads from compliant to dirty trucks. By helping to ensure compliance with ARB's truck rule, the Air District's enforcement efforts have contributed to a major reduction in emissions from drayage trucks in the West Oakland area, as discussed below.

Table 4-4 summarizes MSCP enforcement efforts for calendar years 2010 and 2011.

Table 4-4 Mobile Source Enforcement Summary for Port of Oakland: 1/1/2010-12/31/2011

Inspection Type	# Inspections	# Violations	Compliance Rate
Heavy-Duty Drayage Trucks	3,581	29	99%
Port Truck Terminal Idling	34	0	100%
Commercial Vehicle/Sleeper Berth Idling	267	12	96%
Railroads: Statewide MOU & BAAQMD Protocol	8	0	100%
Off-Road (Construction) Diesel Equipment	7	0	100%
Portable Equipment Registration Program	600	0	100%
Transport Refrigeration Units	434	8	98%
Commercial Harbor Craft	4	0	100%
Oceangoing Vessels: Fuel-Sulfur Limits	41	0	100%
Oceangoing Ships: Onboard Incineration Limits	41	0	100%

Grant and Incentive Programs

To complement ARB's regulations to limit emissions from mobile sources, the Air District provides grants and incentives for projects to reduce emissions from both on-road and off-road vehicles. The purpose of these grant programs is to achieve "surplus" emissions reductions (i.e., over and above regulatory requirements) in order to complement ARB regulations. The Air District awarded a total of $308 million in external grants during a five-year period covering FY 06/07 through FY 11/12. In aggregate, these projects are estimated to reduce emission of over 1,300 tons of PM; 21,000 tons of NOx; 3,100 tons of ROG; and 1,325,000 tons of CO2 over their lifetime. Table 4-5 summarizes key grant programs, projects funded from 2007 through 2011, and the emission benefits of these projects over their lifetime.

Table 4-5 Air District Grant Programs: Projects Funded from 2007 through 2011

Eligible Equipment/Projects	# Projects	$$ Awarded	PM (Tons Reduced)	NOx (Tons Reduced)
Grant Program: Transportation Fund for Clean Air (TFCA)				
Shuttles Ride-Sharing Bicycle Facilities Smart Growth Arterial Management Clean Vehicles Alternative Fuel Infrastructure	499	$111,000,000	470	940
Grant Programs: Carl Moyer Program (CMP), Mobile Source Incentive Fund (MSIF)				
On-road Heavy-Duty Vehicles Off-Road Marine Engines Shore power for Ports/ Ships Locomotives Agricultural Equipment	1,158	$79,373,112	604	13,440
Grant Program: (Vehicle buyback)				
Light-duty scrappage	21,673	$18,094,880	8	565
Grant Program: Goods Movement Emission Reduction Program				
Drayage Trucks Other Trucks Shore power for Ports/ Ships Cargo Handling Equipment Locomotives Marine Engines	1,901	$72,138,878	292	6,606

As discussed later in this chapter, the Air District has developed its Community Air Risk Evaluation (CARE) program to identify communities disproportionately impacted by air pollutants and reduce emissions and health risks in these areas. One of the most direct ways to improve air quality in CARE communities is to replace or retrofit dirty engines and vehicles that operate in these communities. The Air District has made a commitment to target its grant funds to projects in impacted communities. Figure 4-1 summarizes the Air Districts grant funded project allocations over the last five years by project type. Figure 4-2 summarizes the funding awarded for projects in CARE communities in 2011 when approximately $60 million in Air District grant funds were directed to these communities.

Figure 4-1 **Air District Grants Awarded by Project Type from 2007 through 2011 (Total value = $308 million)**

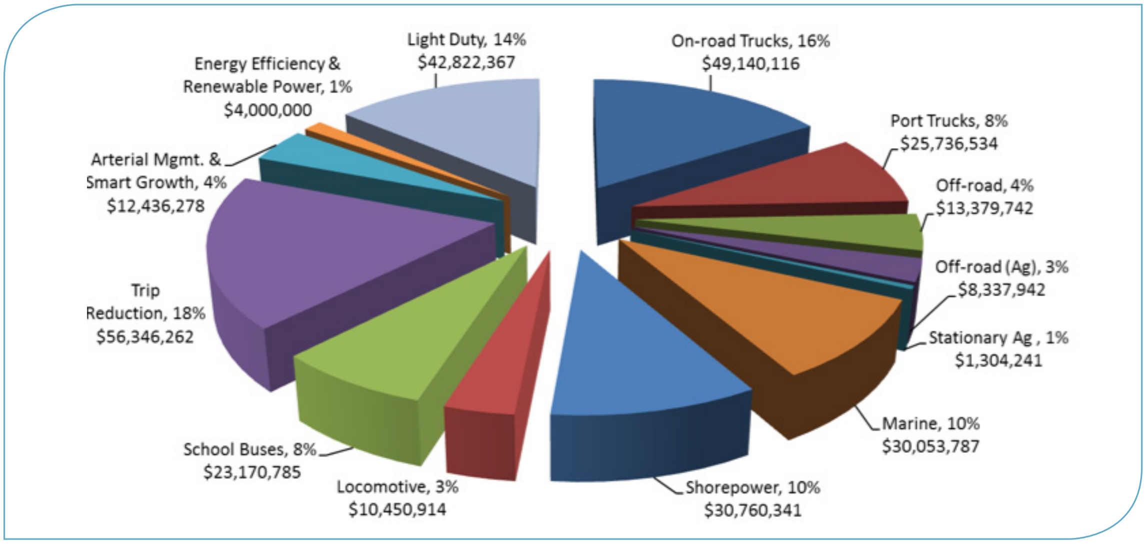
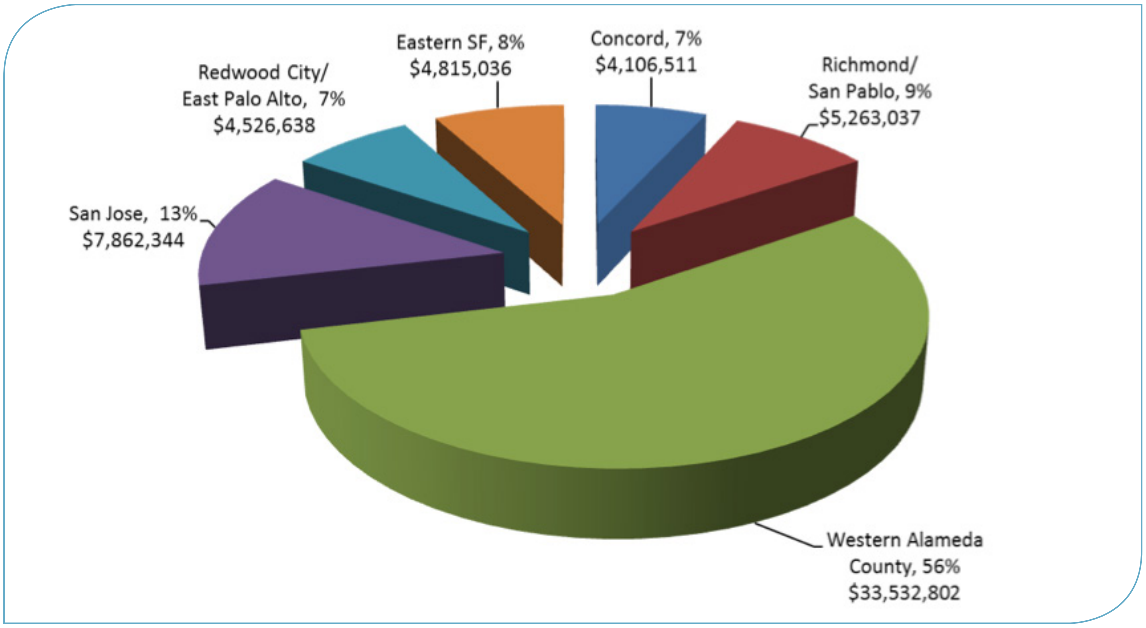


Figure 4-2 **Grant Funds Awarded to Projects in Impacted Communities in 2011 (Total value = $60 million)**



Key projects funded by the Air District to reduce emissions from mobile sources in recent years are briefly described below.

Drayage trucks

The term "drayage trucks" refers to heavy-duty trucks that handle freight at seaports and intermodal rail yards. The Port of Oakland is served by 2,000-3,000 drayage trucks, many of which were equipped with old uncontrolled engines. Collectively, these trucks were a major source of emissions of diesel PM and other pollutants that endangered the health of people who live or work in West Oakland and surrounding areas included in the western Alameda County CARE community. The Air District and partner agencies therefore prioritized the need to reduce diesel PM emissions from the drayage truck fleet that serves the Port of Oakland. In 2008, the Air District accepted applications for drayage truck retrofit and replacement projects as part of its port truck upgrade program. Through this program the Air District received and awarded a total of $25.8 million in funding from Air District, State, and federal sources. These funds were used to upgrade 1,522 trucks operating at the Port of Oakland, including 1,319 truck retrofits and 203 truck replacements. This program reduces over 14 tons of diesel PM per year at the Port of Oakland. This program reduces approximately 0.3 tons of diesel particulate emissions daily at the Port of Oakland and over 14 tons of diesel PM on an annual basis.

In March 2012 the Air District Board of Directors approved a new initiative that will provide additional grant funds to replace drayage trucks with newer trucks that meet the stringent 2007 engine emission standards. Grants of up to $10,000 will be available to eligible Bay Area truck owners toward the purchase of a truck with a cleaner 2007 model year engine or newer. The Air District Board of Directors committed an intial $1.9 million in funding for this initiative, and Alameda County has committed an additional $1.4 million. In addition, $25 million in State grant funding has been awarded for drayage truck replacement in the Bay Area. This new drayage truck initiative is expected to reduce 1.5 tons of PM and 3,401 tons of NOx over the life of the project. This effort will protect public health in communities adjacent to the Port, help local port truck drivers comply with the ARB drayage truck regulation ahead of schedule, and help maintain the economic vitality of the Port.

There is evidence that the efforts to reduce emissions from goods movement have already improved air quality in the West Oakland area. A recent study performed by UC Berkeley (Dallmann et al. 2011) found substantial reductions in exhaust emissions of black carbon (the primary constituent of diesel PM) and NOx from trucks operating in the vicinity of the Port of Oakland as a result of drayage truck retrofit and replacement projects implemented to date. The average black carbon emission factor for this drayage truck fleet decreased by approximately 50%, while the average NOx emission factor was reduced by roughly 40%. Emission reductions for black carbon were driven by the retrofit of trucks with diesel particulate filter systems and the replacement of older model year trucks with newer vehicles; reductions in NOx emissions were mainly the result of truck replacement.

On-Road Trucks

Since 2008 the Air District has issued two major solicitations for grant applications for on-road trucks used to transport goods and freight. The first solicitation resulted in the expenditure of nearly $10 million on 211 truck projects (primarily for truck replacements) and reduced more than 109 tons of PM and more than 2,300 tons of PM. The Air District received applications for over 880 eligible on-road emission reduction projects for the second solicitation, and is currently in the process of contracting with truck owners. As part of the second solicitation, the Air District will allocate approximately $15 million to truck owners to assist in replacing existing trucks. The trucks funded from the second solicitation should be on the road by mid-2013, providing estimated emission reductions of more than 55 tons of PM and 1,400 tons of NOx.

Shore Power

Since 2008 the Air District has invested over $31 million in shore power projects in the Bay Area. By eliminating the need for ships to run their engines while docked, these projects will provide significant reductions in the communities adjacent to the ports of Oakland and San Francisco. The shore power installations at the Port of San Francisco (Pier 27) and at the three berths at the APL terminal at the Port of Oakland have been completed. The installation of shore power at twelve more berths at the Port of Oakland are expected to be completed by the end of 2013. Combined, these shore power projects will reduce over 75 tons of PM and 4,000 tons of NOx over their lifetime.

Cleaner School Buses

As noted in Section 1-B, children who go to school in diesel school buses may be exposed to emissions from the bus they ride in, especially if the buses are old models that lack emission controls. The Lower-Emission School Bus Program is one of the most effective ways to reduce exposure of children to diesel PM. Using a combination of funds from its own grant programs, as well as funding provided by the California Air Resources Board, the Air District has allocated over $47 million since the year 2000 for projects to reduce emissions from over 1,100 school buses throughout the Bay Area. This includes $36.5 million to replace old buses with new ones; $10 million to

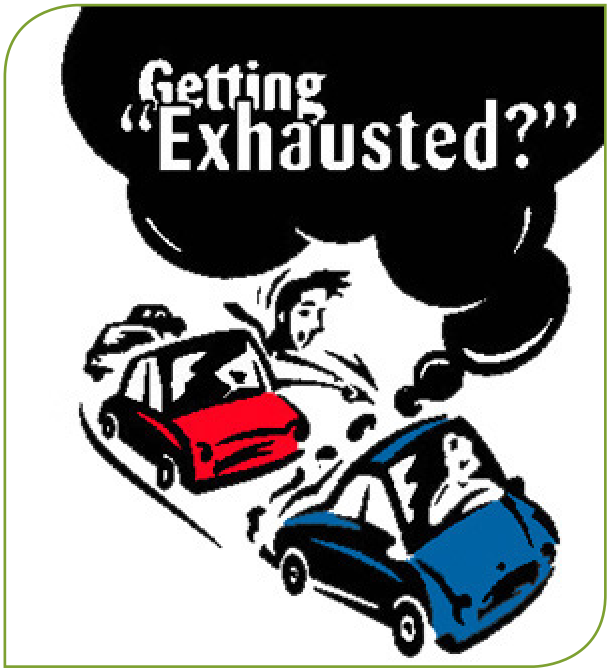
retrofit 694 buses with diesel particulate control devices, and $676,000 to replace the engines in 25 buses.

Reducing Emissions from Light-Duty Vehicles

Reducing population exposure to PM emissions from motor vehicles requires reducing emissions from light-duty passenger vehicles as well as heavy-duty diesel engines. Although PM emissions from light- and medium-duty gasoline vehicles are very low on a per-mile basis, PM emissions from these vehicles are significant on an aggregate basis because light-duty vehicles account for roughly 95% of total motor vehicle travel in the Bay Area. The Air District is working to reduce emissions from light-duty vehicles. Several of these efforts are described below.

Smoking Vehicle Program

High-emitting vehicles, often called "smoking vehicles", make up a small percentage of the vehicle fleet; however, they account for a much bigger share of total emissions of PM and other pollutants. Fortunately, the number of smoking vehicles has declined in recent years, due to turnover in the vehicle fleet as older, dirtier vehicles are replaced by newer, cleaner vehicles that achieve stringent State emission standards. Retirement of older, high-emitting vehicles has been accelerated by programs to purchase and scrap old vehicles. The Air District administered a successful vehicle-scrappage program from 1996 through 2010, which retired over 55,000 old vehicles from Bay Area roads during this period. Cumulatively, the program reduced over 4,600 tons of ROG, over 2,500 tons of NOx, and over 32 tons of PM. The Air District phased out its program, but Bay Area residents can still participate in the statewide Consumer Assistance Program to scrap old vehicles which is administered by the California Bureau of Auto Repair.



Although their numbers have been reduced, smoking vehicles are still a problem, exposing both the driver and members of the public to harmful pollutants. To help identify these vehicles, the Air District established a smoking vehicle assistance program in the early 1990's. Smoking vehicles can be reported via the 1-800-EXHAUST line, or online at www.800exhaust.org, or via an app for iPhones and Android devices. When smoking vehicle reports are received, the Air District sends an informational letter to the owner describing the harmful effects of smoking vehicles and options for vehicle repair or retirement, and requesting that the owner take appropriate action to rectify the problem. In the two-year period 2010-2011, more than 13,000 smoking vehicle reports were submitted to the Air District.

Electric Vehicles

The long-term solution to improving air quality and reducing emissions of greenhouse gases is to transition to zero-emission vehicles (ZEV), such as battery electric vehicles. Therefore, the Air District is playing a key role in funding projects to accelerate the adoption of battery electric vehicles (EVs) in the Bay Area, with the goal of an adoption of 10,000 ZEVs and 100,000 plug-in hybrid electric vehicles in the Bay Area by 2020. Much of this effort is directed at installing a robust EV-charging network throughout the Bay Area. In FY 2009/10, the Air District allocated $1.3 million for projects to install publicly-available Level 2 chargers in up to 250 locations around the region, six direct current (DC) fast chargers to serve taxi fleets, and a Battery-switch station to test the viability of this advanced technology as a pilot project with taxi fleets. In FY 2010/11, the Air District allocated an additional $5 million to expand this effort, with a goal to install up to 3,000 Level 2 chargers in the Bay Area, and up to 50 DC fast chargers by the end of 2013. Looking forward, the Air District is considering other actions to expand the use of electric vehicles, such as offering grants to encourage cities and counties to expand the use of EVs in their fleets, as well as funds to incentivize the early adoption of electric vehicles in medium- and heavy-duty vehicles used in delivery fleets and similar applications, thus reducing emissions of PM and other pollutants from diesel engines.

Bicycle-Sharing

In recent years, major cities in Europe, Asia, and North America have implemented publicly-available bicycle-sharing programs to reduce traffic and air pollution in the urban core. The Air District is leading a partnership to implement a regional bicycle sharing pilot project in five Bay Area cities, in collaboration with transportation agencies in the counties of Santa Clara, San Mateo, and San Francisco. This project will deploy 1,000 bicycles in the cities of San Jose, Palo Alto, Mountain View, Redwood City and San Francisco for a period of at least 12 months. The goal of the pilot is to test and develop a self-funded regional Bike Share System to complement existing transportation options by providing a convenient option for residents, commuters, and visitors making short trips to and from transit facilities, places of employment and residence, and social and recreational destinations. The current schedule is to launch the project by the end of 2012. If successful, the project may be expanded to additional communities within the Bay Area.

Reducing PM from Wood Smoke

Wood smoke from residential wood-burning is a major component of PM in the Bay Area, especially on winter days when high PM concentrations that exceed the 24-hour PM2.5 standard are most likely to occur. At the local scale, in neighborhoods where wood-burning is prevalent during winter months, wood smoke can expose people to high PM levels, especially if topographical features (e.g., a valley or canyon) and/or weather conditions (an inversion) prevent dispersion of the smoke. Therefore, reducing emissions from wood-burning is a key component of the Air District's efforts to reduce PM levels and protect public health in the Bay Area.

The Air District's efforts to reduce residential wood-burning have evolved over the past two decades. Public education and voluntary compliance were the focus of this effort in the 1990's. The Air District began implementing a voluntary Winter Spare the Air program in 1991, requesting that Bay Area residents voluntarily curtail wood-burning on days when an exceedance of PM standards was forecast.

In the mid-1990's, the Air District developed a model wood smoke ordinance as a guidance document for cities and counties that wished to regulate sources of particulate matter in their communities. Along with requesting that residents curtail the use of fireplaces and woodstoves in response to Winter Spare the Air alerts, this model ordinance promoted the use of cleaner technologies to reduce wood smoke pollution. Air District staff worked with health agencies and interested residents throughout the Bay Area to promote adoption of the ordinance. To date, 49 Bay Area cities and counties have adopted wood smoke ordinances.

In 2006, US EPA significantly strengthened the national 24-hour PM2.5 standard, lowering the threshold from 65 to 35 mg/m^3. The Air District recognized that further reductions in PM emissions from wood smoke would be needed to achieve the new PM2.5 standard, especially on days when meteorological conditions are conducive to high PM concentrations. Therefore, in 2008 the Air District adopted a stringent wood-burning rule (Regulation 6-3), and amended another rule which regulates open burning (Regulation 5). The Air District also substantially expanded its public outreach and education program for wood smoke reduction.

Summary of Wood-Burning Rule

Key provisions of the wood smoke rule (Regulation 6, Rule 3: Wood-burning Devices) include the following:

- Prohibits operation of any indoor fireplace, fire pit, wood or pellet stove or fireplace insert on specific winter days when the Air District forecasts that PM2.5 levels may exceed the national 24-hour PM2.5 standard. (Regulation 5: Open Burning prohibits outdoor recreational fires during the same periods when elevated PM2.5 levels are forecast.)
- Prohibits excess visible emissions from wood-burning devices.
- Requires cleaner burning technology (EPA-Phase II certified wood-burning device or pellet stove) when wood-burning devices are sold, resold or installed.
- Requires cleaner burning technology if wood-burning devices are permitted for installation in new building construction and remodels. (Installation of new wood-burning fireplaces is prohibited).
- Prohibits burning of garbage, non-seasoned wood, plastics and other inappropriate materials.

- Requires labeling of moisture content for wood sold for use within Air District boundaries, including instructions on how to dry wood that has moisture content greater than 20 percent.
- Requires a label on packages of wood and other solid fuels (such as pressed logs and pellets) instructing the user to check local air quality status before burning these products.

Promoting Compliance with the Wood-Burning Rule

The Air District relies upon both public education and enforcement to promote compliance with the wood-burning rule. In addition to preventing exceedances of the national 24-hour PM2.5 standard, this effort is intended to reduce wood-burning over the long term by educating the public about the hazards of wood smoke.

The public outreach campaign is intended to educate the public as to the requirements of the rule, ensure that people are aware that they need to check air quality status before burning, and explain the public health benefits from reduced wood smoke pollution. The Winter Spare the Air Alert outreach campaign utilizes a wide variety of media and multiple languages to reach the diverse Bay Area population and notify the public when a Winter Spare the Air Alert has been called.

Recognizing that certain areas experience localized impacts of wood smoke, the Air District has conducted targeted mailings, with information about the wood smoke rule and the negative health effects of wood burning, to neighborhoods with high levels of wood smoke complaints and/or burning. In addition, the Air District recently developed a new wood smoke model ordinance that offers local governments a menu of more stringent and innovative options to choose from to reduce neighborhood wood smoke within their jurisdictions. The Air District provided the new model ordinance to all cities and counties in the Bay Area in April 2012.

The Air District is planning to take the following actions to enhance enforcement of the wood-burning rule for the winter 2012-2013 season:

- Use more conservative forecasting to call Winter Spare the Air alerts that trigger the no-burn provision of Regulation 6, Rule 3;
- Add a requirement for first-time violators to complete “wood smoke awareness school” or pay a $100 fine; and
- Increase fines for repeated violations of Regulation 6, Rule 3 for second-time violators, increase the fine to $500, with progressive penalties for subsequent violations.

Survey data and air quality monitoring data both indicate that the Air District’s efforts have helped to reduce residential wood-burning and avoid exceedances of the 24-hour PM2.5 standard. Responses to surveys that the Air District performs to monitor residential wood-burning suggest that Bay Area residents are burning less wood and are burning less often. Monitoring data shows that the number and severity of high PM days during the winter have decreased, and chemical mass balance data indicate that PM2.5 from wood smoke has decreased by approximately 40% in the past several years.

Reducing Population Exposure to PM

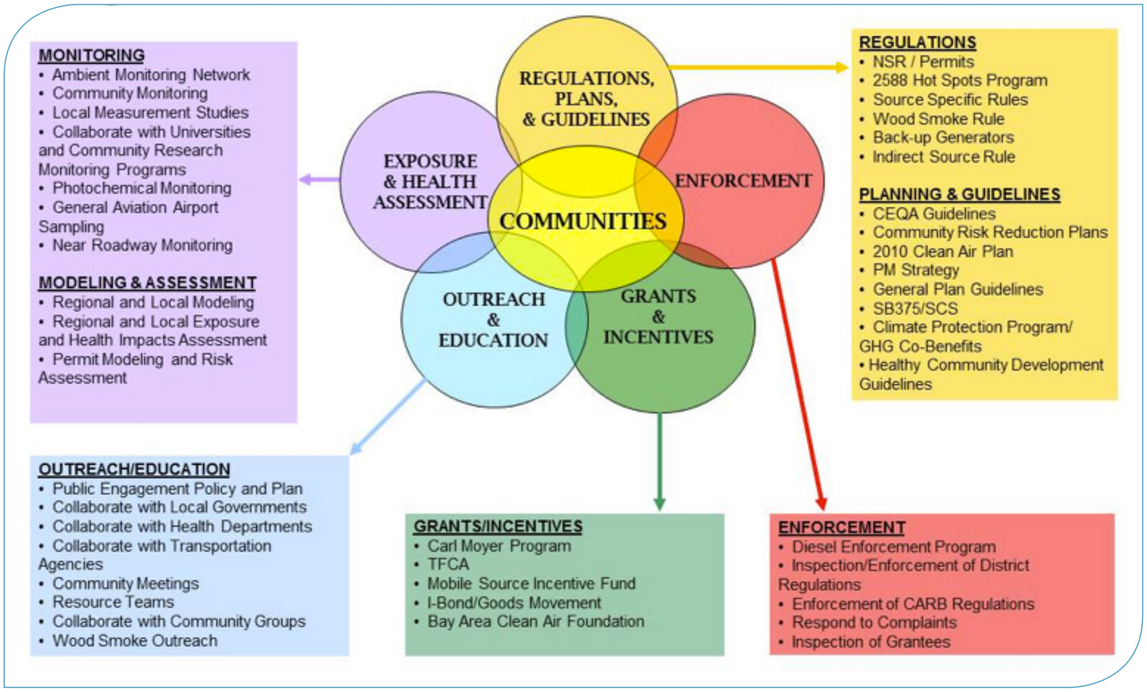
The Air District recognizes that protecting public health means more than just attaining air quality standards at the regional scale. Local concentrations of directly-emitted air pollutants, such as primary PM, may be elevated in proximity to emissions sources such as major roadways, ports and freight distribution hubs, refineries and industrial facilities, airports, and large construction sites. To protect public health, we need to analyze population exposure to air pollution, identify those communities and populations that are most heavily exposed to air pollutants, and develop strategies to reduce population exposure among people who live or work in the most impacted areas.

The Air District's efforts to identify and protect impacted communities have been bundled together under the banner of the multi-faceted **Clean Air Communities Initiative** (CACI). Key elements of the CACI include the following:

- Implementation of the Community Air Risk Evaluation (CARE) program to identify areas in the Bay Area that are disproportionately impacted from transportation and stationary sources.
- New or amended regulations to control emissions from stationary sources that impose disproportionate impacts in CARE communities (e.g., SSM 1, the metal melting rule).
- Implementation of the control strategy in the Bay Area 2010 Clean Air Plan which includes Mobile Source Measures to reduce vehicular emissions; Transportation Control Measures to reduce motor vehicle use, and Land Use and Local Impact Measures to focus on reducing population exposure in impacted areas.
- Performing special monitoring studies to measure ambient concentrations and/or health risks related to PM and/or toxic air contaminants, such as the West Oakland Monitoring Study, the Custom Alloy Scrap Sales (CASS) metals study in West Oakland, and the UC Berkeley study of truck emissions in West Oakland.
- Enforcement of ARB regulations to reduce emissions from diesel engines, via the Mobile Source Compliance Plan described above.
- Providing grants and incentives for projects targeted to reduce emissions within CARE communities, as described above.
- Public education and outreach to encourage compliance with the Air District's wood smoke rule.
- Collaboration with local governments to develop Community Risk Reduction Plans, as described below.
- Collaboration with regional agency partners at MTC, ABAG and BCDC to coordinate regional efforts to promote focused development in a health-protective way via the Air Quality/Priority Development Area working group, as described below.
- Development of on-line analytical tools to help local government agencies to identify and address air quality issues and impacts in their communities.

- Development of a set of standard mitigation strategies to address potential impacts from siting new sensitive receptors near sources of TACs, as described below.
- Reviewing and commenting on air quality analyses in CEQA documents prepared for key plans and projects.
- Development of guidance documents and technical tools to help Bay Area cities and counties address air quality in their General Plans, as described below.

Figure 4-3 BAAQMD Clean Air Communities Initiative



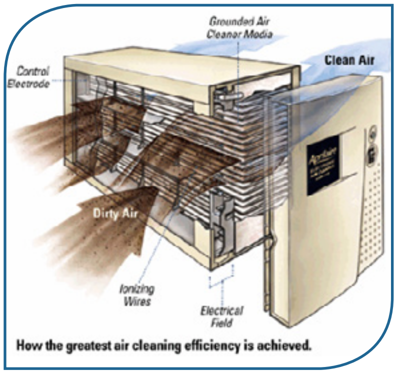
CARE Program to Identify Impacted Communities

Recognizing that certain neighborhoods and communities in the Bay Area are disproportionately impacted by local emission sources, the Air District launched the **Community Air Risk Evaluation** (CARE) program in 2004. The CARE program was initially focused on identifying risks related to Toxic Air Contaminants (TACs). An analysis of the various TACs in the Bay Area found that diesel PM (identified by ARB as a TAC in 1998) is the TAC that poses by far the greatest health risk in the Bay Area. Based on a combination of major emission sources, high population exposures, and sensitive populations, the CARE program identified six impacted areas as impacted communities: Concord; Richmond/San Pablo; western Alameda County; San Jose; Redwood City/East Palo Alto; and eastern San Francisco. In recent years, recognizing that fine PM of all types is harmful to public health, the scope of the CARE program has been expanded to include PM2.5 as well as TACs. (The Air District is also considering adding other air pollutants, such as ozone, for purposes of identifying and defining impacted communities.)

Community Risk Reduction Plans

Addressing air quality issues in local land use and transportation planning also can help reduce exposure to air pollution. The Air District, in cooperation with Bay Area planning and health agencies, is developing a new planning tool, known as a **Community Risk Reduction Plan** (CRRP), to help local jurisdictions identify, evaluate, and reduce risks from local sources of toxic air contaminants (TAC) and fine PM. The Air District is encouraging cities, especially those that have been identified by the Air District's CARE program as disproportionately impacted by local pollutants, to prepare a CRRP. The basic approach to develop a CRRP includes several key steps: (1) developing an inventory of TAC and fine PM emissions within a planning area, which may be a whole city or part of a city; (2) using dispersion modeling to map ambient concentrations and risks from local pollutants within the planning area; (3) developing specific goals and objectives to reduce health risks; and 4) defining implementation actions, such as policies and mitigation measures, in order to achieve the goals and objectives. The Air District is providing financial and technical support in a pilot program to assist the City and County of San Francisco and the City of San Jose in developing CRRPs.

The San Francisco Department of Public Health and the SF Planning Department have collaborated with the Air District to develop a city-wide CRRP that encompasses the impacted areas of eastern San Francisco, with the goal of reducing air pollution exposures and associated health risk on a city-wide basis. City staff worked with the Air District to develop a detailed emissions inventory and in applying local-scale dispersion modeling to identify areas with increased risk from air pollution and to produce maps of TAC risks and PM2.5 concentrations from all emission sources. City staff is developing a range of potential policies and programs to reduce residents' exposure to air pollution, such as expanding current air filtration requirements (Article 38 in the San Francisco Health Code), limiting construction emissions, and more.



San Francisco Health Code Article 38

Model Ordinance for Cities: San Francisco is the first jurisdiction in the country to create a law, known as Article 38, to protect future residents from exposure to roadway air pollution. The law will prevent avoidable lung disease and premature death in residents living near busy roadways, as well as prevent avoidable health care spending, for example, on hospital charges for prevented asthma attacks.

San Francisco Health Code Article 38, adopted in 2008, requires residential projects with more than 10 units located in "Potential Roadway Exposure Zones" (as defined according to maps provided by the San Francisco Department of Public Health) to prepare an air quality assessment, using modeling tools, to determine whether residents would be exposed to unhealthy levels of PM2.5. The Department of Public Health has defined "unhealthy" levels of PM2.5 as roadway concentrations greater than 0.2 μg/m^3. If the air quality assessment indicates that the roadway-attributable PM2.5 would be less than 0.2 μg/m^3, then no further action is required. If the air quality assessment for the residential project indicates that concentrations would be unhealthy, then the project is required to mitigate the traffic-related PM2.5 pollutants, using available technology and design features, to reduce or remove at least 80% of the ambient PM2.5 from indoor spaces.

Meeting the performance standard can be accomplished in several ways, including:

1. Designating lower floors for commercial use and upper for residential use;
2. Setback of buildings from roadway air pollution sources;
3. Locating the intake for fresh air ventilation sources at a non-polluted site;
4. Filtration of fresh air ventilation sources; and/or
5. Recirculation and filtration of indoor air.

Economic Impacts: The City/County of San Francisco's Office of the Controller has determined that the economic impacts of Article 38 on the San Francisco economy, the development community, and future residents of the City are neutral to positive. Although there is a cost associated with implementation of the mitigation measures described above, Article 38 will also prevent avoidable health care spending (for example, hospital charges for emergency room visits for asthma attack) and help to prevent premature mortality associated with exposure to PM. If using a filtration system, the City estimates that costs to install and maintain the system will range from approximately $50-700 per year per unit, while the monetary benefit of the reduction of premature death is estimated to be approximately $2,100 per unit per year. On the basis of this analysis, if installation of a filtration system is required in order to comply with the requirements of Article 38, then the Controller has determined that the net economic benefit of Article 38 would be approximately $1,400 per unit per year.

The Air District has also been providing technical assistance to help the City of San Jose develop a CRRP. The Air District is preparing city-wide emissions inventory for on-road mobile sources on freeways and surface streets, permitted stationary sources, and railroads, airports, and construction projects. Initial air dispersion modeling is underway. The City has also engaged in public outreach, in partnership with the Air District. As a first step on the policy side, the City included several policies in its 2011 General Plan update to analyze and mitigate population exposure from major emissions sources. For example, the air quality section of the General Plan includes policies which (1) require completion of air quality modeling for sensitive land uses such as new residential developments located near emission sources such as freeways and industrial uses; (2) require new residential development projects and projects characterized as sensitive receptors to incorporate effective mitigation into project designs or to be located an adequate distance from sources of toxic air contaminants to avoid significant health risks; and (3) require projects that would emit toxic air contaminants to prepare health risk assessments as part of environmental review and employ effective mitigation to reduce possible health risks to a less-than-significant level. In addition, the General Plan policies mentioned above encourage the use of air filtration devices in existing schools, houses and other sensitive land uses; re-designation of truck routes; and the use of vegetative buffers between emission sources and sensitive receptors.

Promoting Healthy Focused Development

Continued growth in motor vehicle travel could erode the air quality benefits from the ARB and Air District programs described above. We need to better integrate land use, transportation, and air quality planning in order to constrain future increases in vehicle travel and emissions. Therefore, the Air District supports the effort to focus future development in the Bay Area in areas where public transit, biking and walking are viable transportation options. At the same time, however, many of the areas identified as good sites for focused growth already experience high concentrations of air pollutants due to emissions from existing local sources. In fact, a comparison of areas that have been designated as *Priority Development Areas* (PDAs) to date and the impacted communities identified by the Air District's CARE program shows that there is considerable overlap. This emphasizes that we need to plan for focused growth in a way that protects people from exposure to air pollutants, especially local pollutants such as PM and air toxics. To address this issue, the Air District is committing its resources to help planning agencies (cities, counties, MTC, and ABAG) identify, evaluate and mitigate these impacts through the planning and design processes.

The Air District is working actively with partners at both regional and local agencies to support focused development to reduce motor vehicle emissions, while ensuring that development is planned and designed so as to minimize public exposure to air pollutants and protect public health.

At the regional scale, the Air District is engaged with its regional agency partners in the effort to develop *Plan Bay Area*. Plan Bay Area, scheduled for adoption in 2013, will update the Regional Transportation Plan (RTP) and incorporate a **Sustainable Communities Strategy** to better integrate land use and transportation planning, in response to the requirements of Senate Bill (SB) 375. Although SB 375 requirements focus on the need to reduce emissions of greenhouse gases, the Air District worked with its regional agency partners to make sure that the performance targets for Plan

Bay Area include targets to reduce exposure to particulate matter emissions from motor vehicles and to achieve greater reductions of PM in impacted communities.

In addition, Air District staff is participating in the development and environmental review of Station Area Plans where most of the future high-density, transit-oriented development in the Bay Area is projected to occur. The Air District is working to identify potential PM2.5 and TAC impacts and develop plan-level approaches on how to mitigate these impacts, so that subsequent project-level development that conforms to the Station Area Plans is not burdened with costly environmental review.

Since local governments are responsible for land use planning, the Air District is working to develop partnerships and provide technical assistance to the nine counties and 101 cities that comprise the Bay Area to ensure that air quality considerations are addressed in local land use decisions.

Analytical tools: The Air District developed a set of on-line analytical tools to identify and assess the potential impacts from stationary sources, freeways and major roadways in close proximity to a development project (or throughout a plan-area). The stationary source tools can be used to identify all stationary sources permitted by the Air District and to estimate local PM2.5 concentrations associated with each permitted source. The highway tool can be used to identify all highways throughout the Bay Area and to estimate local PM2.5 concentrations associated with each highway. The roadway tool is a set of tables which show the estimated PM2.5 concentrations associated with each major roadway (defined as 10,000 AADT and above) throughout the entire Bay Area. All of the tools are county-specific, meaning the information used in the development of the tool has been customized for each Bay Area county.

Mitigation Strategies: The Air District has also developed a set of recommended mitigation strategies to reduce exposure to PM2.5 concentrations. These mitigation strategies include measures such as:

- Requiring installation of HEPA filtration systems (rated at MERV 16 or higher);
- Designing the project site to minimize population exposure to air pollutants;
- Limiting residential uses on the ground floor;
- Planting trees to buffer and absorb pollutants;
- Reducing emissions, where applicable via measures such as replacing or retrofitting diesel back-up generators; installing electrical hook-ups for diesel trucks; requiring trucks to use transportation refrigeration units that meet EPA Tier 4 emission standards; requiring advanced drive trains or alternative fuels in heavy-duty trucks; and establishing alternative truck routes.

General Plan Guidelines: The Air District is developing a guidance document to help local governments address air quality in their general plans. The general plan is a critically important document for local community planning, because local decisions related to growth and development must be consistent with the policies and objectives contained in the general plan. In addition to elements required by State law, local governments may elect to include additional elements in their general plan, such as health or air quality, to better guide their community's development. The Air District's guidance document provides local agencies with a comprehensive set of air quality-related "model policies" that may be used to build an optional air quality element within their general plan. The guidance document also offers policy recommendations and implementation strategies for community education and outreach, reducing wood burning, improving indoor air quality, and "green" building and contracting practices. The Air District's guidance document should facilitate land use planning to improve air quality and protect public health at both the local and regional scale.

Organizational Changes to Focus on Exposure Assessment & Protecting Public Health: To further its commitment to analyzing air pollution and minimizing population exposure in impacted communities, the Air District reorganized its Planning Division in 2012. The "Special Projects Section", which previously focused on developing emission inventories, was expanded and renamed the "Exposure Assessment and Emission Inventory Section". In addition, the Air District created a Health and Science Officer position in July 2012; this position will bring additional expertise to inform development of Air District policies to protect public health, as well as strengthen the Air District's partnerships with the public health community in the Bay Area. These changes emphasize that the Air District will place a high priority on improving its capabilities in terms of analyzing and reducing population exposure to air pollutants in the coming years.

PM Reductions in the Central Valley: As discussed in Section 2, technical analysis performed by the Air District indicates that transport of PM from the Central Valley contributes significantly to elevated Bay Area PM concentrations during the winter weather pattern typically associated with elevated PM levels in the Bay Area. The San Joaquin Valley Unified Air Pollution Control District is in the process of developing a PM2.5 SIP submittal to demonstrate how it will reduce emissions of primary PM and PM precursors in order to attain the national 24-hour PM2.5 standard. The San Joaquin Valley attainment plan must be submitted to US EPA by December 2012. Monitoring data indicates that the Sacramento air basin recently met the national 24-hour PM2.5 standard. The Sacramento Air Quality Management District plans to develop a re-designation request and maintenance plan to show how it will continue to attain the national 24-hour PM2.5 standard over the next decade. In addition to improving air quality in the Central Valley, implementation of the control strategies set forth in the San Joaquin and Sacramento PM2.5 plans should also help to reduce ambient PM concentrations in the Bay Area during weather patterns that facilitate transport of PM from the Central Valley to the Bay Area.

SECTION 5: LOOKING FORWARD

This final section provides a conceptual framework to guide future Air District efforts to reduce PM in order to protect public health, the climate and the environment. This section identifies several challenges; suggests policy guidelines to inform the development of potential future measures to reduce PM; describes areas where further study and technical enhancements are needed; and provides ideas for how Bay Area residents can reduce their exposure to PM.

As discussed in the preceding sections of this report, the current control programs being implemented by the Air District, the California Air Resources Board, and other partners have reduced substantially ambient PM concentrations in the Bay Area over the past 15-20 years. The Bay Area currently meets the national PM standards and is close to meeting state PM standards. Continued implementation of these programs described in Section 4 is expected to further reduce emissions of primary PM and PM precursors over the next decade. In addition, new initiatives in the final stages of development such as ARB's Low Emission Vehicle (LEV) III standards, should lead to additional PM reductions from targeted emission sectors.

WHY IT'S IMPORTANT TO CONTINUE REDUCING PM

The fact that the Bay Area has made tangible progress in reducing PM levels does not mean that we can rest easy, however. There are compelling reasons why it is important to continue and enhance our efforts to reduce PM.

- Researchers have not been able to establish a safe threshold for population exposure to PM. A robust and growing body of research shows that there are health impacts associated with exposure to PM even below the current standards.
- As new information about the health effects of PM becomes available, the US EPA and/or the ARB may issue more stringent standards in the future.
- Even at the relatively low PM levels that currently prevail in the Bay Area, PM is the air pollutant most harmful to public health, including premature mortality, heart attacks, chronic bronchitis and other key health effects.
- PM levels - and population exposure to PM - can vary significantly at the local scale. Even though the Bay Area currently meets national PM standards (based on the measurements from the regional PM monitoring network), some communities and individuals are exposed to higher concentrations of PM.

- In addition to its detrimental impacts on public health, PM also plays a role in climate change and has negative impacts on ecosystems and visibility.

CHALLENGES WE FACE

Although further PM reductions would benefit the health of Bay Area residents, reducing PM presents challenges, several of which are described below.

- **The Air District's authority is limited**. Emission sources are diverse, and the Air District's authority is limited to a defined set of sources. For example, indoor exposure to ultrafine and fine PM accounts for a significant share of total exposure for many people, but the sources and conditions contributing to indoor PM excposure are complex and the Air District has limited authority to regulate emissions from most indoor sources that contribute to indoor exposures.[5]
- **Low-hanging fruit is sparse**. As described in Section 4, policies and regulations have already been implemented to reduce emissions of primary PM and PM precursors from key sources, such as diesel engines and residential wood burning. It will be a challenge to identify new control measures that achieve significant PM emissions on a cost-effective basis.
- **Wood smoke remains a problem.** The Air District has made a major effort to reduce PM from residential wood burning, as described in Section 4. The evidence indicates that this effort has been effective in reducing PM levels at the regional scale. However, wood smoke is still a significant source of emissions, accounting for roughly one-third of PM2.5 during winter months when the Bay Area experiences its highest PM levels. Moreover, in neighborhoods where wood burning is prevalent during winter months, wood smoke can expose residents to high PM levels, especially if topographical features and/or weather conditions prevent dispersion of the smoke. Achieving further reductions in residential wood burning will be a challenge, however, because the sources are widely dispersed and compliance depends upon public education and cooperation and large-scale behavioral change.
- **Transport of PM from outside the Bay Area**. Analysis shows that on the winter days when the Bay Area experiences its highest PM concentrations, transport of PM by easterly winds from the Central Valley contributes to PM levels in the Bay Area.
- **Coordination challenges**. Many public agencies and other stakeholders have a role to play in reducing PM and protecting public health. The diversity of players and perspectives provides opportunities for collaboration and

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[5] The Air DIstrict does have authority to regulate emissions from certain indoor sources, including wood-burning cooking, water-heaters and furnaces.

partnership, but also highlights the need to coordinate efforts, identify areas of mutual interest, and reconcile competing objectives.

- R**egional and community needs.** Because certain communities are disproportionately impacted by PM and other air pollutants, the Air District needs to reduce PM at both the regional scale and in the communities most impacted by PM.
- **Protecting public health at the regional and community scale**. Monitoring data show that the Bay Area currently meets the national PM standards. However, health studies show that there are health effects even below these standards, and we know that certain communities and individuals may be exposed to higher levels of PM. GIven this context, how do we determine the appropriate objectives in terms of reducing exposure to PM at both the regional and the community scale?

POLICY GUIDELINES TO INFORM FUTURE PM PLANNING

The discussion in this section provides a conceptual framework to guide the Air District's future efforts relating to PM, based on the best available information to date. This is intended to be a "working" framwork; the Air District recognizes that it may need to be revised as new information is available.

- The Air DIstrict will continue to pursue a multi-faceted approach which combines regulations and control measures to reduce emissions of PM and PM precursors from sources under its jursidiction, targeted enforcement of ARB regulations on sources under ARB control, grants and incentives to achieve emission reductions above and beyond regulatory requirements, efforts to reduce population exposure, partnerships with the health community and other stakeholders, and public education.
- Pursuant to the Bay Area 2010 Clean Air Plan, potential future measures to reduce PM will be evaluated on a multi-pollutant basis, to maximize their overall air quality, health and climate protection benefit.
- Since science has not yet determined precisely which components of PM are the most harmful to public health, the Air District will continue its efforts to reduce PM across the board, including all sizes and types of particles. However, the Air District will continue to monitor the latest research on PM health effects to inform its PM reduction efforts, and will collaborate with the health community to provide information on PM emissions and exposure in the Bay Area.
- Results from the Air District's PM modeling for the Bay Area indicate that reducing emissions of primary PM offers the most direct means to reducing ambient PM concentrations. This is especially true in terms of reducing local PM "hot spots"

which are caused mainly by exposure to emissions of primary PM from motor vehicles, residential wood burning, or major point sources.

- The Air District and its partners will also continue efforts to reduce emissions of precursor pollutants that contribute to the formation of secondary PM, especially to help avoid exceedances of the 24-hour PM2.5 standard during winter months when ammonium nitrate is a major component of ambient PM.
- The evidence indicates that reducing emissions of black carbon particles should provide both health and climate protection benefits. Therefore, the Air District will evaluate potential measures to further reduce combustion of fossil fuels and biomass (wood) in order to decrease emissions of black carbon.
- The Air District will continue to enhance its efforts to reduce emissions from sources, such as motor vehicles, that account for high population to exposure to fine and ultrafine particles.
- The Air District will continue its efforts to better understand local PM concentrations and exposures by, for example, expanding monitoring near roadways and major emission sources, undertaking special studies, and improving methods to estimate population exposure to PM.
- The Air District work to strengthen its partnerships with county health departments, local planning agencies, community groups, academic institutions, and other stakeholders with an itnerest in improving air quality and protecting public health to address the challenges and policy issues identified in this report.

AREAS FOR FURTHER EVALUATION

To inform the Air DIstrict's future efforts to control PM and reduce the negative impacts described in previous sections, we need:

- Better information as to which types and sizes of PM are most harmful to public health.
- More precise analysis of personal exposure to PM in various micro-environments to determine which types of exposures pose the highest risk to health.
- Better understanding as to the interplay between PM and climate change, and how the various types of particles promote or inhibit climate heating.

Several specific areas merit further evaluation: (1) ultrafine PM, (2) the role of ammonia as a precursor to secondary PM formation, and (3) the climate change impacts of black carbon. The Air District will consider each of these areas, as discussed below.

Ultrafine PM

As described previously, ultrafine particles (UFPM) present special challenges since they are hard to measure and extremely dynamic, and because ultrafine PM concentrations tend to be elevated and localized in close proximity to emission sources. The current understanding of ultrafine PM in the scientific community is still very limited. Although evidence suggests that exposure to ultrafine PM may be especially harmful to public health because the particles can penetrate deep into body organs and tissues, there are as yet no ambient air quality standards for ultrafine PM nor requirements for monitoring of ambient ultrafine PM concentrations. Because of the gaps in our understanding of ultrafine PM, efforts to characterize these particles and develop appropriate control strategies are still in the early stages. Key constraints in terms of ultrafine PM include the following:

Emissions inventory: As discussed below, the Air District prepared its first ultrafine PM emissions inventory in August 2012. However, further work will be needed to refine the inventory as emission factors are developed and improved for various many source categories.

Ambient concentrations: The technology to measure ambient concentrations (particle numbers) of ultrafine PM is still evolving. Several ultrafine PM monitors have recently been installed in the Bay Area, but we do not yet have a comprehensive ultrafine PM monitoring network.

Modeling: Given its localized and dynamic nature, modeling ultrafine PM may prove especially valuable. Developing good estimates to where and when ultrafine PM concentrations are high will help identify the types of conditions that are conducive to acute human exposure.

Health studies: Research is on-going to analyze the health effects of ultrafine PM and to determine whether there are distinct health effects related to ultrafine PM as opposed to fine particles (PM2.5). However, the lack of ultrafine PM monitoring networks in most urban areas, and the consequent absence of data on ultrafine particle concentrations, makes it difficult to perform epidemiological studies to better elucidate the health effects of ultrafine PM.

Ambient air quality standards: Despite growing concerns about the health effects of ultrafine PM, there are as yet no State or national ambient air quality standards for UFPM. Nor are such standards likely to be adopted until the gaps described above have been addressed. In the meantime, PM2.5 serves as the closest surrogate for ultrafine PM, despite the fact that evidence indicates that the correlation between PM2.5 mass and ultrafine PM numbers is tenuous at best, and may actually be negatively correlated.

To address the gaps in our understanding of ultrafine PM, the Air District will undertake initiatives in regard to the emissions inventory, monitoring and modeling for ultrafine PM, as summarized in the Technical Enhancements section below.

BAAQMD Advisory Council Recommendations

The Air District's Advisory Council has been studying ultrafine PM over the past two years and receiving presentations from leading experts on ultrafine PM measurement and its health effects. In December 2011, the Advisory Council presented recommendations on ultrafine PM to the Board of Directors. Key Advisory Council recommendations to the Board include the following:

- Integrate ultrafine PM considerations into PM2.5 planning.
- Continue its efforts to characterize ultrafine PM sources, chemical composition, and ambient air levels in the Bay Area.
- Consider development of a ultrafine PM emission inventory and monitoring strategy.
- Consider conducting short-term intensive ultrafine PM monitoring to characterize ambient ultrafine PM levels and speciation at selected key locations (e.g., near heavily traveled roadways), possibly integrating those efforts with upcoming near-roadway continuous NO2 monitoring required pursuant to the 1-hour NO2 national standard.
- Maintain a focus on PM2.5 and ultrafine PM emissions from fuel-burning vehicles, with particular attention to PM emissions from unburned and partially burned fuel and lube oil.
- Continue to investigate and evaluate measures to reduce personal exposure to PM2.5 and ultrafine PM.
- Collaborate with the Association of Bay Area Governments and the Metropolitan Transportation Commission to educate the public on the public health effects associated with on-road and near road ultrafine PM and with PM2.5 exposures; and promote strategies to reduce vehicle miles traveled.
- Collaborate with other agencies (e.g., ARB, South Coast Air Quality Management District) in studying ultrafine PM measurements, and health impacts, fashioning effective public policy strategies and focusing policy development on vulnerable populations and highly impacted areas.

PM and Climate Change (Black Carbon)

As discussed in Section 1-C, the relationship between PM and climate change is complex. A variety of physical processes are involved, and different types of particles have differing impacts on climate. Although it is difficult to determine the net effect of PM on climate, there is strong evidence that black carbon (soot) may be a significant contributor to climate heating. Policy actions to reduce heating effects of PM on the climate should focus on reducing black carbon, as well as brown carbon. Such actions would also yield important health benefits by further reducing exposure to fine particles. Future actions may include the following:

- Develop a Bay Area black carbon emissions inventory; work with the Air Resources Board to obtain black carbon emissions factors for motor vehicles and other mobile sources.

- Collaborate with U.S. EPA, ARB, and other agencies as appropriate, to develop a Global Warming Potential (or a substitute metric) value that places proper weight on the near-term climate protection benefits of reducing black carbon.
- Investigate methods to identify and speciate the types of fine particles that will be reduced by potential control measures in order to target the particles that contribute most to climate heating.
- Consider developing additional control measures to reduce black carbon emissions in the next update of the Bay Area Clean Air Plan.
- In developing measures to reduce particulate emissions, prioritize controls on emission sources that have a high ratio of black carbon compared to organic carbon, sulfates, and nitrates.
- Continue to enhance the Air District's efforts to reduce residential wood-burning and to educate Bay Area residents about the detrimental health and climate effects of wood-burning.
- Encourage Bay Area cities (and other local agencies) to address black carbon in their climate protection plans, and provide technical support to assist in this effort.
- Work with local governments and other stakeholders to estimate the public health benefits of climate protection measures to reduce greenhouse gases, as well as short-term climate forcers such as black carbon.
- Monitor on-going research into the climate impacts of PM, and consider how new findings should be reflected in Air District policies and climate protection efforts.
- Facilitate communications between policy-makers and the scientific community to identify research needs regarding fine particles and climate impacts in order to reduce uncertainties in the policy arena.

Ammonia

As noted in Section 2 of this report, ammonia is a precursor to the formation of ammonium nitrate, a compound which accounts for a significant share of PM concentrations in the Bay Area, especially in winter months when PM concentrations are highest. NOx is converted to nitric acid, which then combines with ammonia to form ammonium nitrate. Ammonia is also a precursor to ammonium sulfate, which accounts for over 10% of Bay Area PM2.5 on an annual basis. For purposes of PM2.5 SIP planning requirements, US EPA guidelines[6] state that certain precursors (e.g., SO2 and NOx) are presumed to be significant contributors to the formation of secondary PM2.5, whereas others, such as volatile organic compounds (VOC) and ammonia are presumed **not** to be significant contributors unless the State makes a finding that the precursor contributes significantly to the PM2.5 non-attainment in the air basin or in downwind areas.

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[6] See US EPA Clean Air Fine Particle Implementation Rule, April 25, 2007: www.gpo.gov/fdsys/pkg/FR-2007-04-25/pdf/E7-6347.pdf#page=1

In determining whether ammonia is a significant contributor to PM formation, the key question is which pollutant – ammonia, or NOx (in the form of nitric acid) - is the limiting factor in ammonium nitrate formation. Based on the ratio of ammonia to NOx and the dynamics of ammonium nitrate and ammonium sulfate formation in the Bay Area, PM2.5 modeling performed by Air District staff found that reducing ammonia would be the most effective PM precursor to reduce in order to decrease PM2.5 concentrations, as described in Section 2.[7]

To date, PM2.5 SIP plans prepared by other air districts in the state, including South Coast, San Joaquin Valley, and Sacramento, have not treated ammonia as a significant precursor to PM and have not proposed any regulations or policies to control ammonia emissions. It should also be noted that, even if there is a solid technical rationale for controlling ammonia emissions in the Bay Area, there would be challenges in terms of how to control ammonia emissions. The source categories that account for the bulk of ammonia emissions are very different than for primary PM or the other PM precursors. The menu of available control measures appears to be limited, and some of the major sources (e.g. human perspiration and respiration, domestic animal waste) do not lend themselves to regulation.

To better inform a future decision as to whether to consider ammonia a significant contribute to secondary PM formation and pursue measures to reduce ammonia emissions, Air District staff will analyze future PM2.5 modeling results, seek to improve the Bay Area ammonia emissions inventory, and monitor on-going research and policy guidelines that shed light on this issue.

TECHNICAL ENHANCEMENTS NEEDED TO SUPPORT THE AIR DISTRICT'S EFFORTS

As discussed in the preceding sections of this report, the Air District has been building a solid technical and analytical foundation for its PM control efforts in recent years. The Air District has been performing cutting edge work in its efforts to measure, analyze, and characterize PM emissions, concentrations, population exposure, and health effects. However, because PM is such a complex pollutant, there are a number of gaps to be filled. Opportunities to enhance our technical understanding and capabilities in regard to PM include:

- Better measurements and/or estimates of localized PM concentrations;
- Better information as to population exposure to PM; and
- Technical data to characterize ultrafine PM emissions, concentrations, and population exposure in the Bay Area.

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[7] This is the opposite of results from the Central Valley, where modeling found that the Valley is so rich in ammonia, primarily from agricultural and livestock operations, that reducing ammonia emissions would have little or no effect on decreasing ammonium nitrate formation.

Emissions Inventory

The Air District has a detailed inventory of both primary PM emissions of PM2.5 and PM10, as well as the precursors that contribute to formation of secondary PM. Several potential enhancements to the existing inventory for PM2.5 and PM10 are described below.

Domestic cooking: The Air District PM inventory, like other PM inventories throughout California, currently does not include PM from domestic (residential) cooking. This category would include PM emitted by the both natural gas and electric stoves and ovens, as well as barbeque grills. It is possible that PM from domestic cooking could prove to be a significant source of emissions, given the number of stoves and ovens used in Bay Area homes, as well as the volume of outdoor cooking in summer months. As noted in Section 1-B, indoor PM levels spike when stoves and ovens are in use, and some fraction of this PM finds its way outdoors either by means of stove hood fans, windows, or other types of dispersion. Air District staff will investigate PM emissions from domestic cooking with the objective of developing emissions estimates for this category.

Projections of future wood smoke emissions: As discussed previously, information from speciation of PM collected on filters, as well as Bay Area wood-burning survey results, indicates that the Air District's program has been successful in reducing wood-burning during the winter months. The projections in the PM inventory currently assume that wood-burning emissions will remain constant in future years. However, it seems plausible that PM emissions from residential wood-burning will decrease further in future years in response to continued public education, the gradual phasing out of housing with uncertified wood stoves, conversion of fireplaces to natural gas inserts, and other factors. In order to better understand future wood-burning trends, Air District staff will analyze and research the various factors that influence residential wood-burning, such as the compliance rate during Spare the Air Alerts, the rate at which fireplaces and woodstoves are removed, upgraded, or converted to natural gas inserts, etc. Staff may revise the wood-burning survey to ensure that it provides information to track compliance with all provisions of the wood-burning regulation and overall progress reducing residential wood-burning.

Condensable PM emissions: As mentioned in the emissions inventory discussion, the inventory does not include gaseous emissions that condense to form particles when combustion exhaust cools upon exposure to ambient air. US EPA guidance for PM2.5 planning states that planning efforts should "consider" condensable emissions. This makes sense conceptually; the challenge is to develop test methods to accurately estimate condensable emissions for all the various emission source categories.

Ammonia inventory: As discussed previously, ammonia is one of the precursors to formation of ammonium nitrate, a key component of secondary PM. The Air District has developed an ammonia emissions inventory for purposes of photochemical modeling of PM2.5 in the Bay Area. ARB has developed a separate ammonia inventory for the Bay Area and for other air districts to use in preparing PM2.5 SIP submittals that are due to US EPA by December 2012. There are significant differences between the Air District's ammonia emissions inventory and the one provided by ARB. Air District staff will work to improve the Air District's ammonia inventory and to reconcile the differences with the ARB ammonia inventory, as methodologies to estimate ammonia emissions from the various source categories are improved and refined.

In addition to improving the existing inventory, the Air District will pursue new initiatives for the emissions inventory in regard to ultrafine PM and black carbon.

Regional ultrafine PM emissions inventory: The Air District released its first-ever emissions inventory for ultrafine PM in August 2012. The ultrafine PM inventory will be an important tool to help in analyzing key emission sources, performing photochemical modeling for PM, and laying the groundwork for a potential control strategy to reduce ultrafube PM. Comparing the breakdown of emission sources for ultrafine PM versus PM2.5 will be very instructive for purposes of informing future control efforts. The Air District will refine and revise the ultrafine PM inventory as new emission factors become available.

Develop black carbon inventory: As noted in the section on PM and Climate, the component of PM known as black carbon is a potent climate heating agent produced primarily by combustion of fossil fuels and biomass. Air District staff will consider the utility and feasibility of developing a black carbon inventory in order to inform future efforts to reduce black carbon emissions.

PM Monitoring / Ambient Concentrations

Ultrafine PM Monitoring: The Air District recently installed several ultrafine PM monitors to provide continuous measurements in various Bay Area locations, as described in Section 3-A. The monitors will serve as the preliminary foundation for the District's effort to develop data as to ultrafine particle number concentrations. Looking forward, the District will consider expanding its UFPM monitoring network, subject to the availability of resources to purchase and operate the monitors.

Monitoring Localized and Episodic Concentrations: The Air District will also pursue new technologies, opportunities, and partnerships to increase the density of PM2.5 measurements in the region, especially in most impacted communities, and near freeways and other major emission sources where PM "hot spots" are most likely to occur. In addition, the Air District will also investigate whether the network can be augmented with accurate, real-time, mobile PM measurement capabilities to determine impacts from short-term episodes and to provide better understanding as to how PM concentrations vary over space and time.

PM Photochemical Modeling

Additional PM2.5 Modeling: The Air District performed initial photochemical modeling for PM2.5 in 2009, as described in Section 2. The 2009 modeling was performed on a 4 km by 4 km grid scale using emissions and meteorological data based upon several high PM episodes in winter months. The Air District is currently performing additional regional-scale PM2.5 modeling to simulate PM formation fat the regional scale for a full year, using emissions and meteorological data from all four seasons in order to be able to simulate PM formation for the entire year. Since most epidemiological studies are based on annual average exposure, this will enable the Air

District to more accurately estimate the health effects related to PM in the Bay Area. In addition, it will provide a better understanding of secondary PM formation, and how the formation of secondary PM contributes to ambient PM concentrations for the entire year, rather than just the winter months. This modeling will also analyze potential impacts of reductions in emissions from key PM sources, such as wood smoke. As resources permit, the Air District may consider performing additional PM modeling using a finer grid resolution in order to better identify local areas with the potential for high PM formation.

Ultrafine PM Modeling: Information as to ultrafine particle concentrations in the Bay Area is currently very limited. The Air District will perform its first-ever ultrafine PM modeling on a region-wide basis on a 4 km by 4 km grid scale. The UFPM modeling will simulate concentrations to identify potential hot spot areas in the Bay Area. This modeling will also establish a relationship between UFPM levels and meteorological conditions.

Analyzing Population Exposure to PM

Reducing population exposure to PM in the Bay Area is the key to reducing its health effects. This can be accomplished both by reducing emissions and by avoiding exposures. However, as noted in Section 1-B, analyzing population exposure to PM requires accurate data as to (1) ambient concentrations of PM at a fine-grained spatial scale throughout the region, (2) personal activity patterns; that is, where and when people are exposed to PM, and (3) estimates of PM concentrations in various micro-environments such as homes, schools, cars, and sidewalks, and how these concentrations are related to ambient levels.

The Air District will consider how to improve its PM population exposure assessment capabilities. Improved exposure assessment methods could potentially be used for any or all of the following purposes:

- To evaluate total population exposure to ambient PM in the Bay Area, and to track progress in reducing population exposure at the regional scale or among defined population sub-groups.
- To estimate the PM exposure from outdoor versus indoor environments, as well as the contribution of key micro-environments, such as in-vehicle, near-roadway, etc.
- To inform future efforts to target and implement the Air District’s CARE program and Clean Air Communities Initiative (CACI).
- To inform analyses and policy decisions at the regional and local level to promote focused growth, including the development of station area plans, general plans, specific plans, community risk reduction plans, etc.
- To help Bay Area residents understand the magnitude and the key sources or their personal exposure to PM, and how they reduce their exposure.

Analyzing Health & Climate Impacts

Air District staff will continue to monitor the latest research as to the health and climate effects of PM and incorporate this information into its PM reduction program. In terms of estimating the impact of PM on public health in the Bay Area, the Air District will:

- Update the findings of its September 2011 report on the morbidity and mortality impacts of PM2.5 in Bay Area, and the economic cost of those impacts, based upon the new round of PM2.5 modeling;
- Update the District's Multi-Pollutant Evaluation Method (MPEM) to reflect results of full-year PM2.5 modeling, the latest health risk factors, and the latest information as to the climate impacts of greenhouse gases and the value of reducing GHG emissions; and
- Develop estimates as to the health and economic impacts of ultrafine PM, based on the results of ultrafine PM modeling and health studies.

HOW BAY AREA RESIDENCES CAN REDUCE THEIR PERSONAL EXPOSURE TO PM

In concert with the Air District's efforts, Bay Area residents may decrease their personal exposure to PM by taking simple steps including the following:

- Minimize time spent driving on, or in close proximity to, busy roadways, especially those that carry a high volume of heavy-duty diesel vehicles.
- Avoid opening vents and windows while driving on busy roadways.
- Avoid smoke from tobacco products, incense and candles.
- Avoid exposure to wood smoke. Don't burn wood in a fireplace or stove. Avoid campfires, bonfires, and charcoal fires. Replace wood-burning fireplaces with a natural gas insert.
- Reduce exposure to PM from cooking by ventilating the kitchen when cooking, and switching to electric pilot lights.
- Change filters in furnaces and range hoods on a regular bsais.
- Reduce exposure to PM and other air pollutants from cleaning products by ventilating work areas while cleaning and disposing of used rags promptly.

CONCLUSION

The Air District is committed to analyzing and reducing PM to protect public health, the climate, and the environment. To maintain progress in reducing Bay Area PM levels in the coming years, the Air District will continue to monitor the latest research on PM impacts to public health and the environment, and to enhance its technical capabilities to measure and analyze PM. In addition, the Air District will maintain its efforts to reduce PM by implementing the control measures described in the Bay Area 2010 Clean Air Plan and by considering potential additional measures (to be determined at a future date) in the course of developing future air quality plans.

Appendix A -
Table 1: Bay Area Winter Emissions Inventory for PM & PM Precursors 2010-2030

Appendix A
Bay Area Winter Emissions Inventory for Primary PM + PM Precursors: 2010-2030

Winter Emissions tons/day

SOURCE CATEGORY	PM$_{10}$ 2010	PM$_{10}$ 2015	PM$_{10}$ 2020	PM$_{10}$ 2025	PM$_{10}$ 2030	PM$_{2.5}$ 2010	PM$_{2.5}$ 2015	PM$_{2.5}$ 2020	PM$_{2.5}$ 2025	PM$_{2.5}$ 2030	ROG 2010	ROG 2015	ROG 2020	ROG 2025	ROG 2030	NOx 2010	NOx 2015	NOx 2020	NOx 2025	NOx 2030	SO$_2$ 2010	SO$_2$ 2015	SO$_2$ 2020	SO$_2$ 2025	SO$_2$ 2030
INDUSTRIAL/COMMERCIAL PROCESSES																									
Basic Refining Processes	0.2	0.2	0.2	0.2	0.2	0.1	0.1	0.1	0.1	0.1	0.2	0.2	0.2	0.2	0.2	--	--	--	--	--	0.2	0.2	0.2	0.3	0.3
Wastewater (Oil-Water) Separators	--	--	--	--	--	--	--	--	--	--	1.2	1.2	1.3	1.4	1.4	--	--	--	--	--	--	--	--	--	--
Wastewater Treatment Facilities	--	--	--	--	--	--	--	--	--	--	0.5	0.5	0.5	0.5	0.6	--	--	--	--	--	--	--	--	--	--
Cooling Towers	--	--	--	--	--	--	--	--	--	--	0.9	0.9	1.0	1.0	1.1	--	--	--	--	--	--	--	--	--	--
Flares & Blowdown Systems	--	--	0.1	0.1	0.1	--	--	--	--	--	0.8	0.8	0.9	0.9	1.0	1.2	1.3	1.3	1.4	1.5	0.6	0.7	0.7	0.7	0.8
Other Refining Processes	--	--	--	--	--	--	--	--	--	--	0.3	0.3	0.3	0.3	0.4	--	--	--	--	--	--	--	--	--	--
Fugitives	--	--	--	--	--	--	--	--	--	--	0.5	0.5	0.6	0.6	0.6	--	--	--	--	--	--	--	--	--	--
Subtotal	**0.3**	**0.3**	**0.3**	**0.3**	**0.3**	**0.2**	**0.2**	**0.2**	**0.2**	**0.2**	**4.3**	**4.5**	**4.7**	**5.0**	**5.2**	**1.2**	**1.3**	**1.4**	**1.4**	**1.5**	**0.9**	**0.9**	**1.0**	**1.0**	**1.1**
CHEMICAL MANUFACTURING FACILITIES																									
Coating, Inks, Resins & Other Facilities	0.1	0.1	0.1	0.1	0.1	--	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	2.7	2.9	3.0	3.2	3.3
Pharmaceuticals & Cosmetics	0.4	0.4	0.4	0.5	0.5	0.4	0.4	0.4	0.4	0.5	0.8	0.8	0.9	0.9	1.0	1.4	1.4	1.5	1.6	1.7	3.2	3.3	3.5	3.7	3.9
Fugitives - Valves & Flanges	--	--	--	--	--	--	--	--	--	--	0.6	0.7	0.5	0.6	0.6	--	--	--	--	--	--	--	--	--	--
Subtotal	**0.5**	**0.5**	**0.5**	**0.5**	**0.6**	**0.4**	**0.5**	**0.5**	**0.5**	**0.5**	**1.5**	**1.6**	**1.5**	**1.6**	**1.7**	**1.5**	**1.5**	**1.6**	**1.7**	**1.8**	**5.9**	**6.2**	**6.5**	**6.8**	**7.2**
OTHER INDUSTRIAL / COMMERCIAL PROCESSES																									
Bakeries	--	--	--	--	--	--	--	--	--	--	0.8	0.9	0.9	1.0	1.0	--	--	--	--	--	--	--	--	--	--
Cooking(Charbroiling)	2.0	1.8	2.0	2.2	2.4	1.5	1.4	1.5	1.7	1.9	0.9	1.0	1.1	1.2	1.3	--	--	--	--	--	--	--	--	--	--
Cooking(Deep Fat Frying)	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	--	--	--	--	--	--	--	--	--	--
Cooking(Griddles)	0.6	0.7	0.7	0.8	0.9	0.2	0.2	0.2	0.2	0.2	0.1	0.1	0.1	0.1	0.2	--	--	--	--	--	--	--	--	--	--
Wineries & Other Food & Agr. Processes	0.5	0.6	0.6	0.7	0.7	0.3	0.3	0.4	0.4	0.4	1.1	1.1	1.2	1.2	1.2	--	--	--	--	--	--	--	0.1	0.1	0.1
Metallurgical & Minerals	2.6	2.8	3.0	3.3	3.5	1.8	1.9	2.1	2.2	2.4	0.2	0.2	0.2	0.2	0.2	0.2	0.3	0.3	0.3	0.3	0.2	0.2	0.2	0.2	0.3
Waste Management	2.1	2.1	2.2	2.2	2.2	0.6	0.6	0.6	0.6	0.6	3.1	3.1	3.2	3.2	3.2	--	--	--	--	--	--	--	--	--	--
Semiconductor Mfg.	--	--	--	--	--	--	--	--	--	--	0.2	0.2	0.3	0.3	0.3	--	--	--	--	--	--	--	--	--	--
Fiberglass Products Mfg.	--	--	--	--	--	--	--	--	--	--	0.1	0.2	0.2	0.2	0.2	--	--	--	--	--	--	--	--	--	--
Rubber & Plastic Products	--	--	--	--	--	--	--	--	--	--	0.3	0.3	0.3	0.3	0.3	--	--	--	--	--	--	--	--	--	--
Contaminated Soil Aeration	--	--	--	--	--	--	--	--	--	--	0.1	0.1	0.1	0.1	0.1	--	--	--	--	--	--	--	--	--	--
Other Industrial Commercial	0.8	0.9	1.0	1.0	1.1	0.4	0.5	0.5	0.5	0.6	0.9	1.0	1.1	1.1	1.2	--	--	--	--	--	--	--	--	--	--
Subtotal	**8.8**	**9.0**	**9.6**	**10.3**	**11.0**	**4.9**	**5.0**	**5.3**	**5.8**	**6.2**	**7.9**	**8.3**	**8.6**	**9.1**	**9.5**	**0.3**	**0.3**	**0.3**	**0.4**	**0.4**	**0.3**	**0.3**	**0.3**	**0.3**	**0.4**
PETROLEUM PRODUCT / SOLVENT EVAPORATION																									
PETROLEUM REFINERY EVAPORATION																									
Storage Tanks	--	--	--	--	--	--	--	--	--	--	3.3	3.5	3.6	3.8	4.0	--	--	--	--	--	--	--	--	--	--
Loading Operations	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.2	0.2	0.2	--	--	--	--	--	--	--	--	--	--
Subtotal	**0.1**	**0.1**	**0.1**	**0.1**	**0.1**	**0.1**	**0.1**	**0.1**	**0.1**	**0.1**	**3.4**	**3.6**	**3.8**	**4.0**	**4.2**	**--**	**--**	**--**	**--**	**--**	**--**	**--**	**--**	**--**	**--**
FUELS DISTRIBUTION																									
Natural Gas Distribution	--	--	--	--	--	--	--	--	--	--	0.7	0.8	0.8	0.8	0.8	--	--	--	--	--	--	--	--	--	--
Bulk Plants & Terminals	--	--	--	--	--	--	--	--	--	--	0.7	0.6	0.6	0.6	0.6	--	--	--	--	--	--	--	--	--	--
Trucking	--	--	--	--	--	--	--	--	--	--	2.6	2.6	2.5	2.5	2.5	--	--	--	--	--	--	--	--	--	--
Gasoline Filling Stations	--	--	--	--	--	--	--	--	--	--	5.2	4.9	4.8	4.6	4.5	--	--	--	--	--	--	--	--	--	--
Aircraft Fueling	--	--	--	--	--	--	--	--	--	--	1.1	1.0	1.0	1.0	1.0	--	--	--	--	--	--	--	--	--	--
Recreational Boat Fueling	--	--	--	--	--	--	--	--	--	--	0.9	0.9	1.0	1.0	1.0	--	--	--	--	--	--	--	--	--	--
Portable Fuel Container Spillage	--	--	--	--	--	--	--	--	--	--	3.8	2.8	2.3	2.0	1.8	--	--	--	--	--	--	--	--	--	--
Other Fueling	--	--	--	--	--	--	--	--	--	--	0.4	0.4	0.4	0.5	0.5	--	--	--	--	--	--	--	--	--	--

Appendix A

Bay Area Winter Emissions Inventory for Primary PM + PM Precursors: 2010-2030

SOURCE CATEGORY	Winter Emissions tons/day																								
	PM$_{10}$ 2010	PM$_{10}$ 2015	PM$_{10}$ 2020	PM$_{10}$ 2025	PM$_{10}$ 2030	PM$_{2.5}$ 2010	PM$_{2.5}$ 2015	PM$_{2.5}$ 2020	PM$_{2.5}$ 2025	PM$_{2.5}$ 2030	ROG 2010	ROG 2015	ROG 2020	ROG 2025	ROG 2030	NOx 2010	NOx 2015	NOx 2020	NOx 2025	NOx 2030	SO$_2$ 2010	SO$_2$ 2015	SO$_2$ 2020	SO$_2$ 2025	SO$_2$ 2030
Subtotal	--	--	--	--	--	--	--	--	--	--	**15.3**	**14.0**	**13.4**	**13.0**	**12.8**	--	--	--	--	--	--	--	--	--	--
OTHER ORGANIC COMPOUNDS																									
Cold Cleaning	--	--	--	--	--	--	--	--	--	--	2.7	2.9	3.1	3.3	3.5	--	--	--	--	--	--	--	--	--	--
Vapor Degreasing	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Handwiping	--	--	--	--	--	--	--	--	--	--	2.3	2.5	2.7	2.8	3.0	--	--	--	--	--	--	--	--	--	--
Dry Cleaners	--	--	--	--	--	--	--	--	--	--	0.6	0.7	0.7	0.7	0.8	--	--	--	--	--	--	--	--	--	--
Printing	--	--	--	--	--	--	--	--	--	--	4.1	4.1	4.2	4.4	4.5	--	--	--	--	--	--	--	--	--	--
Adhesives & Sealants	--	--	--	--	--	--	--	--	--	--	9.7	10.6	11.5	12.8	14.0	--	--	--	--	--	--	--	--	--	--
Structures Coating	--	--	--	--	--	--	--	--	--	--	21.2	16.3	17.1	17.8	18.6	--	--	--	--	--	--	--	--	--	--
Industrial/Commercial Coating	--	--	--	--	--	--	--	--	--	--	8.1	6.8	7.3	7.6	8.1	--	--	--	--	--	--	--	--	--	--
Storage Tanks	--	--	--	--	--	--	--	--	--	--	0.8	0.8	0.9	0.9	0.9	--	--	--	--	--	--	--	--	--	--
Lightering & Ballsting	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Other Organics Evaporation	--	--	--	--	--	--	--	--	--	--	1.5	1.6	1.7	1.8	2.0	--	--	--	--	--	--	--	--	--	--
Subtotal	--	--	--	--	--	--	--	--	--	--	**51.0**	**46.2**	**49.0**	**52.2**	**55.4**	--	--	--	--	--	--	--	--	--	--
COMBUSTION - STATIONARY SOURCES																									
FUELS COMBUSTION																									
Natural Gas(Space/Water Heating/Cooking)	1.7	1.8	1.9	2.0	2.1	1.7	1.8	1.9	2.0	2.1	1.0	1.1	1.2	1.2	1.3	18.3	19.8	20.9	22.1	23.3	0.1	0.1	0.2	0.2	0.2
LPG and Liquid Fuel	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	0.5	0.5	0.5	0.5	0.5	0.3	0.3	0.2	0.2	0.2
Solid Fuel (Woodstoves)	4.0	4.0	4.0	4.0	4.0	3.7	3.7	3.7	3.7	3.7	2.8	2.8	2.8	2.8	2.8	0.5	0.5	0.5	0.5	0.5	0.1	0.1	0.1	0.1	0.1
Solid Fuel (Fireplaces)	13.9	13.9	13.9	13.9	13.9	13.1	13.1	13.1	13.1	13.1	5.3	5.3	5.3	5.3	5.3	1.1	1.1	1.1	1.1	1.1	0.2	0.2	0.2	0.2	0.2
Cogeneration	0.7	0.7	0.8	0.8	0.9	0.7	0.7	0.8	0.8	0.9	1.9	2.0	2.1	2.2	2.3	3.9	4.1	4.3	4.6	4.9	0.3	0.4	0.4	0.4	0.4
Power Plants	0.3	0.3	0.3	0.4	0.4	0.3	0.3	0.3	0.4	0.4	0.1	0.1	0.1	0.1	0.1	1.0	1.1	1.2	1.3	1.4	0.1	0.1	0.1	0.1	0.1
Oil Refineries External Combustion	2.5	2.6	2.7	2.8	3.0	2.4	2.5	2.7	2.8	2.9	1.0	1.1	1.1	1.2	1.2	10.5	11.0	11.6	12.2	12.8	14.0	14.7	15.5	16.2	17.1
Glass Melting Furnaces - Natural Gas	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	0.9	0.9	0.9	1.0	1.0	--	--	--	--	--
Reciprocating Engines	0.2	0.1	0.1	0.1	0.2	0.2	0.1	0.1	0.1	0.2	0.5	0.5	0.5	0.5	0.6	2.9	2.7	2.6	2.7	2.9	0.1	0.1	0.1	0.1	0.1
Turbines	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	--	--	--	--	--	0.8	0.8	0.9	0.9	1.0	--	--	--	--	--
Landfill/Cement Plant Combustion	1.6	1.6	1.7	1.7	1.8	1.6	1.6	1.7	1.7	1.8	1.4	1.5	1.5	1.6	1.6	15.4	16.2	17.1	17.8	18.7	3.0	3.2	3.4	3.6	3.8
Subtotal	**24.8**	**25.2**	**25.5**	**25.9**	**26.3**	**23.8**	**24.1**	**24.5**	**24.8**	**25.2**	**14.0**	**14.3**	**14.6**	**15.0**	**15.3**	**55.6**	**58.7**	**61.4**	**64.6**	**67.9**	**18.2**	**19.1**	**20.1**	**21.1**	**22.1**
BURNING OF WASTE MATERIAL																									
Incineration	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.2	0.2	0.2	0.3	--	--	--	--	--
Planned Fires	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.2	0.2	0.2	0.2	0.2	--	--	--	--	--
Subtotal	**0.5**	**0.5**	**0.5**	**0.5**	**0.5**	**0.5**	**0.5**	**0.5**	**0.5**	**0.5**	**0.4**	**0.4**	**0.4**	**0.4**	**0.4**	**0.4**	**0.4**	**0.4**	**0.4**	**0.4**	--	--	--	--	**0.1**
Banked Emissions	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Alternative Compliance Allowance	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Subtotal	**34.9**	**35.5**	**36.5**	**37.7**	**38.8**	**29.8**	**30.3**	**31.0**	**31.9**	**32.8**	**97.9**	**92.9**	**96.1**	**100.2**	**104.5**	**59.0**	**62.2**	**65.1**	**68.4**	**72.0**	**25.3**	**26.5**	**27.9**	**29.3**	**30.8**
COMBUSTION - MOBILE SOURCES																									
ON-ROAD MOTOR VEHICLES																									
Passenger Cars	5.1	5.2	5.4	5.6	5.8	2.3	2.2	2.3	2.4	2.5	42.1	21.5	12.4	10.1	9.3	35.5	18.8	11.6	9.8	9.2	0.4	0.4	0.4	0.4	0.4
Light Duty Trucks<6000lbs	2.3	2.3	2.3	2.4	2.5	1.0	1.0	1.0	1.0	1.1	23.4	15.8	10.9	9.0	7.8	27.1	16.2	9.9	7.1	5.8	0.2	0.2	0.2	0.2	0.2
Medium Duty Trucks 6001-8500 lbs	1.2	1.2	1.2	1.2	1.3	0.5	0.5	0.5	0.5	0.5	10.1	9.2	7.9	6.6	5.9	17.9	13.0	9.1	6.5	5.0	0.2	0.2	0.2	0.2	0.2
Light Heavy Duty Trucks 8501-14000lbs	0.7	0.6	0.6	0.6	0.6	0.4	0.3	0.3	0.3	0.3	6.2	5.1	4.2	3.4	2.8	23.1	18.0	13.8	10.6	8.5	0.1	0.1	0.1	0.1	0.1
Medium Heavy Duty Trucks 14001-33000lbs	1.1	0.7	0.6	0.6	0.6	0.8	0.5	0.3	0.3	0.3	2.4	1.3	0.8	0.7	0.7	21.6	13.8	6.6	3.9	4.0	--	--	--	--	--

Appendix A
Bay Area Winter Emissions Inventory for Primary PM + PM Precursors: 2010-2030

SOURCE CATEGORY	Winter Emissions tons/day																								
	PM$_{10}$ 2010	PM$_{10}$ 2015	PM$_{10}$ 2020	PM$_{10}$ 2025	PM$_{10}$ 2030	PM$_{2.5}$ 2010	PM$_{2.5}$ 2015	PM$_{2.5}$ 2020	PM$_{2.5}$ 2025	PM$_{2.5}$ 2030	ROG 2010	ROG 2015	ROG 2020	ROG 2025	ROG 2030	NOx 2010	NOx 2015	NOx 2020	NOx 2025	NOx 2030	SO$_2$ 2010	SO$_2$ 2015	SO$_2$ 2020	SO$_2$ 2025	SO$_2$ 2030
Heavy Heavy Duty Trucks>33000 lbs	2.0	1.0	0.9	1.0	1.1	1.6	0.7	0.5	0.6	0.6	2.8	1.8	1.8	1.9	2.1	51.7	36.5	23.8	15.6	16.5	0.1	0.1	0.1	0.1	0.1
School/Urban Buses	1.0	0.8	0.8	0.8	0.8	0.6	0.5	0.4	0.4	0.4	1.4	1.0	0.9	0.8	0.6	16.9	14.0	11.3	10.2	8.2	--	--	--	--	--
Motor-Homes	--	--	--	--	--	--	--	--	--	--	1.4	1.2	0.8	0.5	0.4	1.4	1.0	0.8	0.6	0.5	--	--	--	--	--
Motorcycles	0.1	0.1	0.1	0.1	0.1	--	--	--	--	--	17.5	13.7	13.0	13.3	13.8	2.3	2.4	2.5	2.6	2.7	--	--	--	--	--
Subtotal	**13.5**	**11.9**	**11.9**	**12.3**	**12.8**	**7.3**	**5.6**	**5.4**	**5.6**	**5.8**	**107.4**	**70.6**	**52.6**	**46.1**	**43.3**	**197.6**	**133.7**	**89.5**	**66.8**	**60.5**	**0.9**	**0.9**	**1.0**	**1.0**	**1.1**
OFF-HIGHWAY MOBILE SOURCES																									
Lawn and Garden Equipment	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	13.6	11.8	10.8	10.8	11.0	2.1	2.0	2.0	2.1	2.2	--	--	--	--	--
Transportation Refrigeration Units	0.6	0.3	0.1	0.1	0.1	0.6	0.3	0.1	0.1	0.1	1.9	0.9	0.9	1.2	1.5	7.7	7.5	7.1	8.4	10.4	--	--	--	--	--
Agricultural Equipment	0.3	0.2	0.1	0.1	--	0.3	0.2	0.1	0.1	--	1.0	0.7	0.4	0.3	0.3	5.9	4.1	2.7	1.7	1.2	--	--	--	--	--
Construction and Mining Equipment	0.6	0.5	0.3	0.3	0.3	0.6	0.5	0.3	0.3	0.3	1.6	1.4	1.0	1.0	1.1	11.7	9.6	5.2	4.2	3.8	--	--	--	--	--
Industrial Equipment	0.2	0.2	0.1	0.1	0.1	0.2	0.2	0.1	0.1	0.1	1.1	0.9	0.9	0.9	0.9	6.5	5.4	5.0	5.0	5.1	--	--	--	--	--
Light Duty Commercial Equipment	0.6	0.5	0.4	0.4	0.3	0.6	0.5	0.4	0.3	0.3	4.5	3.4	2.8	2.7	2.8	7.4	6.3	4.9	4.1	3.8	--	--	--	--	--
Trains	0.2	0.2	0.2	0.3	0.3	0.2	0.2	0.2	0.3	0.3	0.9	0.9	0.9	0.9	1.0	10.7	12.3	13.0	13.8	14.9	--	--	--	--	--
Off Road Recreational Vehicles	--	--	--	--	--	--	--	--	--	--	1.3	1.2	1.3	1.3	1.4	--	--	--	--	--	--	--	--	--	--
Ships	0.2	0.2	0.2	0.2	0.3	0.2	0.2	0.2	0.2	0.3	0.4	0.5	0.6	0.7	0.7	8.3	8.5	7.5	6.1	5.0	1.8	0.7	0.8	0.8	0.9
Commercial Harborcraft	0.6	0.4	0.3	0.2	0.2	0.6	0.4	0.3	0.2	0.2	1.2	1.1	1.1	1.1	1.1	16.1	11.1	9.2	8.9	8.9	--	--	--	--	--
Recreational Boats	0.5	0.7	0.9	1.1	1.5	0.5	0.7	0.9	1.1	1.5	8.5	8.0	7.9	8.3	9.5	2.3	2.3	2.4	2.4	2.7	--	--	--	--	--
Subtotal	**4.1**	**3.4**	**2.8**	**2.9**	**3.3**	**4.0**	**3.3**	**2.8**	**2.9**	**3.2**	**35.9**	**30.7**	**28.6**	**29.3**	**31.3**	**78.7**	**69.2**	**59.0**	**56.8**	**58.0**	**1.9**	**0.7**	**0.8**	**0.9**	**1.0**
AIRCRAFT																									
Commercial Aircraft	0.1	0.2	0.2	0.2	0.2	0.1	0.2	0.2	0.2	0.2	2.4	3.1	3.7	4.0	4.3	9.1	11.1	14.6	16.6	17.5	0.8	0.9	1.0	1.1	1.3
General Aviation	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.5	0.5	0.6	0.6	0.6	0.1	0.2	0.2	0.2	0.2	--	--	--	--	--
Military Aircraft	0.1	0.2	0.2	0.2	0.2	0.1	0.2	0.2	0.2	0.2	0.7	1.2	1.2	1.2	1.3	1.7	4.0	4.0	4.1	4.1	0.1	0.2	0.2	0.2	0.2
Airport Ground Support Equipment	--	--	--	--	--	--	--	--	--	--	0.1	0.1	0.1	--	0.1	0.8	0.7	0.5	0.3	0.6	--	--	--	0.1	--
Subtotal	**0.4**	**0.4**	**0.4**	**0.5**	**0.5**	**0.3**	**0.4**	**0.4**	**0.4**	**0.5**	**3.7**	**4.9**	**5.5**	**5.8**	**6.3**	**11.7**	**15.8**	**19.2**	**21.2**	**22.3**	**0.9**	**1.2**	**1.3**	**1.5**	**1.5**
MISCELLANEOUS OTHER SOURCES																									
Construction Operations	10.1	11.2	12.2	13.6	14.8	1.0	1.1	1.2	1.4	1.5	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Farming Operations	1.5	1.4	1.4	1.4	1.3	0.2	0.2	0.2	0.2	0.2	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Entrained Road Dust-Paved Roads	23.5	23.5	23.5	23.5	23.5	3.5	3.5	3.5	3.5	3.5	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Entrained Road Dust-Unpaved Roads	3.8	3.8	3.8	3.8	3.8	0.4	0.4	0.4	0.4	0.4	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Wind Blown Dust	3.9	3.9	3.9	3.9	3.9	0.7	0.7	0.7	0.7	0.7	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Animal Waste	7.3	7.4	7.5	7.5	7.6	0.9	0.9	0.9	0.9	1.0	3.9	3.9	3.9	4.0	4.0	--	--	--	--	--	--	--	--	--	--
Agricultural Pesticides	--	--	--	--	--	--	--	--	--	--	0.6	0.6	0.6	0.6	0.6	--	--	--	--	--	--	--	--	--	--
Non-Agricultural Pesticides	--	--	--	--	--	--	--	--	--	--	0.3	0.3	0.3	0.3	0.3	--	--	--	--	--	--	--	--	--	--
Consumer Products(Excluding Pesticides)	--	--	--	--	--	--	--	--	--	--	44.1	45.3	46.5	47.5	48.6	--	--	--	--	--	--	--	--	--	--
Accidental Fires and Cigarette Smoking	1.3	1.2	1.1	1.2	1.2	1.2	1.1	1.1	1.1	1.1	0.4	0.4	0.4	0.4	0.4	0.2	0.2	0.2	0.2	0.2	--	--	--	--	--
Subtotal	**51.3**	**52.4**	**53.3**	**54.8**	**56.1**	**7.9**	**8.0**	**8.0**	**8.2**	**8.3**	**49.2**	**50.5**	**51.8**	**52.8**	**53.9**	**0.2**	**0.2**	**0.2**	**0.2**	**0.2**	**--**	**--**	**--**	**--**	**--**
GRAND TOTAL EMISSIONS	**104**	**104**	**105**	**108**	**111**	**49**	**48**	**48**	**49**	**51**	**294**	**250**	**235**	**234**	**239**	**347**	**281**	**233**	**213**	**213**	**29**	**29**	**31**	**33**	**34**

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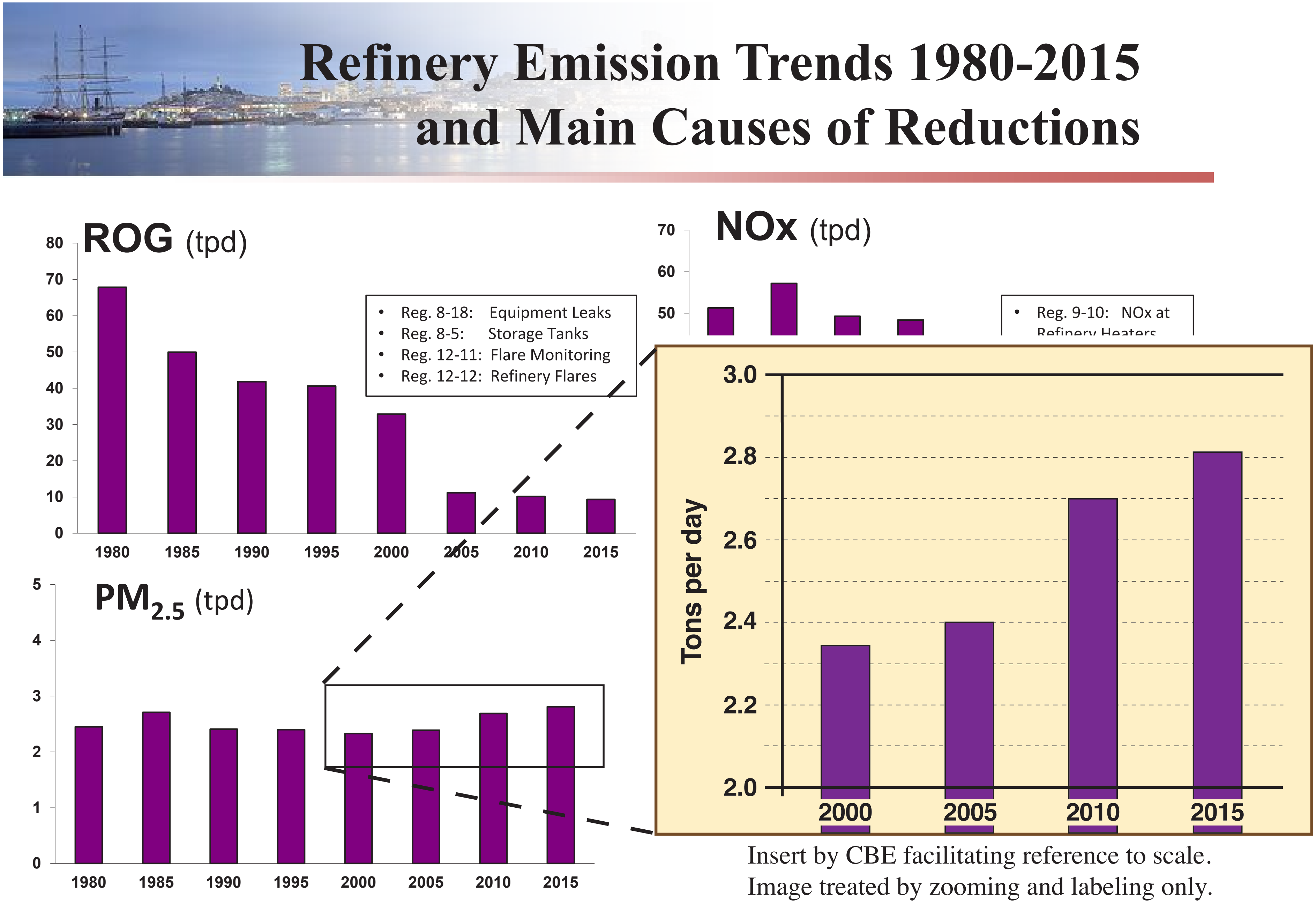
Attachment 5

Attachment 5 to CBE October 2015 Supplemental Comments: Slide excerpted from *Regulations to Track and Mitigate Emissions from Petroleum Refineries Regulation 12, Rules 15 and 16;* March 2015 BAAQMD Staff Worshop Presentation.



Refinery Emission Trends 1980-2015 and Main Causes of Reductions

- Reg. 8-18: Equipment Leaks
- Reg. 8-5: Storage Tanks
- Reg. 12-11: Flare Monitoring
- Reg. 12-12: Refinery Flares

- Reg. 9-10: NOx at Refinery Heaters

Insert by CBE facilitating reference to scale.
Image treated by zooming and labeling only.

Attachment 6

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Size distribution and chemical composition of metalliferous stack emissions in the San Roque petroleum refinery complex, southern Spain

A.M. Sánchez de la Campa[a,*], T. Moreno[b], J. de la Rosa[c], A. Alastuey[b], X. Querol[b]

[a] *Estación Experimental del Zaidín, CSIC, C/Profesor Albareda 1, E18008 Granada, Spain*
[b] *Institute of Environmental Assessment and Water Research IDÆA, CSIC, C/Jordi Girona, 18-26, 08034 Barcelona, Spain*
[c] *Associate Unit CSIC – University of Huelva "Atmospheric Pollution", Center for Research in Sustainable Chemistry (CIQSO), University of Huelva, E21071 Huelva, Spain*

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ABSTRACT

We demonstrate that there is great variation in the size range and chemical composition of metalliferous particulate matter (PM) present within petrochemical complex chimney stacks. Cascade impactor PM samples from seven size ranges (17, 14, 5, 2.5, 1.3, 0.67, and 0.33 μm) were collected from inside stacks within the San Roque complex which includes the largest oil refinery in Spain. SEM analysis demonstrates the PM to be mostly carbonaceous and aluminous fly ash and abundant fine metalliferous particles. The metals with the most extreme concentrations averaged over all size ranges were Ni (up to 3295 $\mu g\,m^{-3}$), Cr (962 $\mu g\,m^{-3}$), V (638 $\mu g\,m^{-3}$), Zn (225 $\mu g\,m^{-3}$), Mo (91 $\mu g\,m^{-3}$), La (865 $\mu g\,m^{-3}$), and Co (94 $\mu g\,m^{-3}$). Most metal PM are strongly concentrated into the finest fraction (<0.33 μm), although emissions from some processes, such as purified terephthallic acid (PTA) production, show coarser size ranges. The fluid catalytic cracking stack shows high concentrations of La (>200 $\mu g\,m^{-3}$ in $PM_{0.67-1.3}$), Cr and Ni in a relatively coarse PM size range (0.7–14 μm). Our unique database, directly sampled from chimney stacks, confirms that oil refinery complexes such as San Roque are a potent source of a variety of fine, deeply inhalable metalliferous atmospheric PM emissions.

1. Introduction

A variety of gaseous pollutants (e.g. SO_2, CO, NO_x, and H_2S), hydrocarbons, fly ashes and metalliferous particles are released into the atmosphere during the refining of crude oil and processing of its downstream products in petrochemical complexes [1]. Despite atmospheric emissions in Europe and North America being under greater governmental control than in other continents, petroleum refineries are still major pollution hot spots which inevitably impact on local ecosystems [2,3] and human health [4], influencing not only workers [5,6] but also surrounding populations (e.g. [7–9]).

This study deals with the size distributions and chemical characterisation of particulate matter emitted by different chimney stacks in the San Roque petrochemical refinery complex, which is situated on the north side of the Bay of Algeciras in southern Spain (Fig. 1). The Bay of Algeciras is a well documented industrial pollution hot spot [10], with relatively high concentrations of metals such as Ni, V, Cr and La in PM_{10} and $PM_{2.5}$ being attributed to both industry and shipping [11–14]. The Gibraltar Strait is accessed by around 80,000 ships per year, leaving or entering the Mediterranean. Abnormally high values of La compared to heavier lanthanoids (e.g. La/Ce > 1) implicate the San Roque FCC refinery as a point source for some of these metals [14]. In the present paper we go directly to this source and report on PM samples collected from inside the chimney stacks of the San Roque refinery. To our knowledge this is the first time that detailed physical and chemical characterisation of such samples has been published.

2. Study location

The San Roque refinery complex lies in a densely populated area that includes two major towns, Algeciras (120,000 inhabitants) and La Linea (65,000 inhabitants), which lie 9 km apart and diametrically opposite each other on the SW and NE sides of the bay (Fig. 1). The refinery has a capacity of 240,000 b/d (annual distillation of 12×10^6 tonnes of oil), making it the largest in Spain, and produces all types of fuel (propane, butane, gasoline, aviation fuel, diesel and fuel oils). Adjacent to the main refinery plants (which include a fluid catalytic converting facility), further petrochemical processing areas with chimney stacks include those of the Guadarranque and Lubrisur plants (Fig. 1), both of which were also incorporated in this study.

Within the refinery there are two distillation units (Crude I and Crude III), these being characterised by the presence of distillation column towers operating at atmospheric pressure, where crude oil

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* Corresponding author. Tel.: +34 959 219855; fax: +34 959 219810.
E-mail address: ana.sanchez@pi.uhu.es (A.M. Sánchez de la Campa).

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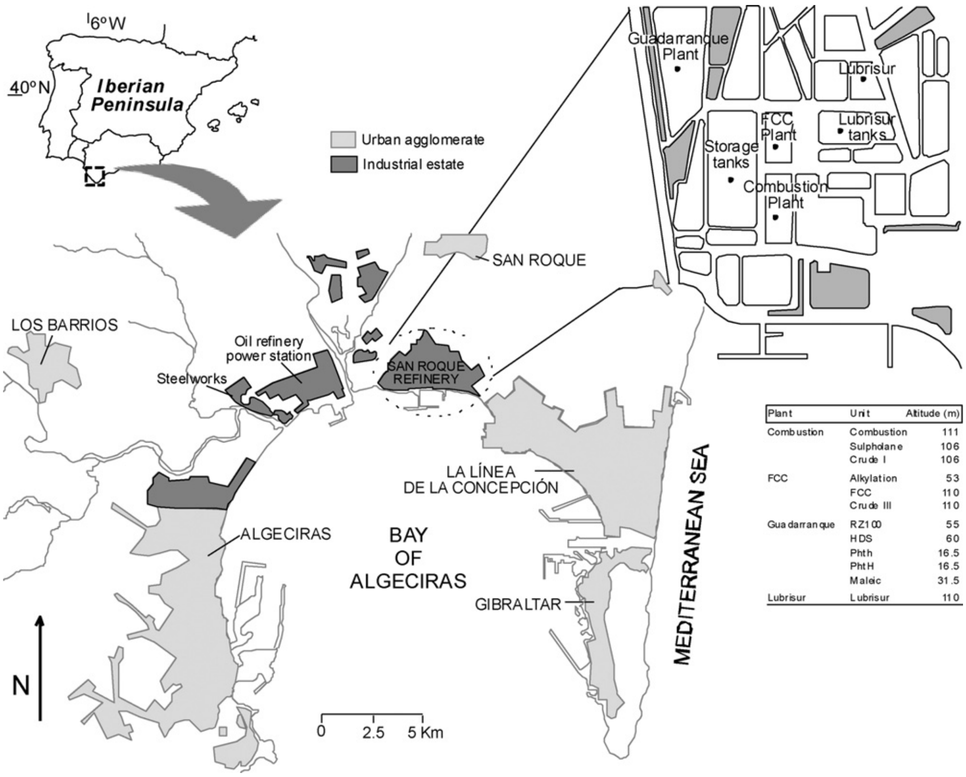

Plant	Unit	Altitude (m)
Combustion	Combustion	111
	Sulpholane	106
	Crude I	106
FCC	Alkylation	53
	FCC	110
	Crude III	110
Guadarranque	RZ100	55
	HDS	60
	Phth	16.5
	PhtH	16.5
	Maleic	31.5
Lubrisur	Lubrisur	110

Fig. 1. Generalised map of the Bay of Algeciras and location of Petrochemical plant and San Roque refinery. Details of the refinery and the altitude of stacks samples have been added.

is heated to the temperature of 379 °C in order to obtain a number of hydrocarbon compounds of different boiling-point ranges. These compounds are separated and treated further to produce gases (butane, propane), gasoline (from naphtha), aviation fuel, and fuel oils. The residual heavy hydrocarbons ("atmospheric bottoms") are further distilled under vacuum, mixed with heavy fuel oil and passed to the Fluid Catalytic Cracking (FCC) plant to be cracked into lighter compounds and processed to form gasoline. The cracking is achieved using a catalyst enriched in Rare Earth Elements (especially La), which can be emitted to the atmosphere during the process [15,16]. The lighter hydrocarbons produced in the FCC plant area are transformed into gasoline after treatment in an alkylation unit. In the Guadarranque plant, other refining processes produce basic materials for the petrochemicals industry, such as purified terephthalic acid (PTA) which is used in the manufacturing of polyester. Finally, the Lubrisur plant, which is located at the north end of the petrochemical complex, produces bases for lubricants and other blended products.

3. Methodology

Twelve chimmey stack's PM samples in the San Roque refinery complex were obtained using the University of Washington Mark III cascade impactor [17] for analysis of particulate size distribution. Of the 12 stacks sampled, 3 were in the area referred to as the combustion plant, 3 in the area of the FCC plant, 5 in the Guadarranque plant, and 1 in the Lubrisur plant (Fig. 1). Samplers in the combustion plant were positioned at over 100 m above ground level within chimneys venting emissions from fuel oil combustion (111 m), from the Crude I distillation unit (106 m), and from a sulpholane plant (106 m) where aromatic hydrocarbons are extracted from hydrocarbon mixtures. The sampled chimney stacks in the FCC plant area included the FCC unit itself (110 m), the Crude III distillation unit (110 m), and the alkylation unit (53 m). Within the Guadarranque plant samples were taken from emissions from the RZ-100 unit (55 m: catalytic reforming of light naphtha), the HDS unit (60 m: catalytic removal of S from naphtha), and emissions during the production of PTA (Phthalic 1 and 2: 16.5 m) and maleic anhydride (Maleic: 31.5 m). The single sample from the Lubrisur plant was taken from a height of 110 m in the chimney.

Seven effective cut off stage diameters (17, 14, 5, 2.5, 1.3, 0.67, and 0.33 μm) and back up (<0.33 μm) were used. Quartz microfiber filters (47 mm filter diameter and 0.45 μm pore diameter) were used for 10-min individual – isokinetic cascade impactor sampling.

Once the mass levels of PM_{10} were obtained by weighing the filters using standard procedures ($T = 20\,^{\circ}C$ and Relative Humidity = 50%), a half fraction of each of them was submitted to an acid digestion (0.25 ml HNO_3: 0.5 ml HF: 0.25 ml $HClO_4$) following the modified method proposed by Querol et al. [18], for the analysis of 43 trace elements (Li, Be, Sc, V, Cr, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Rb, Sr, Y, Zr, Nb, Mo, Cd, Cs, Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Ta, W, Tl, Pb, Bi, Th, y U) by means of ICP-MS (HP 4500®).

Three multi-elemental solutions Spec® 1 (rare earth elements, REE), Spec® 2 (alkalis, earth alkalis, and metals) and Spec® 4 (Nb) were used to construct an external calibration curve. The average precision and the accuracy fall for most of the elements under the normal analytical errors (in the range of 5–10%), and were controlled by repeated analysis of NBS-1633a (fly ash) reference material. The lower detection limit (LDL) for the most of elements in solution was 0.01 ppb.

The size, morphology and chemical composition of individual particles were evaluated by scanning electron microscopy with energy dispersive spectrometer (SEM-EDS). The particles were analysed individually using a LINK-ISIS energy-dispersive spectrometer mounted on a SEM (JEOL-JSM5410). Conditions were fit to 15 kV accelerating voltage and 100 s of effective counting time. Matrix corrections were made following the ZAF procedures, using a combination of silicate, oxides and pure metals as stan-

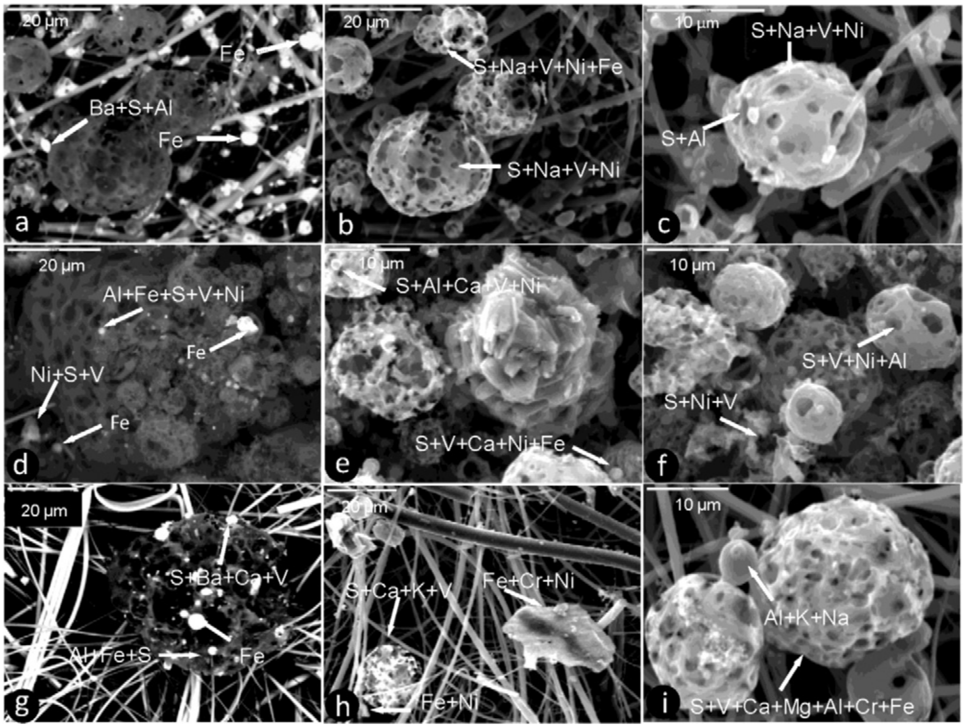

Fig. 2. Secondary and Back-scattered electron images (SEI and BSEI) of representative particles from emissions stacks showing coarse porous individual particles with spherical morphology and fine particles of variable composition.

dards (wollastonite for Ca and Si, jadeite for Na, orthoclase for K, corundum for Al, periclase for Mg, metallic Fe and Ti for Fe and Ti).

4. Results

The PM samples collected from the San Roque chimney stacks are predictably rich in fly ash particles, and representative stack samples examined and analysed by SEM-EDS are shown in Fig. 2. In the combustion sample, coarse (>20 μm) carbonaceous porous fly ash particles are dominant (around 75%). In addition there are aluminous fly ashes (>2 μm), typically rounded to subrounded hollow and composed of Al ± Fe ± Ni ± V. Fine (<2 μm) and ultrafine particles composed of S ± Al ± Fe ± Mg ± Ti ± Ni ± V ± Na ± Ba are seen adhered to the surface of the coarser PM. Similar fly ash particles dominate the PM samples from the FCC and Guadarranque plants, with the largest particles being observed in the HDS unit (>40 μm). Gypsum particles with rose habit are present at the alkylation unit particles (Fig. 2e). The FCC unit sample shows smaller carbonaceous particles (<2 μm) than those from the other units, whereas the particles from the RZ100 unit are irregular in shape, coarse grained (around <30 μm), and composed of Fe + Cr + Ni ± Al.

One of the most striking features of the SEM images is the very fine size of most metalliferous particles. The elemental mass concentrations of ten of the most common metals are provided in Table 1 and displayed graphically in Figs. 3–6. The three samples taken from units in the combustion plant (combustion, sulpholane and Crude I) show a unimodal pattern of ultrafine size PM in the combustion and sulpholane units, and of coarse size in the Crude I unit. Unimodal profiles in the finest (<0.33 μm) size fraction with maximum concentrations of V + Cr + Co + Ni + Cu + Zn + Mo + Pb were observed in the combustion and sulpholane samples, and Cu + Pb in the Crude I sample. Another unimodal size distribution is observed in the fine-coarse size range in La (combustion and Sulpholane) and Ce at the Combustion sample. Only Crude I sample exhibited a bimodal grain size profile (ultrafine and coarse modes), together with the maximum concentration of V + Co + Ni + Zn + La. Finally, a fine unimodal size distribution (0.33–0.67 μm, in all three samples) was observed. The highest metal concentrations in flue gas emissions of this plant were reached by Ni ($104\,\mu\mathrm{g\,m^{-3}}$), V

Table 1
Geochemical profile of main production plants and units in San Roque refinery.

Production plant	Unit	Very fine (<0.67 μm)	Fine to coarse (0.67–5 μm)	Very coarse (5–17 μm)
Combustion	Combustion	V + Cr + Co + Ni + Cu + Zn + Mo + Pb	La + Ce	–
	Sulpholane		La	–
	Crude I	Cu + Pb and V + Co + Ni + Zn		V + Co + Ni + Zn
FCC	Alkylation	V + Cr + Co + Ni + Cu + Mo		
	FCC	–	V + Cr + Co + Ni + Cu + Mo and La + Ce + Zn	La + Ce + Zn
	Crude III	–	V + Co	Cr + La + Ce
Guadarranque	RZ100	V + Cr + Co + Ni + Cu + Zn + Mo + La + Ce + Pb	–	–
	HDS	Cr + Co + Ni + Cu + Mo + Pb	–	La + Ce
	Phthalic h	Zn + Mo + Ce + Pb and Cr + Co + Ni + Cu	–	La and Cr + Co + Ni + Cu
	Phthalic H	Cu and Cr	–	Zn + La + Ce and Cr
	Maleic	V + Pb	–	Cr + Co + Ni + Cu + Zn + Mo + La + Ce and V + Pb
Lubrisur	Lubrisur	V + Co + Ni	–	V + Co + Ni and La + Ce

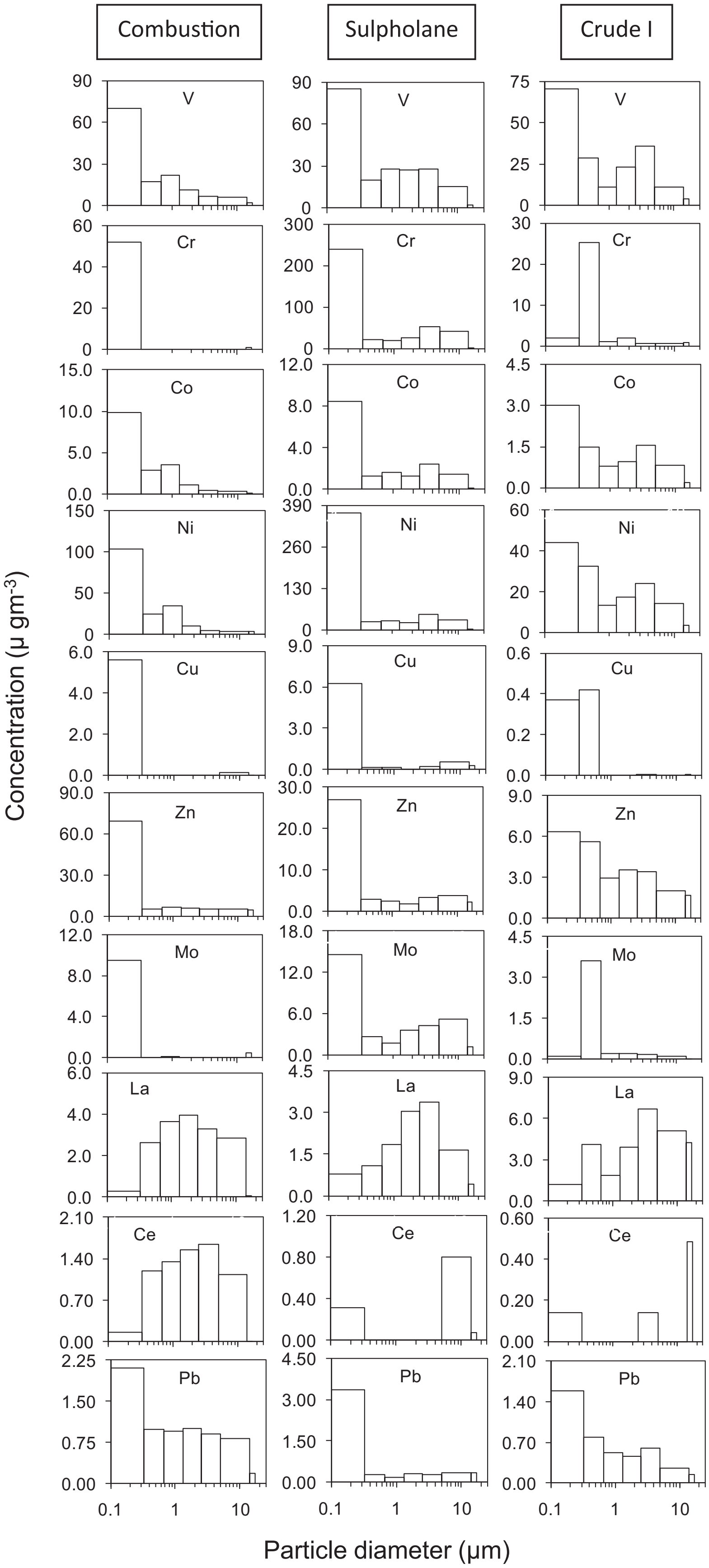

Fig. 3. Concentration and particle size distribution from combustion, sulpholane and Crude I process of combustion plant.

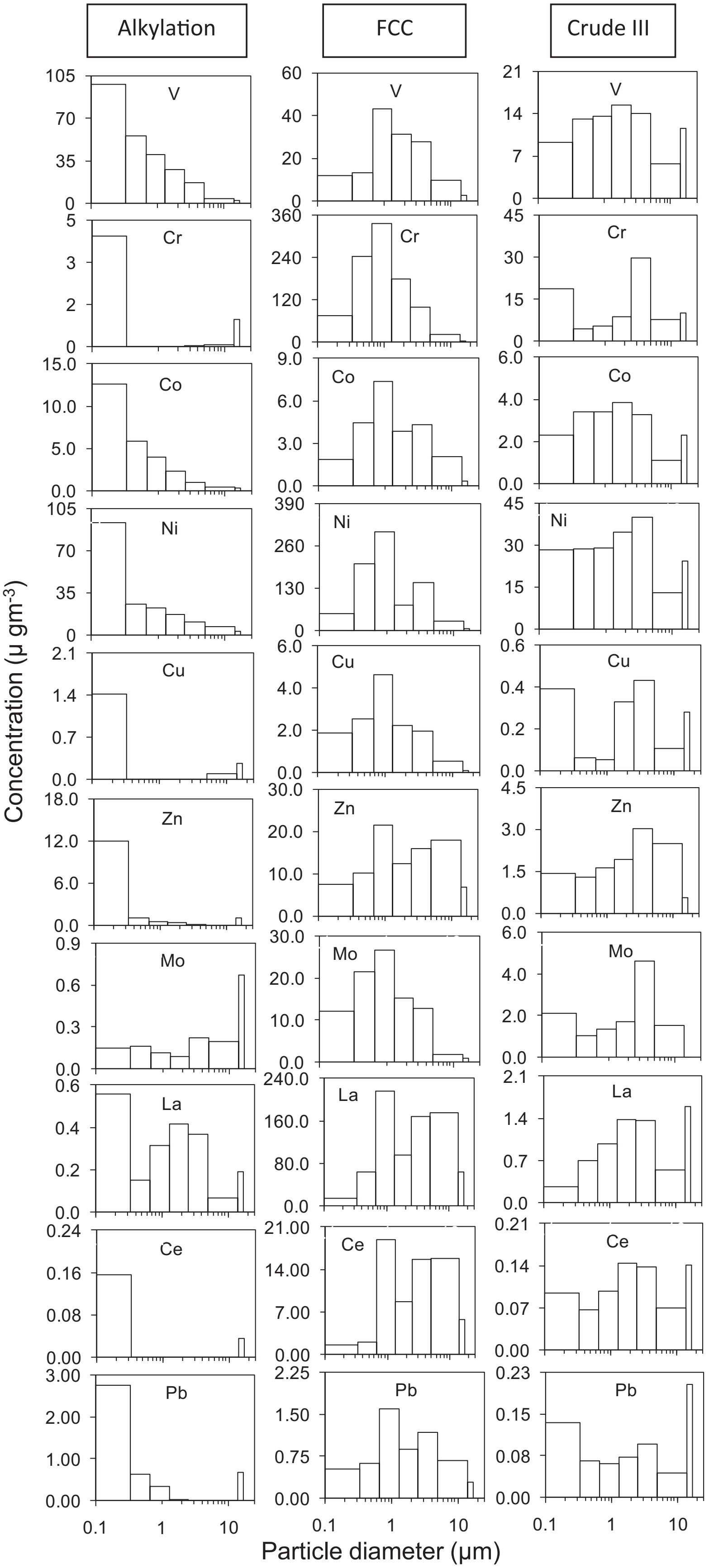

Fig. 4. Concentration and particle size distribution from alkylation, FCC and Crude IIII process of FCC and Crude III plant.

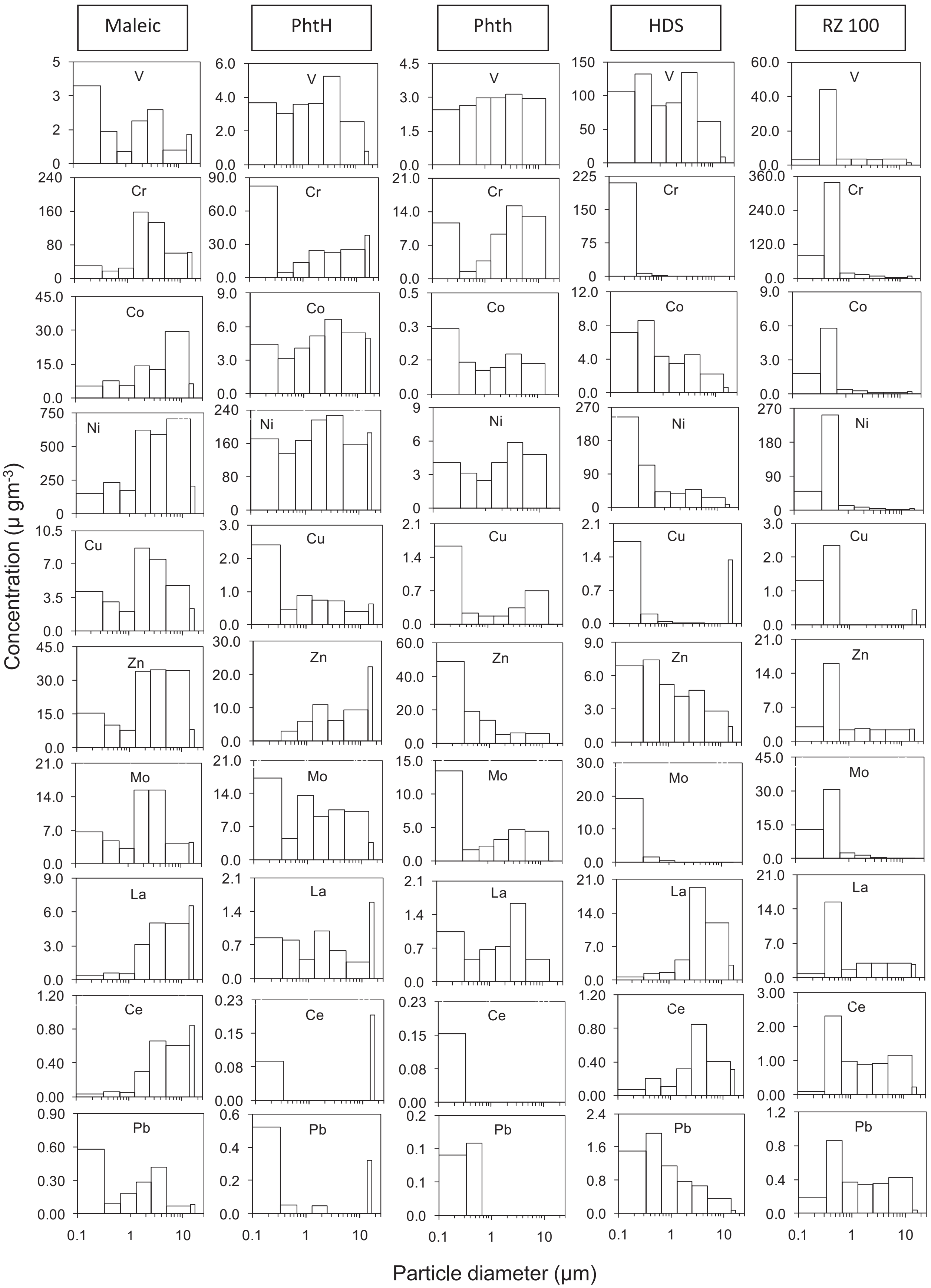

Fig. 5. Concentration and particle size distribution from Maleic, High combustion Phthalic (PhtH), Habitual combustion Phthalic (Phth) HDS and RZ 100 process of Guadarranque plant.

(70 $\mu g\,m^{-3}$) and Zn (70 $\mu g\,m^{-3}$) in the combustion sample; Ni (367 $\mu g\,m^{-3}$), Cr (240 $\mu g\,m^{-3}$), and V (85 $\mu g\,m^{-3}$) in the sulpholane sample; and V (70 $\mu g\,m^{-3}$), Ni (44 $\mu g\,m^{-3}$) and Cr (25 $\mu g\,m^{-3}$) in the Crude I sample.

Of the three samples from the *FCC plant* (alkylation, FCC and Crude III) the size distribution profiles of the alkylation sample are similar to those of the combustion and sulpholane samples of the combustion plant (Fig. 4). The sample obtained specifically from the FCC unit is characterised by an unimodal size distribution (maximum concentration reached intermediately between the fine to coarse modes) for V + Cr + Co + Ni + Cu + Mo, and a bimodal shape profile (fine and coarse size modes) in La, Ce, Zn and Pb. In contrast, in the Crude III sample size distribution profiles have no clear pattern, due to the low emission concentrations close to the analytical lower detection limit. The highest metal emission concentrations in these three chimneys were registered by Ni (302 $\mu g\,m^{-3}$), Cr (336 $\mu g\,m^{-3}$) and La (215 $\mu g\,m^{-3}$) in the FCC sample, V (98 $\mu g\,m^{-3}$) and Ni (93 $\mu g\,m^{-3}$) in the alkylation sample, and Ni (40 $\mu g\,m^{-3}$) Cr (30 $\mu g\,m^{-3}$) in the Crude III sample.

The size distributions of the elemental mass concentrations of *Guadarranque plant* samples (RZ 100, HDS, Phth and PhtH, Maleic) are summarized in Fig. 5. The major constituent elements are Ni + Cr + V + Zn + Co + Mo and La, and both unimodal and bimodal size distributions are evident. With regard to the unimodal patterns, three main size ranges have been distinguished: ultrafine, fine and coarse. Ultrafine (<0.33 μm) is characterised by a maximum concentration of Cr + Co + Ni + Cu + Mo + Pb in the HDS sample, Zn + Mo + Ce + Pb in the Phth sample, and Cu in the PhtH sample. Maximum concentrations of V + Cr + Co + Ni + Cu + Zn + Mo + La + Ce + Pb in the RZ100 sample are recorded in the fine size distribution (0.33–0.67 μm), whereas PM in the coarse size range (>2.5 μm) with a unimodal pattern show maximum concentrations of Cr + Co + Ni + Cu + Zn + Mo + La + Ce in the Maleic sample, Zn + La + Ce in the PhtH sample, La in the at Phth sample, and La + Ce in the HDS sample. Bimodal distribution exhibits both ultrafine (<0.33 μm) and coarse (>2.5 μm) modes, with enrichments in V + Pb (Maleic), Cr (PhtH), and Cr + Co + Ni + Cu (Phth). The highest metal emission concentrations in these five Guadarranque samples were registered by Cr (339 $\mu g\,m^{-3}$) and Ni (252 $\mu g\,m^{-3}$) in the RZ100 sample, Ni (244 $\mu g\,m^{-3}$) and Cr (211 $\mu g\,m^{-3}$) in the HDS sample, Zn (49 $\mu g\,m^{-3}$) and Cr (15 $\mu g\,m^{-3}$) in the Phth sample, Ni (227 $\mu g\,m^{-3}$) and Cr (83 $\mu g\,m^{-3}$) in the PhtH sample, and Ni (713 $\mu g\,m^{-3}$) and Cr (157 $\mu g\,m^{-3}$) in the Maleic sample.

In contrast to the previously described samples, those from the *Lubrisur plant* (Fig. 6) did not show a strongly distinctive size distribution pattern. However, there is a bimodal distribution (ultrafine and coarse sizes) with a maximum concentration of V + Cr + Co + Ni + Pb, and a unimodal distribution of La + Ce.

Table 1 summarizes relevant elements and size grain modes (ultrafine-fine, fine-coarse and coarse) of the stacks emission particles at San Roque Refinery, and clarifies the high diversity of size mode and chemistry between the samples. The unimodal and bimodal grain size distributions imply that elements present in emission particles are controlled by several processes. Most of the analysed units are enriched in metals (e.g. V + Cr + Co + Ni + Cu + Zn + Mo) in the ultrafine-fine size mode, this being attributed to the combustion of petroleum derived products, and proportionally higher from a contaminating effect of tubepipe line erosion [19]. However, FCC emissions are clearly more enriched in V + Cr + Co + Ni + Cu + Mo and La + Ce + Zn in fine to coarse particles, respectively, compared to other units. This bimodality is interpreted as resulting from a mixing of fine particles from petroleum combustion and coarser PM escaping from the FCC catalyst [14,15,20–22].

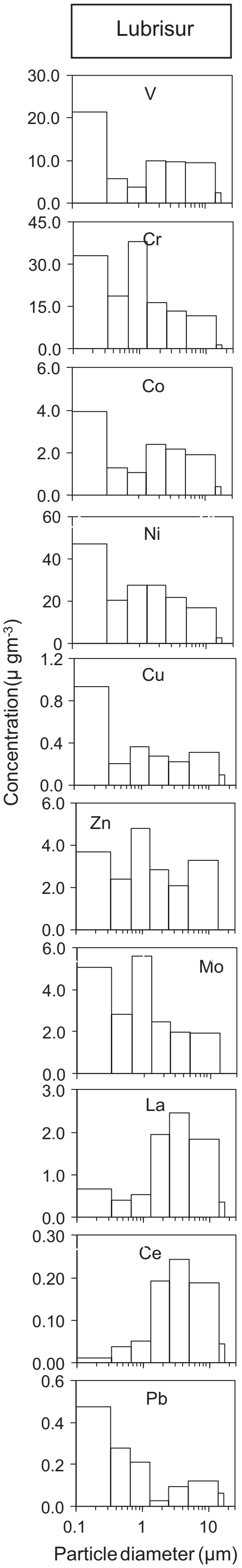

Fig. 6. Concentration and particle size distribution of Lubrisur plant.

Table 2
Chemical composition of sampled unit stacks.

Production area	CEA			FCC			Guadarranque					Lubrisur	Total
Unit	Combustion	Sulpholane	Crude I	Alkylation	FCC	Crude III	RZ-100	HDS	Phtalic h	Phtalic H	Maleic	Lubrisur	
Flow (Nm^3/h)	154,100	173,200	537,000	8000	154,400	87,400	71,700	39,900	3600	23,400	35,700	81,000	
Temperature (°C)	278	263	221	358	234	291	174	249	633	271	40	99	
Volume (Nm^3)	0.52	0.75	0.99	0.53	0.69	0.53	0.97	1.06	0.56	0.56	1.13	0.88	9.17
Concentration ($mg\,m^{-3}$)	18.0	37.6	47.0	33.6	261	3.6	27.4	37.0	59.3	33.0	129	7.50	694
$\mu g\,m^{-3}$													
Li	0.16	5.72	0.13	7.83	3.47	0.12	1.36	0.15	1.21	8.01	0.81	0.09	29.1
Be	0.06	0.10	0.04	0.11	0.15	0.02	0.02	0.05	0.01	0.05	0.01	0.01	0.64
Sc	0.05	0.62	0.33	<0.01	0.49	0.01	0.01	0.14	0.00	0.00	0.02	0.00	1.68
V	140	210	203	253	145	85.9	66.9	638	19.90	25.1	14.5	64.3	1866
Cr	53.2	406	34.9	5.04	962	94.6	476	225	57.7	270	568	144	3296
Co	18.3	16.7	9.53	27.0	24.8	20.3	9.23	31.7	1.10	40.8	93.9	13.6	307
Ni	187	526	160	182	819	208	344	526	26.1	1522	3295	173	7969
Cu	5.77	7.77	0.84	2.06	14.0	1.73	4.12	3.31	4.02	6.96	36.7	2.52	89.8
Zn	108	45.7	29.7	20.0	98.7	14.5	34.2	35.6	117	105	225	21.8	855
Ga	1.24	0.84	1.55	1.15	3.31	0.12	0.40	2.99	<0.01	<0.01	0.27	0.25	12.1
Ge	0.13	0.10	0.04	0.03	0.89	0.05	0.11	0.07	0.08	0.08	0.24	0.06	1.85
As	1.96	1.13	0.72	1.62	1.63	0.76	0.43	1.47	0.16	0.15	0.54	0.75	11.3
Se	0.35	0.79	0.28	0.64	0.58	0.63	0.24	0.53	0.11	0.13	1.00	0.64	5.92
Rb	0.22	1.20	0.19	0.03	0.68	0.10	0.04	0.11	0.01	0.06	6.56	0.05	9.25
Sr	1.81	7.36	0.95	1.71	1.74	1.06	0.93	1.87	0.23	0.20	10.8	1.22	29.9
Y	1.58	0.67	1.00	0.31	2.22	0.11	0.55	0.54	0.08	0.00	0.10	0.09	7.24
Zr	1.82	31.7	0.17	1.69	31.8	0.29	0.83	0.87	1.04	0.67	5.86	2.08	78.8
Nb	1.38	0.98	0.59	0.11	2.91	0.14	0.87	0.43	0.12	0.34	0.68	0.21	8.76
Mo	10.0	33.2	4.50	1.97	91.1	13.84	48.7	21.6	31.5	82.1	66.6	22.2	427
Cd	3.68	0.59	0.18	0.11	2.92	0.04	0.57	0.10	0.1	0.61	2.10	0.07	11.0
Sn	0.21	0.42	0.14	0.18	0.92	0.24	0.26	0.17	0.11	0.40	0.40	0.08	3.53
Sb	0.43	0.35	0.08	0.99	0.41	0.34	0.01	0.37	0.32	0.21	0.29	0.24	4.02
Cs	0.00	0.22	0.00	0.09	0.04	0.33	0.00	0.00	0.01	0.09	0.06	0.00	0.85
Ba	61.3	50.3	56.5	14.2	36.3	2.51	25.5	87.0	0.79	0.00	12.4	3.54	350
La	19.7	13.54	40.9	5.03	865	7.28	41.1	46.7	5.55	6.13	28.0	8.82	1088
Ce	8.43	1.18	1.57	1.94	76.3	0.80	8.63	2.50	0.15	0.28	3.46	0.84	106
$\sum$ REE	36.5	21.4	51.8	8.59	1059	9.59	60.0	58.3	6.14	7.75	38.0	11.3	1368
Hf	0.45	0.57	0.24	0.07	0.63	0.03	0.06	0.16	0.11	0.08	0.23	0.06	2.70
Ta	0.00	0.32	0.03	0.26	0.22	0.02	0.00	0.02	0.11	0.19	0.01	0.01	1.20
W	0.21	1.14	0.09	0.08	1.50	0.19	1.10	0.31	0.23	0.91	1.83	0.31	7.89
Tl	0.00	0.39	0.00	0.00	0.10	0.04	0.00	0.00	0.00	0.00	0.07	0.03	0.63
Pb	7.89	5.58	4.99	5.25	6.41	0.73	3.00	6.84	0.24	1.31	2.13	1.34	45.7
Bi	0.00	0.01	0.00	0.00	0.00	0.04	0.00	0.00	0.00	0.00	0.01	0.02	0.08
Th	5.70	1.05	1.68	0.97	2.14	0.03	3.28	1.86	0.04	0.00	0.02	0.03	16.8
U	0.14	0.36	0.14	0.05	0.55	0.04	0.02	0.07	0.06	0.08	0.03	0.02	1.55

5. Discussion and conclusions

The evaluation of the experimental chemical data on metal emissions at the San Roque refinery (Table 2) suggests that the highest concentrations averaged over all size ranges were registered in the following decreasing order: Ni (up to 3295 $\mu g\, m^{-3}$) > Cr (962 $\mu g\, m^{-3}$) > La (865 $\mu g\, m^{-3}$) > V (638 $\mu g\, m^{-3}$) > Zn (225 $\mu g\, m^{-3}$) > Co (94 $\mu g\, m^{-3}$) > Mo (91 $\mu g\, m^{-3}$). The combustion unit emitted with the highest concentrations of Zn, As and Cd, the sulpholane unit with the highest Se, Sr and Pb, the FCC unit produced the highest concentrations in Cr, Ga, Zr, Mo and REE, whereas Sb and Li emission levels were highest in the alkylation unit. Finally the different stacks in the Guadarraque plants (Fig. 5) emitted especially high concentrations of V (HDS unit), Cr and Mo (RZ-100 unit), and Co, Ni, Cu and Rb (Maleic unit).

The high concentrations of ambient metals recorded in PM_{10} and $PM_{2.5}$ from representative monitoring stations of the Bay of Algeciras have been attributed in part to emissions from the San Roque refinery [14]. In particular, according to Moreno et al. [14], high concentrations of V (28 $ng\, m^{-3}$), Cr (25 $ng\, m^{-3}$), Ni (20 $ng\, m^{-3}$) and La (0.58 $ng\, m^{-3}$) are present within ambient atmospheric PM_{10} in the Algeciras area. Although the exact size ranges of these ambient particles remains unstudied, these authors further note that the metalliferous aerosols typically are extremely fine in size and therefore potentially bioavailable, making a clear case for basing urban background PM characterisation not only on physical parameters such as mass but also on sample chemistry and with special emphasis on trace metal content. The data presented here confirms that many metals are indeed present in high concentrations in the refinery complex chimney stacks, with exceptional concentrations of >100 $\mu g\, m^{-3}$ being reached by V, Cr, Ni, and La. These extreme levels are not uniformly emitted but focused on specific stacks within the petrochemical complex. Taking each of these four metals in turn, in the case of *Vanadium* the highest concentrations are found within emissions from the HDS unit in the Guadarranque plant and from the alkylation, sulpholane, combustion, and Crude I units (Figs. 3–5). A clear difference between these diverse sources is that whereas the HDS unit emits abundant V-rich PM in all sizes ranging up to PM_{10} (Fig. 5), all the other four sources emit mostly very fine PM (<0.33 μm). *Chromium*-rich PM measured in the stack samples is most abundant in the RZ-100 and HDS units of the Guadarranque plant and in the FCC and sulpholane units (Figs. 3–5). Most of these Cr-rich PM are once again fine in size (<0.5 μm), although the FCC unit shows a broader size range peaking around 1 μm. With respect to *Nickel*-rich PM, these are highly abundant in six of the stack samples, especially in the Maleic and sulpholane units, but also in the FCC, PhtH, HDS and RZ-100 units. Once again, although many of the NiPM are very fine in size, some units show a very broad size range (FCC, PhtH: Figs. 4 and 5). Finally, the most spectacular point source contamination of all the metals analysed is that displayed by *Lanthanum* in the FCC unit, with concentrations exceeding 200 $\mu g\, m^{-3}$ and a relatively coarse size distribution of (1–10 μm) confirming derivation from the La-rich catalyst.

Of the 12 samples analysed it is clear the stacks producing the highest concentrations of metalliferous particles are those associated with the following 8 units: combustion (high V, Ni, Zn < 0.33 μm), sulpholane (high V, Cr, Ni < 0.33 μm), Crude I and alkylation (high V < 0.33 μm), FCC (high Cr, Ni, La (0.3–10 μm), Maleic (high Ni, Co 1–10 μm), HDS (V < 10 μm, Cr and Ni < 0.33 μm, La 2–10 μm), and RZ-100 (Cr, Ni, Mo 0.3–0.7 μm. In contrast, relatively low metalliferous concentrations are found in the Crude III and Lubrisur stacks (Figs. 4 and 6).

Most of the more toxic metals concentrate in the finer PM sizes in most samples, reinforcing the visual observations provided by the SEM data (Fig. 2). Thus PM escaping into the atmosphere from these refinery complex stacks will be capable of both proximal and distal contamination, and individual particles will be small enough to be easily inhaled into the deep lung environment [23]. The Bay of Algeciras has consistently presented higher than average rates of cancer incidence and mortality, comprising one of the vertices of a triangle centred between the provinces of Huelva, Cádiz and Seville and contaminated by a range of industrial emissions [10,24–27]. If this increased health risk is linked to air pollution from the refinery complex, then it is the metalliferous component of the emissions which is likely to be at least part of the problem. It clear from our unique database that there is great chemical and size variation in the cocktail of metalliferous pollutants emanating from different emission stacks in refinery complexes, and that some point sources are considerably more contaminating and potentially toxic than others.

Acknowledgments

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Attachment 11

Mercury in Crude Oil Processed in the United States (2004)

S. MARK WILHELM,* LIAN LIANG,† DEBORAH CUSSEN,‡ AND DAVID A. KIRCHGESSNER§

Mercury Technology Services, 23014 Lutheran Church Rd., Tomball, Texas, Cebam Analytical, Seattle, Washington, Frontier Geosciences, Seattle, Washington, and U.S. Environmental Protection Agency, Research Triangle Park, North Carolina

The mean and range of concentrations of mercury in crude oil processed by U.S. refineries in 2004 were determined using two analytical methods. One hundred seventy separate crude oil streams were sampled repetitively to obtain 328 individual samples. Samples were retrieved immediately upstream of refinery tank farms. Losses of mercury during production, separation, and transportation were not examined. The arithmetic mean and median of 170 oil streams were 7.3 and 1.5 μg/kg in total mercury, respectively. The total mercury concentration of oil processed in the United States in year 2004, including all species and both dissolved and suspended forms, expressed as a volume-weighted mean was calculated to be 3.5 ± 0.6 μg/kg. The range of measured concentrations extended from below the analytical detection limit (0.5 μg/kg) to approximately 600 μg/kg. Good agreement was found with other recent and independent studies of mercury in crude oil refined in North America. The total amount of mercury in crude oil processed in the U.S annually is less than five percent of the amount contained in U.S. coal produced annually.

Introduction

The major sources of anthropogenic mercury emissions in the United States are fossil fuel and waste combustion. Mercury in coal has been studied extensively and it contributes the largest amount of atmospheric emissions of any fuel source (*1*, *2*). An obvious analogy exists between mercury in coal and mercury in petroleum; however, the amount of mercury in crude oil, and thus potentially liberated by liquid fuel combustion or other pathways, has remained uncertain because of a paucity of accurate mercury concentration data across the huge range of crude oils entering U.S. refineries.

Studies of mercury in crude oil published before the year 2000 reveal a range of concentrations of total mercury (THg) in crude oil between approximately 0.1 and 20 000 μg/kg (*3*). The breadth of the range reflects the wide variability of natural geological conditions. Calculations of a mean concentration from measured concentration data published before year 2000 were biased high because of poor analytical detection limits and because analysts tended to select oils with higher concentrations for investigations (*3*). A consistent and broadly focused study of mercury in crude oil has not been available to allow comparison to coal and other fuels.

In 2003, the United States Environmental Protection Agency's Office of Research and Development initiated a study directed to determining the mean and range of concentrations of mercury in crude oil processed in the U.S. for the point in time accessible. The research program was organized into three parts, all now complete. Part 1 examined analytical procedures (*4*). Part 2 examined sampling and species effects (*5*, *6*). In Part 3 and reported here, total mercury concentrations were measured for a wide variety of crudes made available to the study by the U.S. oil refining industry.

Mercury in Crude Oil. A number of mercury species have been identified in crude oil (*7*−*9*). The list includes elemental mercury (typically the major component) and dialkylmercury (typically a trace component), both volatile and reactive and mercuric sulfide and asphaltene mercury (major component in some oils), both nonvolatile and relatively nonreactive. Complexed ionic mercury species are also present in some oils as a minor component.

In measuring total mercury concentration (THg) in crude oil matrix, samples must reflect the homogeneous amounts of all mercury species that are present in the stream being sampled and analytical methods must avoid losses of volatile species in aliquot removal procedures. Because some mercury compounds are associated with oil sediment, some loss of mercury is expected in stagnant tanks that allow settling or stratification of suspended material. Elemental and organic mercury are volatile and losses are also expected when volatile components escape heated tanks or in transportation. Elemental mercury also reacts with steel surfaces irreversibly and some losses likely occur in pipelines. In the present study, such possible losses were not investigated. The focus of the study was not the wellhead concentration of mercury but rather the "as delivered" concentration in crude entering the refining process.

Crude Oil Processed in the United States. A wide variety of materials are processed by U.S. refineries to make liquid fuels and other products derived from crude oil (asphalt, coke, wax, lubricants). The raw material categories include condensates, crude oil, heavy crude oil, and synthetic crudes. Samples in all crude oil categories were donated and analyzed in the project. Oils having trade names are blends of oils that come from several or many individual wells and are typically consistent in composition and characteristics. The conscious desire to maintain consistency of oil properties likely constrains the range of mercury concentration in trade-named oils as well. The variation of measured mercury concentrations of trade-named crude oil streams over time is not well studied, but it was postulated that the range of concentrations exhibited by a single trade-named oil was much less than the range of mercury concentrations found in crude oils generally.

This study relied on industry volunteers to donate samples of crude to the project for analysis. Considerable efforts were made to ensure that the sample ensemble reflected both domestic production and imports in proportion to what is actually processed. No major crude sources are known that were left out of the group of oils analyzed, but certainly not all of the oil streams processed in U.S. refineries were included.

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* Corresponding author phone: 281-255-3775; e-mail: smw@hgtech.com.

† Cebam Analytical.

‡ Frontier Geosciences.

§ U.S. Environmental Protection Agency.

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TABLE 1. Laboratory Performance Parameters

	reagent blank	control CRM recovery	average RPD (THg>0.5)	averageRPD (all)	RPD > 20% (THg>0.5)	average spike recovery
CAF	88.5% <0.1 μg/kg 6.2% 0.1 μg/kg 5.3% 0.2 μg/kg	mean = 98.1% σ = 3.9% range: 87.4–107.7%	7.8%	8.4%	2.1%	mean: 96.0% σ = 8.5% range: 75.1–117%
number of measurements	113	56	289	335	289	136
DAF	mean = 0.07 μg/kg σ = 0.181 μg/kg	mean = 85.0% σ = 6.8 range 78.9–102.3%	6.8%	41.4%	0	mean = 98.8 σ = 7.6 range 85.2–116.2%
number of measurements	24	10	26	43	26	24

Experimental Section

Analytical Methods. Analytical methods targeted the total concentration of mercury (THg) in crude oil. Because several mercury species exist in oil, the analytical methods needed to be robust so that the chemically inert species (HgS and asphaltene–Hg) can be retrieved quantitatively while at the same time avoiding losses of the volatile mercury species. Techniques to retrieve and retain volatile species were investigated early on in the project and incorporated into the analytical methods and procedures (*5*). Because mercury is associated with material suspended in oil, sampling procedures and homogenization of samples were deemed important and were optimized to retrieve suspended mercury components.

Two independent laboratories, each using a different method, were selected to analyze crude oil samples. Methods were selected based on literature pedigree, performance comparison, and measured method detection limit (MDL). MDLs for the two methods that were selected were less than 0.5 μg/kg. The methods and procedures employed to analyze mercury in oil have been described previously, but a brief summary is offered here. Table 1 summarizes each laboratory's performance relative to data quality objectives.

Laboratory A (Cebam Analytical, Seattle, Washington) used a combustion method developed and constructed in-house (*10, 11*). The combustion system consisted of a quartz temperature-controlled combustion column, a gas filter, and gas flow apparatus. The combustion column was divided into two independently controlled heating zones. The initial segment vaporized the hydrocarbon and mercury compounds and the second segment combusted the hydrocarbon. Liquid aliquots were drawn from the sample vial by penetrating the septum using a needle preconditioned microsyringe without opening the vial. Aliquots were then injected directly into the vaporization section of the instrument. Purified air, from which mercury was removed by passing through a gold-coated sand trap, was used as the carrier and combustion gas. Mercury vapor in combustion product gas exiting the combustion chamber was filtered to remove partially combusted hydrocarbons and then collected by amalgamation on a gold trap (single amalgamation). The mercury on the gold trap was released by heating in flowing argon that passed to the atomic fluorescence (AF) detector (Brooks Rand). This method is referred to as "Combustion–AF" or "CAF".

Laboratory B (Frontier Geosciences, Seattle Washington) used a digestion – atomic fluorescence method in which homogenized oil aliquots were placed in quartz vials containing concentrated nitric acid. The vials were capped with quartz lids and heated to 300 °C under approximately 130 bar pressure in an autoclave (Anton-Paar high-pressure asher) until the sample dissolved. After cooling and removal from the autoclave, the samples were diluted with reagent water, and aliquots of sample digests were transferred by pipet into a bubbler to which a stannous chloride ($SnCl_2$) solution was then added. Argon was passed sequentially through the bubbler, through soda-lime traps, and finally through a gold sand amalgam trap. The first gold trap containing the collected mercury was heated in flowing argon to transfer the collected mercury to a second gold amalgam trap. Mercury collected on the second trap was likewise desorbed by heating in flowing argon and passed into an atomic fluorescence (AF) detector (Tekran 2500). The detector response was recorded and integrated. This method is referred to as "Digestion–AF or DAF".

For the CAF method, detector calibrations were accomplished using dilutions of constructed standards. A 1000 mg/L stock solution was prepared by dissolving CH_3HgCl (99%, Johnson Mattey, U.S.) into isopropanol. Working standard solutions (10, 20, and 50 ng/mL) were prepared by serial dilution of the stock solution with toluene. The stock standard was traced to NIST standards using the EPA1631 method (*12*). The working standards in glass vials were found to be stable at 20 °C for at least a month, but were prepared twice monthly. The CAF method used calibration factors (CFs) for calculation of concentrations. A CF was calculated as the standard value in pg divided by the difference of the standard peak area (PA) and the mean PA of method blanks (unit pg/PA). At least four nonzero CF points were analyzed daily. Samples were processed only when RSD of CFs was less than 15%. 80% of RSDs were within 5–10% during the project. The verification of a daily calibration was performed by analyzing a lab control sample (LCS) in duplicate following the standard curve. Two LCSs, no. 5-01 heavy oil and a Conostan (Ponca City, Oklahoma) oil standard (100 ppm as Hg), were used. The no. 5-01 heavy oil was certified as 288.0 ng/g for THg as Hg by three independent labs using three independent techniques. The sample was found to be stable for past years since it was certified in 2001. The two LCSs were serially diluted to 10.0 ng/mL with toluene prior to use. Diluted standards were prepared monthly. Analyses of matrix spikes and spike duplicates (MS/MSD) for the CAF method were performed by injecting a spiked oil sample aliquot using the sample syringe. Spikes were methylmercury in toluene working standards. The criteria for MS/MSD recovery was 75–125%.

For the DAF method, certified reference materials (CRMs) were purchased (high purity standards, 1000 ± 3 μg/mL and absolute standards, 100.4 ± 0.378 μg/mL). A working calibration standard (10 ng/mL) was constructed by dilution using reagent water containing 2% BrCl and used for 3 months. Daily calibrations of the Hg analyzer were performed using a 5-point (0.05, 0.10, 0.50, 2.00, 4.00 ng) standard calibration curve. Linearity was better than 0.999 for all calibrations. Calibration was verified using BrCl water blanks (mean = 0.06 μg/kg, $n = 38$, $\sigma = 0.055$ μg/kg) and separately constructed aqueous verification standards (mean > 99.1% recovery, $n = 38$, $\sigma = 6.3\%$). Mercury in oil certified reference

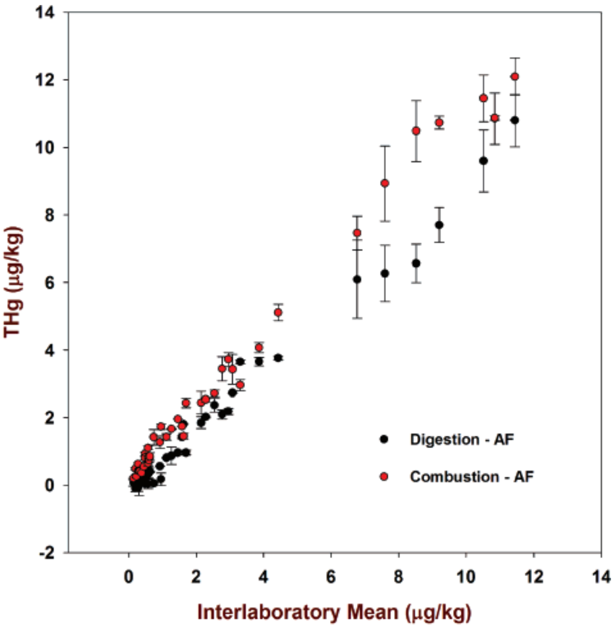

FIGURE 1. Laboratory and method comparison (CAF and DAF).

material (5000 µg/kg THg; Spex Certiprep Inc.) was used as a control (each sample batch) to verify method performance. A 1000× dilution of the 100 µg/mL CRM (100 ng/mL Hg) was used for matrix spikes.

Method detection limits were calculated according to procedures set forth in EPA Method 40 CFR 136 (Appendix B) and found to be 0.11 and 0.47 µg/kg for CAF and DAF, respectively.

Crude Oil Sampling. Crude oil samples were retrieved at the entry point to refineries prior to crude oil tanks. For oils that arrived by pipeline, the sample point typically was that used for refinery quality assurance sampling. For oil tankers, the sampling point was typically the discharge and metering point. Refineries used their normal sampling procedure, modified as needed to accommodate sample containers for mercury analysis. Techniques for sampling crude oils were investigated early in the project and details have been discussed previously (*5*).

Sample Reproducibility. An oil "sample" consisted of four vials filled simultaneously with vials labeled "A, B, C, D". Sample vials were homogenized prior to removal of aliquots for analysis by ultrasonication (CAF) or manual shaking (DAF). In the normal procedure, one vial (usually B) was analyzed in duplicate (two aliquots from one vial) by the CAF laboratory. Fifteen percent of samples were analyzed more than twice to check vial-to-vial homogeneity and sample age variation. Reproducibility of measurements was judged by calculation of relative percent difference (RPD = average of the sum of differences expressed as a percentage of the mean). Reproducibility of aliquots of a single sample is a measure of laboratory and method performance, as opposed to oil mercury concentration variability, assuming homogeneous samples (all vials the same) and uniform aliquots were obtained from vials. As shown in Table 1, RPD for the CAF and DAF laboratories averaged 8 and 7%, respectively.

Interlaboratory Comparison. Laboratory/method bias was assessed by examination of the differences of duplicate means as a percentage of the interlaboratory mean. Laboratories analyzed the same sample but different vials. Forty-three samples, selected at random, were analyzed in parallel using CAF and DAF methods. The data are shown graphically in Figure 1. The error bars shown are simply the range of duplicate measurements. In the graph, two points at interlaboratory means of 77 (RPD = 18.8) and 80 µg/kg (RPD = 10.4) are not shown so as to expand the scale of the plot. Interlaboratory reproducibility above MDL met data quality objectives for the project. Systematic bias averaged 24% (range = 0.1−94%) for samples where the interlaboratory mean was greater than 0.5 µg/kg. The bias was not investigated but is thought to derive from minor losses of volatile mercury in the DAF procedure. Oil aliquots are exposed to the atmosphere and have opportunity for slight evaporation prior to digestion in the DAF procedure. As shown in Table 1, the DAF method also demonstrated slightly poorer CRM recovery.

Results and Discussion

An "oil stream" was defined as a trade-named oil received by an individual refinery. For each oil stream, THg was measured in one or more discreet sampling events. An independent sampling event retrieved four 40 mL vials of oil, either from a tanker discharge or from a pipeline. Samples were independent if taken days or weeks apart from pipelines or from different tankers. Obtaining three independent samples was the goal but numerous exceptions occurred because of supply dynamics. The mean concentration of an oil stream ($\bar{C}^{\mathrm{OS}}$) was calculated as the average of n individual sample measurements with each sample result (c_i) the mean of two or more replicates.

$$\bar{C}^{\mathrm{OS}} = \frac{1}{n}\sum_{i=1}^{n} c_i \qquad (1)$$

The number of samples (n) averaged to obtain stream concentrations was between 1 and 9. The standard deviation (s^{os}) of oil stream sample concentrations was calculated according to eq 2.

$$s^{\mathrm{os}}_{n>2} = \sqrt{\sum_{i=1}^{n}\frac{(c_i - \bar{C}^{\mathrm{OS}})^2}{(n-1)}} \qquad (2)$$

The mean mercury concentration of 170 oil streams treated independently was calculated to be 7.3 µg/kg (median = 1.5 µg/kg, SD = 46.1 µg/kg, range = 593 µg/kg). Fifty-seven percent of streams in the range of 1−10 µg/kg and 93% were less than 10 µg/kg. A very few oils were measured with concentrations over 50 µg/kg. All of these oils (THg > 50 µg/kg) came from Asia, however, which is known in the industry to be a "mercury prone" region. Few Asian oils reach the U.S. refineries because of transportation costs and Asian market demands. Asian imports represented less than 1% of oil processed in the U.S. in 2004, but this percentage may increase in the future.

Inadvertent volume weighting affects the calculation of the simple mean. Because several participating refineries may have received the same trade-named oil, some trade-named oils are represented in the "oil stream" ensemble (170 oil streams) more than once. Popular oils that are desired due to characteristics and high volume oils are represented by several donated oil streams and thus contribute to the calculated mean in proportion to the replication of their donation.

Volume Weighting. To obtain a mean concentration of mercury in oil processed in the 50 United States, concentrations of market-named oils were weighted by the volume of U.S. refined oil they represented in 2004. This was accomplished in the following manner. Statistics are not kept, nor therefore available, of the amount of oil refined according to individual trade names. Statistics on production volume

TABLE 2. Mercury in Oil by Country (Imports) and by State (Production)

country	country volume (1000 barrels)	% U.S. supply	country average (μg/kg)	$\sum n$	N	s^{CS} (μg/kg)
Algeria	78 719	1.31	13.3	2	2	4.4
Angola	112 018	1.86	1.6	2	2	0.6
Cameroon	6 756	0.11				
Chad	20 805	0.35	1.2	3	2	0.5
Congo	8 019	0.13				
Gabon	52 061	0.87	0.5	2	1	0.2
Guinea	24 212	0.40	0.3	1	1	0.1
Ivory Coast	1 840	0.03	0.3	1	1	0.2
Libya	6 724	0.11				
Nigeria	394 560	6.57	1.8	20	12	0.1
Africa	**705 714**	**11.75**	**2.7**	**31**	**21**	
China	5 273	0.09				
Indonesia	12 360	0.21				
Kazakhstan	3 228	0.05				
Australia	7 855	0.13	0.8	1	1	0.3
Brunei	5 616	0.09				
Malaysia	6 551	0.11				
Thailand	194	0.003	593.1	2	1	184
Viet Nam	9 256	0.15	66.5	5	2	4.9
Asia	**50 333**	**0.84**	**220.1**	**7**	**4**	
Canada	**591 489**	**9.845**	**2.1**	**72**	**32**	0.2
Denmark	821	0.01				
Norway	52 365	0.87	19.5	3	2	8.2
Russia	58 010	0.97	3.1	5	4	0.2
UK	87 193	1.45	3.6	10	3	1.0
Europe	**198 389**	**3.30**	**8.7**	**18**	**9**	
Mexico	**585 023**	**9.74**	**1.3**	**25**	**9**	0.1
Iraq	239 758	3.99	0.7	10	6	0.1
Kuwait	88 359	1.47	0.8	7	4	0.2
Qatar	1 383	0.02				
UAE	1 885	0.03				
Oman	3 570	0.06				
Syria	501	0.01				
Yemen	1 365	0.02				
Saudi Arabia	547 125	9.11	0.9	28	14	0.1
Middle East	**883 946**	**14.71**	**0.8**	**45**	**24**	
Argentina	21 499	0.36	16.1	1	1	7.1
Brazil	18 733	0.31	1.1	2	2	0.2
Columbia	52 049	0.87	3.4	10	7	0.6
Ecuador	84 937	1.41	1.8	11	6	0.3
Bolivia	311	0.01				
Guatemala	6 699	0.11				
Peru	383	0.01				
Trinidad	18 027	0.30				
Venezuela	474 531	7.90	4.2	18	12	13
S. America	**677 169**	**11.27**	**5.3**	**42**	**28**	0.0
AK	332 464	5.53	3.7	16	6	0.5
CA	240 206	4.00	11.3	6	3	3.6
GOM	531 900	8.85	2.1	19	7	0.2
LA	72 824	1.21	9.9	7	7	2.6
MT	24 724	0.41	3.1	2	2	1.0
OK	62 502	1.04	1.4	4	2	0.4
TX	392 865	6.54	3.4	6	3	22
UT	14 628	0.24	2.2	1	1	0.9
WY	51 621	0.86	2.7	15	8	0.4
X	592 026	9.85	3.1	12	3	0.6
U.S.	**2 315 760**	**38.55**	**4.3**	**88**	**42**	
total	**6 007 823**			**328**	**170**	

are available but typically only for some major oil streams. Records are compiled of oil produced in each U.S. state and, for imports, by country of origin. Imported oil cargoes have trade names that identify the country of origin, hence attaching country import volumes to oil concentrations is straightforward. Likewise oil produced and traded in the U.S. is identified by origin such that state volumes can be associated with most streams. Many of the smaller U.S. refineries tap local production and, in some cases, the identity and volume of local domestic sources are elusive to any association. Import, production and processing statistics were obtained from the Energy Information Agency (U.S. Department of Energy) website (*13*). Year 2004 oil volumes were chosen for the statistical base. Samples were acquired in years 2004, 2005, and 2006, but the majority of samples were gathered in 2004.

In Table 2, country and state THg averages are compiled along and attached to volumes of oil processed in U.S refineries from countries and volumes of oil produced within states. The country or state average was calculated as the arithmetic mean of *N* oil stream concentrations ($\bar{C}^{OS}$, eq 1) obtained from that country or state.

$$\bar{C}^{CS} = \frac{1}{N}\sum_{j=1}^{N} \bar{C}_j^{OS} \tag{3}$$

As with the calculation of the simple mean, inadvertent weighting occurred in this procedure also. Oil streams (eq 3) were treated independently within countries and states. More than one oil stream may have derived from a single trade-named oil. Larger streams contributed a stream average from each refinery that received the oil and donated samples. Larger volume streams were, therefore, more likely to contribute more streams in calculating country/state averages because they were processed by more refineries during the time samples were collected. In Table 2, "∑n" is the total number of samples from a state or country and "*N*" is the number of oil streams from that country or state.

The weighted mean was calculated from country and state averages weighted by their associated fraction of oil contributed using the following formula:

$$\bar{C}^{US} = \sum_{CS}\left[\bar{C}^{CS}\frac{V^{CS}}{0.9V^{US}}\right] \tag{4}$$

where $\bar{C}^{US}$ is the mean total mercury concentration of U.S. processed oil; $\bar{C}_{CS}$ is the average concentration in a country or state obtained from oil streams and $V^{CS}/0.9V^{US}$ is the ratio of country/state volume to total U.S. oil volume. The sum was taken over all countries and states that contribute to the U.S. total processed oil volume and for which concentrations of THg were measured. The factor of 0.9 comes from the fact that only 90% of U.S. oil volume was accessed in the set of samples obtained in the program. In weighting, state and country average oil density was assumed to be approximately equal to the U.S. average oil density. The calculation yields a value of $\bar{C}^{US} = 3.5$ μg/kg for the mean concentration of mercury in oil refined in the U.S.

Uncertainty. Ten percent of oil processed in the U.S. was not accessed in the project. The 10% number derives from states and countries, known to contribute to U.S. totals, but for which no oil mercury concentrations were measured, and thus no concentrations were available to be associated to the volumes known to be imported (countries) or produced (states). Within individual countries and states, some oil streams likely to contribute to U.S. totals also were not accessed, but it was not possible to know exactly how many, or the corresponding volume excluded, because import and production volumes are not compiled for all individual trade-named oils. It is thought that the greatest uncertainty in the calculated mean concentration of mercury refined in the U.S. lies with calculated country mean concentrations due to inaccessibility, not with analytical uncertainty. Analytical uncertainty, estimated as the average difference from the mean of duplicates, was about 8% overall for measured values above 0.5 μg/kg. It was hypothesized that individual oil

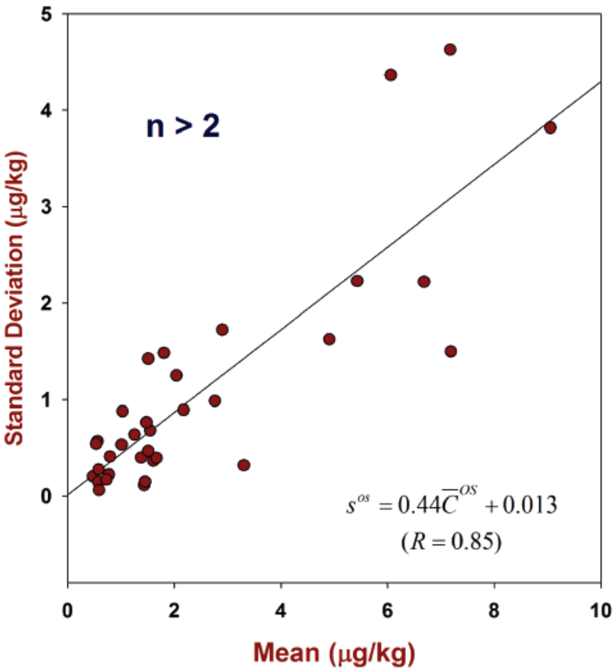

FIGURE 2. Oil stream standard deviation versus mean mercury concentration.

streams exhibited normal distributions of mercury content but the number of samples obtained from many streams was not sufficient to judge.

Oil stream estimated standard deviation (s^{os}) was found to depend on the mean (eqs 1 and 2) in the manner shown in Figure 2 for oil streams with more than two samples.

The U.S. average mercury in oil concentration was calculated according to eq 4 where country/state averages of N oil streams have the following estimated variance:

$$(s^{cs})^2 \approx \frac{1}{N^2}\sum_{j=1}^{N}[(s_j^{os})^2/n_j] \tag{5}$$

where $(s_j^{os})^2$ is the variance of each of the n_j samples of the jth oil stream in a country or state. To calculate country/state variances, oil stream variances at sample numbers $n = 1$ and $n = 2$ used the proportionality shown in Figure 2. The volume weighted variance in the U.S. mean (S_{US}^2) was estimated in the same manner as the volume weighted mean:

$$S_{US}{}^2 \approx \sum_{CS}\left\{\left(\frac{V^{CS}}{0.9V^{US}}\right)^2 (s^{cs})^2\right\} \tag{6}$$

The estimated uncertainty in the U.S. mean mercury concentration was therefore estimated to be

$$\bar{C}^{US} \pm 1.96S_{US} \tag{7}$$

Using the stated methods and approximations, the estimated uncertainty in the volume (2004) weighted mean mercury concentration is 3.5 ± 0.6 μg/kg. While more sophisticated ANOVA methods might improve the accuracy of the estimate of uncertainty, the calculated variance likely suffices for practical purposes.

Comparison to Recent Studies. Long and Kelly (*14*) reported concentrations of mercury in crude oil obtained using an isotope dilution method. The sample ensemble derived from oils delivered to the strategic petroleum reserve (SPR). The average concentration measured for the limited set of samples obtained from SPR deliveries was less than 2 μg/kg.

Magaw et al. (*15*) reported data on 26 crude oils that were regionally identified and described as purchased by U.S. refineries. Magaw's data span the major U.S. west coast crude streams and include concentrations for both domestically produced and imported crudes. Magaw reported total mercury concentrations below 10 ug/kg (the detection limit of the CVAA instrument) for all oils except one. Magaw reported one California crude oil (Cymric) as having 1.5 mg/kg THg, which is the single exception to all of the 25 other crude oils that tested below the instrumental detection limit.

The New Jersey Department of Environmental Protection reported concentrations of total mercury in oil delivered to U.S East Coast refineries (*16*). The methods of analysis were similar to those employed in this study, i.e., combustion and digestion with CVAF detection. Approximately 25 oil streams were analyzed with replication. The oils originated in the Middle East, South America, Africa, North Sea, Canada, and Mexico. The range of THg was 0.1−12.3 μg/kg. The mean and median were reported as 3.5 and 2.7 μg/kg, respectively.

In parallel to the study reported here, Environment Canada investigated mercury in crude oils refined in Canada (*17*). In the Canadian study, samples of crude oil were collected from refineries for crude types covering approximately 70% of the total crude volume processed in Canada in 2002. The sampling procedures and methods of analysis in the Canadian study were similar (combustion, atomic fluorescence and atomic absorption detection) to those employed in this investigation. The volume weighted average for Canadian refined oils is reported preliminarily as less than 4 μg/kg (95% < 10 μg/kg).

Nothing is obvious in the literature to suggest that analytical methods other than those employed in this study and used to measure total mercury concentrations in crude oil provide consistently higher THg values. One can make a case for low method bias in some literature studies when precautions against loss of volatile mercury or procedures to homogenize samples were not incorporated into the method procedures. No ensemble bias was obvious or suspected. It should be noted, however, that the calculated mean is sensitive to even a few high mercury oil streams that may arrive to U.S. refineries now or in the future. If refineries processed an additional 30 million barrels per year (0.5% of the U.S. total) having an average 1000 μg/kg total mercury concentration, the calculated U.S. volume weighted average would more than double.

Significance. The mass of coal burned in the U.S. annually (~10^{12} kg/y) is approximately the same as the mass of oil refined in the U.S. annually (*13*). The concentration of mercury in all U.S coal (coal rank volume corrected) is approximately 100 μg/kg (*1, 2*). From the measured mean concentration for total mercury in oil and total annual volume (2004), oil that passes through U.S. refineries contains approximately 3 metric tons of mercury. The maximum amount of mercury released to the ecosphere from oil processed in the U.S. is, therefore, approximately less than 5% of that which may be derived from burning coal in any given year.

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Environmental Protection Agency. Mention of trade names, products, or services does not convey, and should not be interpreted as conveying, official EPA approval, endorsement, or recommendation. The participation and cooperation of the American Petroleum Institute (Karin Ritter) and the National Petrochemical and Refiners Association (Norbert Dee) are gratefully acknowledged. The donation of crude oil samples by API and NPRA member companies is sincerely appreciated.

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10 Commercial Hydrotreating and Hydrocracking

Paul R. Robinson and Geoffrey E. Dolbear

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10.1 INTRODUCTION

Hydrotreaters are the most common process units in modern petroleum refineries. As shown in Table 10.1, the world's hydrotreating capacity is nearly half as large as the world's crude distillation capacity (Stell, 2003). In more than 700 refineries around the globe, there are more than 1300 hydrotreating units. A typical Western petroleum refinery (Figure 10.1) uses at least three hydrotreaters — one for naphtha, one or two for light gas oil, and one or two for heavy gas oil or vacuum gas oil.

Hydrocracking is far less common than hydrotreating, but the number of partial conversion mild hydrocrackers is increasing as refiners build new units to meet clean fuel regulations.

10.2 HYDROPROCESSING UNITS: SIMILARITIES AND DIFFERENCES

Process flow schemes for hydrotreating and hydrocracking are similar. Both use high-pressure hydrogen to catalytically remove contaminants from petroleum fractions. Both achieve at least some conversion, and they use the same kinds of hardware. Therefore, to avoid redundancy, we will discuss them together. As is common in the refining industry, we use the term ***hydroprocessing*** when a statement applies to both hydrotreating and hydrocracking.

TABLE 10.1
Worldwide Refining Process Units (as of January 1, 2004)

	Crude Distillation	Coking + Visbreaking	FCC	Catalytic Reforming	Hydrotreating	Hydrocracking
Number of units	>710	>330	360	550	1316	168
Total world capacity[a]	82.0	8.0	14.3	11.3	40.3	4.6
Average capacity[b]	114,000	45,700	39,700	20,500	30,600	27,400

[a] Million barrels per calendar day.
[b] Barrels per calendar day.

Source: Stell, J. *Oil & Gas J.* 101(49), December 22, 2003. (With permission.)

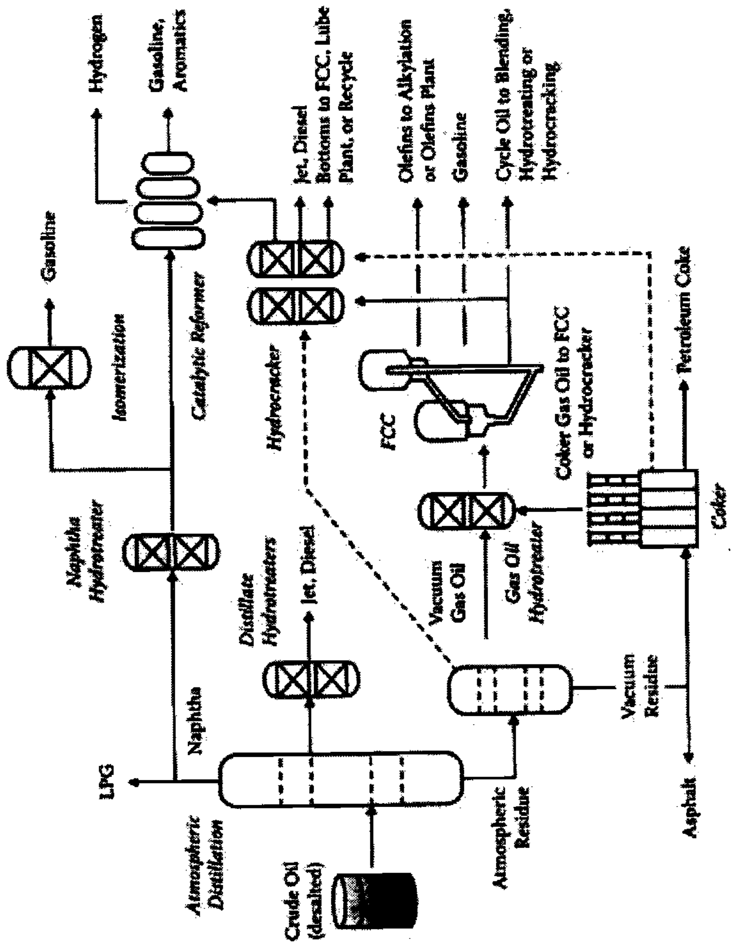

FIGURE 10.1 Layout of a typical high-conversion oil refinery.

TABLE 10.2
Hydrotreating and Hydrocracking: Ranges of H_2 Partial Pressure and Conversion

Process, Feedstock Types	H_2 Partial Pressure psig	H_2 Partial Pressure kPa	Conversion wt%
Hydrotreating			
Naphtha	250–450	1825–3204	0.5–5
LGO (kerosene)	250–600	1825–4238	0.5–5
HGO (diesel), LCO	600–800	4238–5617	5–15
VGO, VBGO, DAO, CGO, HCO	800–2000	5617–13,891	5–15
Residual oil	2000–3000	13,891–20,786	5–15
Mild Hydrocracking			
VGO, VBGO, DAO, CGO, LCO, HCO	800–1200	5617–8375	20–40
Once-Through Hydrocracking			
VGO, VBGO, DAO, CGO, LCO, HCO	1500–2000	10,443–13,891	60–90
Residual oil	2000–3000	13,891–20,786	15–25
Recycle Hydrocracking			
VGO, VBGO, DAO, CGO, LCO, HCO	1500–2000	10,443–13,891	80–99
Ebullated-Bed Hydrocracking			
VGO, VBGO, DAO, HCO	2000	13,891	80–99
Residual oil	2000–3000	13,891–20,786	>50

Note: LGO = light gas oil; HGO = heavy gas oil; LCO = FCC light-cycle oil; HCO = FCC heavy-cycle oil; VGO = vacuum gas oil; VBGO = visbreaker gas oil; DAO = deasphalted oil; CGO = coker gas oil.

As shown in Table 10.2, the extent of conversion is the most significant difference between hydrotreating and hydrocracking. In this context, the term *conversion* is defined as the amount of unconverted oil in the product divided by the amount of unconverted oil in the feed. Unconverted oil is defined as material that boils above a specified temperature. For vacuum gas oil (VGO), a typical specified temperature is 650°F (343°C). Conversion in hydrotreaters is less than 15 wt%, while conversion in hydrocrackers and mild hydrocrackers exceeds 20 wt%.

In hydrotreating units, reactions that convert organic sulfur and nitrogen into H_2S and NH_3 also produce light hydrocarbons. The removal of sulfur from dibenzothiophene (boiling point = 630°F, 332°C) generates biphenyl (492.6°F, 255.9°C). This reaction does not break any carbon-to-carbon bonds, but it does convert a molecule that boils above 600°F (315.5°C) into one that boils below 600°F (315.5°C).

Hydrotreating and hydrocracking differ in other ways. For a given amount of feed, hydrocrackers use more catalyst and operate at higher pressures. They also use different catalysts. Because they make large amounts of light products, hydrocracker fractionation sections must be more complex. In some hydrocrackers, unconverted oil from the fractionation section is recycled, either back to the front of the unit or to a separate cracking reactor.

Many mild hydrocrackers contain at least one bed of cracking catalyst, which allows them to achieve higher conversion — between 20 and 40 wt%. The unconverted bottoms can go to a fluid catalytic cracking (FCC) unit, a lube plant, or fuel oil blender. Due to its high value in other applications, the bottoms are blended into fuel oil only when there is no other feasible option.

In hydrocrackers that process vacuum gas oils or other feeds with similar boiling ranges, the typical once-through conversion exceeds 60 wt%. If the unconverted oil is recycled, the overall conversion can exceed 95 wt%. As with mild hydrocracking, the unconverted bottoms are high-value oils, which usually are sent to FCC units, lube plants, or olefin plants. For heavier feeds — atmospheric and vacuum residues — conversions are much lower, especially in fixed-bed units. In ebullated-bed units, the conversion of 1050°F+ (566°C+) residue can exceed 60 wt%.

Catalytic isomerization and dewaxing is a special kind of hydrocracking used to make high-quality lube base stocks.

10.3 PROCESS OBJECTIVES

Table 10.3 presents a list of feeds and product objectives for different kinds of hydrotreaters and hydrocrackers. In the 1950s, the first hydrotreaters were used to remove sulfur from feeds to catalytic reformers. In the 1960s, the first hydrocrackers were built to convert gas oil into naphtha.

Today, in addition to naphtha, hydrotreaters process kerosene, gas oil, vacuum gas oil, and residue. Hydrocrackers process vacuum gas oil, coker gas oil, visbreaker gas oil, FCC heavy cycle oil, and other feeds that boil between 650 and 1050°F (343 and 566°C). Most residue hydrocrackers use fluidized-bed or ebullated-bed technology.

For hydroprocessing units, product specifications are set to meet plant-wide objectives. For example, the naphtha that goes to catalytic reforming and isomerization units must be (essentially) sulfur-free. Before it can be sold as jet fuel, the aromatics content of kerosene must be low enough to meet smoke-point specifications (American Society for Testing and Materials [ASTM] D1655). Heavier distillates cannot be sold as diesel fuel unless they meet stringent sulfur specifications.

10.3.1 Clean Fuels

As mentioned in Chapter 1, on-road diesel in the United States must contain <15 wppm sulfur by 2006. The sulfur limit for nonroad diesel will be 500 wppm in 2007.

TABLE 10.3
Feeds and Products for Hydroprocessing Units

Feeds	Products from Hydrotreating	Products from Hydrocracking
Naphtha	Catalytic reformer feed	LPG
Straight-run light gas oil	Kerosene, jet fuel	Naphtha
Straight-run heavy gas oil	Diesel fuel	Naphtha
Atmospheric residue	Lube base stock, low-sulfur fuel oil, RFCC[a] feed	Naphtha, middle distillates, FCC feed
Vacuum gas oil	FCC feed, lube base stock	Naphtha, middle distillates, FCC feed, lube base stock, olefin plant feed
Vacuum residue	RFCC feed	Naphtha, middle distillates, RFCC feed
FCC light-cycle oil	Blend stocks for diesel, fuel oil	Naphtha
FCC heavy-cycle oil	Blend stock for fuel oil	Naphtha, middle distillates
Visbreaker gas oil	Blend stocks for diesel, fuel oil	Naphtha, middle distillates
Coker gas oil	FCC feed	Naphtha, middle distillates, FCC feed, lube base stock, olefin plant feed
Deasphalted oil	Lube base stock, FCC feed	Naphtha, middle distillates, FCC feed, lube base stock

[a] RFCC = residue FCC unit or reduced crude FCC unit, both of which are specially designed to process feeds that contain high concentrations of carbon-forming compounds.

The present U.S. specification for gasoline is <30 wppm sulfur. In the European Union, the sulfur content of both gasoline and diesel must be <50 wppm by 2005 and <10 wppm by 2008.

To meet clean fuel specifications, refiners in North America and Europe are increasing their hydroprocessing capabilities and adjusting operations. Two real-world examples are described below.

Example 10.1

A U.S. refinery is planning to produce diesel fuel that contains <15 wppm sulfur by June 2006. At present, the hydrocracker makes 39,000 barrels/day of middle distillate that is nearly sulfur-free. The existing 60,000 barrels/day distillate hydrotreater (DHT) gives a product with 600 to 700 wppm sulfur. Mixing the two streams yields a blend containing 425 to 485 wppm sulfur, which meets existing specifications for low-sulfur diesel fuel (per ASTM D975). To make ultra-low-sulfur diesel (ULSD), the refiner is adding a reactor and a high-pressure amine absorber to the existing DHT, enabling the unit to make a stream with 12 to 18 wppm sulfur. Blending this with distillate from the hydrocracker will give a final product containing 7 to 12 wppm sulfur.

Example 10.2

A European refiner now runs a mild hydrocracker (MHC) to maximize conversion of VGO and to pretreat the feed to its FCC unit. The plant cannot posttreat its FCC gasoline, so the sulfur content of the MHC bottoms must be less than 500 wppm to guarantee that the sulfur content of the FCC gasoline is less than 150 wppm. Other low-sulfur streams (reformate, alkylate, and hydrotreated gas oil) go into the final gasoline blend, so sulfur in the FCC gasoline can exceed the final product limit of 50 wppm.

10.3.2 The Process in Between

As shown in Figure 10.1, hydrocracking often is an in-between process. The required hydrogen comes from catalytic reformers, steam/methane reformers, or both. Liquid feeds can come from atmospheric or vacuum distillation units, delayed cokers, fluid cokers, visbreakers, or FCC units. Middle distillates from a hydrocracker usually meet or exceed finished product specifications, but the heavy naphtha from a hydrocracker usually is sent to a catalytic reformer for octane improvement. The fractionator bottoms can be recycled or sent to an FCC unit, an olefins plant, or a lube plant.

10.4 PROCESS MODELING

During the past 20 years, academic and industrial researchers have developed composition-based kinetic models with hundreds or even thousands of lumps and pure compounds. The quantitative structure-reactivity correlation (QSRC) and linear free energy relationship (LFER) lumping techniques are discussed by Klein and Hou (2006). The structure-oriented lumping (SOL) approach of Quann and Jaffe (1996) yields models rigorous enough for use in closed-loop real-time optimizers (CLRTOs), which automatically adjust setpoints for commercial process units several times each day.

In the composition-based model developed by Lapinas et al. (1991) and applied to a commercial hydrocracker by Pedersen et al. (1995) rate equations are based on the Langmuir–Hinshelwood–Hougen–Watson (LHHW) mechanism for heterogeneous reactions. In brief, the LHHW mechanism describes (1) the adsorption of reactants to acid and metal sites on a catalyst surface; (2) reactions between the reactants, including saturation, cracking, ring opening, dealkylation, hydrodesulfurization (HDS); hydrodenitrogenation (HDN); and so forth, and (3) desorption of products. Inhibition effects are modeled, too. These include the adsorption of organic nitrogen to acid sites and the inhibition of HDS reactions by H_2S.

Rigorous, flow-sheet-based models for hydrocrackers include submodels for furnaces, pumps, compressors, reactors, quench zones, flash drums, recycle gas scrubbers, fractionation towers, and — importantly — economic data. As discussed by Mudt et al. (2006) such models can comprise hundreds of reactions and hundreds of thousands of equations. The model grows when inequalities are

TABLE 10.4
Supports Used in Hydroprocessing Catalysts

Support	Major Use	Acidity
γ-Alumina	Hydrotreating catalysts	Low
Amorphous aluminosilicates	Distillate-selective hydrocracking catalysts	High
Zeolites (X, Y, or mordenite)	High-stability hydrocracking catalysts	Very high

included to ensure a feasible solution that honors process constraints. To solve such models in real time (that is, in less than 1 h), open-equation mathematics and high-powered solvers are used.

10.5 HYDROPROCESSING CATALYSTS

Recent books by Magee and Dolbear (1998) and Scherzer and Gruia (1996) are superb sources of technical information on hydroprocessing catalysts. The hydroprocessing catalyst business is big, with annual sales approaching U.S.$800 million per year. The materials most commonly used to make these catalysts are shown in Table 10.4 and Table 10.5.

In fixed-bed hydroprocessing units, the catalysts must be able to drive the desired reactions, but they also must possess a high surface area and great physical strength, enough to resist crushing under the forces imposed by rapidly flowing high-pressure fluids and the weight of the catalyst itself. A single bed can contain several hundred tons of catalyst.

Chemical reactions take place inside small pores, which account for most of the catalyst surface area. The diameters of these pores range from 75 to 85 Å for catalysts that process light and heavy gas oils. For catalysts that process residue, the average pore size ranges from 150 to 250 Å.

TABLE 10.5
Active Metals Used in Hydroprocessing Catalysts

Metals	Major Use	Activation Method	Hydrogenation Activity
CoMo	HDS	Sulfiding	Moderate
NiMo	HDN, hydrocracking	Sulfiding	High
NiW	HDN, hydrocracking	Sulfiding	Very high
Pd, Pt[a]	Hydrocracking	Reduction by H_2	Highest

[a] Pd and Pt are poisoned by sulfur and can only be used in low-H_2S environments.

10.5.1 CATALYST PREPARATION

The following steps may be used to prepare the supported metal catalysts used in hydrotreaters and hydrocrackers (Magee and Dolbear, 1998; Scherzer and Gruia, 1996):

- Precipitation
- Filtration (or centrifugation), washing, and drying
- Forming
- Calcining
- Impregnation
- Activation

Other steps, such as kneading, mulling, grinding, and sieving, may also be used. For some catalysts, some of the above-listed steps are eliminated or additional steps are added. For example, if mulling is used to mix active metals with a support, precipitation and impregnation may not be needed.

10.5.1.1 Precipitation

In the catalyst world, precipitation involves combining two solutions to form a desired solid. For example, mixing an aqueous solution of aluminum nitrate [$Al(NO_3)_3$] with sodium aluminate [$Na_2Al_2O_4$] yields aluminum hydroxide [$Al(OH)_3$], which forms a gelatinous solid. As the gel ages, tiny crystals grow larger and a pore structure starts to develop.

The zeolites used in hydrocracking catalysts are also prepared by precipitation. Zeolites occur naturally, but the ones used for catalysis are synthetic. Figure 10.2 outlines a common procedure for synthesizing Na-Y and H-Y zeolites.

These remarkable aluminosilicates can be used as drying agents, ion exchangers, and molecular sieves for gas separation. Their microporosity provides them with high surface area, and they can be converted into solid acids with superb catalytic activity.

The Al(III) atoms in zeolites replace Si(IV) atoms in a SiO_2 superstructure. To maintain a neutral charge, every aluminum atom must be accompanied by a counter-ion such as Na^+, K^+, H^+, NH_4^+, and so forth. Counter-ions can be swapped via ion exchange. When Na-Y zeolite is exchanged with an ammonium salt, the Na^+ ion is replaced by NH_4^+. When NH_4-Y is heated to the right temperature, the ammonium ion decomposes, releasing NH_3 (gas) and leaving behind the highly acidic H-Y zeolite.

The synthetic zeolites used in catalysts for hydrocracking include X, Y, mordenite, and ZSM-5. The latter is made by including a soluble organic template, such as a quaternary ammonium salt, in the mix of raw materials. ZSM-5 is used for catalytic dewaxing. Due to its unique pore structure, it selectively cracks waxy n-paraffins into lighter molecules. It is also used in FCC catalysts to increase propylene yields.

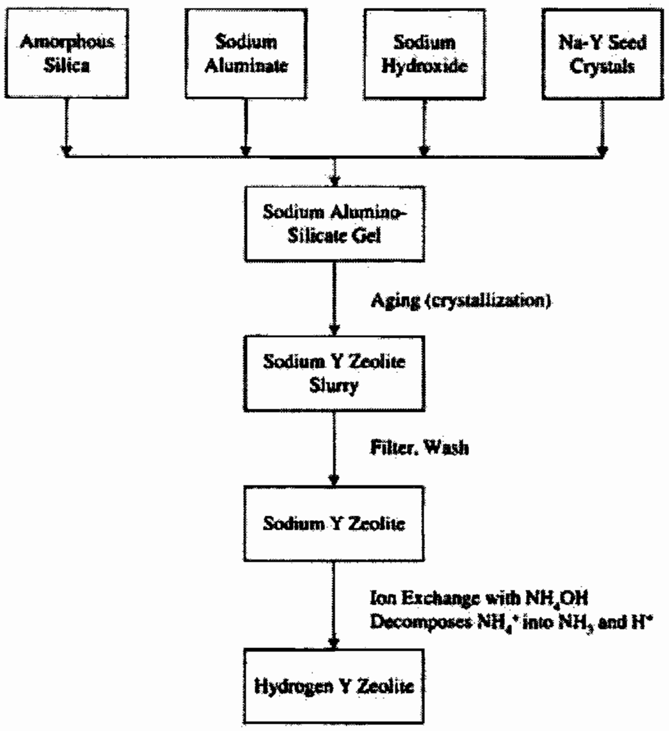

FIGURE 10.2 Synthesis procedure for H-Y zeolite.

10.5.1.2 Filtration, Washing, and Drying

Filtration, washing, and drying remove undesired impurities. In our $Al(OH)_3$ example, sodium nitrate is washed away with water. Sometimes ammonium hydroxide is added to expedite sodium removal. Subsequent air- and oven-drying removes most of the excess water and initiates the transformation of $Al(OH)_3$ into alumina (Al_2O_3).

10.5.1.3 Forming

Catalyst and support precursors can be formed into extrudates, spheres, or pellets. Extrudates are generated by forcing a paste (for example, formed by mixing powdered-alumina with water) through a die. Adding peptizing agents such as nitric acid increases the average pore size of the product. Raising the extrusion pressure tends to decrease the average pore size.

The resulting spaghetti-like strands are dried and broken into short pieces with a length/diameter ratio of 3 to 4. The particles are dried and then calcined. In our alumina example, calcination decomposes residual ammonium nitrate. It also hardens the particles and completes the conversion of $Al(OH)_3$ into Al_2O_3. The preferred alumina for catalyst supports is γ-alumina, also known as bohemite. This material has a high surface area, great physical strength, and a well-defined network of pores. If the calcination temperature gets too high, γ-alumina transforms into α-alumina or β-alumina, whose physical properties are far less desirable.

An extrudate cross section can be circular or shaped like a three- or four-leaf clover without the stem (Gruia, 2006). Compared to cylindrical extrudates, clover-leaf (multilobe) catalysts have a higher surface-to-volume ratio. In trickle-bed hydroprocessing reactors, they have less resistance to diffusion and a lower pressure drop. Spherical catalysts are made by (1) spray-drying slurries of catalyst precursors, (2) spraying liquid onto powders in a tilted rotating pan, or (c) dripping a silica-alumina slurry into hot oil (Magee and Dolbear, 1998). Pellets are made by compressing powders in a dye.

10.5.1.4 Impregnation

Impregnation is a common technique for distributing active metals within the pores of a catalyst support. Calcined supports are especially porous. Like sponges, they use capillary action to suck up aqueous solutions containing active metals. For some catalysts, the support is soaked in excess metal-containing solution, which saturates the pores fully.

In the incipient wetness method, precise amounts of solution are added — just enough to leave the support dry to the touch. After a drying step, additional solution may be added to increase loading of the same or different active metal.

10.5.1.5 Activation

Prior to use, most nonnoble-metal catalysts are activated (sulfided) by circulating hydrogen and a light, sulfur-containing start-up oil through the catalyst. Often, the start-up oil is spiked with dimethyl sulfide (CH_3–S–CH_3) or dimethyl disulfide (CH_3–S–S–CH_3). The temperature is raised slowly to the decomposition temperature of the sulfiding agent. The process continues until breakthrough, that is, the point at which significant amounts of H_2S appear in the recycle gas.

During dry sulfiding, a mixture containing 2 to 5 vol% H_2S in hydrogen is circulated through the catalyst. The temperature is increased slowly to the temperature at which the unit is expected to operate. The process continues until the exit gas contains the same amount of H_2S as the inlet gas.

Most manufacturers offer presulfided catalysts, which allow a refiner to shorten the start-up of a unit by 2 or 3 days. That may not seem like much, but for a 40,000 barrel/day FCC feed pretreater, it can generate up to U.S.$500,000 in extra income.

10.5.2 NOBLE-METAL CATALYSTS

Some hydrocracking catalysts contain small amounts of highly dispersed platinum or palladium. These noble metals are expensive, but their loading is low — 0.6

to 1.0 wt% — and their high hydrogenation activity justifies the cost. They are added to hydrocracking catalysts by impregnation with tetraammine complexes — $Pt(NH_3)_4^{2+}$ or $Pd(NH_3)_4^{2+}$. When the catalysts are heated in air to about 840°F (450°C), the complexes decompose, giving off ammonia and leaving behind divalent metal oxides.

In commercial hydrocrackers, catalysts containing noble-metal oxides are activated by direct reduction with high-pressure hydrogen at 700°F (350°C).

10.5.3 HYDROTREATING CATALYSTS

Hydrotreating catalysts comprise oxides of either Mo or W and either Co or Ni on a support comprised of γ-alumina. Usually, CoMo catalysts are better for HDS while NiMo catalysts are better for HDN. NiW catalysts are especially active for the saturation of aromatics. Typical physical properties are shown in Table 10.6.

Hydrotreating catalyst particles are surprisingly small, with diameters of 1.5 to 3.0 mm and length/diameter ratios of 3 to 4. In many units, ceramic balls or successively larger catalyst particles are loaded on top of the first catalyst bed. This graded bed protects the bulk of the catalyst by filtering particulate matter out of the feed.

10.5.4 HYDROCRACKING CATALYSTS

Commercial hydrocracking catalysts comprise active metals on solid, highly acidic supports. The active metals are Pd, NiMo, and NiW, all of which catalyze both hydrogenation and dehydrogenation reactions. The most common supports are synthetic crystalline zeolites and amorphous silica-aluminas.

Hydrocracking catalyst shapes can be spherical or cylindrical, with gross dimensions similar to those for hydrotreating catalysts.

As already mentioned, in most hydrocrackers, the first few catalyst beds contain a high-activity HDN catalyst, which also is active for HDS, saturation of olefins, and saturation of aromatics. Other hydrocrackers use a bifunction catalyst — one that is active for both hydrotreating and hydrocracking — in all catalyst beds.

TABLE 10.6
Physical Properties for Hydrotreating Catalysts

Property	Low	High
Surface area, m^2/g	150	250
Pore volume, ml/g	0.5	1.0
Pore diameter (average), Å	75	250
Bulk density, lb/ft^3	30	60
Bulk density, kg/m^3	490	980
Co or Ni (as CoO or NiO), wt%	3	8
Mo or W (as MoO_3 or WO_3), wt%	10	30

10.5.5 CATALYST CYCLE LIFE

Catalyst cycle life has a major impact on the economics of fixed-bed refinery units, including hydrotreaters and hydrocrackers. Cycles can be as short as 12 months and as long as 60 months. Two-year cycles are typical. At the start of a cycle, average reactor temperatures are low — 620 to 660°F (327 to 349°C). As the cycle proceeds, the catalyst deactivates and refiners must raise temperatures to maintain conversion. A catalyst cycle is terminated for one of the following reasons, whichever occurs first. Note that only one of the listed events relates directly to catalyst activity.

1. *The temperature required to achieve the unit's main process objective hits a metallurgical limit.* Or alternatively, the main process objective can be met only at reduced feed rate. To ensure safe operation, the maximum average reactor temperature is about 760°F (404°C) and the maximum peak temperature is about 800°F (427°C).
2. *Side reactions are starting to cause process or economic problems.* If the production of light gases exceeds the capacity of one or more towers in the downstream gas plant, operators must decrease feed rate or reduce conversion. Both options are expensive. Excess gas production consumes expensive hydrogen and converts it into low-value liquefied petroleum gas (LPG), which also is expensive. Running at high temperature decreases selectivity to middle distillates and increases aromatics in middle distillates. At some point, due to one or more of these factors, refinery-wide economics show that it is better to shut down for a catalyst change vs. trying to keep limping along — even though metallurgical limits have not yet been reached.
3. *The recycle compressor cannot overcome pressure drop across the unit.* The overall pressure drop is the difference in pressure between the recycle compressor suction and the recycle compressor discharge. At start-of-run, the pressure drop across the catalyst is low — 3 to 10 psi (0.2 to 0.7 bar) for each bed — but it increases as the run proceeds. Usually, the increase is largest in the first catalyst bed, which is most susceptible to fouling. Attempts to continue running a unit despite very high pressure drop can deform the quench-deck support beams inside a reactor.
4. *A related unit has to shut down for more than a few weeks.* Related units might include an upstream vacuum distillation unit, an upstream hydrogen source, or a downstream FCC unit. In refineries with enough intermediate tankage, hydroprocessing units can continue to run for a few days despite an interruption in the supply of liquid feed, but a loss of hydrogen supply can cause an immediate shutdown. At best, if the unit gets hydrogen from multiple sources, the feed rate must be reduced.

5. *Major process upsets.* Most process upsets are caused by sudden changes in feed quality. For a fixed-bed VGO hydrotreater, a slug of residue can poison part of the catalyst with trace metals such as Fe, Ni, V, and Si, or foul it with particulates, asphaltenes, or refractory carbon. In fixed-bed units, poisoning and fouling usually are confined to the top few feet of the first catalyst bed. If so, the ruined catalyst can be skimmed off the top and replaced during a brief, scheduled shutdown. A brief, scheduled shutdown does not require a cycle-ending catalyst change-out.
6. *Equipment failure.* Hardware problems occur most frequently in rotating equipment — pumps and compressors. Fortunately, many problems can be detected in advance, allowing operators to schedule a brief shutdown for preventive maintenance.

Process variables that increase or decrease the rate of catalyst deactivation are shown in Table 10.7.

Hydrogen keeps the catalyst clean by inhibiting coke formation. This explains why increasing the hydrogen partial pressure decreases the rate of catalyst deactivation.

Raising the temperature increases the rates of most hydrocracking reactions, including coke formation. Raising the hydrogen/oil ratio increases heat removal, which limits temperature rise.

If the feed rate goes up and targets for HDS, HDN, or conversion remain the same, the temperature must go up. If the feed rate goes up and the temperature does not, then HDS, HDN, or conversion will decrease.

TABLE 10.7
Factors Affecting Catalyst Cycle Life

	Effect on Cycle Life	Comment
Higher-H_2 partial pressure	+	
Higher recycle gas rate	+	Increases H_2 partial pressure
Higher makeup gas purity	+	Increases H_2 partial pressure
Increased purge of recycle gas	+	Increases H_2 partial pressure
Higher fresh feed rate	–	
Higher conversion	–	
Higher fresh feed endpoint	–	Increases rate of catalyst coking; can increase pressure-drop buildup rate
Higher fresh feed impurities[a]	–	Related to feed type and feed endpoint
Process upsets[b]	–	

[a] Deleterious feed impurities include sulfur, nitrogen, refractory carbon, asphaltenes, metals (nickel, vanadium, iron, silicon), and particulate matter (coke fines, FCC catalyst fines).

[b] Process upsets include "burps" in upstream units that feed the hydrocracker, equipment failures (typically loss of a feed pump or compressor), or temperature excursions requiring depressuring.

Increasing the feed endpoint or density tends to increase the amount of coke precursors in the feed. The precursors include asphaltenes, refractory carbon, and polynuclear aromatic hydrocarbons (PAH).

10.5.5.1 Catalyst Regeneration and Rejuvenation

After working 24/7 for a year or two (or in some cases five) in a fixed-bed hydroprocessing unit, the catalyst is spent. The entire unit is shut down and catalyst is removed. During the shutdown, which typically lasts 3 to 4 weeks, refiners inspect and repair equipment. Meanwhile, the catalyst is shipped to an off-site facility, where it is regenerated by controlled combustion in air, air plus oxygen, or air plus steam. During combustion, accumulated coke is converted to CO_2 and CO plus small amounts of SO_2 and NOx, which are formed from the sulfur and nitrogen in the coke. Typically, the temperature used for regeneration in air is 750 to 930°F (400 to 500°C).

The regenerated catalyst may also undergo rejuvenation, a wet process in which the active metals are chemically redispersed. A combination of regeneration and rejuvenation can restore a catalyst to more than 95% of its original activity.

Inevitably, some particles break apart during the unloading, transportation, regeneration, and rejuvenation of spent catalysts. If part of the catalyst is contaminated with Fe, Ni, V, or Si, that part cannot be regenerated. Typically, losses due to fragmentation and fouling amount to 10 to 15%.

In the bad old days, regeneration meant burning coke off the catalyst while it was still inside the reactor. Today, *in situ* regeneration is rare because it is hard to control and often gives poor results. A poor regeneration is costly, because afterwards the unit's performance will be poor. With a crippled catalyst, the unit may have to limp along for several months at lower feed rates and lower severity. Worst of all, the catalyst will not last long, which means that it will have to be regenerated or replaced sooner rather than later.

10.5.5.2 Catalyst Reclamation

Even though noble-metal hydrocracking catalysts contain only small amounts of Pd or Pt, these metals are so expensive that recovering the metals is more cost-effective than throwing them away. Other hydroprocessing catalysts contain Mo or W, Ni, or Co. Spent hydrotreating catalysts — especially those used to hydrotreat residue — can be very rich in vanadium, richer than many ores.

Reclamation companies convert these materials into salable products using different combinations of oxidation, pyrolysis, dissolution in acid or alkali, precipitation, extraction, or ion exchange. Depending on the process used, the salable products may include several of the materials shown in Table 10.8.

The book by Scherzer and Gruia (1996) provides a well-written description of catalyst reclamation processes used by four major companies — CRI-MET, Eurecat, Gulf Chemical, and TNO/Metrex.

TABLE 10.8
Some of the Materials Sold by Catalyst Reclamation Companies

Material	Formula
Palladium metal or chloride salt	Pd or Na_2PdCl_4
Platinum metal or chloride salt	Pt of Na_2PtCl_4
Molybdenum trisulfide	MoS_3
Molybdenum oxide	MoO_3
Ammonium molybdate	$(NH_4)_2Mo_4O_{13}\cdot 2H_2O$
Sodium molybdate	$Na_2MoO_4\cdot 2H_2O$
Tungsten trioxide	WO_3
Ammonium para-tungstate	$(NH_4)_{10}W_{12}O_{41}\cdot 5\ H_2O$
Sodium tungstate	$Na_2WO_4\cdot 2H_2O$
Vanadium pentoxide	V_2O_5
Sodium (meta) vanadate	$NaVO_3$
Nickel metal or chloride	Ni or $NiCl_2$
Cobalt metal or chloride	Co or $CoCl_2$
Nickel-cobalt concentrate	Ni_xCo_y
Iron-molybdenum concentrate	Fe_xMo_y
Alumina hydrate	$Al_2O_3\cdot 3H_2O$

10.6 PROCESS FLOW

10.6.1 Trickle-Bed Units

Most hydrotreaters and hydrocrackers are trickle-bed units. A classic article by Satterfield describes the fundamental behavior of such units, in which mixtures of liquid and gaseous reactants pass down over fixed beds of catalyst. In hydroprocessing units, the liquid reactants are petroleum fractions, and the gaseous reactant is hydrogen.

Figure 10.3 shows a flow scheme for a once-through unit designed to process heavy gas oil feeds. Designs offered by major process licensors can differ in several areas, which correspond to the bold numbers on the diagram.

1. *Heaters.* Units with gas-only heaters mix hot gas with preheated liquid feed just before the reactants enter the first reactor. Other designs use a gas-plus-oil heater to bring the mixed fluids up to reaction temperature.
2. *Reactors, catalyst beds, and quench zones.* Addition of hydrogen typically occurs with heat release, and most hydroprocessing reactions are exothermic. The heat released in naphtha and kerosene hydrotreaters is relatively low, so units designed for these feeds may use just one reactor that contains a single catalyst bed. However, for heavier feeds or feeds that contain large amounts of sulfur, aromatics, or olefins, the total increase in temperature can exceed 180°F (100°C). It is unsafe to allow that much temperature rise in a single bed of catalyst. To divide the heat release into smaller, safer portions, commercial units use multiple catalyst beds with cooling in between. A unit can have one bed per reactor, or multiple beds in each reactor with quench zones in between. For simplicity, Figure 10.4 shows only 4 catalyst beds, but most hydrocrackers have more; some have as many as 30.

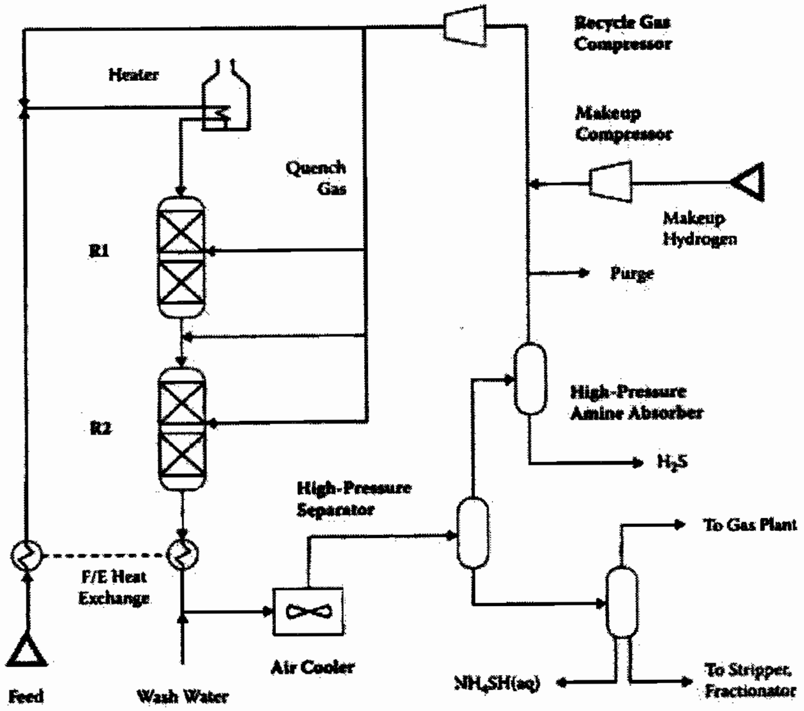

FIGURE 10.3 Once-through hydroprocessing unit: two separators and recycle gas scrubber.

In a quench section (Figure 10.4), hot process fluids from the preceding bed are combined with relatively cold hydrogen-rich quench gas before the mixture passes into the next bed. We can think of a catalyst bed as a stack of thin, horizontal discs. Ideally, the top disc is the coolest, the bottom disc is the hottest, and at every point in each given disc, temperatures are identical. But in real units, the downward flow of reactants is never perfectly uniform, so the temperatures within the discs are different, especially near the bottom.

The difference between the highest and lowest temperature at the bottom of a catalyst bed is called the radial temperature difference (RTD).

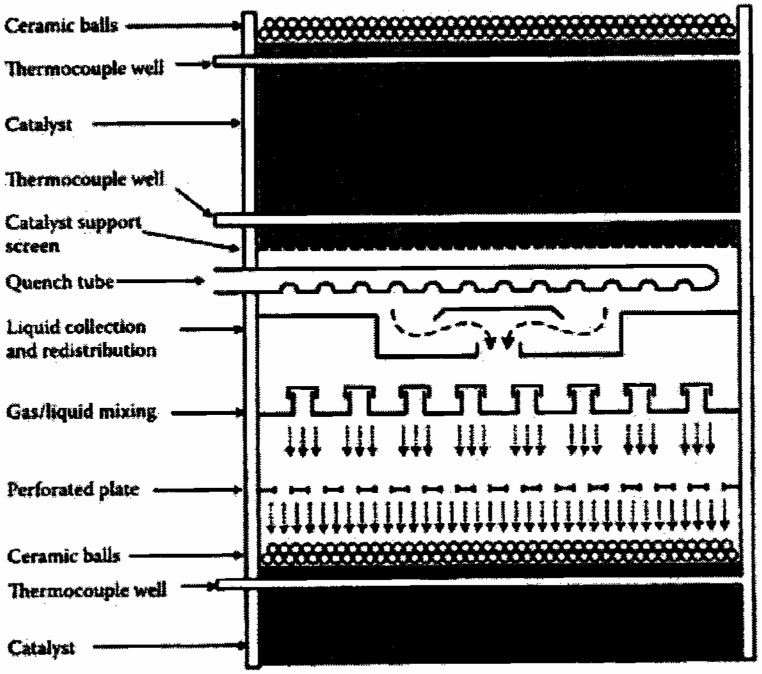

FIGURE 10.4 Hydroprocessing reactor: quench zone.

The truth is, we never know the actual highest and lowest temperatures, because we cannot place thermocouples everywhere. But if the measured RTD is small — less than 5°F (3°C) — we can assume that the actual RTD is also small, and that flow through the bed is nearly uniform. If the measured RTD is large, the actual RTD is almost certainly larger, and we have to be concerned about hot spots, flow blockages, and other potentially dangerous symptoms of maldistribution.

Modern quench sections are designed to do three things: (1) lower the overall temperature of the reacting fluids, (2) reduce radial maldistribution with radial mixing, and (3) redistribute the reactants and deliver them to the next bed. The major parts of a quench deck are the quench tube, the liquid collector and redistributor, the gas/liquid mixing zone, and the final distributor.

Quench tubes bring quench gas into the reactor. Some are very simple — just a tube with a series of holes in it. Others, such as the ExxonMobil spider vortex design, are more complex, distributing gas horizontally through several spokes to different parts of the quench deck.

In the liquid collector and redistributor, liquids are forced to flow down two angled slides into a raceway. The slides give the liquids some angular momentum, and the raceway gives them time to mix. More than anything else, this part of the quench deck reduces RTD.

In the gas/liquid mixing zone, a bubble-cap tray or similar device provides intimate contact between gases and liquids from the redistribution zone. The final distributor sends a fine spray of fluids down to the catalyst bed below.

In residue hydroprocessing units, heat release is high, but some licensors avoid using intrareactor quench because residue feeds often form lumps of coke-bonded catalyst in fixed-bed units. In reactors with complex internals, such lumps are very hard to remove during a catalyst change-out. Therefore, fixed-bed residue units often comprise three or more one-bed reactors in series with quench in between. In many cases, the first reactor is a guard bed filled with one or more catalysts designed to remove metals.

3. *Catalysts.* Hydrotreaters are loaded with either a CoMo HDS or NiMo HDN catalyst, or both. NiMo catalysts are better for the saturation of aromatics, which is required for the removal of hindered sulfur compounds during deep desulfurization. Therefore, some refiners load a layer of NiMo catalyst on top of a CoMo catalyst in diesel desulfurization units. Recently, catalyst manufacturers have been offering trimetallic (CoNiMo) hydrotreating catalysts.

   Most of the cracking in hydrocracking units is driven by catalysts with high acidity. The acidic sites are inhibited by organic nitrogen, so the first several catalyst beds in a hydrocracking unit typically contain a high-activity HDN catalyst. In a few units, all beds in a hydrocracker are filled with an amorphous dual-function catalyst, which catalyzes both HDN and cracking. This type of catalyst has a high selectivity for producing middle distillates from VGO.

   The last bed in a hydrocracker often contains a final layer of posttreat catalyst to remove mercaptans.

4. *Makeup and recycle hydrogen.* Compressors for makeup hydrogen are reciprocating machines, most of which are driven by electric motors. Recycle gas compressors can be reciprocating or centrifugal; the latter are often driven by steam. In naphtha hydrotreaters, the high-pressure off-gas can be purer than the makeup gas, because (a) conversion is nil and (b) liquids in the makeup gas are absorbed by the naphtha. In most other units, the makeup gas is purer than the recycle gas.

   Makeup hydrogen can enter the unit at the cold high-pressure separator (CHPS), at the suction of the recycle gas compressor, or at the discharge of the recycle gas compressor. If the makeup comes in at the CHPS, the makeup compressor discharge pressure is lower, which can reduce electricity costs. However, if part of the recycle gas is purged after leaving the CHPS, part of the incoming makeup gas goes right back out again. If the makeup comes in at the discharge of the recycle gas compressor, the discharge pressure of the makeup compressor is higher, but none of the high-purity makeup is lost with purge gas.

5. *High-pressure amine absorption.* Prior to the advent of ultra-low-sulfur fuels, it was rare to find hydroprocessing units with a high-pressure amine absorber to remove H_2S from the recycle gas. H_2S inhibits HDS reactions and lowers the purity of the recycle gas. For both of these reasons, high-pressure amine absorbers are now included in most new and revamped diesel hydrotreaters and mild hydrocrackers.
6. *Product cooling and separation.* Commercial units comprise a number of different product cooling and flash drum configurations. The simplest comprises a feed/effluent heat exchanger train, a large air- or water-cooled heat exchanger, and one or two flash drums.

   Heavy-feed units have at least a CHPS and a low-pressure separator (LPS). The CHPS overhead stream can go directly to the recycle gas system or through a high-pressure amine absorber for removal of H_2S. The CHPS bottoms go to the CLPS. Sometimes the pressure differential between the CHPS and CLPS is used to drive a power recovery turbine. As shown in Figure 10.5, some units include a hot high-pressure separator (HHPS) upstream from the CHPS. The HHPS overhead goes through a cooler to the CHPS, and HHPS bottoms go through a cooler to the LPS. This arrangement provides better heat recovery. In single-stage hydrocrackers with recycle of unconverted oil, hot separation minimizes fouling caused by the accumulation of PAH in the recycle oil.
7. *Wash water addition.* As mentioned above, HDS and HDN reactions produce H_2S and NH_3, respectively. Wash water is injected into the effluent from the last reactor to convert almost all of the NH_3 and some of the H_2S into aqueous ammonium bisulfide, $NH_4HS(aq)$. The $NH_4HS(aq)$ is rejected from the unit as sour water in the low-pressure flash drum.
8. *Fractionation.* For product fractionation, HDS units that treat naphtha or light gas oil may use a simple steam stripper to remove H_2S and traces of light hydrocarbons from the liquid product (CLPS bottoms). An absorber may be used to recover C_3+ compounds from the CLPS overhead.

   Conversion units may employ a full-fledged fractionation train, with a preflash tower to remove light ends; an atmospheric fractionator to separate light naphtha, heavy naphtha, middle distillates, and unconverted oil; and a vacuum tower to maximize the recovery of diesel. Some hydrocrackers use the atmospheric tower to produce full-range naphtha, which is then separated into light and heavy fractions in a naphtha splitter.
9. *Recycle of fractionator bottoms.* In full-conversion hydrocrackers, unconverted oil from the fractionator is recycled. Single-stage units with multiple reactors (Figure 10.5) send the recycled oil either to the hydrotreating reactor (R1) via the feed surge drum or to the hydrocracking reactor (R2). Recycle to R1 means that R1 must be larger, but recycle to R2 eliminates an expensive and troublesome high-pressure pump.

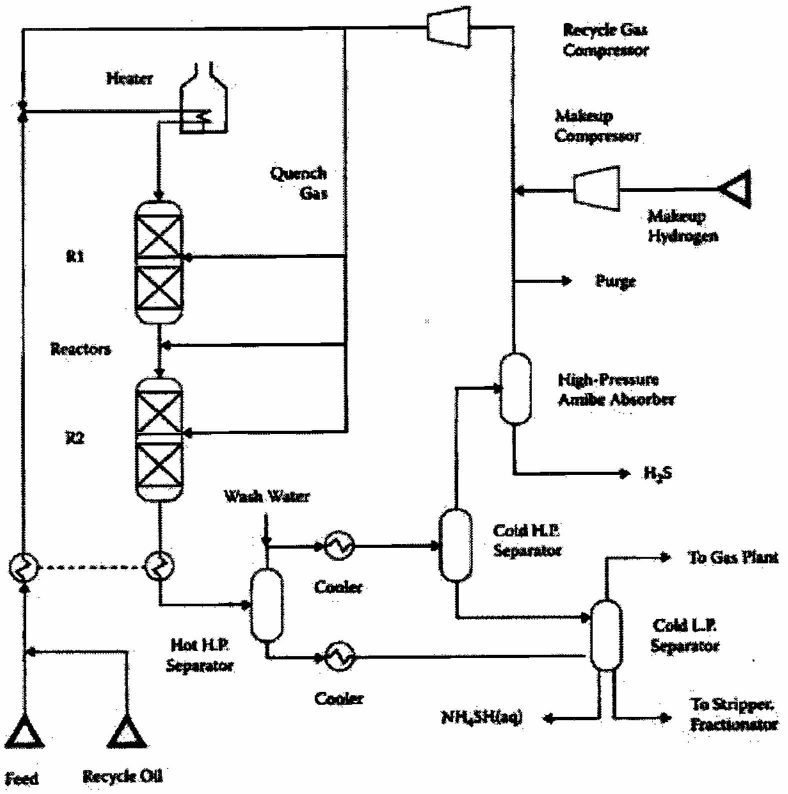

FIGURE 10.5 Single-stage hydrocracker: HHPS, recycle to R1.

Figure 10.6 shows a two-stage hydrocracker. In these units, unconverted oil goes to a separate cracking reactor (R3) with its own high-pressure separator.

The unit shown in Figure 10.6 uses a single makeup and recycle gas system to supply all reactors. In other units, the second stage has a separate gas system. Units with a common recycle gas system need only one recycle compressor, but in units with two gas systems, the second stage can operate at lower pressure, which can reduce both investment and operating costs. Also, the second stage can use sweet gas (no H_2S) rather than sour, allowing the refiner to employ a wider range of catalysts.

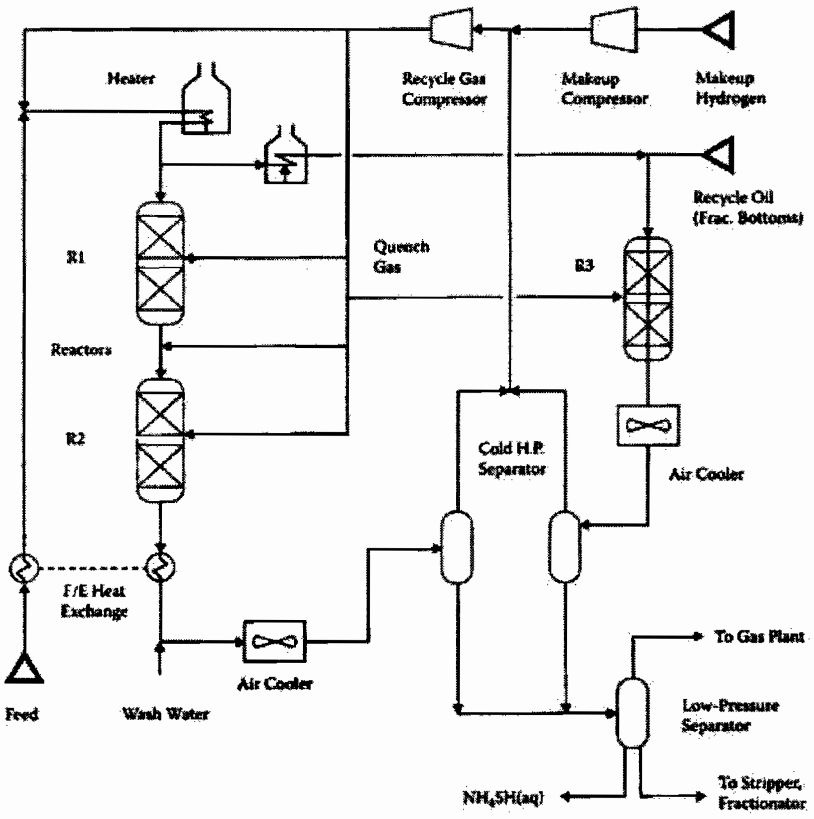

FIGURE 10.6 Two-stage hydrocracker: common recycle gas system.

Early fixed-bed hydrocrackers used a separate hydrotreat flow scheme, which resembles a two-stage design with nothing but hydrotreating catalyst in the first stage. This flow scheme is discussed in further detail by Gruia (2006).

10.6.2 SLURRY-PHASE HYDROCRACKING

Slurry-phase hydrocracking converts residue in the presence of hydrogen under severe process conditions — more than 840°F (450°C) and 2000 to 3000 psig (13,891 to 20,786 kPa). To prevent excessive coking, finely powdered additives made from carbon or iron salts are added to the liquid feed. Inside the reactor, the liquid/powder mixture behaves as a single phase due to the small size of the additive particles. Residue conversion can exceed 90%, and the quality of converted products is fairly good.

Unfortunately, the quality of the unconverted pitch is poor, so poor that it cannot be used as a fuel unless it is blended with something else — coal or heavy fuel oil. Even then, its high metals and sulfur content can create problems.

At the 5000 barrels/day CANMET demonstration plant in Canada, the pitch is sent to a cement kiln for use as a clinker. Other slurry-phase processes include COMBIcracking (developed by Veba Oel), Aurabon (UOP), and HDH Cracking (Intevep). Although several slurry-phase demonstration plants have been built, the pitch-disposal problem has kept it from gaining industry-wide acceptance.

10.6.3 EBULLATING-BED UNITS

In contrast to fixed-bed VGO hydrocrackers, ebullating-bed units can (and do) process residual oils. In ebullating-bed units (Figure 10.7), hydrogen-rich recycle

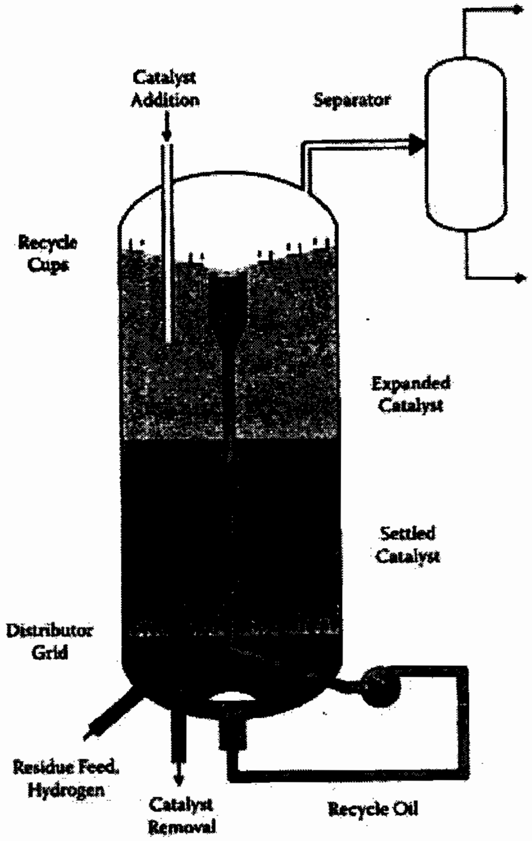

FIGURE 10.7 Ebullating-bed hydrocracking reactor.

gas is bubbled up through a mixture of oil and catalyst particles. This provides three-phase turbulent mixing, which is needed to ensure a uniform temperature distribution. At the top of the reactor, catalyst is disengaged from the process fluids, which are separated in downstream flash drums. Most of the catalyst is returned to the reactor. Some is withdrawn and replaced with fresh catalyst. The two major ebullating-bed processes are H-Oil, which is offered for license by Axens (IFP), and LC-Fining, which is offered by Chevron Lummus Global. Their main advantages are:

- High conversion of atmospheric residue, up to 90 vol%.
- Better product quality than many other residue conversion processes, especially delayed coking.
- Long run length. Catalyst life does not limit these units. Fresh catalyst is added and spent catalyst is removed continuously. Therefore, barring any mechanical problems, the units can run for a much longer time than fixed-bed residue units.

10.7 PROCESS CONDITIONS

For fixed-bed hydroprocessing units, the process conditions — pressure, temperature, space velocity, and catalyst — are determined by feed quality and process objectives. Table 10.9 shows typical process conditions for the hydrotreating of different feeds in fixed-bed hydrotreating units. The values shown are approximate.

TABLE 10.9
Typical Process Conditions for Hydrotreating Different Petroleum Fractions

	Naphtha	Kerosene	Diesel	VGO	Residue
WART[a]					
°F	530	550	575–600	680–700	700–725
°C	277	288	300–315	360–370	370–385
H_2 pressure[b]					
psig	250–450	250–600	600–800	800–2000	>2000
kPa	1825–3204	1825–4238	4238–5617	5617–13,891	>13,891
LHSV	5	4	2–3	0.8–1.5	0.5
H_2/oil ratio[c]					
scf/bbl	350	450	800	1200	>3000
M^3/m^3	60	80	140	210	>525

[a] Approximate weighted average reactor temperature at start of run.
[b] Approximate hydrogen partial pressure at the high-pressure separator.
[c] Approximate hydrogen-to-oil ratio at the first reactor inlet.

The H_2/oil ratios are for units in which off-gas from the high-pressure separator is recycled. For once-through naphtha hydrotreaters associated with catalytic reformers, the H_2/oil ratio can be much higher than 350 scf/bbl (60 m^3/m^3). For units that treat olefinic cracked stocks from FCC or coking units, H_2/oil ratios are higher to control the extra heat released by olefin saturation.

10.8 YIELDS AND PRODUCT PROPERTIES

Table 10.10 illustrates the yield flexibility of recycle hydrocracking. The ability to swing in just a day or two from 90 vol% full-range naphtha to >75 vol% full-range diesel provides unparalleled capability to respond to short-term changes in market conditions — if the refinery has sufficient blending, storage, and distribution capacity. To shift the product slate, operators adjust reactor temperatures and change cut points in the fractionation section.

For all process units, product specifications are set to meet refinery-wide objectives. For example, if a refinery wants to produce diesel fuel containing <15 wppm sulfur, and if its hydrocracker makes 40,000 barrels/day of sulfur-free middle distillate, the product sulfur specification for its 20,000 barrels/day distillate hydrotreater (DHT) could be as high as 45 wppm — if a blend of the two streams satisfies the requirements of ASTM D975, which is the standard specification for heavy-duty diesel fuel in the United States. In practice, the DHT sulfur target would be lower than 45 wppm to cushion the refinery against upsets and measurement error. For a diesel fuel containing 10 wppm sulfur, the analytical reproducibility for ASTM D5453 is ±1.8 wppm. For a diesel containing 50 wppm sulfur, the reproducibility is ±8.1 wppm. ASTM D5453 is an x-ray fluorescence method for measuring sulfur in distillate fuels, including ultra-low-sulfur diesel.

10.9 OVERVIEW OF ECONOMICS

10.9.1 COSTS

Throughput, operating pressure, and process configuration — once-through or recycle of unconverted oil — are the major factors affecting construction costs for hydroprocessing units, which range from $1000 to $4000 per daily barrel. On this basis, a fully installed 25,000 barrels/day hydrocracker can cost between U.S.$40 million and U.S.$100 million. These estimates do not include costs for a hydrogen plant and off-site utilities.

For hydrotreaters, operating costs are roughly U.S.$1.7 per barrel. The cost of producing and compressing hydrogen accounts for 60 to 70% of this. For high-conversion hydrocrackers, operating costs are roughly U.S.$4.0 to U.S.$4.5 per barrel, of which 75 to 80% is due to hydrogen.

10.9.2 BENEFITS

Many hydrotreaters are stay-in-business investments, so it is difficult to quantify their upgrade value, which is the value of products minus costs — labor, materials

TABLE 10.10
Feed and Product Properties for a Flexible Single-Stage Hydrocracker

Feedstock Type	**Straight-Run Vacuum Gas Oil**		
Boiling range, °C	340–550		
Boiling range, °F	644–1022		
API gravity	22.0		
Specific gravity	0.9218		
Nitrogen, wppm	950		
Sulfur, wt%	2.5		
Product Objective	Naphtha	Jet	Diesel
Weighted average reactor temperature, °C	Base	–6	–12
Weighted average reactor temperature, °F	Base	–11	–22
Yields, vol% Fresh Feed			
C_4	11	8	7
C_5, 82°C (C_5, 180°F)	25	18	16
82°C+ (180°F+) Naphtha	90	29	21
Jet A-1 or diesel	—	69	77
Total C_4+	126	124	121
Chemical H_2 Consumption			
Nm^3/m^3	345	315	292
Scf/bbl	2050	1870	1730
Product Qualities			
C_5, 82°C			
RONC	79	79	80
Heavy Naphtha			
P/N/A	45/50/5	44/52/4	—
RONC	41	63	67
Endpoint, °C (°F)	216 (421)	121 (250)	118 (244)
Jet A-1			
Flash point, °C (°F)	—	38 (100)	—
Freeze point, °C (°F)	—	–48 (–54)	—
Smoke point, mm	—	34	—
FIA aromatics, vol%	—	7	—
Endpoint, °C (°F)	—	282 (540)	—
Diesel			
Cloud point, °C (°F)	—	—	–15 (5)
API gravity	—	—	44
Cetane number	—	—	55
Flash point, °C (°F)	—	—	52 (126)
Endpoint, °C (°F)	—	—	349 (660)

(liquid feed, hydrogen, catalysts, and chemicals), utilities, maintenance, and investment amortization. In some plants, the refinery planning linear program (LP) assigns equal value to treated and untreated naphtha, and even to treated and untreated distillates. This reflects the underlying assumption that the increase in value across a hydrotreater is equal to the cost of running the unit, that is, the upgrade value is zero. In other LPs, the naphtha hydrotreater (NHT) that pretreats catalytic reformer feed is lumped in with the reformer. Certainly, if a key naphtha or distillate hydrotreater shuts down, the refinery may have to run at a reduced rate, but that can be said of most units.

For an FCC feed pretreater, the upgrade value can be more than U.S.$3 per barrel if the calculation includes its positive impact on FCC yields. Usually, benefits to the FCC are greater than the value of conversion and volume swell in the hydrotreater itself. Typically, the upgrade value for a high-conversion VGO hydrocracker is U.S.$3 to U.S.$4 per barrel.

With hydroprocessing units, most refiners try to maximize feed rate while (1) meeting other process objectives and (2) maintaining a high on-stream factor. Some try to maximize conversion, while others just want to hit a key process target at minimum cost.

10.9.3 CATALYST CYCLE LIFE

For fixed-bed units, catalyst cycle life dominates economics. Catalysts can not be changed if the units are operating, so shorter catalyst cycles mean decreased production. For a typical 25,000 barrels/day unit, 1 day of lost production can cost U.S.$100,000.

Here are some of the many economic trade-offs that must be considered when setting hydrocracker process targets:

- Higher feed rates and higher conversion are desirable economically, but they increase consumption of hydrogen and decrease catalyst cycle life.
- In units that can recycle fractionator bottoms, higher recycle oil rates can increase selectivity, but they may impose limits on fresh feed rate.
- For many recycle units, switching to once-through (zero recycle) operation is attractive economically if the unconverted oil (that is, the fractionator bottoms) goes to an FCC, olefins plant, or lube plant for further upgrading. Conversion goes down in the hydrocracker, but it may be possible to increase fresh feed rates without decreasing catalyst cycle life, and operating costs may go down due to decreased hydrogen consumption.

10.10 HYDROCRACKER–FLUID CATALYTIC CRACKER (FCC) COMPARISON

In a petroleum refinery, heavy molecules with low hydrogen-to-carbon ratios (H/C) are converted into light molecules with higher H/C ratios. The FCC process

TABLE 10.11
Comparison of Hydrocracking with FCC

	FCC	Hydrocracking
Operating pressure	Low	High, 1500–2800 psi
Operating temperature	High, 900–1000°F	Moderate, 600–780°F
Construction costs	Moderate	High
Volume swell	112–118 vol%	115–140 vol%
	Includes fuel gas FOEB	Fresh feed basis
Product olefins	High	Nil
Light naphtha octane (RONC)	>100	78–81
Heavy naphtha octane (RONC)	95–100	40–64
Distillate cetane index	Low	56–60
Distillate sulfur content	Moderate to high	Very low
Bottoms' sulfur content	Moderate to high	Very low

Note: FOEB = fuel oil equivalent barrels; RONC = research octane number clear (without tetraethyl lead).

increases H/C by rejecting carbon, while hydrocracking increases H/C by adding hydrogen. Consequently, FCC and hydrocracking have marked differences in operating conditions, volume swell, product yields, and product properties. Table 10.11 summarizes some of these differences.

10.11 OPERATIONAL ISSUES

Hydroprocessing — especially hydrocracking — is exothermic. Effective control of produced heat is the primary concern of designers, owners, and operators of hydrocracking units. In modern units, a high flux of recycle gas provides a sink for process heat. It also promotes plug flow and the transport of heat through the reactors. Most licensors recommend that the ratio of recycle gas to makeup gas should exceed 4:1.

During design, limits on temperature rise ($T_{rise} = T_{out} - T_{in}$) set the size of catalyst beds and determine the number and location of quench zones. During operation, when feeds (and maybe catalysts) are different, the T_{rise} is also different — sometimes dangerously so. A sudden spike in T_{rise} can lead to a temperature runaway or temperature excursion. These are dangerous. The rates of cracking reactions increase exponentially with temperature — the hotter they get, the faster they get hot. In a few cases, temperature runaways have melted holes in the stainless steel walls of hydrocracking reactors. This is remarkable, because the walls were more than 8 in. (20 cm) thick.

The best way to stop a temperature excursion is to depressure the unit by venting recycle gas through a special valve at the CHPS. This decelerates all hydrocracking reactions by rapidly reducing H_2 partial pressure in the reactors. Depressuring can also lead to catalyst maldistribution, decreased catalyst activity, or increased pressure drop. For these reasons, operators are extremely careful when restarting a unit after a temperature excursion.

Due to the presence of hydrogen, leaks in hydroprocessing units often cause fires. Such fires can be devastating, if not deadly. The replacement of a reactor and the reconstruction of other equipment damaged by the accident can take 12 months. The cost of lost production can exceed U.S.$50 million.

Safety concerns are responsible for several operating constraints, such as:

- An upper limit on temperature in the reactors. This and other temperature constraints prevent damage to the reactor.
- Upper limits on the T_{rise} in each bed and each reactor, and upper limits on the rate at which T_{rise} changes. These are designed to decrease the likelihood of temperature excursions.
- An upper limit on the velocity of fluid flow through elbows in high-pressure piping. This constraint emerged after erosion–corrosion cut a hole in a high-pressure pipe in a hydrocracker, causing a major accident.
- A lower limit on reserve quench gas — usually 15% of the total flow of recycle gas. Reserve quench provides a way to react quickly to nonemergency changes in T_{rise}.
- A lower limit on wash water injection. This ensures the near-total removal of ammonia from the system.

10.12 LICENSORS

Leading licensors of hydroprocessing technology are listed in Table 10.12.

Many engineering contractors will gladly build unlicensed hydrotreaters. However, for hydrocrackers and special-application hydrotreaters, especially those designed to meet clean-fuel specifications, refiners almost always select licensed technology from an experienced vendor willing to offer guarantees.

10.13 CONCLUSION

Advances in hydroprocessing are driven by competitive forces and clean-fuel regulations. These advances include improved catalysts, better reactor design, advanced process control, and online optimization. As clean-fuel regulations migrate from North America and the EU into the rest of the world, and as globalization of the oil industry continues apace, the need will continue for new (and better) hydroprocessing units. Hopefully, within a few years, this chapter will be obsolete and we will have to write an update.

TABLE 10.12
Leading Licensors of Hydroprocessing Technology

Company	Process Name	Description
Axens (IFP)	Prime-G	Gasoline desulfurization
	IFP hydrotreating	Naphtha, distillate, VGO hydrotreating
	IFP Hydrocracking	High-conversion fixed-bed hydrocracking
	T-Star	Ebullating-bed hydrotreating
	H-Oil	Ebullating-bed hydrocracking
CDTECH	CDHydro	Hydrotreating with catalytic distillation
	CDHDS	
Chevron Lummus	Isocracking	High-conversion hydrocracking
	RDS	Atmospheric residue hydrotreating
	VRDS	Vacuum residue hydrotreating
	OCR	On-stream catalyst replacement
	Isodewaxing	Catalytic dewaxing
	LC-Fining	Ebullating-bed hydrocracking
Criterion/ABB/Shell Global	SynSat	Distillate hydrotreating; aromatics saturation
	Deep gasoil HDS	Hydrotreating to make ultra-low-sulfur diesel
ExxonMobil	SCANfining	Hydrotreating to make low-sulfur gasoline
	OCTGAIN	Hydrotreating to make low-sulfur gasoline
	ULSD-Fining	Hydrotreating to make ultra-low-sulfur diesel
	MAXSAT	Saturation of aromatics in distillate streams
	LCO-fining	LCO hydrotreating
	GO-fining	FCC feed pretreating
	RESIDfining	Residue hydrotreating
	MIDW	Lube isomerization/dewaxing
Haldor Topsoe		Naphtha, distillate, VGO hydrotreating
KBR	MAK hydrotreating	Distillate and VGO hydrotreating
	MAK hydrocracking	Mild hydrocracking; FCC feed pretreatment
UOP	ISAL	Gasoline desulfurization
	Unifining	Naphtha hydrotreating
	Unionfining	Distillate, VGO, residue hydrotreating
	Unicracking	High-conversion VGO hydrocracking

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Attachment 13

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Combustion Emissions from Refining Lower Quality Oil: What Is the Global Warming Potential?

GREG KARRAS*

Communities for a Better Environment (CBE), 1904 Franklin Street, Suite 600, Oakland, California 94612, United States

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The greenhouse gas emission intensity of refining lower quality petroleum was estimated from fuel combustion for energy used by operating plants to process crude oils of varying quality. Refinery crude feed, processing, yield, and fuel data from four regions accounting for 97% of U.S. refining capacity from 1999 to 2008 were compared among regions and years for effects on processing and energy consumption predicted by the processing characteristics of heavier, higher sulfur oils. Crude feed density and sulfur content could predict 94% of processing intensity, 90% of energy intensity, and 85% of carbon dioxide emission intensity differences among regions and years and drove a 39% increase in emissions across regions and years. Fuel combustion energy for processing increased by approximately 61 MJ/m^3 crude feed for each 1 kg/m^3 sulfur and 44 MJ/m^3 for each 1 kg/m^3 density of crude refined. Differences in products, capacity utilized, and fuels burned were not confounding factors. Fuel combustion increments observed predict that a switch to heavy oil and tar sands could double or triple refinery emissions and add 1.6–3.7 gigatons of carbon dioxide to the atmosphere annually from fuel combustion to process the oil.

Introduction

Replacing limited conventional crude oil (*1*) with heavy oil and natural bitumen (tar sands) resources could have substantial energy and environmental costs (*2*). Physical and chemical properties of the lower quality, heavier, more contaminated oils predict the combustion of more fuel for the energy necessary to convert them into product slates dominated by light hydrocarbon liquids (*3–8*). Preliminary estimates from fuel cycle analyses suggest that a switch to heavy oil and tar sands could increase the greenhouse gas emission intensity of petroleum energy by as much as 17–40%, with oil extraction and processing rather than tailpipe emissions accounting for the increment (*3, 4*). This raises the possibility that a switch to these oils might impede or foreclose the total reduction in emissions from all sources that is needed to avoid severe climate disruption. Accurate prediction of emissions from substitutes for conventional petroleum is therefore critical for climate protection. However, estimates of the emissions from processing lower quality oils have not been verified by observations from operating refineries.

Crude oils are extremely complex, widely ranging mixtures of hydrocarbons and organic compounds of heteroatoms and metals (*2, 7*). Refiners use many distinct yet interconnected processes to separate crude into multiple streams, convert the heavier streams into lighter products, remove contaminants, improve product quality, and make multiple different products in varying amounts from crude of varying quality (*5–11*). Factors that affect emissions from refinery process energy consumption include crude feed quality, product slates, process capacity utilization, fuels burned for process energy, and, in some cases, preprocessing of refinery feeds near oil extraction sites. Estimates that construct process-by-process allocations of emissions among these factors have not been verified by observations from operating refineries in part because publicly reported data are limited for refinery-specific crude feeds and unavailable for process-level material and energy inputs and outputs (*4–6*). Research reported here distinguishes effects of crude feed quality on processing from those of the other factors using refinery-level data from multiple operating plants to estimate and predict the process energy consumption and resultant fuel combustion emissions from refining lower quality oil.

Experimental Section

Refinery crude feed volume, density, and sulfur content, process capacity, capacity utilization, yield, and fuels were reported annually for each U.S. Petroleum Administration Defense District from 1999 to 2008 (*9, 10*). See the Supporting Information for this data (Table S1, Supporting Information). Districts 1 (East Coast-Appalachia), 2 (Midwest), 3 (Gulf Coast and vicinity), and 5 (West Coast, AK, and HI) each refined diverse crude feeds (19–41 source countries) at multiple facilities. Smaller, landlocked District 4 (Rocky Mountain states) refined nondiverse crude feeds (2–3 source countries).

At concentrations 4–8 times those of nitrogen and 160–500 times those of nickel and vanadium, sulfur is the major process catalyst poison in crude by mass (*2, 11*). In addition, for diverse blends of whole crude oils from many locations and geologic formations, distillation yield, and asphaltic, nitrogen, nickel, and vanadium content are roughly correlated with density and sulfur (*2, 7*). Variability in the effects of unreported crude feed characteristics on processing is thus constrained by the density and sulfur content of well-mixed crude feeds. Mixing analysis suggested that density and sulfur are reasonably reliable predictors of natural variability in unreported characteristics for annual crude feeds processed in Districts 1, 2, 3 and 5 but could not exclude the potential for unpredicted effects in processing the poorly mixed District 4 feed (Table S2, Supporting Information). The District 4 feed also was proportionately higher in synthetic crude oil (SCO) than those of other districts (Table S3, Supporting Information), and variant hydrogen production that was not predicted by crude feed density was found in District 4 (Table S4, Supporting Information). SCO may increase refinery hydroprocessing requirements (*12, 13*). High hydrogen capacity coincided with SCO refining in Districts 2 and 4 during 1999–2008, but the effect on refinery energy was minimal in District 2, while it was significant and more variable in District 4; other anomalies in the District 4 feed might cause this effect (Tables S2 and S4, Supporting Information). For these reasons, District 4 data were excluded from analysis of refinery observations and used only in estimates including upgrading for SCO. Districts 1, 2, 3, and 5 accounted collectively for 97% of U.S. refining capacity, 1999–2008. Analysis compared the reported data among these districts and years for interactions of the variables defined below.

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* Corresponding author e-mail: gkatcbe@gmail.com.

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Oil quality *(OQ)* was defined as the density (*d*) and sulfur content (*S*) of crude feeds in mass per cubic meter (1 m^3, 6.29 barrels oil; 264 gallons). The density of crude oils is proportional to the fraction of higher molecular weight, higher boiling point, larger hydrocarbon compounds in the oils that are distilled in a vacuum, then cleaved (cracked) into fuel-size compounds to make light hydrocarbon fuels. The larger hydrocarbons have lower hydrogen/carbon ratios that require hydrogen addition to improve product quality and higher concentrations of sulfur and other catalyst poisons that are freed by cracking and bonded with hydrogen to remove them from the oil and protect process catalysts (*2, 11*). This hydrocracking and hydrotreating of gas oil and residua uses several times more hydrogen than does hydrotreating of lighter streams such as naphtha (*11*). These processing characteristics require increased capacity for vacuum distillation, cracking, and hydroprocessing of gas oil and residua in refineries designed to make light liquid products from heavier, higher sulfur crude oils (*4, 8, 14*).

Crude processing intensity (*PI*) was thus defined as the ratio by volume of vacuum distillation capacity, conversion capacity (catalytic, thermal, and hydrocracking), and crude stream (gas oil and residua) hydrotreating capacity to atmospheric crude distillation capacity. These processes account for the primary processing acting on the crude and "reduced crude" that *Speight* distinguishes from secondary processes acting on product streams such as gasoline, naphtha, and distillate oils (*7*). PI measures the increasing portion of the crude input fed to these processes that is predicted by worsening *OQ* (increasing *d*, *S*, or both) and indicates the additional energy needed for heat, pressure, and reactants such as hydrogen to process those increasing feed volumes. It also defines an operational distinction between "crude stream" processing that acts on crude, gas oils, and residua and the subsequent "product stream" processing that acts on the unfinished products from crude stream processing. This distinction was useful in the absence of reported data for more detailed process-level analyses of material and energy flows. *PI* was analyzed with refinery-level crude feed, fuel, capacity utilization, and product yield data to verify the refinery process energy predicted by *OQ*.

Energy intensity (*EI*) was defined as total refinery process energy consumed per volume crude feed, based on reported fuels consumed (Table S1, Supporting Information). Purchased fuels consumed by refiners, such as electric power from the transmission grid, were included in *EI*. Energy used by hydrogen production plants was estimated based on 90% of production capacity and data for new natural gas-fed steam methane reforming facilities (*10, 15*, Table S1, Supporting Information). *EI* integrates all factors in refineries that consume fuel energy, allowing analysis of *EI* with *OQ* and processing to account for refinery capacity utilized and yield.

Effects of variable product slates on refinery energy consumption were distinguished from those of *OQ* in five ways. First, product slate effects on the relationships observed among crude feed quality, crude stream processing, and energy were estimated directly. This was done by including the products ratio, defined as the volume of gasoline, kerosene, distillate, and naphtha divided by that of other refinery products, as an explanatory variable in comparisons of *OQ*, *PI*, and *EI*. Second, the products ratio, combined yield of gasoline and distillate, and combined yield of petroleum coke and fuel gas were analyzed with *EI* and *OQ*. This quantified changes in refinery energy with yield and changes in yield with crude feed quality for key conversion products and byproducts. Third, energy use was analyzed with product stream process capacities to estimate changes in *EI* that could be explained by changes in product processing rates. Fourth, effects of product stream processing on energy for hydrogen were compared with those of crude stream processing by analyzing hydrogen production capacity with product hydrotreating capacity, hydrocracking capacity, and *OQ*. Finally, estimated total energy for processing product slates (Eproducts) was analyzed with *OQ*. Eproducts was estimated based on product-specific factors developed by Wang et al. (*6*) and yield data (Tables S1 and S5, Supporting Information). Refinery capacity utilization was included as an explanatory variable in all comparisons.

Analysis was by partial least squares regression (PLS, XLSTAT 2009). PLS was used based on the expectation that explanatory (*x*) variables may be correlated, the primary interest in prediction of *y* (e.g., *EI*) and a secondary interest in the weights of *x* variables (e.g., *S* and *d*) in predicting *y*. Distributions of PLS residuals appeared normal (Shapiro-Wilk; Anderson-Darling; Lilliefors; Jarque-Bera tests, α 0.05).

Synthetic Crude Oil (SCO). Coking- and hydrocracking-based upgrading of bitumen in Western Canada uses energy to yield SCO that has poor gas oil and distillate qualities but lower density and sulfur than the bitumen (*12, 13*). Refinery crude feeds and energy consumption do not reflect the original bitumen quality for this SCO or the energy used in its upgrading. SCO comprised appreciable fractions of annual crude feeds in Districts 2 (2−8%) and 4 (2−12%), based on limited estimates that may exclude SCO in some blended oil streams (Table S3, Supporting Information). Process modeling data for energy consumed and density and sulfur lost in coking- and hydrocracking-based upgrading (*16*) were applied to the estimated SCO volume in refinery feeds (Table S3, Supporting Information). Districts and years were compared for total processing (upgrading and refining) energy estimated and that predicted by including estimated original oil quality (*d, S*) in the prediction mode of the PLS model based on refinery observations (Table S6, Supporting Information).

Emissions. Emissions were assessed for carbon dioxide (CO_2), the predominant greenhouse gas emitted by refineries (Table S7, Supporting Information). Direct measurements for all emission vents were not reported. Observed fuel consumption and fuel-specific emission factors developed by the U.S. Energy Information Administration (*17, 18*) were used to estimate "observed" emissions, and estimation details were documented (Table S1, Supporting Information). Fuel energy consumed ranged more widely among districts and years than the emission intensity of the fuel mix. Emissions predicted by *OQ* were based on *EI* predicted by *OQ* results from PLS and the emission intensity of the fuel mix. Observed and predicted emissions were compared among districts and years by PLS. Emissions estimates by government agencies (*5, 19−21*) that could be matched to data for *OQ* were superimposed on this comparison by including their *OQ* and predicted *EI* values in the prediction mode of the PLS models for the districts data (Tables S8 and S9, Supporting Information).

For heavy oil and natural bitumen, *OQ* data reported by the U.S. Geological Survey (*2*) and the average (1999−2008) U.S. refinery capacity utilization and products ratio were used in the prediction mode of the PLS model for observed *EI* versus *OQ* to predict *EI* (Table S8, Supporting Information). Predicted emissions from heavy oil and natural bitumen were derived from the products of these *EI* predictions (95% confidence for observations) and the emission intensity of the average (1999−2008) U.S. refinery fuel mix.

Results

Figure 1 shows results from comparisons of *OQ*, *PI*, and *EI* among districts and years from 1999 to 2008. Observed *OQ* ranges by 7.85 kg/m^3 crude feed (kg/m^3) for *S* and 37.6 kg/m^3 for *d*. Observed *PI* ranges by 0.42, or 42% of atmospheric crude distillation capacity. Observed *EI* ranges by 1.89 GJ/m^3 crude feed. *PI* is strongly and positively associated with

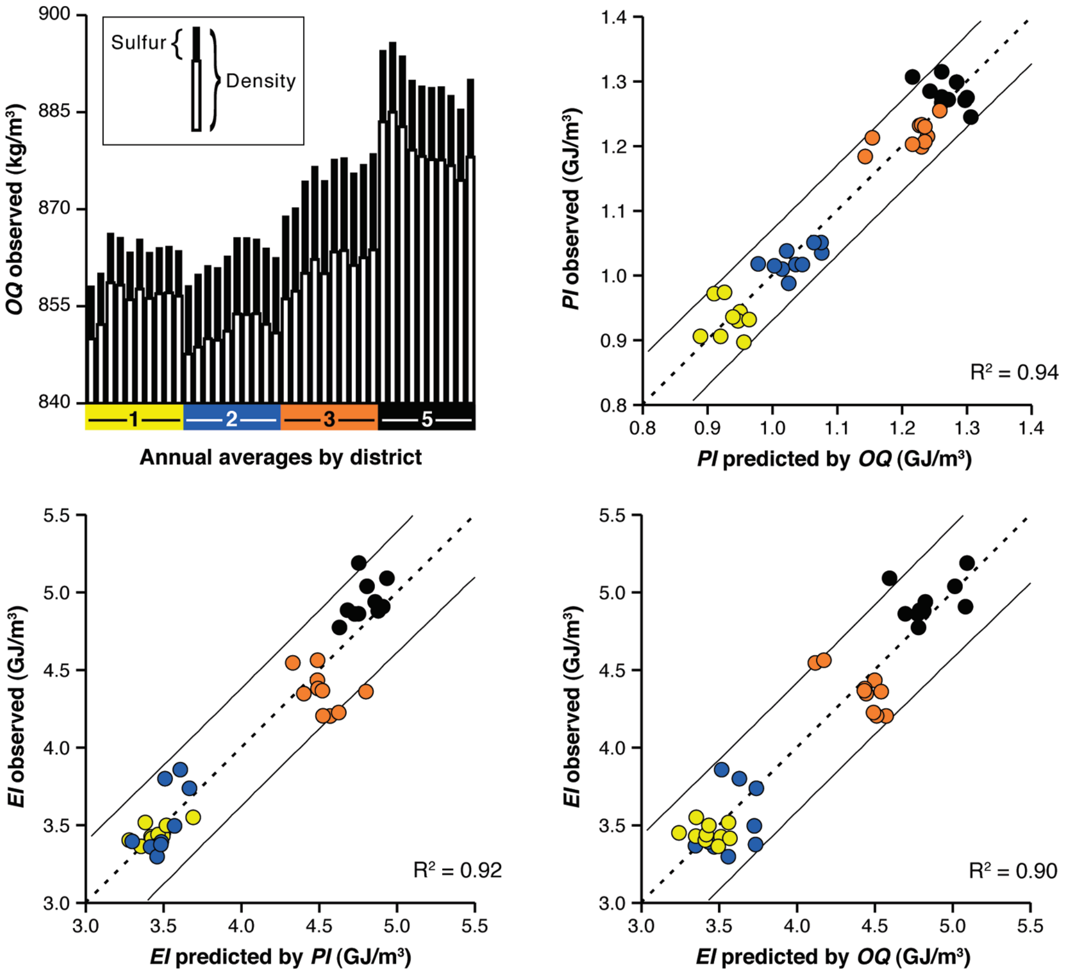

FIGURE 1. Increasing crude processing intensity and energy intensity with worsening oil quality. *OQ:* Crude feed oil quality. *PI:* Crude processing intensity. *EI:* Refinery energy intensity. Observations are annual weighted averages for districts 1 (yellow), 2 (blue), 3 (orange), and 5 (black) in 1999–2008. Diagonal lines bound the 95% confidence of prediction for observations.

worsening *OQ* (increasing *d*, *S*, or both). *EI* is strongly and positively associated with worsening *OQ* and increasing *PI*. *EI* increases by approximately 44 MJ/m^3 for each 1 kg/m^3 *d* and 61 MJ/m^3 for each 1 kg/m^3 *S* based on the PLS regression analysis for *EI* versus *OQ*. The equation of the model (*EI* vs *OQ*) can be expressed as

$$EI = 0.044d + 0.061S + 0.010(\text{Capacity utilized}) - 0.159(\text{Products ratio}) - 35.092 \quad (1)$$

where *EI* is the central prediction in GJ/m^3, *d* is in kg/m^3, *S* is in kg/m^3, capacity utilized is in percent, products ratio is expressed as a quotient, and the last term is the coefficient for the intercept.

Table 1 shows additional results from analysis of refinery observations. *PI* increases strongly with *d* and *S* (95% confidence for observations). *EI* increases strongly with *d* and *S* and with vacuum distillation, conversion, and crude stream hydrotreating capacities. Hydrogen production capacity increases strongly with *d* and hydrocracking capacity. Sulfur recovery capacity increases strongly with *S*. These observations describe increasing portions of crude feeds processed by crude stream capacity and resultant effects on total refinery energy consumption as crude density and sulfur content increase.

In contrast to crude stream processing, except for cracking byproducts and two processes that treat them, product slate indicators are not significant or decrease with increasing *OQ* and *EI*. The products ratio is not significant in the strong relationships among *EI*, *PI*, and *OQ*, perhaps in part because light liquids yield is less variable than *S* or *EI* among these districts and years. However, the ratio of light liquids to other products decreases with increasing *d* (products ratio vs *OQ)* and *EI* (*EI* vs products processing), and yield shifts, from gasoline and distillate to coke and fuel gas, as *OQ* worsens and *EI* increases.

Products processing reflects this shift from light liquids to cracking byproducts. Product stream hydrotreating, reforming, asphalt, aromatics, and polymerization/dimerization capacities decrease as *EI* increases. Those five processes account for 83–90% of total product stream processing capacity among districts (Table S1, Supporting Information). Among products processes, only alkylation and isomerization (7–13% of products capacity), which receive light streams from conversion processes, are positively associated with *EI*. Product hydrotreating cannot explain the observed increase in hydrogen production with increasing *d*. Estimated refinery energy use for products processing (Eproducts) decreases with increasing *d*. These results appear to measure the decreasing fraction of crude inputs converted to light liquid product streams and increasing creation of cracking byproducts such as coke and fuel gas that result from incomplete conversion as crude feed density and sulfur increase.

A weak inverse association of hydrogen production with product hydrotreating capacity (Table 1) results from a strong increase in H_2 capacity with *d* and hydrocracking, a steady decrease in the hydrotreating/hydrocracking ratio with increasing H_2 capacity, and lower hydrotreating at high

TABLE 1. Results from Refinery Crude Feed Quality, Processing, Energy, Yield, and Emission Comparisons[a]

effects of crude feed oil quality (*OQ*)

		standardized coefficients of *x* variables (coeff)			
y vs *x*	R^2	density	sulfur	cap. utilized	products ratio
process intensity *(PI)* vs *OQ*	0.94	**0.73**	**0.42**	**0.09**	−0.02
energy intensity *(EI)* vs *OQ*	0.90	**0.80**	**0.23**	0.05	−0.10
hydrogen production vs *OQ*	0.91	**1.09**	−0.01	0.05	**0.35**
sulfur recovery vs *OQ*	0.94	−0.01	**0.95**	−0.06	**−0.15**
pet. coke + fuel gas vs *OQ*	0.95	**0.80**	**0.34**	−0.04	
gasoline + distillate vs *OQ*	0.75	**−0.85**	−0.07	−0.04	
products ratio vs *OQ*	0.26	**−0.40**	−0.12	0.17	
Eproducts vs *OQ*	0.74	**−0.61**	0.13	**0.49**	

effects of oil quality (*OQ*) and fuels on CO_2 emissions

		standardized coefficients of *x* variables (coeff)	
y vs *x*	R^2	*EI* predicted by *OQ*	fuel mix emission intensity
observed vs predicted CO_2	0.85	**0.88**	−0.04

effects of processing and products yield

y vs *x*	R^2	coeff.	*y* vs *x*	R^2	coeff.
EI vs *PI*	0.92		*EI* vs yield	0.93	
vacuum distillation		**0.35**	pet. coke + fuel gas		**0.59**
conversion capacity		**0.35**	gasoline + distillate		**−0.42**
csHydrotreating		**0.22**	capacity utilized		−0.01
capacity utilized		**−0.16**	products ratio		−0.02
products ratio		−0.14			
			EI vs psProcessing	0.91	
H_2 production vs hydrocracking	0.97		psHydrotreating		**−0.17**
hydrocracking		**1.02**	reforming		**−0.19**
capacity utilized		−0.06	asphalt		**−0.30**
products ratio		**0.14**	aromatics		**−0.33**
			polym./dimerization		**−0.25**
H_2 production vs product-stream hydrotreating			lubricants		0.04
	0.18		alkylation		**0.30**
psHydrotreating		**−0.33**	isomerization		**0.24**
capacity utilized		−0.09	capacity utilized		−0.06
products ratio		**−0.17**	products ratio		**−0.33**

[a] *R*-squared values and standardized coefficients from PLS regressions on annual data from refining districts 1, 2, 3 and 5, 1999−2008. **Boldface**: significant at 95% confidence. Eproducts: estimated energy use to process a given product slate. Prefix cs (ps): crude stream (product stream) processing.

H_2 capacity among these districts and years (Figure S1, Supporting Information). Refinery capacity utilization was not significant in the effects of *OQ* on *EI* and affected the relationships between *PI* and *OQ* and between *PI* and *EI* only marginally, possibly because capacity utilization varied little among districts and years (Table S1, Supporting Information). Significant capacity utilization results are consistent with marginally increased energy consumption and decreased flexibility to process lower quality crude when refineries run closer to full capacity.

Rough estimates including the energy, *d*, and *S* lost in bitumen upgrading for SCO refined reveal greater effects of total processing for crude feeds refined in Districts 2 and 4 and follow the relationships observed in refining (Figure 2). Estimated total processing energy falls within the prediction based on *OQ* from refinery observations in 43 of 50 cases and exceeds the 95% confidence of prediction by more than 2% only in two cases explained by District 4 hydrogen anomalies discussed above. Oil quality−energy relationships observed in refining can predict those for total processing because upgrading and refining use similar carbon rejection, hydrogen addition, and utility technology.

Emissions calculated from observed fuels consumed are strongly and positively associated with *EI* predicted by *OQ* (Table 1) and range by 39%, from 257 to 358 kg/m^3 crude feed (Figure 3). Observed emissions fall within the 95% confidence of prediction based on *OQ* in 36 of 40 cases and are within 3% of the confidence of prediction in all cases. Despite emission differences among fuels, the fuel mix is not significant in this prediction. The emission intensity of the fuel mix varies much less than *EI* and decreases slightly with decreasing petroleum coke contributions and a shift in cracking processes as *EI*, *d*, and *S* increase (Table S1 and Figure S1, Supporting Information). Refinery emission estimates by government agencies that could be matched to *OQ* differ from each other by as much as 12−30% but fall within 2% of the central prediction based on *OQ* or within 4% of its confidence interval (*5, 19*−*21*, Table S8, Supporting Information). The 2008 San Francisco Bay Area estimate in Figure 3 (360 kg/m^3) is close to estimated 2008 California refinery emissions (354 kg/m^3) (*21*), for which matching *OQ* data were not available. California gasoline and diesel production may account for 56% (197.2 kg) and 22% (78.7 kg) of this 354 kg/m^3, respectively, based on fuel-specific estimates for the average California crude feed (*21*−*23*, Table S8, Supporting Information).

Predictions for heavy oil (957.4 kg/m^3 *d;* 27.8 kg/m^3 *S*) and natural bitumen (1 033.6 kg/m^3 *d;* 45.5 kg/m^3 *S*) (USGS average) (*2*) reflect their low quality compared with crude feeds observed (Figure 1). On the basis of the PLS model for

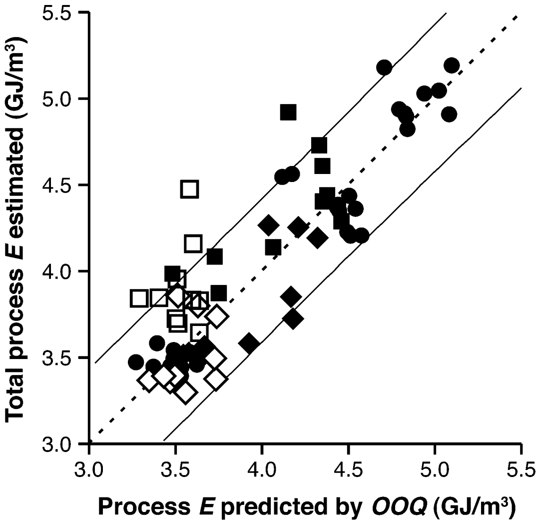

FIGURE 2. Estimated process energy for bitumen upgrading and refining versus that predicted by oil quality (GJ/m^3 crude), 1999–2008. *OOQ:* original oil quality including bitumen quality for synthetic oil inputs. Black diamonds: District 2. Black squares: District 4. Black circles: Districts 1, 3, and 5. White diamonds (squares): District 2 (District 4) refinery energy and oil quality only. Diagonal lines bound the 95% confidence of prediction for refinery observations.

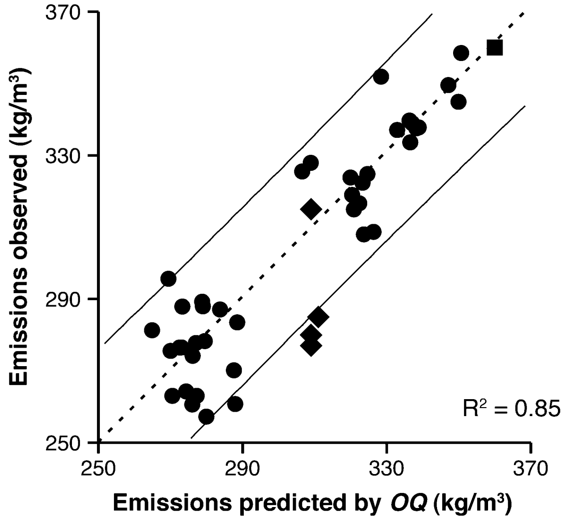

FIGURE 3. Refinery CO_2 emission intensity observed versus predicted by oil quality. *OQ:* Oil quality. Black circles: District 1, 2, 3, or 5 annually, 1999–2008. Black diamonds: United States in 2002, 2005, 2006, 2007. Black square: San Francisco Bay Area in 2008. Diagonal lines bound the 95% confidence of prediction for observations. R^2 value shown is for the comparison among districts and years.

observations from Districts 1, 2, 3, and 5 (*EI* vs *OQ*) and the emission intensity of the U.S. refinery fuel mix (73.8 kg/GJ), processing the range of heavy oil/bitumen blends could use 8.23–14.13 GJ/m^3 fuel (Table S8, Supporting Information) and emit 0.61–1.04 t/m^3 CO_2.

Discussion

Strongly coupled increases in energy and crude stream processing intensities with worsening oil quality (Figure 1) describe energy for carbon rejection, aggressive hydrogen addition, and supporting processes acting on larger portions of heavier, higher sulfur crude feeds to yield light liquid product streams. The creation of cracking reaction byproducts that limits conversion of heavier oils to light liquid product streams is observed in the shift from gasoline and distillate to coke and fuel gas yield as *OQ* worsens and *EI* increases. Observed decreases in light liquids yield and most major product stream processes as *EI* increases are consistent with this rising reliance on incomplete conversion. Differences in product slates cannot explain increasing *EI* as *OQ* worsens because capacities of processes comprising 83–90% of product stream processing capacity decrease as *EI* increases, and estimated energy use for products processing decreases as *OQ* worsens. Hydrogen production increases with crude density and hydrocracking. *EI* drives emissions variability. *OQ* predicts 94% of *PI*, *PI* predicts 92% of *EI*, and *OQ* predicts 90% of *EI* and 85% of emissions variability. These observations from operating plants across the four largest U.S. refining districts over 10 years provide evidence that crude feed density and sulfur content predict processing, energy, and CO_2 emission intensities for large groups of refineries with diverse feeds.

Slight, unexpected decreases in product hydrotreating at high hydrogen production and in fuel mix emission intensity with increasing *d* and *S* can be explained by a coincident shift from hydrotreating and catalytic cracking to hydrocracking with worsening *OQ*. Refiners can substitute hydrocracking for hydrotreating and catalytic cracking to some extent. *OQ*, along with other factors beyond this study scope, may influence those business decisions.

Energy increments predicted by density (44 MJ/kg) and sulfur (61 MJ/kg) in crude feeds (eq 1) compare to energy inputs of 40–70 MJ/kg density (including sulfur) lost from bitumen upgrading for SCO, based on process modeling of coking- and hydrocracking-based upgraders ((*16*), Table S6, Supporting Information). At an energy cost of 16.4 MJ/m^3 (Table S1, Supporting Information), hydrogen for density reduction by hydrocracking could account for 44 MJ/kg, based on the H_2/oil feed ratio of 308 m^3/m^3 Robinson and Dolbear report for 22°API feed and 44°API yield (*11*).

Results help to explain differences among government estimates of refinery emissions (Figure 3) and support the high case fuel cycle emission increments from a switch to heavy and tar sands oils reported for gasoline by Brandt and Farrel (+40%) (*3*) and for diesel by Gerdes and Skone (+17%) (*4*). Predicted emissions from processing heavy oil/natural bitumen blends (0.61–1.04 t/m^3) are 2–3 times the average of observed and estimated emissions in Figure 3 (0.30 t/m^3). Assuming this 0.30 t/m^3 refining average and 2007 world petroleum emissions (11.27 Gt) (*24*) as a baseline, processing heavy oil/bitumen blends at 2009 world refining capacity (5.06×10^9 m^3) (*10*) could increase annual CO_2 emissions by 1.6–3.7 gigatons and total petroleum fuel cycle emissions by 14–33%. Extraction emissions would add to these percentages.

This prediction applies to average CO_2 emissions from large, multiplant refinery groups with diverse, well-mixed crude feeds and appears robust for that application. However, the method used here should be validated for other applications. If it is applied to different circumstances, the potential for significantly different product slates, poorly mixed crude feeds, synthetic crude oil impacts on refining, and effects on fuel mix emission intensity and hydrotreating resulting from choices among carbon rejection and hydrogen addition technologies should be examined.

Several issues suggest future work. Other properties of crude feeds and incremental efficiencies from modernization of equipment and catalyst systems might explain up to 10% of the variability in *EI* observed among U.S. refining districts and years and could be more important for single plants and nondiverse crude feeds. Burning more fuel to refine lower quality oil emits toxic and ozone–precursor combustion products along with CO_2. Pastor et al. estimate that refinery emissions of such "co-pollutants" dominate health risk in nearby communities associated with particulate matter emitted by the largest industrial sources of greenhouse gases in California and identify racial disparities in this risk as important in emission assessment (*25*). Better facility-level *OQ* data could improve local-scale pollutant assessment. Better crude quality predictions could improve energy, and climate protection, forecasts. Assessments of the need, scope, and timing for transition to sustainable energy should account for emissions from lower quality oil.

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Supporting Information Available

Data and details of methods, analyses, and results. This material is available free of charge via the Internet at http://pubs.acs.org.

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Supporting information for the manuscript:

Combustion emissions from refining lower quality oil: What is the global warming potential?

Greg Karras

Communities for a Better Environment (CBE)
1904 Franklin Street, Suite 600
Oakland, CA 94612
(510) 302-0430
Gkatcbe@gmail.com

Supporting information given in the following 49 pages includes tables S1–S9, Figure S1, and references. Each table includes a legend and notes narrative that follows the values given, as does the figure. References identify sources of data cited. This information appears as follows:

Pages S2-14: **Table S1.** Data.

Pages S15-19: **Table S2.** Simplified mixing analysis for potential effects of anomalous oils on crude feeds.

Pages S20-23: **Table S3.** Estimate calculation for Canadian synthetic crude oil (SCO) exports to districts and years.

Pages S24-26: **Table S4.** Evidence for effects of synthetic oil (SCO) on refinery processing during 1999-2008 in District 4.

Page S27: **Table S5.** Efficiency factors for processing refinery products.

Pages S28-34: **Table S6.** Estimate calculation, oil quality and processing *EI* including bitumen upgrading.

Page S35: **Table S7.** Contribution of CO_2 to CO_2e emitted by oil refineries.

Pages S36-40: **Table S8.** PLS inputs for emissions predicted by *OQ,* and comparison emission estimates.

Page S41: **Table S9.** Estimate calculation, San Francisco Bay Area crude feed *OQ* in 2008.

Pages S42-43: **Figure S1.** Some shifts among hydrogen addition and carbon rejection technologies affecting relationships between (A) hydrotreating and hydrogen production, and (B) fuel mix emission intensity and crude feed density, across districts 1, 2, 3 and 5, 1999-2008.

Pages S44-49: **References.**

Table S1

US		Refinery crude inputs[a] ------------------				Refinery process capacity[b] --	-------------	-------------	-----------
District	Year	Feed volume	Density	Sulfur	Source	Atm. dist.	Vacuum dist.	Coking & therm.	Cat. cracking
PADD		(m^3/d•10^4)	(kg/m^3)	(kg/m^3)	countries	(m^3/d•10^4)	(m^3/d•10^4)	(m^3/d•10^4)	(m^3/d•10^4)
1	1999	24.436	858.199	8.239	24	24.365	9.802	1.420	10.476
1	2000	24.754	860.182	8.000	23	24.592	9.721	1.440	10.798
1	2001	23.546	866.344	7.710	19	24.958	9.658	1.409	9.924
1	2002	24.246	865.708	7.445	20	25.222	9.742	1.442	9.899
1	2003	25.184	863.436	7.426	21	25.075	9.975	1.448	9.827
1	2004	24.961	865.443	7.789	21	25.025	9.974	1.448	9.827
1	2005	25.422	863.384	7.166	22	25.263	10.150	1.448	9.970
1	2006	23.626	864.122	7.172	21	25.263	10.149	1.448	9.970
1	2007	23.419	864.333	7.260	24	25.263	10.149	1.448	9.970
1	2008	22.115	863.647	7.082	24	25.263	10.149	1.448	9.970
2	1999	53.626	858.252	10.642	15	57.095	23.272	5.880	19.325
2	2000	54.215	860.025	11.352	16	56.984	23.625	6.098	19.189
2	2001	52.609	861.334	11.370	15	56.427	22.989	6.131	18.822
2	2002	51.162	861.019	11.279	20	55.775	22.592	5.698	18.688
2	2003	51.258	862.804	11.648	16	55.587	22.669	5.612	18.475
2	2004	52.482	865.655	11.859	20	55.528	22.961	5.818	18.268
2	2005	52.688	865.655	11.946	23	56.465	23.689	5.962	18.555
2	2006	52.609	865.443	11.597	20	56.506	23.895	5.948	18.538
2	2007	51.480	864.069	11.838	17	57.873	23.169	6.032	18.010
2	2008	51.575	862.594	11.731	16	57.980	23.466	5.923	18.676
3	1999	111.689	869.004	12.861	33	123.434	57.573	15.493	43.165
3	2000	113.024	870.287	12.967	31	123.436	59.107	16.498	43.434
3	2001	115.600	874.428	14.341	28	123.625	58.157	17.318	44.964
3	2002	112.786	876.703	14.466	33	125.817	57.449	18.717	46.010
3	2003	116.013	874.482	14.429	30	126.876	58.417	19.390	45.821
3	2004	119.145	877.791	14.396	33	128.032	60.442	20.047	46.126
3	2005	114.534	878.009	14.399	36	132.323	59.682	19.897	46.475
3	2006	117.253	875.673	14.361	41	133.383	59.850	20.190	46.632
3	2007	117.682	876.975	14.470	37	134.189	61.054	20.938	46.728
3	2008	111.879	878.663	14.937	36	133.771	61.411	21.046	47.311
5	1999	41.973	894.607	11.093	24	49.484	23.172	9.594	12.630
5	2000	43.086	895.853	10.840	23	49.836	23.152	9.714	12.717
5	2001	44.262	893.759	10.993	26	49.542	23.692	9.757	12.695
5	2002	44.787	889.993	10.858	27	48.422	23.419	9.834	12.768
5	2003	45.661	889.098	10.936	29	48.924	23.597	9.671	12.604
5	2004	45.486	888.874	11.200	28	48.723	23.478	9.695	12.717
5	2005	46.090	888.986	11.379	27	49.104	23.538	9.735	12.762
5	2006	45.693	887.648	10.918	30	49.441	23.930	9.759	13.026
5	2007	44.373	885.537	11.069	30	49.609	24.031	10.003	13.332
5	2008	44.739	890.161	12.106	30	49.730	24.411	9.793	13.170
4	1999	8.029	854.468	11.706	3	8.603	3.464	0.663	2.826
4	2000	8.156	859.346	12.031	2	8.094	3.130	0.663	2.705
4	2001	8.077	859.190	11.084	2	8.802	3.549	0.663	2.768
4	2002	8.363	860.234	12.043	2	9.054	3.616	0.676	2.898
4	2003	8.442	861.229	12.488	2	9.019	3.596	0.687	2.906
4	2004	8.856	862.594	11.645	2	9.296	4.255	0.695	2.950
4	2005	8.935	862.910	11.218	2	9.129	3.502	0.711	2.920
4	2006	8.856	860.496	11.359	2	10.018	3.560	0.711	3.121
4	2007	8.681	862.384	11.728	2	10.016	3.472	0.727	3.151
4	2008	8.585	863.120	12.170	2	9.555	3.305	0.989	2.832
US	1999	239.753	869.111	11.559	--	262.981	117.283	33.050	88.422
US	2000	243.235	870.822	11.669	--	262.942	118.735	34.413	88.844
US	2001	244.077	873.510	12.404	--	263.354	118.046	35.278	89.173
US	2002	241.343	873.888	12.322	--	264.289	116.819	36.368	90.263
US	2003	246.558	872.864	12.482	--	265.481	118.253	36.809	89.633
US	2004	250.930	875.185	12.515	--	266.604	121.109	37.703	89.887
US	2005	247.670	875.077	12.426	--	272.284	120.561	37.753	90.682
US	2006	248.052	873.780	12.320	--	274.612	121.385	38.056	91.286
US	2007	245.635	873.888	12.497	--	277.389	124.553	39.148	91.191
US	2008	238.910	875.023	12.863	--	276.299	122.742	39.198	91.959
Energy factor[c]		--	--	--	--	--	--	--	--
CO_2 emission factor (kg/GJ)[c]			--	--	--	--	--	--	--

Table S1 *continued*

US		Refinery process capacity[b] ---							
District	Year	Hydrocracking	csHydrotreating	psHydrotreating	Reforming	Alkylation	Pol./Dim.	Aromatics	Isomerization
PADD		(m^3/d•10^4)	(m^3/d•10^4)	(m^3/d•10^4)	(m^3/d•10^4)	(m^3/d•10^4)	(m^3/d•10^4)	(m^3/d•10^4)	(m^3/d•10^4)
1	1999	0.666	1.320	12.826	4.567	1.282	0.284	0.861	0.447
1	2000	0.666	1.320	12.460	4.468	1.346	0.284	0.852	0.431
1	2001	0.680	0.715	13.030	4.483	1.281	0.212	0.852	0.526
1	2002	0.602	2.131	12.214	4.528	1.292	0.212	0.852	0.611
1	2003	0.602	1.473	13.779	4.548	1.290	0.212	0.852	0.868
1	2004	0.603	1.477	13.513	4.649	1.290	0.212	0.852	0.878
1	2005	0.603	1.477	13.227	4.681	1.335	0.212	0.852	0.878
1	2006	0.615	0.704	13.993	4.681	1.335	0.212	0.852	0.878
1	2007	0.615	0.704	14.057	4.681	1.335	0.212	0.852	0.878
1	2008	0.615	0.704	14.057	4.681	1.335	0.212	0.852	0.878
2	1999	2.533	7.126	29.912	13.533	3.927	0.208	0.924	2.796
2	2000	2.533	6.099	31.548	13.770	3.959	0.208	0.924	2.764
2	2001	2.386	5.401	32.961	13.435	3.940	0.208	0.924	2.757
2	2002	2.434	7.177	31.440	13.357	3.892	0.136	0.888	2.698
2	2003	2.410	7.355	34.844	13.339	3.835	0.136	0.888	2.863
2	2004	2.191	8.214	35.157	13.247	3.807	0.129	0.876	2.900
2	2005	2.798	8.330	38.089	13.368	3.984	0.128	0.838	2.908
2	2006	3.065	7.937	39.013	13.347	3.991	0.128	0.919	2.940
2	2007	3.701	7.929	38.528	13.460	3.911	0.128	0.657	2.944
2	2008	3.652	8.440	36.890	12.972	3.871	0.130	0.657	2.784
3	1999	11.265	18.638	64.038	27.308	8.602	0.310	4.081	4.523
3	2000	11.513	19.190	65.900	27.730	8.599	0.297	4.202	4.347
3	2001	11.842	15.900	70.483	26.840	8.514	0.297	4.260	4.291
3	2002	12.138	18.588	70.415	27.234	9.806	0.353	4.310	4.551
3	2003	11.359	21.356	76.385	27.088	8.982	0.355	4.072	4.572
3	2004	11.868	22.256	82.382	27.517	10.514	0.378	4.386	4.472
3	2005	11.439	22.191	87.486	26.859	9.144	0.347	4.354	4.345
3	2006	11.447	22.301	90.603	26.857	9.253	0.345	4.239	4.312
3	2007	12.059	24.717	91.006	27.458	8.907	0.646	5.026	3.923
3	2008	11.843	22.910	94.039	27.091	9.179	0.646	5.786	4.284
5	1999	8.089	9.630	21.588	8.763	2.928	0.224	0.040	2.097
5	2000	8.119	8.347	22.626	8.849	4.181	0.234	0.040	2.142
5	2001	8.192	8.614	22.642	8.950	2.933	0.234	0.045	2.142
5	2002	8.192	9.472	21.821	8.833	2.999	0.234	0.045	2.147
5	2003	8.043	8.053	23.957	8.847	3.114	0.235	0.045	2.716
5	2004	8.138	8.151	24.765	8.895	3.119	0.238	0.040	2.659
5	2005	8.259	8.154	24.643	8.946	3.153	0.250	0.036	2.727
5	2006	8.896	7.932	25.742	9.400	3.359	0.280	0.021	2.937
5	2007	9.221	8.274	26.024	9.634	3.362	0.228	0.019	3.258
5	2008	9.124	8.123	26.175	9.473	3.337	0.228	0.019	3.171
4	1999	0.079	0.965	4.702	1.901	0.578	0.073	0.000	0.245
4	2000	0.079	0.744	4.368	1.770	0.525	0.067	0.000	0.245
4	2001	0.278	0.437	5.062	1.905	0.586	0.083	0.000	0.236
4	2002	0.079	0.783	4.784	1.889	0.612	0.083	0.000	0.236
4	2003	0.087	0.783	5.090	1.901	0.622	0.083	0.000	0.238
4	2004	0.254	0.836	4.673	1.772	0.566	0.076	0.000	0.239
4	2005	0.087	0.852	5.123	1.917	0.583	0.097	0.000	0.239
4	2006	0.254	1.092	5.444	1.940	0.596	0.097	0.000	0.258
4	2007	0.280	1.092	5.607	1.953	0.604	0.097	0.000	0.264
4	2008	0.087	1.302	5.720	1.816	0.612	0.083	0.000	0.264
US	1999	22.632	37.678	133.066	56.072	17.317	1.099	5.906	10.108
US	2000	22.910	35.699	136.901	56.585	18.609	1.090	6.017	9.929
US	2001	23.379	31.067	144.178	55.613	17.254	1.034	6.080	9.952
US	2002	23.446	38.151	140.674	55.840	18.602	1.018	6.093	10.243
US	2003	22.502	39.021	154.054	55.723	17.842	1.020	5.856	11.258
US	2004	23.054	40.935	160.490	56.081	19.295	1.034	6.154	11.148
US	2005	23.186	41.005	168.568	55.771	18.200	1.033	6.079	11.097
US	2006	24.278	39.967	174.794	56.226	18.534	1.062	6.032	11.324
US	2007	25.876	42.717	175.222	57.186	18.119	1.311	6.554	11.268
US	2008	25.322	41.479	176.881	56.034	18.333	1.299	7.314	11.381
Energy factor[c]		--	--	--	--	--	--	--	--
CO$_2$ emission factor (kg/GJ)[c]		--	--	--	--	--	--	--	--

Table S1 ***continued***

US		Refinery process capacity[b] ---			-------------- Fuels consumed in refineries[a] -------------				
District	Year	Lubes	Asphalt	Sulfur	H_2 production	Crude oil	LPG	Distillate	Res. fuel oil
PADD		(m^3/d•10^4)	(m^3/d•10^4)	(kg/d•10^6)	(m^3•10^8)	(m^3•10^4)	(m^3•10^4)	(m^3•10^4)	(m^3•10^4)
1	1999	0.368	1.033	0.921	11.783	0.000	2.766	2.035	37.012
1	2000	0.300	0.461	0.921	14.056	0.000	5.008	4.165	38.904
1	2001	0.300	0.461	0.856	11.576	0.000	5.819	8.967	44.675
1	2002	0.299	0.445	1.265	10.232	0.000	4.483	7.631	29.190
1	2003	0.299	0.445	1.301	15.090	0.000	7.854	9.921	28.014
1	2004	0.300	0.445	1.301	15.090	0.000	7.870	7.409	18.013
1	2005	0.300	0.445	1.319	15.297	0.000	11.479	5.819	18.220
1	2006	0.300	0.445	1.319	17.364	0.000	5.231	0.366	14.627
1	2007	0.300	0.445	1.285	13.333	0.000	2.941	0.350	13.132
1	2008	0.300	0.445	1.285	13.333	0.000	0.827	0.461	6.344
2	1999	0.264	3.493	4.436	44.237	0.000	27.123	0.986	43.531
2	2000	0.264	3.763	4.402	44.030	0.000	14.484	0.763	34.166
2	2001	0.264	3.617	4.425	47.751	0.000	13.975	1.288	38.888
2	2002	0.277	3.668	4.672	43.926	0.000	16.439	1.081	29.747
2	2003	0.277	3.727	4.818	40.619	0.000	25.804	0.588	9.380
2	2004	0.277	3.705	4.631	41.032	0.000	17.155	0.588	3.100
2	2005	0.269	3.814	5.140	49.611	0.000	12.385	0.795	2.591
2	2006	0.269	3.897	5.243	77.000	0.000	9.015	0.715	3.275
2	2007	0.269	3.151	4.600	77.931	0.000	13.387	0.747	3.005
2	2008	0.135	3.608	5.200	78.551	0.000	12.783	0.700	3.084
3	1999	1.786	1.930	14.092	146.456	0.159	12.560	1.892	0.191
3	2000	1.801	1.967	15.297	148.833	0.000	13.085	2.798	0.032
3	2001	1.772	1.848	15.266	155.655	0.000	11.018	2.178	0.000
3	2002	1.745	1.904	16.516	160.512	0.000	13.450	1.335	0.000
3	2003	1.793	2.569	17.134	160.512	0.000	17.489	0.700	0.000
3	2004	1.982	2.409	19.395	174.362	0.000	5.898	1.304	0.000
3	2005	2.343	1.936	19.135	172.398	0.000	5.708	1.367	0.064
3	2006	2.351	1.914	19.393	162.269	0.000	4.404	1.765	0.016
3	2007	2.282	1.938	19.013	160.822	0.000	3.307	1.828	0.048
3	2008	2.281	1.938	19.243	164.233	0.000	8.204	1.701	0.048
5	1999	0.437	1.191	4.152	126.301	0.000	18.649	4.086	9.015
5	2000	0.437	1.215	4.152	151.934	0.000	34.150	3.736	11.081
5	2001	0.437	1.078	4.152	149.247	0.000	47.251	4.436	13.609
5	2002	0.342	0.742	4.230	151.004	0.000	19.587	3.307	14.341
5	2003	0.342	0.979	4.331	148.523	0.000	34.484	3.911	11.558
5	2004	0.286	0.920	4.286	147.903	0.000	24.627	3.657	11.495
5	2005	0.286	0.940	4.520	149.557	0.000	36.424	4.022	11.558
5	2006	0.318	0.916	4.911	159.169	0.000	23.339	4.054	12.242
5	2007	0.318	0.940	4.539	162.786	0.000	22.497	3.752	11.813
5	2008	0.318	0.940	5.011	162.786	0.000	23.991	4.642	11.845
4	1999	0.000	0.688	0.381	8.889	0.000	0.636	0.095	3.450
4	2000	0.000	0.671	0.382	8.992	0.000	0.890	0.048	4.786
4	2001	0.000	0.838	0.367	9.612	0.000	0.620	0.111	3.482
4	2002	0.000	0.738	0.368	9.612	0.000	0.700	0.000	3.259
4	2003	0.000	0.738	0.538	9.199	0.000	0.779	0.000	2.671
4	2004	0.000	0.743	0.612	9.509	0.000	1.065	0.016	2.337
4	2005	0.000	0.576	13.577	13.953	0.000	0.382	0.000	2.655
4	2006	0.000	0.796	0.593	13.953	0.000	0.238	0.000	1.924
4	2007	0.000	0.783	0.599	18.191	0.000	0.207	0.000	1.320
4	2008	0.000	0.807	0.595	20.878	0.000	0.779	0.000	0.779
US	1999	2.856	8.335	23.982	337.665	0.159	61.735	9.094	93.198
US	2000	2.803	8.077	25.154	367.845	0.000	67.617	11.511	88.969
US	2001	2.774	7.842	25.066	373.840	0.000	78.683	16.980	100.655
US	2002	2.662	7.498	27.051	375.287	0.000	54.660	13.355	76.536
US	2003	2.710	8.458	28.122	373.943	0.000	86.410	15.120	51.623
US	2004	2.845	8.222	30.225	387.896	0.000	56.615	12.973	34.945
US	2005	3.199	7.712	43.691	400.816	0.000	66.377	12.004	35.088
US	2006	3.239	7.967	31.459	429.756	0.000	42.227	6.900	32.084
US	2007	3.169	7.256	30.036	433.063	0.000	42.338	6.677	29.317
US	2008	3.035	7.737	31.334	439.781	0.000	46.583	7.504	22.099
Energy factor[c]		--	--	--	16.4 MJ/m^3	38.49 GJ/m^3	25.62 GJ/m^3	38.66 GJ/m^3	41.72 GJ/m^3
CO_2 emission factor (kg/GJ)[c]		--	--	--	52.70	78.53	65.76	77.18	83.14

Table S1 *continued*

US		Fuels consumed in refineries[a]						
District	Year	Fuel gas (bl)	Pet. coke	Other product	Natural gas	Coal	Electricity purchased	Steam purchased
PADD		($m^3 \bullet 10^5$)	($m^3 \bullet 10^5$)	uct ($m^3 \bullet 10^4$)	($m^3 \bullet 10^8$)	(Gg)	(TWh)	(Tg)
1	1999	32.387	20.538	6.964	11.501	28.123	3.180	1.599
1	2000	31.990	19.093	6.105	12.553	27.216	3.084	1.897
1	2001	32.322	18.975	5.406	9.915	29.030	3.450	1.797
1	2002	33.987	18.805	5.851	11.086	28.123	3.282	1.865
1	2003	35.329	19.649	7.059	8.032	29.030	3.415	1.674
1	2004	35.419	20.377	2.242	9.177	26.308	3.410	2.352
1	2005	35.481	20.369	2.242	10.082	29.937	3.520	2.228
1	2006	33.756	17.541	0.859	10.258	28.123	3.576	2.593
1	2007	36.392	19.036	0.334	8.129	29.030	3.984	2.624
1	2008	33.909	19.393	0.461	7.892	28.123	4.192	2.360
2	1999	76.667	29.697	22.560	26.317	0.000	8.956	1.262
2	2000	77.341	29.335	19.047	30.038	1.814	8.949	0.890
2	2001	76.697	27.643	20.382	26.510	6.350	8.728	2.060
2	2002	73.293	27.689	19.555	27.235	0.000	8.933	2.368
2	2003	72.970	27.357	16.392	26.727	8.165	8.885	2.577
2	2004	79.249	25.339	27.855	29.254	7.257	9.486	2.863
2	2005	79.832	27.572	26.805	30.152	7.257	9.875	2.283
2	2006	78.834	26.236	31.177	32.485	2.722	10.488	3.310
2	2007	78.586	24.963	6.280	33.993	6.350	10.555	4.871
2	2008	77.716	23.856	0.286	39.330	10.886	10.804	4.999
3	1999	181.263	66.223	31.177	147.683	0.000	13.762	8.968
3	2000	184.163	67.454	34.405	147.541	0.000	14.501	11.455
3	2001	177.565	66.822	30.923	138.325	0.000	15.868	13.142
3	2002	181.193	66.891	21.479	129.876	0.000	16.145	14.670
3	2003	194.971	67.972	29.874	121.706	0.000	15.682	14.456
3	2004	190.864	69.595	22.544	111.896	0.000	17.044	14.827
3	2005	177.745	65.660	20.668	112.129	0.000	16.620	15.757
3	2006	198.807	72.481	31.336	112.029	0.000	18.612	17.690
3	2007	192.263	67.964	24.007	102.791	0.000	20.433	28.790
3	2008	181.956	62.598	26.996	107.893	0.000	20.675	28.919
5	1999	72.803	21.174	25.851	34.754	0.000	5.389	8.469
5	2000	74.282	22.314	26.185	38.268	0.000	4.809	8.268
5	2001	77.031	22.827	22.576	34.867	0.000	4.695	7.881
5	2002	70.694	22.640	22.672	38.733	0.000	4.780	7.589
5	2003	74.354	23.823	25.740	37.477	0.000	4.520	8.595
5	2004	73.964	24.441	31.305	35.335	0.000	4.871	8.732
5	2005	72.657	24.438	27.028	34.906	0.000	4.978	8.145
5	2006	71.543	23.133	34.961	35.733	0.000	4.973	8.164
5	2007	72.423	23.087	27.282	37.863	0.000	5.113	8.091
5	2008	68.973	19.651	32.227	39.629	0.000	5.125	8.064
4	1999	11.585	4.442	11.415	6.145	0.000	1.422	0.424
4	2000	11.465	4.153	13.132	5.502	0.000	1.486	0.384
4	2001	11.946	4.302	12.655	5.686	0.000	1.446	0.419
4	2002	11.639	4.262	13.260	6.024	0.000	1.581	0.337
4	2003	13.827	4.040	13.752	5.319	0.000	1.515	0.402
4	2004	13.541	4.372	8.649	5.472	0.000	1.583	0.504
4	2005	13.050	4.496	7.981	6.112	0.000	1.601	0.432
4	2006	13.508	4.480	2.258	7.031	0.000	1.704	0.343
4	2007	13.202	4.884	0.986	6.375	0.000	1.744	0.540
4	2008	14.501	4.571	1.081	6.445	0.000	1.886	0.458
US	1999	374.706	142.074	97.968	226.399	28.123	32.709	20.722
US	2000	379.240	142.348	98.874	233.902	29.030	32.829	22.894
US	2001	375.561	140.570	91.942	215.304	35.380	34.187	25.299
US	2002	370.806	140.287	82.816	212.953	28.123	34.721	26.830
US	2003	391.451	142.841	92.817	199.261	37.195	34.017	27.705
US	2004	393.037	144.125	92.594	191.134	33.566	36.394	29.278
US	2005	378.765	142.535	84.724	193.381	37.195	36.594	28.844
US	2006	396.448	143.871	100.591	197.536	30.844	39.353	32.100
US	2007	392.867	139.933	58.889	189.152	35.380	41.829	44.916
US	2008	377.056	130.069	61.051	201.191	39.009	42.682	44.801
Energy factor[c]		39.82 GJ/m^3	39.98 GJ/m^3	38.66 GJ/m^3	38.27 MJ/m^3	25.80 MJ/kg	3.6 MJ/kWh	2.18 MJ/kg
CO_2 emission factor (kg/GJ)[c]		67.73	107.74	73.20	55.98	99.58	187.78	91.63

Table S1 ***continued***

US		Refinery product yields[a] --							
District PADD	Year	LPG (%)	Fin. motor gasoline (%)	Aviation gasoline (%)	Kerosine jet fuel (%)	Kerosine (%)	Distillate fuel oil (%)	Residual fuel oil (%)	Naphtha for chem FS (%)
1	1999	2.5	46.6	0.2	7.0	0.8	26.3	6.5	0.8
1	2000	2.8	45.2	0.2	6.3	0.8	27.9	6.8	0.8
1	2001	2.9	45.8	0.2	5.3	0.8	29.1	6.6	0.8
1	2002	3.0	46.7	0.3	5.3	0.8	28.1	5.7	0.9
1	2003	3.0	46.4	0.2	5.2	0.8	27.2	7.8	0.8
1	2004	2.6	46.5	0.4	6.1	0.7	26.6	6.9	0.8
1	2005	2.4	46.6	0.3	5.7	0.7	28.8	6.2	0.8
1	2006	2.6	45.8		5.1	0.4	29.2	7.1	1.1
1	2007	3.2	45.5	0.1	5.0	0.5	29.4	7.2	1.1
1	2008	3.3	44.6		5.7	0.6	29.6	7.1	1.1
2	1999	3.7	51.1	0.1	6.6	0.5	24.8	1.6	0.6
2	2000	3.7	50.4	0.1	6.9	0.4	25.7	1.8	0.5
2	2001	3.6	51.1	0.1	6.6	0.4	26.0	2.0	0.6
2	2002	3.5	52.0	0.1	6.7	0.3	25.4	1.8	0.6
2	2003	3.3	51.5	0.1	6.2	0.3	26.0	1.7	0.5
2	2004	3.3	51.6	0.1	6.4	0.3	25.7	1.8	0.8
2	2005	3.1	50.4	0.1	6.5	0.3	27.1	1.6	0.8
2	2006	4.0	49.4	0.1	6.2	0.3	27.3	1.7	0.9
2	2007	3.9	49.8	0.1	6.1	0.1	28.2	1.7	0.9
2	2008	3.5	48.5	0.1	6.3	0.0	30.0	1.6	0.8
3	1999	6.1	44.8	0.2	11.1	0.4	21.1	4.3	2.1
3	2000	6.0	44.7	0.1	11.1	0.4	21.9	4.6	2.2
3	2001	5.6	44.3	0.1	10.5	0.6	22.8	4.8	1.7
3	2002	5.8	45.4	0.1	10.3	0.4	22.3	3.7	2.7
3	2003	5.5	44.8	0.1	9.9	0.4	23.0	4.1	2.6
3	2004	5.3	44.6	0.1	10.0	0.5	23.5	3.9	2.8
3	2005	4.7	43.8	0.1	10.2	0.6	24.5	3.9	2.3
3	2006	4.8	43.5	0.2	9.7	0.4	25.2	3.8	1.9
3	2007	5.0	43.2	0.1	9.4	0.3	26.0	4.1	1.9
3	2008	5.1	41.6	0.1	9.6	0.0	28.4	4.0	1.5
5	1999	2.6	44.7	0.1	15.8	0.2	18.3	8.5	0.2
5	2000	3.1	45.7	0.1	16.2	0.2	18.5	6.8	0.1
5	2001	2.7	45.5	0.1	16.0	0.1	19.2	6.9	0.1
5	2002	2.7	47.3	0.1	16.0	0.1	19.0	6.2	0.1
5	2003	2.9	47.2	0.1	16.0	0.0	19.5	5.8	0.1
5	2004	2.6	47.3	0.1	16.2	0.0	19.5	6.1	0.0
5	2005	2.5	47.3	0.1	16.2	0.0	20.4	5.8	0.0
5	2006	2.8	47.7	0.1	15.3	0.0	20.3	5.8	0.0
5	2007	2.8	46.6	0.1	15.6	0.0	20.8	6.3	0.0
5	2008	2.8	45.6	0.1	17.5	0.0	21.6	5.5	0.0
4	1999	1.3	47.8	0.1	5.4	0.5	28.7	2.3	
4	2000	1.3	47.1	0.1	5.8	0.3	29.1	2.0	0.0
4	2001	1.3	47.4	0.1	5.3	0.3	29.8	2.3	
4	2002	1.1	48.0	0.1	4.8	0.4	29.9	2.1	
4	2003	0.8	47.9	0.1	4.9	0.4	29.5	2.4	
4	2004	0.8	47.5	0.1	4.9	0.3	30.4	2.5	
4	2005	0.7	46.0	0.1	5.4	0.3	30.6	2.7	
4	2006	1.3	46.4	0.1	5.3	0.4	30.6	2.8	
4	2007	1.5	46.3	0.1	5.4	0.3	29.8	2.6	
4	2008	1.6	47.4	0.1	4.8	0.2	31.6	2.2	
US	1999	4.5	46.5	0.2	10.2	0.4	22.3	4.6	1.3
US	2000	4.5	46.2	0.1	10.3	0.4	23.1	4.5	1.3
US	2001	4.3	46.2	0.1	9.8	0.5	23.8	4.6	1.1
US	2002	4.3	47.3	0.1	9.8	0.4	23.2	3.9	1.6
US	2003	4.2	46.9	0.1	9.5	0.4	23.7	4.2	1.5
US	2004	4.0	46.8	0.1	9.7	0.4	23.9	4.1	1.6
US	2005	3.6	46.2	0.1	9.8	0.4	25.0	4.0	1.4
US	2006	3.9	45.8	0.1	9.3	0.3	25.4	4.0	1.2
US	2007	4.1	45.5	0.1	9.1	0.2	26.1	4.2	1.3
US	2008	4.1	44.2	0.1	9.7	0.1	27.8	4.0	1.0
Energy factor[c]		--	--	--	--	--	--	--	--
CO_2 emission factor (kg/GJ)[c]		--	--	--	--	--	--	--	--

Table S1 ***continued***

US District PADD	Year	Refinery product yields[a]								Utilization of operable ref. capacity[a] (%)
		Oth. oils for chem FS (%)	Special naphtha (%)	Lubricants (%)	Waxes (%)	Petroleum coke (%)	Asphalt & road oil (%)	Fuel gas (%)	Miscellaneous products (%)	
1	1999		0.1	1.0	0.0	3.1	5.4	3.7	0.1	90.9
1	2000		0.1	0.9	0.1	3.0	6.1	3.5	0.1	91.7
1	2001		0.1	0.9	0.0	3.3	6.0	3.8	0.1	87.2
1	2002		0.1	1.0	0.0	3.1	6.0	3.9	0.1	88.9
1	2003		0.1	1.0	0.0	2.9	5.7	3.8	0.1	92.7
1	2004		0.1	1.1	0.0	3.1	6.2	3.9	0.1	90.4
1	2005		0.1	1.0	0.0	2.9	5.7	3.8	0.1	93.1
1	2006		0.1	1.1	0.0	3.0	5.6	3.6	0.2	86.7
1	2007		0.0	1.0	0.0	3.2	5.0	3.9	0.2	85.6
1	2008		0.0	1.1	0.1	3.3	5.1	3.8	0.2	80.8
2	1999	0.7	0.7	0.6	0.1	4.2	5.6	3.9	0.3	93.3
2	2000	0.4	0.7	0.5	0.1	4.3	5.5	3.9	0.3	94.2
2	2001	0.0	0.6	0.4	0.1	4.3	5.1	4.0	0.3	93.9
2	2002	0.0	0.5	0.5	0.1	4.1	5.3	4.0	0.4	90.0
2	2003	0.0	0.6	0.5	0.1	4.2	5.6	4.1	0.4	91.6
2	2004	0.3	0.1	0.4	0.1	4.3	5.7	4.1	0.4	93.6
2	2005	0.3	0.2	0.4	0.1	4.5	5.7	4.1	0.5	92.9
2	2006	0.2	0.2	0.5	0.1	4.4	6.1	4.1	0.5	92.4
2	2007	0.2	0.1	0.4	0.1	4.3	5.3	4.2	0.4	90.1
2	2008	0.2	0.1	0.4	0.1	4.3	5.3	4.0	0.4	88.4
3	1999	2.5	0.8	1.7	0.2	4.8	1.7	4.1	0.4	94.7
3	2000	2.3	0.4	1.7	0.2	4.8	1.8	4.1	0.4	93.9
3	2001	2.1	0.4	1.6	0.1	5.3	1.6	4.1	0.5	94.8
3	2002	1.9	0.4	1.6	0.1	5.7	1.6	4.2	0.5	91.5
3	2003	2.3	0.4	1.5	0.1	5.7	1.6	4.4	0.5	93.6
3	2004	2.4	0.5	1.6	0.1	5.9	1.5	4.3	0.4	94.1
3	2005	2.1	0.4	1.6	0.1	6.0	1.6	4.3	0.4	88.3
3	2006	2.4	0.4	1.7	0.1	6.2	1.5	4.6	0.5	88.7
3	2007	2.4	0.5	1.7	0.1	6.0	1.3	4.3	0.5	88.7
3	2008	2.3	0.5	1.7	0.1	6.0	1.1	4.4	0.6	83.6
5	1999	0.3	0.1	1.0	0.0	6.1	2.4	5.8	0.2	87.1
5	2000	0.3	0.1	0.9	-0.1	6.3	2.4	5.6	0.3	87.5
5	2001	0.3	0.1	1.0	0.0	6.0	2.1	5.8	0.3	89.1
5	2002	0.3	0.1	0.8	0.0	6.0	2.1	5.5	0.3	90.0
5	2003	0.3	0.1	0.8		6.2	1.9	5.6	0.3	91.3
5	2004	0.3	0.0	0.7		6.1	1.9	5.4	0.3	90.4
5	2005	0.4	0.0	0.7		6.2	1.7	5.1	0.3	91.7
5	2006	0.4	0.1	0.7		6.0	1.8	5.2	0.4	90.5
5	2007	0.3	0.0	0.6		5.8	1.8	5.4	0.4	87.6
5	2008	0.1	0.0	0.8		6.1	1.4	5.1	0.5	88.1
4	1999	0.1	0.0		0.7	3.4	8.8	4.1	0.4	95.7
4	2000	0.1	0.0		0.6	3.3	9.3	3.9	0.4	94.7
4	2001	0.2	0.0		0.6	3.3	8.6	4.1	0.4	90.7
4	2002	0.1			0.5	3.2	9.2	3.8	0.4	91.6
4	2003	0.1			0.4	3.2	9.1	4.5	0.4	91.9
4	2004	0.1			0.4	3.2	9.3	4.2	0.4	95.7
4	2005	0.1	0.0		0.4	3.3	9.5	4.1	0.4	95.5
4	2006	0.1	0.0		0.3	3.3	8.5	4.2	0.4	93.5
4	2007	0.1	0.0		0.0	3.4	8.9	4.2	0.3	91.3
4	2008		0.0		0.0	4.6	6.1	4.6	0.5	89.4
US	1999	1.4	0.6	1.2	0.1	4.7	3.3	4.3	0.3	92.6
US	2000	1.3	0.4	1.2	0.1	4.7	3.4	4.2	0.4	92.6
US	2001	1.1	0.3	1.1	0.1	4.9	3.1	4.3	0.4	92.6
US	2002	1.0	0.3	1.1	0.1	5.1	3.2	4.3	0.4	90.7
US	2003	1.2	0.3	1.1	0.1	5.1	3.2	4.5	0.4	92.6
US	2004	1.3	0.3	1.1	0.1	5.2	3.2	4.4	0.4	93.0
US	2005	1.1	0.2	1.1	0.1	5.3	3.2	4.3	0.4	90.6
US	2006	1.2	0.2	1.2	0.1	5.3	3.2	4.5	0.4	89.7
US	2007	1.3	0.3	1.1	0.1	5.2	2.9	4.4	0.4	88.5
US	2008	1.2	0.3	1.1	0.1	5.3	2.7	4.3	0.5	85.3
Energy factor[c]		--	--	--	--	--	--	--	--	--
CO_2 emission factor (kg/GJ)[c]		--	--	--	--	--	--	--	--	--

Table S1 ***continued***

US		Energy consumed/volume crude feed (GJ/m^3) and CO_2 emitted/vol. crude feed (kg/m^3) for refinery fuels[c]											
District	Year	Hydrogen prod.		Crude oil consmd.		LPG consumed		Distillate consmd.		Res. fuel oil cons.		Fuel gas (bl)	
PADD		(GJ/m^3)	(kg/m^3)	(GJ/m^3)	(kg/m^3)	(GJ/m^3)	(kg/m^3)	(GJ/m^3)	(kg/m^3)	(GJ/m^3)	(kg/m^3)	(GJ/m^3)	(kg/m^3)
1	1999	0.195	10.28	0.000	0.00	0.008	0.52	0.009	0.68	0.173	14.39	1.446	97.93
1	2000	0.230	12.10	0.000	0.00	0.014	0.93	0.018	1.38	0.180	14.94	1.410	95.49
1	2001	0.199	10.48	0.000	0.00	0.017	1.14	0.040	3.11	0.217	18.03	1.498	101.43
1	2002	0.171	8.99	0.000	0.00	0.013	0.85	0.033	2.57	0.138	11.44	1.529	103.58
1	2003	0.242	12.77	0.000	0.00	0.022	1.44	0.042	3.22	0.127	10.57	1.530	103.66
1	2004	0.244	12.88	0.000	0.00	0.022	1.46	0.031	2.43	0.082	6.86	1.548	104.85
1	2005	0.243	12.82	0.000	0.00	0.032	2.08	0.024	1.87	0.082	6.81	1.523	103.13
1	2006	0.297	15.66	0.000	0.00	0.016	1.02	0.002	0.13	0.071	5.88	1.559	105.58
1	2007	0.230	12.13	0.000	0.00	0.009	0.58	0.002	0.12	0.064	5.33	1.695	114.82
1	2008	0.244	12.85	0.000	0.00	0.003	0.17	0.002	0.17	0.033	2.73	1.673	113.30
2	1999	0.334	17.58	0.000	0.00	0.036	2.33	0.002	0.15	0.093	7.71	1.560	105.64
2	2000	0.328	17.31	0.000	0.00	0.019	1.23	0.001	0.12	0.072	5.99	1.556	105.41
2	2001	0.367	19.34	0.000	0.00	0.019	1.23	0.003	0.20	0.084	7.02	1.590	107.72
2	2002	0.347	18.30	0.000	0.00	0.023	1.48	0.002	0.17	0.066	5.53	1.563	105.85
2	2003	0.320	16.89	0.000	0.00	0.035	2.32	0.001	0.09	0.021	1.74	1.553	105.19
2	2004	0.316	16.66	0.000	0.00	0.023	1.51	0.001	0.09	0.007	0.56	1.647	111.58
2	2005	0.381	20.07	0.000	0.00	0.016	1.09	0.002	0.12	0.006	0.47	1.653	111.96
2	2006	0.592	31.19	0.000	0.00	0.012	0.79	0.001	0.11	0.007	0.59	1.635	110.72
2	2007	0.612	32.26	0.000	0.00	0.018	1.20	0.002	0.12	0.007	0.55	1.665	112.80
2	2008	0.616	32.46	0.000	0.00	0.017	1.14	0.001	0.11	0.007	0.57	1.644	111.34
3	1999	0.530	27.94	0.000	0.01	0.008	0.52	0.002	0.14	0.000	0.02	1.771	119.92
3	2000	0.533	28.06	0.000	0.00	0.008	0.53	0.003	0.20	0.000	0.00	1.778	120.40
3	2001	0.545	28.70	0.000	0.00	0.007	0.44	0.002	0.15	0.000	0.00	1.676	113.50
3	2002	0.576	30.33	0.000	0.00	0.008	0.55	0.001	0.10	0.000	0.00	1.753	118.71
3	2003	0.559	29.49	0.000	0.00	0.011	0.70	0.001	0.05	0.000	0.00	1.833	124.18
3	2004	0.592	31.19	0.000	0.00	0.003	0.23	0.001	0.09	0.000	0.00	1.748	118.37
3	2005	0.609	32.08	0.000	0.00	0.003	0.23	0.001	0.10	0.000	0.01	1.693	114.67
3	2006	0.560	29.49	0.000	0.00	0.003	0.17	0.002	0.12	0.000	0.00	1.850	125.28
3	2007	0.553	29.12	0.000	0.00	0.002	0.13	0.002	0.13	0.000	0.00	1.782	120.72
3	2008	0.594	31.28	0.000	0.00	0.005	0.34	0.002	0.12	0.000	0.00	1.774	120.17
5	1999	1.217	64.13	0.000	0.00	0.031	2.05	0.010	0.80	0.025	2.04	1.892	128.17
5	2000	1.426	75.15	0.000	0.00	0.056	3.66	0.009	0.71	0.029	2.44	1.881	127.39
5	2001	1.364	71.86	0.000	0.00	0.075	4.93	0.011	0.82	0.035	2.92	1.899	128.59
5	2002	1.363	71.85	0.000	0.00	0.031	2.02	0.008	0.60	0.037	3.04	1.722	116.63
5	2003	1.315	69.32	0.000	0.00	0.053	3.49	0.009	0.70	0.029	2.41	1.776	120.32
5	2004	1.315	69.29	0.000	0.00	0.038	2.50	0.009	0.66	0.029	2.40	1.774	120.15
5	2005	1.312	69.15	0.000	0.00	0.055	3.65	0.009	0.71	0.029	2.38	1.720	116.48
5	2006	1.409	74.24	0.000	0.00	0.036	2.36	0.009	0.73	0.031	2.55	1.708	115.69
5	2007	1.484	78.18	0.000	0.00	0.036	2.34	0.009	0.69	0.030	2.53	1.781	120.60
5	2008	1.471	77.54	0.000	0.00	0.038	2.48	0.011	0.85	0.030	2.52	1.682	113.92
4	1999	0.448	23.59	0.000	0.00	0.006	0.37	0.001	0.10	0.049	4.08	1.574	106.62
4	2000	0.446	23.50	0.000	0.00	0.008	0.50	0.001	0.05	0.067	5.58	1.534	103.86
4	2001	0.481	25.36	0.000	0.00	0.005	0.35	0.001	0.11	0.049	4.10	1.614	109.29
4	2002	0.465	24.49	0.000	0.00	0.006	0.39	0.000	0.00	0.045	3.70	1.518	102.84
4	2003	0.441	23.22	0.000	0.00	0.006	0.43	0.000	0.00	0.036	3.01	1.787	121.02
4	2004	0.434	22.88	0.000	0.00	0.008	0.56	0.000	0.01	0.030	2.51	1.668	112.99
4	2005	0.631	33.28	0.000	0.00	0.003	0.20	0.000	0.00	0.034	2.82	1.593	107.92
4	2006	0.637	33.58	0.000	0.00	0.002	0.12	0.000	0.00	0.025	2.06	1.664	112.71
4	2007	0.847	44.66	0.000	0.00	0.002	0.11	0.000	0.00	0.017	1.44	1.659	112.38
4	2008	0.983	51.82	0.000	0.00	0.006	0.42	0.000	0.00	0.010	0.86	1.843	124.81
US	1999	0.570	30.01	0.000	0.01	0.018	1.19	0.004	0.31	0.044	3.69	1.705	115.48
US	2000	0.612	32.23	0.000	0.00	0.020	1.28	0.005	0.39	0.042	3.48	1.701	115.21
US	2001	0.619	32.64	0.000	0.00	0.023	1.49	0.007	0.57	0.047	3.92	1.679	113.70
US	2002	0.629	33.14	0.000	0.00	0.016	1.05	0.006	0.45	0.036	3.01	1.676	113.53
US	2003	0.613	32.32	0.000	0.00	0.025	1.62	0.006	0.50	0.024	1.99	1.732	117.31
US	2004	0.625	32.94	0.000	0.00	0.016	1.04	0.005	0.42	0.016	1.32	1.709	115.74
US	2005	0.654	34.49	0.000	0.00	0.019	1.24	0.005	0.40	0.016	1.35	1.668	113.00
US	2006	0.701	36.92	0.000	0.00	0.012	0.79	0.003	0.23	0.015	1.23	1.744	118.10
US	2007	0.713	37.57	0.000	0.00	0.012	0.80	0.003	0.22	0.014	1.13	1.745	118.18
US	2008	0.744	39.23	0.000	0.00	0.014	0.90	0.003	0.26	0.011	0.88	1.722	116.62
Energy factor[c]		16.4 MJ/m^3		38.49 GJ/m^3		25.62 GJ/m^3		38.66 GJ/m^3		41.72 GJ/m^3		39.82 GJ/m^3	
CO_2 emission factor (kg/GJ)[c]		--	52.70	--	78.53	--	65.76	--	77.18	--	83.14	--	67.73

Table S1 *continued*

US		Energy consumed/volume crude feed (GJ/m^3) and CO$_2$ emitted/vol. crude feed (kg/m^3) for refinery fuels^c											
District	Year	Petroleum coke		Other products		Natural gas		Coal consumed		Electricity purch.		Steam purch.	
PADD		(GJ/m^3)	(kg/m^3)	(GJ/m^3)	(kg/m^3)	(GJ/m^3)	(kg/m^3)	(GJ/m^3)	(kg/m^3)	(GJ/m^3)	(kg/m^3)	(GJ/m^3)	(kg/m^3)
1	1999	0.921	99.186	0.030	2.21	0.493	27.63	0.008	0.81	0.128	24.10	0.039	3.58
1	2000	0.845	91.022	0.026	1.91	0.532	29.76	0.008	0.77	0.123	23.07	0.046	4.19
1	2001	0.883	95.103	0.024	1.78	0.442	24.72	0.009	0.87	0.145	27.14	0.046	4.18
1	2002	0.850	91.531	0.026	1.87	0.479	26.84	0.008	0.82	0.134	25.07	0.046	4.21
1	2003	0.855	92.078	0.030	2.17	0.334	18.72	0.008	0.81	0.134	25.11	0.040	3.64
1	2004	0.894	96.342	0.010	0.70	0.386	21.58	0.007	0.74	0.135	25.30	0.056	5.16
1	2005	0.878	94.557	0.009	0.68	0.416	23.28	0.008	0.83	0.137	25.64	0.052	4.80
1	2006	0.813	87.620	0.004	0.28	0.455	25.48	0.008	0.84	0.149	28.03	0.066	6.01
1	2007	0.890	95.924	0.002	0.11	0.364	20.37	0.009	0.87	0.168	31.51	0.067	6.13
1	2008	0.961	103.488	0.002	0.16	0.374	20.95	0.009	0.90	0.187	35.11	0.064	5.84
2	1999	0.607	65.353	0.045	3.26	0.515	28.80	0.000	0.00	0.165	30.93	0.014	1.29
2	2000	0.593	63.855	0.037	2.72	0.581	32.52	0.000	0.02	0.163	30.57	0.010	0.90
2	2001	0.576	62.009	0.041	3.00	0.528	29.58	0.001	0.08	0.164	30.73	0.023	2.14
2	2002	0.593	63.869	0.040	2.96	0.558	31.24	0.000	0.00	0.172	32.34	0.028	2.53
2	2003	0.585	62.985	0.034	2.48	0.547	30.60	0.001	0.11	0.171	32.10	0.030	2.75
2	2004	0.529	56.979	0.056	4.11	0.584	32.72	0.001	0.10	0.178	33.48	0.033	2.99
2	2005	0.573	61.755	0.054	3.94	0.600	33.59	0.001	0.10	0.185	34.71	0.026	2.37
2	2006	0.546	58.853	0.063	4.59	0.647	36.24	0.000	0.04	0.197	36.92	0.038	3.44
2	2007	0.531	57.224	0.013	0.95	0.692	38.76	0.001	0.09	0.202	37.97	0.057	5.18
2	2008	0.507	54.586	0.001	0.04	0.800	44.76	0.001	0.15	0.207	38.80	0.058	5.30
3	1999	0.649	69.972	0.030	2.16	1.386	77.61	0.000	0.00	0.122	22.82	0.048	4.39
3	2000	0.654	70.430	0.032	2.36	1.369	76.62	0.000	0.00	0.127	23.76	0.061	5.55
3	2001	0.633	68.217	0.028	2.07	1.255	70.23	0.000	0.00	0.135	25.42	0.068	6.22
3	2002	0.650	69.991	0.020	1.48	1.207	67.59	0.000	0.00	0.141	26.51	0.078	7.12
3	2003	0.642	69.143	0.027	2.00	1.100	61.57	0.000	0.00	0.133	25.04	0.074	6.82
3	2004	0.640	68.933	0.020	1.47	0.985	55.12	0.000	0.00	0.141	26.49	0.074	6.81
3	2005	0.628	67.654	0.019	1.40	1.026	57.46	0.000	0.00	0.143	26.88	0.082	7.53
3	2006	0.677	72.950	0.028	2.07	1.002	56.08	0.000	0.00	0.157	29.40	0.090	8.26
3	2007	0.633	68.154	0.022	1.58	0.916	51.27	0.000	0.00	0.171	32.16	0.146	13.39
3	2008	0.613	66.029	0.026	1.87	1.011	56.60	0.000	0.00	0.182	34.23	0.154	14.15
5	1999	0.553	59.534	0.065	4.78	0.868	48.60	0.000	0.00	0.127	23.78	0.121	11.04
5	2000	0.567	61.118	0.064	4.71	0.931	52.13	0.000	0.00	0.110	20.67	0.115	10.50
5	2001	0.565	60.863	0.054	3.95	0.826	46.24	0.000	0.00	0.105	19.65	0.106	9.74
5	2002	0.554	59.655	0.054	3.92	0.907	50.76	0.000	0.00	0.105	19.77	0.101	9.27
5	2003	0.571	61.570	0.060	4.37	0.861	48.17	0.000	0.00	0.098	18.33	0.112	10.30
5	2004	0.589	63.411	0.073	5.34	0.814	45.60	0.000	0.00	0.106	19.83	0.115	10.51
5	2005	0.581	62.572	0.062	4.55	0.794	44.45	0.000	0.00	0.107	20.00	0.106	9.67
5	2006	0.555	59.745	0.081	5.93	0.820	45.90	0.000	0.00	0.107	20.16	0.107	9.78
5	2007	0.570	61.399	0.065	4.77	0.895	50.08	0.000	0.00	0.114	21.34	0.109	9.98
5	2008	0.481	51.835	0.076	5.58	0.929	51.99	0.000	0.00	0.113	21.22	0.108	9.86
4	1999	0.606	65.292	0.151	11.02	0.802	44.92	0.000	0.00	0.175	32.80	0.032	2.89
4	2000	0.558	60.087	0.171	12.48	0.707	39.60	0.000	0.00	0.180	33.74	0.028	2.57
4	2001	0.583	62.862	0.166	12.15	0.738	41.32	0.000	0.00	0.177	33.16	0.031	2.84
4	2002	0.558	60.150	0.168	12.29	0.755	42.28	0.000	0.00	0.186	35.01	0.024	2.21
4	2003	0.524	56.473	0.173	12.63	0.661	36.98	0.000	0.00	0.177	33.24	0.028	2.61
4	2004	0.541	58.265	0.103	7.57	0.648	36.27	0.000	0.00	0.176	33.11	0.034	3.12
4	2005	0.551	59.384	0.095	6.93	0.717	40.15	0.000	0.00	0.177	33.19	0.029	2.64
4	2006	0.554	59.705	0.027	1.98	0.832	46.60	0.000	0.00	0.190	35.64	0.023	2.12
4	2007	0.616	66.398	0.012	0.88	0.770	43.10	0.000	0.00	0.198	37.21	0.037	3.41
4	2008	0.583	62.831	0.013	0.98	0.787	44.07	0.000	0.00	0.217	40.69	0.032	2.92
US	1999	0.649	69.932	0.043	3.17	0.990	55.43	0.001	0.08	0.135	25.27	0.052	4.73
US	2000	0.641	69.064	0.043	3.15	1.008	56.44	0.001	0.08	0.133	25.00	0.056	5.15
US	2001	0.631	67.966	0.040	2.92	0.925	51.78	0.001	0.10	0.138	25.94	0.062	5.67
US	2002	0.637	68.598	0.036	2.66	0.925	51.79	0.001	0.08	0.142	26.65	0.066	6.08
US	2003	0.635	68.369	0.040	2.92	0.847	47.44	0.001	0.11	0.136	25.55	0.067	6.15
US	2004	0.629	67.782	0.039	2.86	0.799	44.71	0.001	0.09	0.143	26.86	0.070	6.39
US	2005	0.630	67.916	0.036	2.65	0.819	45.83	0.001	0.11	0.146	27.36	0.070	6.37
US	2006	0.635	68.447	0.043	3.14	0.835	46.74	0.001	0.09	0.156	29.38	0.077	7.08
US	2007	0.624	67.229	0.025	1.86	0.807	45.20	0.001	0.10	0.168	31.54	0.109	10.01
US	2008	0.596	64.249	0.027	1.98	0.883	49.43	0.001	0.11	0.176	33.09	0.112	10.26
Energy factor^c		39.98 GJ/m^3		38.66 GJ/m^3		38.27 MJ/m^3		25.80 MJ/kg		3.60 MJ/kWh		2.18 MJ/kg	
CO$_2$ emission factor (kg/GJ)^c		--	107.74	--	73.20	--	55.98	--	99.58	--	187.78	--	91.63

Table S1 ***continued***

US District PADD	Year	Refinery energy consumed *(EI)*[d] (GJ/m^3)	Fuel mix emission intensity (CO$_2$)[d] (kg/GJ)	Refinery carbon dioxide emissions[d] (kg/m^3)
1	1999	3.451	81.53	281.3
1	2000	3.430	80.34	275.6
1	2001	3.518	81.85	288.0
1	2002	3.426	81.08	277.8
1	2003	3.364	81.51	274.2
1	2004	3.416	81.46	278.3
1	2005	3.404	81.23	276.5
1	2006	3.440	80.40	276.5
1	2007	3.499	82.28	287.9
1	2008	3.551	83.26	295.7
2	1999	3.368	78.10	263.1
2	2000	3.361	77.56	260.6
2	2001	3.396	77.46	263.1
2	2002	3.393	77.90	264.3
2	2003	3.298	78.00	257.3
2	2004	3.376	77.25	260.8
2	2005	3.496	77.27	270.2
2	2006	3.738	75.84	283.5
2	2007	3.800	75.55	287.1
2	2008	3.858	74.97	289.3
3	1999	4.546	71.61	325.5
3	2000	4.563	71.87	327.9
3	2001	4.348	72.43	315.0
3	2002	4.434	72.71	322.4
3	2003	4.381	72.81	319.0
3	2004	4.204	73.43	308.7
3	2005	4.205	73.24	308.0
3	2006	4.367	74.15	323.8
3	2007	4.226	74.93	316.7
3	2008	4.361	74.48	324.8
5	1999	4.908	70.27	344.9
5	2000	5.189	69.09	358.5
5	2001	5.039	69.38	349.6
5	2002	4.881	69.15	337.5
5	2003	4.885	69.40	339.0
5	2004	4.861	69.89	339.7
5	2005	4.774	69.88	333.6
5	2006	4.862	69.32	337.1
5	2007	5.091	69.12	351.9
5	2008	4.939	68.39	337.8
4	1999	3.843	75.90	291.7
4	2000	3.698	76.25	282.0
4	2001	3.846	75.80	291.6
4	2002	3.726	76.06	283.4
4	2003	3.833	75.56	289.6
4	2004	3.644	76.10	277.3
4	2005	3.830	74.80	286.5
4	2006	3.955	74.48	294.5
4	2007	4.159	74.43	309.6
4	2008	4.475	73.61	329.4
US	1999	4.211	73.46	309.3
US	2000	4.261	73.09	311.5
US	2001	4.172	73.51	306.7
US	2002	4.170	73.62	307.0
US	2003	4.126	73.74	304.3
US	2004	4.052	74.08	300.2
US	2005	4.065	73.98	300.7
US	2006	4.222	73.94	312.1
US	2007	4.221	74.34	313.8
US	2008	4.289	73.90	317.0
Energy factor[c]		--	--	--
CO$_2$ emission factor (kg/GJ)[c]		--	--	--

Legend and notes for Table S1.

Observations of operating refineries that support the central analysis reported in the main text are based on the data given in Table S1.

a. Refinery crude inputs, fuels consumed, products yield, and capacity utilization are from the U.S. Energy Information Administration (USEIA) *(S1-6)*. Fuel energy consumption for hydrogen production is discussed below. Blank entries for yield of some minor products in some districts and years were blank in the original data reported *(S5)* and were assigned a value of zero in the analysis.

b. Process capacities are volumes that can be processed during 24 hours after making allowances for types and grades of inputs and products, environmental constraints and scheduled downtime, from *Oil & Gas Journal (S7)*. The prefix “cs” or “ps” denotes processing of crude streams (including gas oil and residua) or of product streams, respectively (csHydrotreating thus includes hydrotreating of gas oil, residua and catalytic cracking feeds). Atmospheric and vacuum distillation capacities reported for the BP Ferndale, WA, and Carson, CA, refineries in 2007 are higher than those in 2006 or 2008 although no distillation upgrades are reported at those plants in 2006 or 2007, and reported vacuum distillation capacity exceeded total crude capacity reported at the Ferndale plant *(S7)*. The reported data for those four entries are replaced by the average of 2006 and 2008 atmospheric, and vacuum distillation, capacities for each of those two plants. This results in $49.609 \bullet 10^4$ instead of $50.047 \bullet 10^4$ m^3/day for atmospheric distillation, and $24.031 \bullet 10^4$ instead of $26.709 \bullet 10^4$ m^3/day for vacuum distillation, in those District 5 entries shown for 2007. Analyses including the reported data, including the corrected data, and excluding the observation (for District 5 in 2007), showed that this correction did not affect the results significantly.

c. Contributions of refinery fuels to refinery energy consumption/m^3 crude feed (GJ/m^3) and refinery mass emissions of CO_2 (kg/m^3 crude feed) are shown. These contributions are calculated using the fuel consumption reported and the energy and emission factors shown below each fuel in the table. The energy factor for hydrogen is for an efficient natural gas-fueled steam methane reforming unit as discussed below. Steam energy is based on latent heat of evaporation at 153 kPa/126 ºC. All other factors for conversions to common energy units (HHV) are from the California Air Resources Board *(S8)*. Emission factors (except for H_2 production) are the fuel emission factors for CO_2 emission from stationary combustion established by USEIA for its voluntary reporting of greenhouse gases program *(S9)*. These emission factors are based on carbon content and oxidation estimates for U.S. fuels quality that the agency derived and documented for its estimates of greenhouse gas emissions in the U.S. *(S10)*. The U.S. grid average factor is applied to purchased electricity. The average of distillate, LPG, and waste oil blended with distillate fuel factors is applied to the "other products" category.

Energy consumed by hydrogen production cannot be calculated from the USEIA fuels data *(S11, S12)*. However, the strong trend of hydroprocessing and hydrogen plant capacity addition shown in Table S1 suggests that U.S. refineries were generally hydrogen-limited, and used most of their available H_2 capacity, during 1999-2008. Energy requirements are assigned to 90% of the hydrogen production capacity reported *(S7)* for these reasons. Energy use for steam reforming of natural gas ranges by approximately 15-18 MJ/m^3 H_2 produced *(S12-15),* and is greater for less efficient designs and for plants using heavier feeds such as naphtha. The energy factor used here (16.4 MJ/m^3) is for a modern steam methane reformer using pressure swing absorption and natural gas feed *(S13)*. The CO_2 emissions factor (52.7 kg/GJ) is derived from the same source *(S13)* and is virtually identical to USEPA's estimate of 0.053 t/MM Btu *(S15)*. Steam reformer CO_2 emissions are primarily from the shift reaction rather than direct combustion, and increase with the use of heavier feeds and less efficient hydrogen production methods *(S12, S15)*. Because many refinery hydrogen plants use less efficient technology, naphtha feed or both, the factors used are conservative.

d. Refinery energy intensity (*EI*) (GJ/m^3 crude feed), fuel mix emission intensity (kg/GJ), and emissions (kg/m^3) are shown in the last three columns of the table. *EI* ranges by 57%, from 3.30 to 5.19 GJ/m^3 crude feed, while fuel mix emission intensity ranges from 68.4 to 83.3 kg/GJ (22%) among districts and years. The much larger percentage range for *EI* indicates that differences in total amounts of fuel energy used per volume crude processed have a greater impact on total emissions than differences in the emission intensity of the fuel mix, for these districts and years.

Fuel gas, natural gas, petroleum coke and hydrogen (assumed to be natural gas-fueled herein) account for the vast majority of energy and emissions in all cases but the fuel mix varies between districts and years. Fuel gas accounts for 34% of total energy and emissions in District 5 during 2008, but it accounts for 49% of total energy and 43% of total emissions in District 2 during 2004. Natural gas excluding H_2 production accounts for 10% of energy and 7% of emissions in District 1 during 2003 but 30% of energy and 24% of emissions in District 3 during 1999. Hydrogen accounts for 5% of energy and 3% of emissions in District 1 during 2002, but 30% of energy and 23% of emissions in District 5 during 2008.

Petroleum coke accounts for 10% of energy and 15% of emissions in District 5 during 2008, but it accounts for 27% of energy and 35% of emissions in District 1 during both 1999 and 2008.

Fuel mix emission intensity generally increases with the portion of fuel mix emissions accounted for by coke, which increases with the catalytic cracking/ atmospheric distillation ratio, among districts and years. Petroleum coke is a byproduct of cracking reactions that is burned in cracking catalyst regeneration. Catalytic cracking generally decreases with increasing hydrocracking (capacities/atm. capacity). At the same time, hydrogen production capacity increases with hydrocracking capacity, and with crude feed density. (Other variables also relate to crude density and sulfur content as described in the main text.) Although it varies much less than *EI,* fuel mix emission intensity decreases as *EI,* crude feed density, and crude feed sulfur content increase, among these districts and years.

Table S2. Simplified mixing analysis for potential effects of anomalous oils on crude feeds.

		Refinery crude feed volume data reported[a]				Anomalous oil assumption[c]		Potential crude feed effect[d]	
		Potentially anomalous streams[b]			Other streams				
PADD	Year	Stream 1 (% vol.)	Stream 2 (% vol.)	Stream 3 (% vol.)	Other streams (% vol.)	Predicted by density, sulfur (factor)	Excess in anomalous oil (factor)	Crude feed predicted (factor)	Crude feed with anomaly (factor)
1	1999	16.59	14.62	10.82	57.97	1	2	1.00	1.27
1	2000	19.73	11.91	11.51	56.85	1	2	1.00	1.29
1	2001	20.49	12.87	11.51	55.13	1	2	1.00	1.30
1	2002	17.28	12.96	12.32	57.44	1	2	1.00	1.27
1	2003	21.93	14.15	13.46	50.46	1	2	1.00	1.32
1	2004	27.74	12.61	11.06	48.59	1	2	1.00	1.37
1	2005	29.46	13.42	11.68	45.44	1	2	1.00	1.39
1	2006	29.89	14.12	12.27	43.72	1	2	1.00	1.40
1	2007	26.88	17.86	11.21	44.05	1	2	1.00	1.39
1	2008	23.23	18.71	10.97	47.09	1	2	1.00	1.35
2	1999	24.01	5.50	4.49	66.00	1	2	1.00	1.28
2	2000	26.90	5.78	4.00	63.32	1	2	1.00	1.31
2	2001	29.08	5.84	3.33	61.75	1	2	1.00	1.33
2	2002	29.40	5.50	1.93	63.17	1	2	1.00	1.33
2	2003	30.82	5.57	2.52	61.09	1	2	1.00	1.34
2	2004	32.02	4.66	2.26	61.06	1	2	1.00	1.35
2	2005	31.35	3.99	2.46	62.20	1	2	1.00	1.34
2	2006	34.76	4.83	1.63	58.78	1	2	1.00	1.38
2	2007	34.73	4.97	2.17	58.13	1	2	1.00	1.38
2	2008	36.35	4.52	1.94	57.19	1	2	1.00	1.39
3	1999	16.50	14.22	11.78	57.50	1	2	1.00	1.27
3	2000	16.77	14.99	13.60	54.64	1	2	1.00	1.28
3	2001	17.72	15.26	14.84	52.18	1	2	1.00	1.29
3	2002	19.61	14.82	14.71	50.86	1	2	1.00	1.31
3	2003	20.18	14.82	14.64	50.36	1	2	1.00	1.31
3	2004	20.21	15.55	12.22	52.02	1	2	1.00	1.31
3	2005	20.52	14.40	11.24	53.84	1	2	1.00	1.31
3	2006	20.53	13.07	10.73	55.67	1	2	1.00	1.30
3	2007	18.39	13.28	11.69	56.64	1	2	1.00	1.28
3	2008	16.61	13.08	12.52	57.79	1	2	1.00	1.26
4	1999	29.57	70.13	0.30	0.00	1	2	1.00	1.65
4	2000	33.07	66.93	0.00	0.00	1	2	1.00	1.67
4	2001	38.31	61.69	0.00	0.00	1	2	1.00	1.69
4	2002	43.61	56.39	0.00	0.00	1	2	1.00	1.72
4	2003	47.16	52.84	0.00	0.00	1	2	1.00	1.74
4	2004	46.77	53.23	0.00	0.00	1	2	1.00	1.73
4	2005	48.29	51.71	0.00	0.00	1	2	1.00	1.74
4	2006	49.87	50.13	0.00	0.00	1	2	1.00	1.75
4	2007	50.99	49.01	0.00	0.00	1	2	1.00	1.75
4	2008	49.10	50.90	0.00	0.00	1	2	1.00	1.75
5	1999	31.84	5.02	3.25	59.89	1	2	1.00	1.35
5	2000	33.00	5.21	3.80	57.99	1	2	1.00	1.37
5	2001	31.84	5.44	4.25	58.47	1	2	1.00	1.36
5	2002	30.86	3.89	3.59	61.66	1	2	1.00	1.34
5	2003	27.61	8.74	3.75	59.90	1	2	1.00	1.33
5	2004	26.28	8.95	5.50	59.27	1	2	1.00	1.32
5	2005	25.14	10.90	6.48	57.48	1	2	1.00	1.32
5	2006	24.26	10.05	6.88	58.81	1	2	1.00	1.31
5	2007	24.68	9.16	5.92	60.24	1	2	1.00	1.31
5	2008	24.34	10.23	7.58	57.85	1	2	1.00	1.31

Legend and notes for Table S2.

Density and sulfur content can predict unreported characteristics of crude oils more reliably in well-mixed crude feeds than in poorly mixed crude feeds. When multiple streams each comprise a small portion of the feed, if an oil stream of divergent quality is present, it will have less potential to change the quality of the total crude feed. Table S2 presents results from a simplified four-component mixing analysis for potential effects of anomalous oils on the crude feeds processed in each district and year. These results indicate that the District 4 crude feed is less well mixed than those of other districts.

a. Refinery crude feed component streams, shown in percent of total crude feed volume for simplicity of presentation, are from USEIA data on gross crude oil inputs to atmospheric distillation and refinery crude oil imports *(S1, S3),* and California Energy Commission data on refinery inputs of crude produced in California *(S16).*

b. Potentially anomalous streams <u>might</u> be dominated by oils in which unreported characteristics that affect processing occur in anomalously high amounts. The three streams with highest potential to effect the crude feed in this way are shown for each district and year. Component streams of crude feeds are ranked based on their potential for anomalous oil and their volume. Oils from the Western Canadian Sedimentary Basin (WCSB) dominate the highest-ranked stream (stream 1) for districts 2 and 4. The WCSB oil stream includes substantial heavy oil and bitumen sources, which tend to be high in nitrogen and vanadium *(S17-19),* and some of this stream is partially pre-processed *(Table S3).* The other streams are ranked based on their volume and the assumption that oils from a single country of origin or U.S. region may originate from similar geology and have similar anomalies. This assumption is made to assess the reliability of predictions based on density and sulfur for these crude feeds where more complete data for specific crude feeds are not available, and may overstate the potential for anomalies in the crude feeds processed by districts 1, 2, 3 and 5. The origins *(S3, S16)* and ranks of streams are as follows.

District 1 streams are ranked by volume for country of origin, with Nigeria supplying the largest volume (stream 1) in all years. Stream 2 was from Canada, Angola or Saudi Arabia, stream 3 was from Saudi Arabia, Venezuela, Angola or Norway, and 17-21 countries supplied other streams processed in District 1 annually. District 2 processed Canadian crude as its largest import (stream 1) each year, and its other streams are ranked by volume for foreign country of origin. Stream 2 was from Saudi Arabia in all years, stream 3 was from Nigeria, Venezuela or Algeria, and 12-20 countries supplied other streams refined in District 2 annually. District 3 streams are ranked by volume for foreign country of origin, and Mexico supplied the largest of these inputs (stream 1) in all years. Streams 2 and 3 were from Saudi Arabia or Venezuela, and 25-38 countries supplied other streams refined in District 3 annually.

District 4 processed Canadian crude as its largest import stream in all years, with virtually all of the balance from the U.S., and little or none of its crude feed came from any other country. The Canadian stream (stream 1) is dominated by oils from the WCSB, which have known potential for anomalies. Specific origins of the equal or larger U.S. stream are not reported, however, parts of the WCSB and other oil deposits with similar geology are located in District 4 *(S17).* Limiting crude transport logistics in the landlocked Rocky Mountain states, which are unique to District 4 and help to explain the limited scope of its imports relative to those of other districts, might also result in reliance on locally produced U.S. feeds. This circumstantial evidence suggests, but does not confirm, the possibility that both the imported and domestic oils refined here might have similar anomalies. Because of this possibility the U.S. stream refined in District 4 is ranked second (stream 2).

District 5 processes substantial amounts of crude from California and Alaska. The California stream (stream 1) is larger than that from any single foreign country, and includes oils from the San Joaquin Valley, which tend to have high density relative to their sulfur content *(Table S9)*. The other streams are ranked by volume for foreign country of origin. Stream 2 was from Iraq or Saudi Arabia, stream 3 was from Ecuador, Iraq or Saudi Arabia, and 20-27 countries supplied other crude oil streams refined in District 5 annually.

c. An unreported characteristic that affects processing is assumed twice as abundant in the anomalous oil as predicted by the density and sulfur content of that oil. The assumed factor of two appears plausible based on the variability observed for nitrogen, vanadium and nickel in whole crude oils. For example, among all assays of crude oils by NETL after 1969 where density, sulfur, nitrogen and residua yield are reported (N = 728) *(S20),* the highest-divergent 1% of oils had 1.85 times as much nitrogen by weight as predicted by density and sulfur (nonparametric regression by LOWESS, $R^2 = 0.71$). Real anomalies could vary from this factor, but since it is applied to all districts and years, results will scale in proportion to the factor chosen. A lower or higher factor would thus decrease or increase values for all results, but would not change the results for any differences between districts and years. The predicted and (assumed) excess abundance of the unreported characteristic are shown, for the anomalous oil, in the columns under note (c).

d. These results estimate, for each district and year, the potential for crude feeds to have anomalous high content for unreported characteristics that are not predicted by crude feed density and sulfur. They do not show that any such anomaly actually occurred. Potential effects in the total refinery crude feed assume that the anomalous oil is 100% of stream 1, 50% of stream 2, and 25% of stream 3 for each district and year. The percentages are discounted sequentially because of the decreasing likelihood of the same anomaly in multiple separate streams. The predicted factor is assigned to the balance of the streams for each district and year. Results are shown as increases from the predicted crude feed factor of 1.00 on the right of the table.

Relatively well-mixed crude feeds limit the effect of the anomaly in districts 1, 2, 3 and 5 to less than half of its assumed magnitude in the anomalous oil stream. This compares with crude sulfur concentrations four to eight times those of nitrogen and 160 to 500 times those of nickel and vanadium *(S17)*. The ranges of annual estimates for these districts overlap, or adjoin for districts 3 and 5. However, the estimates for District 4 are significantly larger (range: 1.65-1.75) than those for the other districts (combined range: 1.26-1.40). Further, although estimates for the other districts represent an extreme case, the assumption that anomalous oil is 50% of stream 2 might understate the potential effects on the District 4 crude feed, in the event that its Canadian and U.S. inputs both have the same anomaly.

This estimate is limited by the simplified four-component blending analysis and anomalous oil stream assumptions described above, and although it shows that unpredicted anomalies are possible in the District 4 crude feed, it represents an extreme and unlikely scenario for districts 1, 2, 3 and 5.

Table S3. Estimate calculation for Canadian synthetic crude oil (SCO) exports to districts and years.

NR = Not reported

	units	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008
SCO yield from bitumen upgraders											
NEB Canada estimate[a]	$(m^3 \bullet 10^6)$	18.8	18.3	20.0	25.2	29.0	34.3	31.0	37.7	39.5	37.9
ERCB Alberta estimate[b]	$(m^3 \bullet 10^6)$	18.8	18.6	20.3	25.6	29.5	34.7	31.7	38.2	39.9	37.9
Upgrading method[a,b]											
Hydrocracking-based	$(m^3 \bullet 10^6)$	3.2	3.0	3.3	3.4	9.3	11.4	12.7	11.9	12.7	11.7
Coking-based	$(m^3 \bullet 10^6)$	15.6	15.6	16.9	22.1	21.9	23.3	19.0	26.3	27.1	26.2
SCO to Canadian refineries[c]											
All Canadian refining	$(m^3 \bullet 10^6)$	13.3	12.5	12.9	12.7	12.1	16.0	14.8	15.6	17.2	17.0
Alberta refineries	$(m^3 \bullet 10^6)$	10.1	9.9	10.4	9.6	8.6	11.8	11.8	12.3	13.8	13.0
Other refineries	$(m^3 \bullet 10^6)$	3.2	2.6	2.5	3.1	3.4	4.2	3.1	3.3	3.4	3.9
SCO removals from Alberta[b]	$(m^3 \bullet 10^6)$	8.8	7.4	8.9	14.2	17.4	21.1	18.9	24.1	25.0	25.0
Supply-demand balance											
Yield (NEB)-all refining	$(m^3 \bullet 10^6)$	5.5	5.8	7.1	12.5	16.9	18.3	16.2	22.0	22.3	20.9
Removals-other ref.	$(m^3 \bullet 10^6)$	5.6	4.8	6.4	11.1	14.0	16.9	15.8	20.8	21.6	21.1
Excess supply estimate	$(m^3 \bullet 10^6)$	5.6	5.8	7.1	12.5	16.9	18.3	16.2	22.0	22.3	21.1
Total SCO exports											
Estimated by NEB[d]	$(m^3 \bullet 10^6)$	6.5	NR	NR	9.4	NR	NR	17.5	NR	NR	19.5
SCO exports to U.S.											
Estimated by NEB[d]	$(m^3 \bullet 10^6)$	6.5	NR	NR	9.4	NR	NR	17.4	NR	NR	19.3
% of total exports	(%)	100.0			100.0			99.3			99.2
Estimated by interpolation with recent supply/export ratio[e]	$(m^3 \bullet 10^6)$	--	5.9	6.3	--	14.5	17.7	--	22.5	21.7	--
Consolidated estimate	$(m^3 \bullet 10^6)$	6.5	5.9	6.3	9.4	14.5	17.7	17.4	22.5	21.7	19.3
Supply-export balance	$(m^3 \bullet 10^6)$	-0.9	-0.1	0.8	3.2	2.4	0.6	-1.3	-0.5	0.7	1.8
SCO exports to U.S. refining districts estimated by NEB[d]											
PADD 1	$(m^3 \bullet 10^6)$	0.37	NR	NR	0.26	NR	NR	0.77	NR	NR	0.46
PADD 2	$(m^3 \bullet 10^6)$	5.36	NR	NR	6.02	NR	NR	11.89	NR	NR	13.68
PADD 3	$(m^3 \bullet 10^6)$	0.00	NR	NR	0.27	NR	NR	0.07	NR	NR	0.09
PADD 4	$(m^3 \bullet 10^6)$	0.77	NR	NR	2.36	NR	NR	3.25	NR	NR	2.49
PADD 5	$(m^3 \bullet 10^6)$	0.00	NR	NR	0.45	NR	NR	1.44	NR	NR	2.62
SCO exports to districts estimated by interpolation with to recent U.S. SCO portions[e]											
PADD 1	$(m^3 \bullet 10^6)$	--	0.28	0.24	--	0.49	0.69	--	0.84	0.66	--
PADD 2	$(m^3 \bullet 10^6)$	--	4.52	4.41	--	9.53	11.86	--	15.56	15.15	--
PADD 3	$(m^3 \bullet 10^6)$	--	0.06	0.12	--	0.30	0.22	--	0.10	0.10	--
PADD 4	$(m^3 \bullet 10^6)$	--	0.97	1.30	--	3.35	3.70	--	3.77	3.21	--
PADD 5	$(m^3 \bullet 10^6)$	--	0.10	0.20	--	0.87	1.26	--	2.26	2.55	--
U.S. exports–PADDs balance	$(m^3 \bullet 10^6)$	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Legend and notes for Table S3.

Table S3 shows data, reported exports, and calculated estimates for synthetic crude oil (SCO) volume exported from Canada and processed in each district and year. Reported SCO exports are estimates, and these are reported as annual volumes at three-year intervals. Values for the years 2000, 2001, 2003, 2004, 2006 and 2007 are estimated by interpolation based on reported data for the two proximate years. For example, reported data for 1999 and 2002 are used to estimate exported SCO processed in 2000. These estimates thus assume there was no unknown factor that changed the relationship of exports to supply or refinery capacity greatly between the estimated year and the years immediately before and after that year. Results indicate differences between districts in SCO inputs, increasing SCO inputs with time for districts 2 and 4, and that, especially in the earlier years, the SCO came mainly from coking-based upgraders. However, the exact volume and refining characteristics of SCO processed in specific districts and years is uncertain. Notes cited in the table further discuss the sources, data quality, and methods for estimates below.

a. The first estimate of annual SCO yield for 1999-2008 is from the National Energy Board of Canada (NEB) *(S21).*

b. The second estimate of SCO yield for 1999-2008, and yield by upgrading method for 2000-2008, are from the Energy Resources Conservation Board of Alberta (ERCB) *(S22).* Yield by upgrader in 1999 is from the NEB *(S21).* The exact volumes from coking- and hydrocracking-based upgrading are uncertain. One major upgrader that primarily uses the coking method also uses hydrocracking *(S22).* Most (75%) of the SCO yield from this upgrader is assigned to coking and 25% is assigned to hydrocracking in the table.

c. SCO inputs to Canadian refineries are from Statistics Canada *(S23)*. The agency reports these inputs for light SCO, however, some intermediate and heavy crude streams from the Western Canadian Sedimentary Basin (WCSB) are delivered as blends that may contain SCO. The SCO in such blends may not be reported, for some exports or refinery inputs.

d. SCO exports, including exports the U.S. and to each U.S. district, are estimated by the NEB for 1999 *(S24),* 2002 *(S25),* 2005 *(S26),* and 2008 *(S27).* The U.S. receives nearly all these exports, however, estimated exports do not balance exactly with the excess supply of SCO estimated to be available after Canadian usage of these oils. NEB export estimates appear to exceed available supply by 0.9 and 1.3 million m^3 in 1999 and 2005, while supply appears to exceed NEB export estimates by 3.2 and 1.8 million m^3 in 2002 and 2008, respectively. This is shown in the “supply-export balance” line of the table. These differences are small for some estimation purposes, but they approach or exceed the total exports estimated for some districts and years. Refining characteristics of the SCO exports are not reported.

e. Although reported only at three-year intervals, exports increase steadily with supply, and their apportionment among the districts changes little over these intervals. This is explained by the need for disposition of the SCO created, and the unique logistical constraints posed by transport and refining of SCO from the WCSB in each district. These constraints allow a rough estimate of the relative SCO volumes exported and refined in the intervening years.

   First, total U.S. exports are estimated for years when they are not reported. The excess supply estimate for each such year is multiplied by the weighted average fraction of supply exported in the two nearest reported years. This weighted average is calculated using a 2:1 ratio to give twice as much weight to the proximate year (e.g., 1999 for the 2000 estimate) and half as much weight to the year more distant in time (2002 in this example). The supply-export balance line of the table shows that these interpolated estimates generally compare more closely with excess supply than do the reported estimates.

SCO exports to districts are then estimated by apportioning the estimated total U.S. exports for the year to be estimated based on the weighted average of each district's share of total SCO exports in the two nearest reported years. This weighted average is calculated using a 2:1 ratio to give twice as much weight to the proximate year (e.g., 1999 for the 2000 estimate) and half as much weight to the year more distant in time (2002 in this example). The bottom line of the table shows that these SCO estimates for districts balance with total estimated SCO exports to the U.S. for each year.

These estimates should be interpreted with caution as discussed above. Nevertheless, they provide evidence that SCO comprised an appreciable portion of crude refined during some years in District 2, and especially District 4, which refines much less oil in total than other districts *(Table S1).* The estimates suggest that SCO accounts for more than 10% of District 4 crude feeds and up to 8% of District 2 feeds, in some years. Reported and estimated Canadian SCO accounted for less than 2% of the crude feeds processed in districts 1, 3 and 5 during 1999-2008.

Table S 4. Evidence for effects of synthetic oil (SCO) on refinery processing during 1999-2008 in District 4.

		Refinery observations for selected parameters[a]					SCO % vol. of refinery crude feed[b]	Predictions based on non-SCO feeds[c]				Energy for excess H_2 production/m^3 crude feed[d]	
								H_2 predicted by crude fd. density		csHydrotreating pred. by conv. cap.			
PADD	Year	Crude feed density (kg/m^3)	H^2 production capacity (m^3/m^3)	Conversion capacity (m^3/m^3)	Crude stream hydrotreating capacity (m^3/m^3)	Refinery energy intensity (*EI*) (GJ/m^3)	(%)	Predicted (m^3/m^3)	Excess (m^3/m^3)	Predicted (m^3/m^3)	Excess (m^3/m^3)	(GJ/m^3)	(% *EI*)
1	1999	858.20	13.25	0.516	0.054	3.451	0.41	19.60	--	0.122	--	--	--
1	2000	860.18	15.66	0.525	0.054	3.430	0.31	24.22	--	0.130	--	--	--
1	2001	866.34	12.71	0.481	0.029	3.518	0.28	38.66	--	0.094	--	--	--
1	2002	865.71	11.11	0.474	0.084	3.426	0.30	37.16	--	0.087	--	--	--
1	2003	863.44	16.49	0.474	0.059	3.364	0.53	31.83	--	0.087	--	--	--
1	2004	865.44	16.52	0.475	0.059	3.416	0.76	36.54	--	0.088	--	--	--
1	2005	863.38	16.59	0.476	0.058	3.404	0.83	31.70	--	0.089	--	--	--
1	2006	864.12	18.83	0.476	0.028	3.440	0.98	33.44	--	0.090	--	--	--
1	2007	864.33	14.46	0.476	0.028	3.499	0.77	33.93	--	0.090	--	--	--
1	2008	863.65	14.46	0.476	0.028	3.551	0.57	32.32	--	0.090	--	--	--
2	1999	858.25	21.23	0.486	0.125	3.368	2.74	19.73	1.50	0.097	0.028	0.022	0.66
2	2000	860.03	21.17	0.488	0.107	3.361	2.28	23.85	--	0.099	0.008	--	--
2	2001	861.33	23.18	0.485	0.096	3.396	2.30	26.91	--	0.096	--	--	--
2	2002	861.02	21.58	0.481	0.129	3.393	3.22	26.17	--	0.093	0.035	--	--
2	2003	862.80	20.02	0.477	0.132	3.298	5.09	30.35	--	0.090	0.043	--	--
2	2004	865.65	20.25	0.473	0.148	3.376	6.19	37.04	--	0.087	0.061	--	--
2	2005	865.65	24.07	0.484	0.148	3.496	6.18	37.04	--	0.096	0.052	--	--
2	2006	865.44	37.33	0.488	0.140	3.738	8.10	36.54	0.79	0.099	0.042	0.012	0.31
2	2007	864.07	36.89	0.479	0.137	3.800	8.06	33.31	3.58	0.092	0.045	0.053	1.39
2	2008	862.59	37.12	0.487	0.146	3.858	7.27	29.85	7.26	0.098	0.047	0.107	2.78
3	1999	869.00	32.51	0.566	0.151	4.546	0.00	44.95	--	0.165	--	--	--
3	2000	870.29	33.03	0.579	0.155	4.563	0.01	47.99	--	0.175	--	--	--
3	2001	874.43	34.50	0.600	0.129	4.348	0.03	57.86	--	0.193	--	--	--
3	2002	876.70	34.95	0.611	0.148	4.434	0.07	63.32	--	0.203	--	--	--
3	2003	874.48	34.66	0.604	0.168	4.381	0.07	57.99	--	0.196	--	--	--
3	2004	877.79	37.31	0.610	0.174	4.204	0.05	65.94	--	0.201	--	--	--
3	2005	878.01	35.69	0.588	0.168	4.205	0.02	66.46	--	0.183	--	--	--
3	2006	875.67	33.33	0.587	0.167	4.367	0.02	60.85	--	0.182	--	--	--
3	2007	876.98	32.83	0.594	0.184	4.226	0.02	63.97	--	0.188	--	--	--
3	2008	878.66	33.64	0.600	0.171	4.361	0.02	68.04	--	0.193	--	--	--
4	1999	854.47	28.31	0.415	0.112	3.843	2.64	10.96	17.34	0.040	0.073	0.256	6.66
4	2000	859.35	30.44	0.426	0.092	3.698	3.25	22.27	8.17	0.049	0.043	0.121	3.26
4	2001	859.19	29.92	0.421	0.050	3.846	4.43	21.91	8.01	0.045	0.005	0.118	3.07
4	2002	860.23	29.09	0.404	0.087	3.726	7.73	24.34	4.75	0.031	0.056	0.070	1.88
4	2003	861.23	27.94	0.408	0.087	3.833	10.86	26.66	1.28	0.034	0.053	0.019	0.49
4	2004	862.59	28.02	0.419	0.090	3.644	11.44	29.85	--	0.043	0.047	--	--
4	2005	862.91	41.87	0.407	0.093	3.830	9.98	30.59	11.28	0.034	0.060	0.167	4.35
4	2006	860.50	38.16	0.408	0.109	3.955	11.67	24.95	13.21	0.034	0.075	0.195	4.93
4	2007	862.38	49.76	0.415	0.109	4.159	10.13	29.36	20.39	0.040	0.069	0.301	7.24
4	2008	863.12	59.86	0.409	0.136	4.475	7.94	31.09	28.78	0.035	0.101	0.425	9.49
5	1999	894.61	69.93	0.613	0.195	4.908	0.00	107.06	--	0.204	--	--	--
5	2000	895.85	83.53	0.613	0.167	5.189	0.06	110.15	--	0.204	--	--	--
5	2001	893.76	82.53	0.619	0.174	5.039	0.13	104.95	--	0.209	--	--	--
5	2002	889.99	85.44	0.636	0.196	4.881	0.28	95.65	--	0.224	--	--	--
5	2003	889.10	83.17	0.620	0.165	4.885	0.52	93.45	--	0.210	--	--	--
5	2004	888.87	83.17	0.627	0.167	4.861	0.76	92.90	--	0.216	--	--	--
5	2005	888.99	83.44	0.626	0.166	4.774	0.86	93.18	--	0.216	--	--	--
5	2006	887.65	88.20	0.641	0.160	4.862	1.35	89.89	--	0.228	--	--	--
5	2007	885.54	89.90	0.656	0.167	5.091	1.58	84.73	5.17	0.242	--	0.076	1.50
5	2008	890.16	89.68	0.645	0.163	4.939	1.60	96.07	--	0.232	--	--	--

Legend and notes for Table S4.

Table S4 presents results from analysis of synthetic crude oil (SCO) effects on refining. Canadian export estimates *(Table S3)* suggest that during 1999-2008 SCO from Western Canada was 2-8% and 2-12% of crude feeds in districts 2 and 4, respectively. This SCO stream yields more and lower quality gas oil as compared with typical whole crude oils, and can require more hydroprocessing in refineries *(S24, S25).* Crude density correlates with hydrogen demand for crude oils generally but does not correlate well for some SCO *(S14).* Reported hydrogen capacity is compared with that predicted by crude feed density, and reported crude stream hydrotreating capacity is compared with that predicted by conversion capacity, among districts and years. Crude stream hydrotreating processes gas oil, residua and catalytic cracking feeds *(Table S1).* These comparisons provide information about the relationship of hydrogen production to hydrogen use in processing gas oil, including gas oil from refinery SCO inputs. Hydrogen production in excess of that predicted by crude feed density is then compared with total refinery processing requirements on an energy basis.

Results suggest that SCO affects hydroprocessing and hydrogen production in refineries and may have increased refinery energy intensity significantly during some years in District 4. Hydrogen excesses are found only when SCO was present in crude feeds, and are found during four years in District 2 and nine years in District 4. Hydrotreating excesses are found only when estimated SCO inputs exceeded 2% of crude feeds and occurred during nine years in District 2 and ten years in District 4. The magnitude of hydrogen excesses generally increased with that of hydrotreating excesses and both were larger in District 4 than in District 2. Energy use for excess hydrogen production was minimal in District 2, but in District 4 it exceeded 5% of total refinery energy consumed during three years, and exceeded 9% of total refinery energy in 2008. The magnitude of hydrogen excesses is not well correlated with the estimated percentage of SCO in crude feeds, especially in District 4. The extent to which this poor correlation reflects unreported changes in the quality of SCO inputs, unreported changes in the quality of the balance of the poorly-mixed District 4 crude feed *(Table S2),* or errors in SCO volume estimates *(Table S3),* could not be determined with available data.

a. Refinery observations shown on the left of the table are based on the data given in Table S1. Capacities/m^3 atmospheric distillation capacity are shown.
b. The percentage of total refinery crude feed volume comprised of SCO is estimated based on estimated SCO exports from Table S3 and reported total crude inputs from Table S1. The SCO export estimates are uncertain, as detailed in Table S3.
c. Predictions shown are from PLS regression on all data for districts where estimated SCO inputs never exceeded 2% of total crude feeds during 1999-2008 (districts 1, 3 and 5). R-squared values are 0.88 for hydrogen production capacity predicted by crude feed density, and 0.85 for crude stream hydrotreating capacity predicted by conversion capacity. These predictions are "blind" to the presence of SCO in that it was not included as a variable in either of these two PLS models. Predictions and excesses shown are based on the upper 95% confidence for observations. Observed values exceed the lower 95% confidence (not shown) for all comparisons.
d. Energy consumed for the excess in hydrogen production capacity, which is shown as cubic meters H_2/m^3 atmospheric distillation capacity in this table, is calculated using the energy (16.4 MJ/m^3 H_2) and capacity utilization (90%) factors from Table S1.

Table S5. Efficiency factors for processing refinery products.

Product	Efficiency factor (%)	Average specific gravity
Light liquids		
Gasoline	86.4	0.737
Diesel	91.0	0.845
Kerosine	92.2	0.814
Naphtha	92.7	0.756
Other products		
Lube stocks	80.5	0.889
Waxes	80.5	0.799
Asphalt	84.9	1.038
Coke	86.3	0.967
Fuel gas	90.0	0.844
Heavy fuel oil	91.0	0.946
LPG	92.7	0.539
Residual oil	94.1	0.946

Legend and notes for Table S5.

Product-specific processing energy efficiency factors for a current typical U.S. refinery (mass-based) from reference *S11,* and average specific gravities of North American products from reference *S28*. These values were used with yield data from Table S1 to estimate energy use for products processing (“Eproducts”). The Eproducts estimates for refining districts and years are used, with *S, d,* capacity utilized, and products ratio observations from data in Table S1, in the Eproducts v. *OQ* comparison reported in Table 1 of the main text.

Table S6. Estimate calculation, oil quality and processing *EI* including bitumen upgrading.

		Refinery observations[a]						Synthetic crude oil input estimate[b]		Coking : hydrocracking[b]
PADD	Year	Crude input (m^3/d•10^4)	Density (kg/m^3)	*S* (kg/m^3)	*EI* (GJ/m^3)	Cap. utilized (%)	Prod. ratio (ratio)	(m^3/d•10^4)	(%)	(ratio)
1	1999	24.436	858.20	8.24	3.451	90.9	3.668	0.101	0.41	4.8
1	2000	24.754	860.18	8.00	3.430	91.7	3.489	0.077	0.31	5.2
1	2001	23.546	866.34	7.71	3.518	87.2	3.479	0.065	0.28	5.2
1	2002	24.246	865.71	7.45	3.426	88.9	3.605	0.073	0.30	6.5
1	2003	25.184	863.44	7.43	3.364	92.7	3.321	0.134	0.53	2.4
1	2004	24.961	865.44	7.79	3.416	90.4	3.398	0.190	0.76	2.0
1	2005	25.422	863.38	7.17	3.404	93.1	3.756	0.212	0.83	1.5
1	2006	23.626	864.12	7.17	3.440	86.7	3.522	0.231	0.98	2.2
1	2007	23.419	864.33	7.26	3.499	85.6	3.443	0.181	0.77	2.1
1	2008	22.115	863.65	7.08	3.551	80.8	3.400	0.125	0.57	2.2
2	1999	53.626	858.25	10.64	3.368	93.3	4.077	1.469	2.74	4.8
2	2000	54.215	860.03	11.35	3.361	94.2	4.132	1.238	2.28	5.2
2	2001	52.609	861.33	11.37	3.396	93.9	4.313	1.210	2.30	5.2
2	2002	51.162	861.02	11.28	3.393	90.0	4.345	1.648	3.22	6.5
2	2003	51.258	862.80	11.65	3.298	91.6	4.281	2.611	5.09	2.4
2	2004	52.482	865.65	11.86	3.376	93.6	4.167	3.250	6.19	2.0
2	2005	52.688	865.65	11.95	3.496	92.9	4.207	3.258	6.18	1.5
2	2006	52.609	865.44	11.60	3.738	92.4	3.907	4.264	8.10	2.2
2	2007	51.480	864.07	11.84	3.800	90.1	4.161	4.152	8.06	2.1
2	2008	51.575	862.59	11.73	3.858	88.4	4.333	3.747	7.27	2.2
3	1999	111.689	869.00	12.86	4.546	94.7	3.120	0.000	0.00	4.8
3	2000	113.024	870.29	12.97	4.563	93.9	3.120	0.015	0.01	5.2
3	2001	115.600	874.43	14.34	4.348	94.8	3.128	0.033	0.03	5.2
3	2002	112.786	876.70	14.47	4.434	91.5	3.251	0.073	0.07	6.5
3	2003	116.013	874.48	14.43	4.381	93.6	3.160	0.081	0.07	2.4
3	2004	119.145	877.79	14.40	4.204	94.1	3.228	0.060	0.05	2.0
3	2005	114.534	878.01	14.40	4.205	88.3	3.316	0.020	0.02	1.5
3	2006	117.253	875.67	14.36	4.367	88.7	3.176	0.027	0.02	2.2
3	2007	117.682	876.98	14.47	4.226	88.7	3.205	0.027	0.02	2.1
3	2008	111.879	878.66	14.94	4.361	83.6	3.229	0.026	0.02	2.2
5	1999	41.973	894.61	11.09	4.908	87.1	2.952	0.001	0.00	4.8
5	2000	43.086	895.85	10.84	5.189	87.5	3.160	0.027	0.06	5.2
5	2001	44.262	893.76	10.99	5.039	89.1	3.231	0.056	0.13	5.2
5	2002	44.787	889.99	10.86	4.881	90.0	3.460	0.124	0.28	6.5
5	2003	45.661	889.10	10.94	4.885	91.3	3.487	0.238	0.52	2.4
5	2004	45.486	888.87	11.20	4.861	90.4	3.551	0.345	0.76	2.0
5	2005	46.090	888.99	11.38	4.774	91.7	3.700	0.394	0.86	1.5
5	2006	45.693	887.65	10.92	4.862	90.5	3.615	0.618	1.35	2.2
5	2007	44.373	885.54	11.07	5.091	87.6	3.551	0.700	1.58	2.1
5	2008	44.739	890.16	12.11	4.939	88.1	3.803	0.717	1.60	2.2
4	1999	8.029	854.47	11.71	3.843	95.1	3.910	0.212	2.64	4.8
4	2000	8.156	859.35	12.03	3.698	94.7	3.943	0.265	3.25	5.2
4	2001	8.077	859.19	11.08	3.846	90.7	3.986	0.357	4.43	5.2
4	2002	8.363	860.23	12.04	3.726	91.6	4.078	0.647	7.73	6.5
4	2003	8.442	861.23	12.49	3.833	91.9	3.962	0.917	10.86	2.4
4	2004	8.856	862.59	11.65	3.644	95.7	3.981	1.013	11.44	2.0
4	2005	8.935	862.91	11.22	3.830	95.5	3.887	0.892	9.98	1.5
4	2006	8.856	860.50	11.36	3.955	93.5	3.962	1.033	11.67	2.2
4	2007	8.681	862.38	11.73	4.159	91.3	3.900	0.879	10.13	2.1
4	2008	8.585	863.12	12.17	4.475	89.4	4.291	0.682	7.94	2.2

Table S6. Estimate calculation, oil quality and processing *EI* including bitumen upgrading.
Continued

		Bitumen upgrading estimate[c]			Upgrading and refining estimate				Estimate compared to *OQ* prediction[k]
PADD	Year	Density_{add}[d] (kg/m^3)	S_{add}[e] (kg/m^3)	EI_{add}[f] (GJ/m^3)	Density_{adj}[g] (kg/m^3)	S_{adj}[h] (kg/m^3)	EI_{adj}[i] (GJ/m^3)	EI_{tp}[j] (GJ/m^3)	(±% 95% Conf.)
1	1999	0.45	0.17	0.022	858.65	8.41	3.473	3.271	--
1	2000	0.34	0.13	0.017	860.52	8.13	3.447	3.372	--
1	2001	0.30	0.11	0.015	866.65	7.82	3.533	3.579	--
1	2002	0.33	0.12	0.016	866.04	7.57	3.442	3.533	--
1	2003	0.57	0.22	0.030	864.01	7.65	3.394	3.531	--
1	2004	0.81	0.32	0.043	866.25	8.11	3.459	3.623	--
1	2005	0.87	0.35	0.048	864.26	7.52	3.452	3.470	--
1	2006	1.04	0.41	0.055	865.17	7.58	3.495	3.488	--
1	2007	0.82	0.32	0.044	865.16	7.58	3.543	3.489	--
1	2008	0.60	0.24	0.032	864.25	7.32	3.583	3.393	--
2	1999	3.00	1.12	0.148	861.26	11.76	3.516	3.546	--
2	2000	2.51	0.93	0.123	862.53	12.28	3.484	3.634	--
2	2001	2.52	0.94	0.124	863.86	12.31	3.520	3.662	--
2	2002	3.56	1.30	0.172	864.58	12.58	3.565	3.667	--
2	2003	5.45	2.12	0.285	868.25	13.77	3.583	3.925	--
2	2004	6.58	2.59	0.349	872.24	14.45	3.725	4.179	-2%
2	2005	6.48	2.62	0.355	872.14	14.57	3.852	4.168	--
2	2006	8.65	3.39	0.455	874.09	14.98	4.193	4.321	--
2	2007	8.59	3.37	0.454	872.66	15.21	4.254	4.210	--
2	2008	7.75	3.03	0.408	870.35	14.76	4.266	4.038	--
3	1999	0.00	0.00	0.000	869.00	12.86	4.546	4.117	2%
3	2000	0.02	0.01	0.001	870.30	12.97	4.563	4.173	1%
3	2001	0.03	0.01	0.002	874.46	14.35	4.350	4.446	--
3	2002	0.07	0.03	0.004	876.78	14.49	4.437	4.504	--
3	2003	0.07	0.03	0.004	874.56	14.46	4.385	4.440	--
3	2004	0.05	0.02	0.003	877.84	14.42	4.207	4.575	--
3	2005	0.02	0.01	0.001	878.03	14.41	4.206	4.512	--
3	2006	0.02	0.01	0.001	875.70	14.37	4.369	4.434	--
3	2007	0.02	0.01	0.001	877.00	14.48	4.227	4.493	--
3	2008	0.02	0.01	0.001	878.69	14.95	4.362	4.541	--
5	1999	0.00	0.00	0.000	894.61	11.09	4.909	5.082	--
5	2000	0.07	0.03	0.003	895.92	10.87	5.192	5.097	--
5	2001	0.14	0.05	0.007	893.90	11.04	5.046	5.023	--
5	2002	0.31	0.11	0.015	890.30	10.97	4.896	4.834	--
5	2003	0.56	0.22	0.029	889.65	11.15	4.914	4.825	--
5	2004	0.81	0.32	0.043	889.68	11.52	4.903	4.830	--
5	2005	0.90	0.36	0.049	889.88	11.74	4.824	4.841	--
5	2006	1.44	0.57	0.076	889.09	11.48	4.938	4.793	--
5	2007	1.68	0.66	0.089	887.22	11.73	5.180	4.707	2%
5	2008	1.71	0.67	0.090	891.87	12.78	5.029	4.939	--
4	1999	2.89	1.08	0.143	857.36	12.78	3.986	3.482	4%
4	2000	3.57	1.32	0.175	862.91	13.35	3.873	3.750	--
4	2001	4.86	1.80	0.239	864.05	12.88	4.085	3.726	--
4	2002	8.54	3.13	0.414	868.78	15.17	4.139	4.065	--
4	2003	11.62	4.53	0.608	872.85	17.01	4.441	4.377	--
4	2004	12.16	4.79	0.645	874.76	16.44	4.289	4.459	--
4	2005	10.46	4.23	0.574	873.37	15.45	4.404	4.352	--
4	2006	12.45	4.87	0.655	872.94	16.23	4.610	4.349	--
4	2007	10.79	4.24	0.570	873.17	15.96	4.729	4.331	1%
4	2008	8.47	3.31	0.446	871.59	15.48	4.921	4.152	9%

Legend and notes for Table S6.

Table S6 presents an estimate of oil quality and processing energy for total oil processing, including refining and pre-processing for that portion of refinery crude feeds comprised of synthetic crude oil (SCO), for each district and year. Coking- and hydrocracking-based bitumen upgrading uses energy to yield SCO of lower density and sulfur content than the bitumen. SCO imported from Western Canada accounts for an estimated 2-8% of total District 2 crude feeds and 2-12% of total District 4 feeds during 1999-2008. Refinery crude feeds and energy consumption do not reflect the original bitumen quality for this SCO or the energy consumed in its upgrading. The estimate shown in this table relates initial oil quality to process energy for total processing. The energy consumed and density and sulfur lost in upgrading is estimated based on process modeling data and added "back" to the refinery crude feed and energy consumption observed. The estimated total process energy is then compared to that predicted by the initial oil quality. Results suggest that in general, total process energy increases with worsening initial oil quality consistent with the prediction based on observed refinery data. The exception involves two results for District 4. This is discussed in note (k).

a. Refinery feed volume, density, sulfur content (*S*), capacity utilization, and products ratio (calculated as described in the main paper) are from data in Table S1.
b. Synthetic crude oil (SCO) inputs and sources by upgrader type are from the estimates detailed in Table S3. The volume, percentage of total refinery crude feed volume, and ratio of coking- to hydrocracking-based upgrading for the SCO are shown.
c. SCO was produced from bitumen in Western Canada by coking-based and hydrocracking-based upgrading *(S22)*. Both upgrading schemes typically also use atmospheric and vacuum

distillation and significant hydrotreating, sulfur recovery and hydrogen production. Material and energy inputs and outputs were estimated using process modeling of typical coking-based and hydrocracking-based upgraders yielding SCO from Athabasca bitumen by Keesom et al. *(S14).* Modeled parameters included, among others, bitumen feed density (1.011 t/m^3) and sulfur content (48.64 kg/m^3), SCO yield (22,259 m^3/d), and SCO density and sulfur content for the coking-based (881.07 kg/m^3 *d,* 3.23 kg/m^3 *S*) and hydrocracking-based (921.82 kg/m^3 *d,* 3.23 kg/m^3 *S*) schemes. Carbon rejection, hydrogen addition and utility energy inputs estimated by process modeling on these parameters were 4,773 GJ/h for the coking-based scheme and 6,155 GJ/h for the hydrocracking-based scheme *(S14).* This indicates energy inputs of approximately 0.04 GJ per kg density (including sulfur) lost from the feed in the SCO from the coking-based scheme, and 0.07 GJ/kg for that from the hydrocracking scheme. Energy inputs were not allocated to sulfur removal separately from density reduction in the reported results.

Bitumen feed to the coking- and hydrocracking-based schemes was modeled at 1.15 times and 0.97 times the SCO volume yield, respectively *(S14).* Thus, on a product volume basis, estimated energy use was approximately 5.15 and 6.64 GJ per m^3 SCO produced for the coking- and hydrocracking-based upgraders, respectively. SCO from the coking- and hydrocracking-based schemes was 130.22 and 89.47 kg/m^3 lighter than the bitumen feed, respectively, and both schemes produced SCO with 45.41 kg/m^3 less sulfur than the bitumen feed. These estimates are applied to the shares of SCO from coking- and hydrocracking-based upgrading each year to estimate initial oil quality and total process energy. Notes d through f detail the calculations.

d. Density lost in upgrading the bitumen ($\text{Density}_{\text{add}}$) is added to the total refinery crude feed density to account for the bitumen input processed upstream to produce the SCO. $\text{Density}_{\text{add}}$ is calculated as:

$\text{Density}_{\text{add}} = \text{SCOvol} \bullet (\text{DR} \div \text{VC})$

Where

SCOvol is the percentage of SCO in the total refinery crude feed; DR is the density reduction from bitumen from note (c) in kg/m^3; VC is the volume change from bitumen to SCO from note (c); and the result is in kg/m^3 refinery crude feed.

e. Sulfur lost in upgrading the bitumen (S_{add}) is added to the total refinery crude feed sulfur to account for the bitumen input processed upstream to produce the SCO. S_{add} is calculated as:

$S_{\text{add}} = \text{SCOvol} \bullet (45.41 \div \text{VC})$

Where

SCOvol is the percentage of SCO in the total refinery crude feed; 45.41 is the sulfur content reduction from bitumen from note (c) in kg/m^3; VC is the volume change from bitumen to SCO from note (c); and the result is in kg/m^3 refinery crude feed.

f. Energy lost in upgrading the bitumen (EI_{add}) is added to the refinery energy intensity calculated from the data in Table S1 (EI) to estimate the total energy intensity of processing the oil feed. EI_{add} is calculated as:

$EI_{\text{add}} = \text{SCOvol} \bullet \text{EC}$

Where

SCOvol is the percentage of SCO in the total refinery crude feed; EC is the energy consumed by upgrading in GJ/m^3 SCO from note (c); and the result is expressed as GJ/m^3 refinery crude feed.

g. $\text{Density}_{\text{adj}}$ is the sum of crude feed density and $\text{Density}_{\text{add}}$ and is an estimate of initial crude feed quality accounting for the bitumen feed upgraded to produce SCO refined.

h. S_{adj} is the sum of S and S_{add} and is an estimate of initial crude feed quality accounting for the quality of the bitumen feed upgraded to produce SCO processed in a refinery.

i. EI_{adj} is the sum of EI and EI_{add} and is an estimate of the total energy intensity of processing including upgrading and refining.

j. EI_{tp} is the total *predicted* energy intensity of upgrading and processing and is an estimate of the total energy intensity predicted by the relationship of EI to crude feed density and sulfur based on the refinery observations. EI_{tp} is the result from inputting S_{adj}, $\text{Density}_{\text{adj}}$, product ratio and capacity utilized to the prediction mode of the PLS model, which is run on the observations from districts 1, 2, 3 and 5. EI_{tp} is compared with EI_{adj} in the final column of the table (note k) and in Figure 2.

k. The final column of the table compares estimated total processing energy (EI_{adj}) with total processing energy predicted by initial oil quality (EI_{tp}). Dashed lines (--) show that the result for estimated energy falls within the 95% confidence of prediction for observations. Negative values (e.g., -1%) show the percentage by which any result falls below the 95% confidence of prediction. Positive values (e.g., 1%) show the percentage by which any result exceeds the 95% confidence of prediction.

Estimated EI_{adj} is within the prediction based on oil quality or within 3% of its confidence interval in 48 of 50 cases. The exceptions are excesses for the years 1999 and 2008 in District 4. These excesses can be attributed to high excess hydrogen production in District 4 during those years *(Table S4).* It is possible that those high hydrogen values were related to increased hydroprocessing needs for SCO, or for some other anomaly, in the District 4 crude feed during those years. The need for hydrogen addition to address the poor gas oil and distillate product qualities of SCO *(S24, S25)* and its variable quality *(S14, S24)* support this possibility. This possibility cannot be confirmed or excluded, because the SCO input volume is uncertain *(Table S3),* its quality is unknown, and there is a potential for other sources of variability in the poorly-mixed District 4 crude feed *(Table S2).*

Table S7. Contribution of CO_2 to CO_2e emitted by oil refineries.

	Units	CO_2	CH_4	N_2O
Refinery emissions mass				
Scope				
U.S. (NETL)	Mt/y	257.90	0.1656	0.0040
U.S. (EPA)	Mt/y	228.53	0.0873	0.0007
California	Mt/y	35.54	0.0015	0.0001
Global warming potential				
20-yr. horizon	Factor	1	62	275
100-yr. horizon	Factor	1	23	296
500-yr. horizon	Factor	1	7	156
20-yr. horizon CO_2e				
U.S. (NETL)	Mt/y	257.90	10.27	1.11
U.S. (EPA)	Mt/y	228.53	5.41	0.19
California	Mt/y	35.54	0.09	0.03
100-yr. horizon CO_2e				
U.S. (NETL)	Mt/y	257.90	3.81	1.19
U.S. (EPA)	Mt/y	228.53	2.01	0.21
California	Mt/y	35.54	0.03	0.03
500-yr. horizon CO_2e				
U.S. (NETL)	Mt/y	257.90	1.16	0.63
U.S. (EPA)	Mt/y	228.53	0.61	0.11
California	Mt/y	35.54	0.01	0.02
Range of percent total CO_2e				
20-yr. horizon	Percent	95.78-99.66	0.26-3.81	0.08-0.41
100-yr. horizon	Percent	98.10-99.82	0.10-1.45	0.08-0.45
500-yr. horizon	Percent	99.31-99.93	0.03-0.45	0.04-0.24

Legend and notes for Table S7. (Mt/y, megatons per year.) U.S. refinery emission estimates are reported as mass emitted (NETL) *(S25)* and as CO_2e emitted (EPA) *(S29)*. California refinery emissions are reported as mass emitted *(S30)*. Global warming potential is from the Intergovernmental Panel on Climate Change *(S31)*. The U.S. (EPA) emissions mass estimate is calculated from reported CO_2e *(S29)* and 100-year global warming potential *(S31)*. The percent of total CO_2e from CO_2 and the small differences between estimates shown in Table S7 support the finding that CO_2 dominates refinery greenhouse gas emissions.

Table S8. PLS inputs for CO_2 emissions predicted by *OQ,* and comparison emission estimates.
(NA, not applicable; value predicted by *OQ*)

PADD	Year	Density (kg/m^3)	Sulfur (kg/m^3)	Cap. utlzd. (%)	Prod. ratio (ratio)	Observed *EI* (GJ/m^3)	Predicted *EI* (95% conf.) Lower (GJ/m^3)	Predicted *EI* (95% conf.) Central (GJ/m^3)	Predicted *EI* (95% conf.) Upper (GJ/m^3)	Fuel mix em. intensity (kg/GJ)
1	1999	858.20	8.24	90.9	3.668	3.451	2.877	3.241	3.604	81.53
1	2000	860.18	8.00	91.7	3.489	3.430	2.987	3.349	3.711	80.34
1	2001	866.34	7.71	87.2	3.479	3.518	3.198	3.559	3.919	81.85
1	2002	865.71	7.45	88.9	3.605	3.426	3.152	3.511	3.870	81.08
1	2003	863.44	7.43	92.7	3.321	3.364	3.133	3.493	3.853	81.51
1	2004	865.44	7.79	90.4	3.398	3.416	3.209	3.568	3.927	81.46
1	2005	863.38	7.17	93.1	3.756	3.404	3.048	3.410	3.772	81.23
1	2006	864.12	7.17	86.7	3.522	3.440	3.054	3.417	3.780	80.40
1	2007	864.33	7.26	85.6	3.443	3.499	3.067	3.433	3.800	82.28
1	2008	863.65	7.08	80.8	3.400	3.551	2.972	3.352	3.733	83.26
2	1999	858.25	10.64	93.3	4.077	3.368	2.984	3.347	3.711	78.11
2	2000	860.03	11.35	94.2	4.132	3.361	3.104	3.468	3.832	77.56
2	2001	861.33	11.37	93.9	4.313	3.396	3.126	3.495	3.863	77.46
2	2002	861.02	11.28	90.0	4.345	3.393	3.068	3.432	3.796	77.90
2	2003	862.80	11.65	91.6	4.281	3.298	3.195	3.558	3.922	78.00
2	2004	865.65	11.86	93.6	4.167	3.376	3.369	3.733	4.098	77.25
2	2005	865.65	11.95	92.9	4.207	3.496	3.362	3.725	4.089	77.27
2	2006	865.44	11.60	92.4	3.907	3.738	3.380	3.738	4.095	75.84
2	2007	864.07	11.84	90.1	4.161	3.800	3.270	3.629	3.989	75.55
2	2008	862.59	11.73	88.4	4.333	3.858	3.154	3.515	3.875	74.97
3	1999	869.00	12.86	94.7	3.120	4.546	3.759	4.117	4.476	71.61
3	2000	870.29	12.97	93.9	3.120	4.563	3.813	4.172	4.531	71.87
3	2001	874.43	14.34	94.8	3.128	4.348	4.085	4.444	4.803	72.43
3	2002	876.70	14.47	91.5	3.251	4.434	4.140	4.499	4.859	72.71
3	2003	874.48	14.43	93.6	3.160	4.381	4.076	4.435	4.794	72.81
3	2004	877.79	14.40	94.1	3.228	4.204	4.213	4.572	4.930	73.43
3	2005	878.01	14.40	88.3	3.316	4.205	4.149	4.511	4.873	73.24
3	2006	875.67	14.36	88.7	3.176	4.367	4.067	4.432	4.798	74.15
3	2007	876.98	14.47	88.7	3.205	4.226	4.127	4.491	4.856	74.93
3	2008	878.66	14.94	83.6	3.229	4.361	4.165	4.540	4.915	74.48
5	1999	894.61	11.09	87.1	2.952	4.908	4.713	5.082	5.451	70.27
5	2000	895.85	10.84	87.5	3.160	5.189	4.725	5.092	5.460	69.09
5	2001	893.76	10.99	89.1	3.231	5.039	4.648	5.014	5.380	69.38
5	2002	889.99	10.86	90.0	3.460	4.881	4.450	4.814	5.178	69.15
5	2003	889.10	10.94	91.3	3.487	4.885	4.422	4.788	5.153	69.40
5	2004	888.87	11.20	90.4	3.551	4.861	4.410	4.775	5.140	69.89
5	2005	888.99	11.38	91.7	3.700	4.774	4.409	4.780	5.151	69.88
5	2006	887.65	10.92	90.5	3.615	4.862	4.331	4.695	5.060	69.32
5	2007	885.54	11.07	87.6	3.551	5.091	4.235	4.594	4.953	69.12
5	2008	890.16	12.11	88.1	3.803	4.939	4.456	4.824	5.191	68.39
Other inputs										
US	2002	873.89	12.32	90.7	3.534	NA	3.838	4.194	4.549	73.62
US	2005	875.08	12.43	90.6	3.597	NA	3.885	4.241	4.597	73.98
US	2006	873.78	12.32	89.7	3.458	NA	3.835	4.191	4.547	73.94
US	2007	873.89	12.50	88.5	3.485	NA	3.833	4.190	4.547	74.34
SFBA	2008	899.66	11.91	90.8	3.469	NA	4.938	5.307	5.676	68.39
Heavy oil		957.40	27.80	90.8	3.469	NA	8.228	8.795	9.363	73.77
Nat. bitumen		1 033.60	45.50	90.8	3.469	NA	12.266	13.200	14.135	73.77

Table S8. PLS inputs for CO_2 emissions predicted by *OQ,* and comparison emission estimates, *continued.*

		Central *EI* prediction	Fuel mix em. intensity	Observed emissions	Predicted emissions (95% conf.)			Comp-arison
PADD	Year	(GJ/m^3)	(kg/GJ)	(kg/m^3)	Lower (kg/m^3)	Central (kg/m^3)	Upper (kg/m^3)	(± % CI)
1	1999	3.241	81.53	281	243	265	287	--
1	2000	3.349	80.34	276	249	270	292	--
1	2001	3.559	81.85	288	257	279	301	--
1	2002	3.511	81.08	278	255	277	299	--
1	2003	3.493	81.51	274	254	276	298	--
1	2004	3.568	81.46	278	258	279	301	--
1	2005	3.410	81.23	277	251	272	294	--
1	2006	3.417	80.40	277	252	273	294	--
1	2007	3.433	82.28	288	251	273	295	--
1	2008	3.352	83.26	296	247	269	292	+1.4%
2	1999	3.347	78.11	263	249	271	292	--
2	2000	3.468	77.56	261	254	276	298	--
2	2001	3.495	77.46	263	256	277	299	--
2	2002	3.432	77.90	264	253	274	296	--
2	2003	3.558	78.00	257	259	280	301	-0.5%
2	2004	3.733	77.25	261	267	288	309	-2.2%
2	2005	3.725	77.27	270	266	288	309	--
2	2006	3.738	75.84	284	267	289	310	--
2	2007	3.629	75.55	287	262	284	306	--
2	2008	3.515	74.97	289	256	279	301	--
3	1999	4.117	71.61	326	285	307	328	--
3	2000	4.172	71.87	328	287	309	331	--
3	2001	4.444	72.43	315	300	321	342	--
3	2002	4.499	72.71	322	302	323	345	--
3	2003	4.435	72.81	319	299	320	342	--
3	2004	4.572	73.43	309	305	326	348	--
3	2005	4.511	73.24	308	302	324	345	--
3	2006	4.432	74.15	324	299	320	341	--
3	2007	4.491	74.93	317	301	322	344	--
3	2008	4.540	74.48	325	303	325	346	--
5	1999	5.082	70.27	345	328	350	372	--
5	2000	5.092	69.09	358	329	351	373	--
5	2001	5.014	69.38	350	325	347	369	--
5	2002	4.814	69.15	338	317	338	360	--
5	2003	4.788	69.40	339	315	337	359	--
5	2004	4.775	69.89	340	315	336	358	--
5	2005	4.780	69.88	334	315	337	358	--
5	2006	4.695	69.32	337	311	333	354	--
5	2007	4.594	69.12	352	307	328	350	+0.5%
5	2008	4.824	68.39	338	317	339	361	--
Other inputs								
US	2002	4.194	73.62	315	288	309	331	--
US	2005	4.241	73.98	285	290	311	333	-1.7%
US	2006	4.191	73.94	277	288	309	330	-3.9%
US	2007	4.190	74.34	280	288	309	330	-2.6%
SFBA	2008	5.307	68.39	360	338	360	383	--

Legend and notes for Table S8.

Table S8 shows inputs for emissions predicted by crude feed quality and compares the predictions with observed or estimated emissions. Observed crude feed density and sulfur, capacity utilized and products ratio were compared with observed *EI* among districts and years. Predicted *EI* values are the results from this PLS analysis, and are shown for the central prediction and the 95% confidence of prediction for observations. The central *EI* prediction and the observed fuel mix emission intensity were then compared with observed emissions among districts and years. Predicted emissions are the results from this PLS analysis, and are shown for the central prediction and the 95% confidence of prediction for observations. The observations compared among districts and years are from the data in Table S1. Other inputs shown at the bottom of the table were used in the prediction mode of these PLS models.

For U.S. refineries in 2002, 2005, 2006 and 2007, all data except estimated annual emissions are from Table S1. USEIA estimated that U.S. refineries emitted 277.6 megatons (Mt) of CO_2 in 2002 *(S32).* The National Energy Technology Laboratory estimated that U.S. refineries emitted 257.9 Mt in 2005 *(S12).* USEIA estimated that U.S. refineries emitted 250.7 Mt in 2006 and 251.3 Mt in 2007 *(S33).* U.S. refinery crude feed volumes in 2002, 2005, 2006 and 2007 totaled $241.3 \bullet 10^4$, $247.7 \bullet 10^4$, $248.0 \bullet 10^4$ and $245.6 \bullet 10^4$ m^3/day respectively *(Table S1).*

OQ inputs for San Francisco Bay Area (SFBA) refineries in 2008 were estimated as detailed in Table S9. The domestic component of SFBA crude feeds was more limited and better characterized than that of refinery crude feeds statewide, and this allowed a more reliable *OQ* estimate for SFBA refining than that which could be derived from publicly reported data for California refineries statewide. Although it has less capacity than Southern California, the SFBA has greater total crude capacity than other refining centers in District 5 *(S7).* The District 5 fuel mix during 2008 is used for the SFBA prediction to account for fuel mix differences observed among districts *(Table S1)*. SFBA inputs for capacity utilized and products ratio were the US averages for 1999-2008 from Table S1. Third party-certified estimates of emissions from SFBA refineries and adjacent plants supplying them hydrogen, as reported by the California Air Resources Board *(S34),* total 17.18 Mt in 2008. Crude feed volume was estimated as the total crude capacity of SFBA refineries in 2008 ($13.07 \bullet 10^4$ m^3/day) reported by *Oil & Gas Journal (S7)*. This SFBA emissions estimate (360 kg/m^3) compares with estimated California emissions of 354 kg/m^3 based on estimated emissions (36.88 Mt) and crude feed volume ($28.5 \bullet 10^4$ m^3/day) for refineries statewide in 2008 *(S34, S35)*.

The California Air Resources Board *(S36, S37)* reported estimated CO_2 emissions from refining the average crude feed in California, including those from bulk vents and refinery fuels acquisition, of 13.34 g/MJ gasoline (CARBOB) and 11.19 g/MJ diesel (ULSD) for 30.10 GJ/m^3 gasoline and 33.86 GJ/m^3 diesel. The California Energy Commission *(S35)* reported 2008 California refinery crude inputs, gasoline (RBOB, CBOB) yield, and diesel (≤ 15 ppm sulfur) yield of 104.04, 51.11 and 21.61 $m^3 \bullet 10^6$ respectively (total gasoline and diesel yield was 61.05 and 23.06 $m^3 \bullet 10^6$ respectively). These reports suggest refinery emissions of 197.2 and 78.7 kg/m^3 crude refined for California-grade gasoline and diesel production, respectively.

OQ inputs for heavy oil and natural bitumen are the average densities and sulfur contents of heavy oil and natural bitumen reported by the U.S. Geologic Survey *(S17)*. Other inputs for heavy oil and natural bitumen assume the 1999-2008 U.S. averages based on the data from Table S1. The 1999-2008 fuel mix assumption may be conservative for future emissions from refining lower quality oil, which tends to create more byproduct gases and petroleum coke that could replace some of the natural gas now burned as fuel. Refinery emissions observations were not available for these oils.

The columns on the right of the table compare predicted and observed emissions. Horizontal lines (—) indicate that the result is within the 95% confidence of prediction. Emissions observed among districts and years vary consistently with those predicted by *OQ,* fall within the 95% confidence of prediction in 36 of 40 cases, and fall within 3% of the confidence of prediction in all cases. Emissions estimated by government agencies fall within the prediction in 2 of 5 cases and fall within 4% of its confidence interval in all cases. The agency estimates differ from each other by 12% to 30% while they differ from the central prediction based on *OQ* by 0.1% to 10%.

Table S9. Estimate calculation, San Francisco Bay Area crude feed *OQ* in 2008.

Crude feed vol. (m^3/d)	Foreign[a]	SJV[b]	ANS[c]	Subtotal[d]
Benicia Plant	$8.870 \bullet 10^{3}$	$5.323 \bullet 10^{3}$	$7.987 \bullet 10^{3}$	$2.218 \bullet 10^{4}$
Golden Eagle Plt.	$9.683 \bullet 10^{3}$	$7.987 \bullet 10^{3}$	$7.930 \bullet 10^{3}$	$2.560 \bullet 10^{4}$
Martinez Plt.	$4.837 \bullet 10^{3}$	$1.992 \bullet 10^{4}$	$4.592 \bullet 10^{2}$	$2.522 \bullet 10^{4}$
Richmond Plt.	$2.992 \bullet 10^{4}$	0	$8.710 \bullet 10^{3}$	$3.863 \bullet 10^{4}$
Rodeo/S. Maria Plt.	$1.611 \bullet 10^{3}$	$1.450 \bullet 10^{4}$	$2.968 \bullet 10^{3}$	$1.908 \bullet 10^{4}$
Crude feed mass (kg/d)	Foreign[a]	SJV[e]	ANS[f]	Total
Whole crude	$4.827 \bullet 10^{7}$	$4.540 \bullet 10^{7}$	$2.392 \bullet 10^{7}$	$1.176 \bullet 10^{8}$
Sulfur in crude	$7.592 \bullet 10^{5}$	$5.901 \bullet 10^{5}$	$2.076 \bullet 10^{5}$	$1.557 \bullet 10^{6}$
		OQ	*S* (kg/m^3)	**11.91**
			d (kg/m^3)	**899.66**

Legend and notes for Table S9.

The *OQ* input for the San Francisco Bay Area refineries prediction (*S* and *d, Table S8)* is an estimate based on crude feed from foreign, Alaskan North Slope (ANS) and California oils that assumes transport logistics result in California supply from San Joaquin Valley crude delivered by pipeline (SJV) *(S16, S38)*. SJV portions of refinery feeds *(S39)* are used with refinery capacities *(S7)* and foreign crude feed volumes *(S40)* to estimate SJV volume processed. ANS volume is then estimated by difference. Weighted average crude feed *OQ* is estimated using these feed volumes and foreign *(S40),* SJV *(S38, S41)* and ANS *(S42)* crude quality data. Superscript notes in Table S9 identify the usage of these data in the estimate calculation specifically:

(a) Foreign crude feed volume, density and sulfur content reported for each plant *(S40)*.

(b) San Joaquin Valley pipeline crude volume based on SJV percentage of refinery feed reported *(S39)* and crude charge capacities *(S1)*.

(c) Alaskan North Slope (ANS) volume estimated by difference.

(d) Refinery crude charge capacities from *Oil & Gas Journal (S7)*.

(e) Based on SJV volume processed by Bay Area refineries, weighted average density (951.0 kg/m^3) from available data *(S38),* and sulfur content (12.36 kg/m^3) *(S41)*.

(f) From ANS volume calculated, and density (860.18 kg/m^3) and sulfur content (7.40 kg/m^3) of ANS crude at the Richmond Plant *(S42)*.

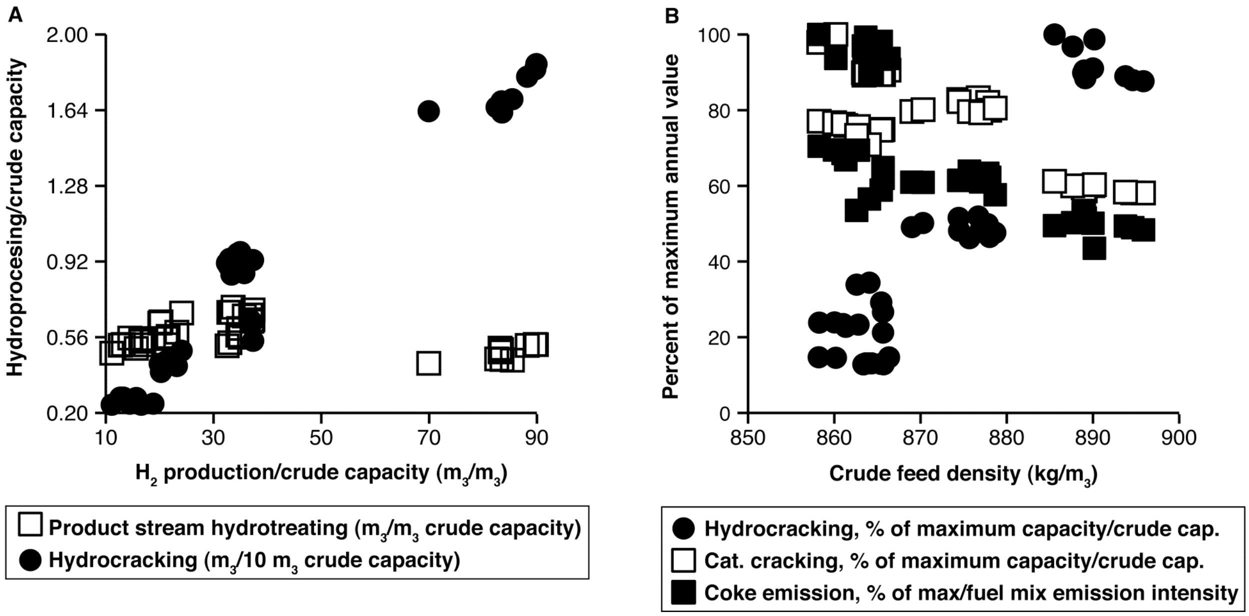

Figure S1. Some shifts among hydrogen addition and carbon rejection technologies affecting relationships between (A) hydrotreating and hydrogen production, and (B) fuel mix emission intensity and crude feed density, across refining districts 1, 2, 3 and 5, 1999-2008. All observations shown are from the data in Table S1.

A. Decreasing hydrotreating/hydrocracking ratio with increasing hydrogen production. Capacities are shown per volume atmospheric crude distillation capacity. Hydrocracking capacities are much smaller than total hydrotreating capacities and are shown at ten-times scale to reveal trends for both types of hydroprocessing. Hydrocracking uses much more hydrogen per volume oil feed than hydrotreating *(S43),* though actual unit H_2 requirements vary by type and quality of feed, unit design, catalyst type and condition, firing rate and quench rate of process units. Hydrocracking increases steadily with hydrogen production while product hydrotreating does not. Hydrotreating increases with H_2 production at lower H_2 production but is lowest at highest H_2 production. Relative to hydrocracking capacity, hydrotreating capacity decreases steadily with increasing H_2 production, from the largest capacity relative to hydrocracking in

District 1 (H_2 capacity 13-19 m^3/m^3 crude capacity) to the smallest relative to hydrocracking in District 5 (H_2 capacity 69-90 m^3/m^3).

B. Decreasing petroleum coke contribution to total fuel mix emissions with increasing crude feed density. The portion of total fuel mix emissions accounted for by petroleum coke and the process capacities/volume crude capacity are shown as percentages of the maximum (100%) for each value. The observed increase in hydrocracking with density is consistent with the strong positive associations of hydrogen production with both hydrocracking and density *(Table 1, main text).* Coke accounts for a decreasing portion of fuel mix emissions as crude feed density and hydrocracking increase. This change for coke, which has higher emission intensity than other major refinery fuels, can explain why the fuel mix emission intensity decreases slightly with worsening oil quality *(Table S1).* Despite increasing total conversion capacity (hydrocracking, catalytic cracking, and thermal coking), catalytic cracking capacity per vol. crude capacity decreases as crude feed density and hydrocracking increase. The ratio of catalytic cracking to hydrocracking decreases across districts, following the hydrotreating pattern noted above. Decreasing catalytic cracking explains decreasing coke emissions because cracking catalyst regeneration is a major cause of coke combustion in refineries.

The shifts from hydrotreating and catalytic cracking to hydrocracking observed can explain the coincidence of slightly lower hydrotreating at high hydrogen production, and of slightly decreasing fuel mix emission intensity as crude feed density increases, for these districts and years. Refiners can choose to substitute hydrocracking for hydrotreating and catalytic cracking to some extent, but the relative importance of crude feed quality among the factors that influenced such business decisions is beyond the scope of this study.

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Environmental Science & Technology

ADDITION/CORRECTION

pubs.acs.org/est

Combustion Emissions from Refining Lower Quality Oil: What Is the Global Warming Potential? [*Environmental Science & Technology* **2010**, *44*, 9584–9589]. Greg Karras

A typographic error has been corrected in Figure 1. The axis labels of the chart comparing observed and predicted *PI* (crude processing intensity) stated the wrong units. *PI* was defined, measured, analyzed, and reported as the ratio of specified "crude stream" process capacities to atmospheric crude distillation capacity. See the Experimental Section, paragraph 4; the Results section, paragraph 1; and the values shown graphically for observed and predicted *PI* in Figure 1. The error has been corrected by replacing "(GJ/m^3)" with "(ratio)" in the axis labels of this chart. The corrected Figure 1 is provided below. The correction clarifies the results reported but does not change the results, analysis, or findings of this work.

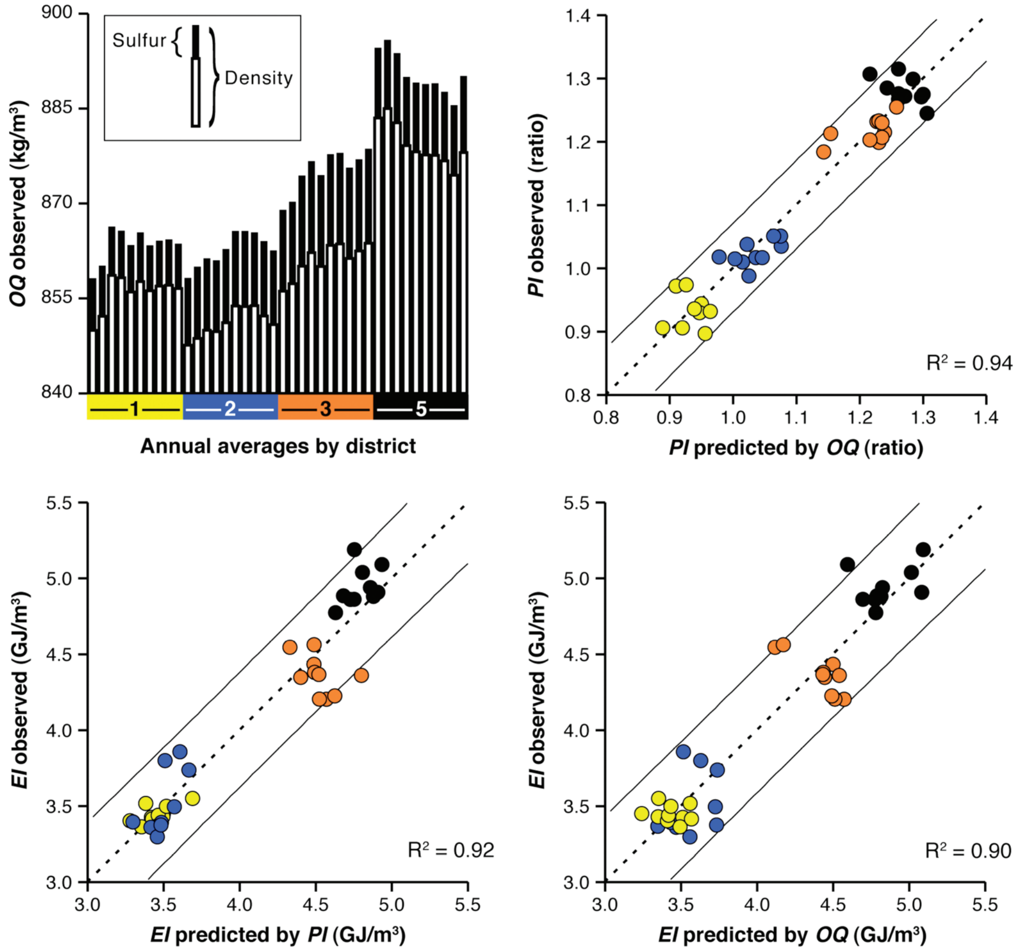

Figure 1. Increasing crude processing intensity and energy intensity with worsening oil quality. *OQ*: Crude feed oil quality. *PI*: Crude processing intensity. *EI*: Refinery energy intensity. Observations are annual weighted averages for districts 1 (yellow), 2 (blue), 3 (orange), and 5 (black) in 1999−2008. Diagonal lines bound the 95% confidence of prediction for observations.

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Attachment 14

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CARBON FOOTPRINTING

Factors driving refinery CO_2 intensity, with allocation into products

**Larry Bredeson · Raul Quiceno-Gonzalez ·
Xavier Riera-Palou · Andrew Harrison**

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Abstract

Background and scope Attempts to develop adequate allocation methods for CO_2 emissions from petroleum products have been reported in the literature. The common features in those studies are the use of energy, mass, and/or market prices as parameters to allocate the emissions to individual products. The crude barrel is changing, as are refinery complexities and the severity of conversion to gasoline or diesel leading to changes in the emissions intensity of refining. This paper estimates the consequences for CO_2 emissions at refineries of allowing these parameters to vary.

Materials and methods A detailed model of a typical refinery was used to determine CO_2 emissions as a function of key operational parameters. Once that functionality was determined, an allocation scheme was developed which calculated CO_2 intensity of the various products consistent with the actual refinery CO_2 functionality.

Results The results reveal that the most important factor driving the refinery energy requirement is the H_2 content of the products in relation to the H_2 content of the crude. Refinery energy use is increased either by heavier crude or by increasing the conversion of residual products into transportation fuels. It was observed that the total refinery emissions did not change as refinery shifted from gasoline to diesel production.

Discussion The energy allocation method fails to properly allocate the refinery emissions associated with H_2 production. It can be concluded that the reformer from a refinery energy and CO_2 emissions standpoint is an energy/CO_2-equalizing device, shifting energy/CO_2 from gasoline into distillates. A modified allocation method is proposed, including a hydrogen transfer term, which would give results consistent with the refinery behavior.

Conclusions The results indicate that the refinery CO_2 emissions are not affected by the ratio of gasoline to distillate production. The most important factors driving the CO_2 emissions are the refinery configuration (crude heaviness and residual upgrading) which link to the refinery H_2 requirement. Using the H_2-energy equivalent allocation proposed in this study provides a more reliable method to correctly allocate CO_2 emissions to products in a refinery in a transparent way, which follows the ISO recommendations of cause-effect and physical relationship between emissions and products.

Recommendations and perspectives Regulatory activity should recognize that there is no functional relationship between refinery CO_2 emissions and the production ratio of gasoline, jet, and diesel, and adopt a methodology which more accurately mirrors actual refinery behavior.

Keywords CO_2 allocation methods · Carbon footprint · Crude oil refining CO_2 emissions · Diesel production · Energy-consuming products · Gasoline production · H_2 effects on refinery CO_2 · Life cycle assessment · Refining emissions

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L. Bredeson
Westhollow Technology Center, Shell Global Solutions (USA),
Houston, TX 77082-3101, USA

R. Quiceno-Gonzalez (✉) · X. Riera-Palou · A. Harrison
Shell Global Solutions, Shell Technology Centre, Thornton,
PO Box 1, Chester CH1 3SH, UK
e-mail: raul.quiceno@shell.com

1 Background, aim, and scope

Policy makers and regulators are seeking to impose greenhouse gases (GHG) performance standards on fuel lifecycles, e.g., California's Low Carbon Fuel Standard (LCFS 2007) and the European Union's Fuels Quality and Renewables Directives (COD 2008). The common feature of these regulations is that fuel providers will be required to track the lifecycle (i.e., well to wheels) GHG emissions intensity of their products, measured per unit of fuel energy, and reduce this value over time. Furthermore, the US Environmental Protection Agency is assessing fuel lifecycle GHG emissions intensities for the Energy Information and Security Act. Models describing emissions in the fuel lifecycle, which were designed to meet academic scenario forecasting needs, now have to be redesigned to suit regulatory applications, with the associated legal and commercial implications.

Crude oil based transport fuels are produced concurrently with other fuel and non-fuel products. Consequently, overall CO_2 emissions generated by the refining process can be distributed between the individual products through "allocation" rules. Historically, such rules have reflected the scope and goals of the study, the modeler's understanding of the process, the available data and end-use options for the products because there is no theoretical basis for choosing one allocation scheme over another. When some refining products are regulated on their carbon content but not others, it is important to ensure that the allocation rules reflect the actual climate impacts of the regulated products as fairly as possible, whilst at the same time, minimizing incentives to transfer responsibility for the impacts onto unregulated products.

The International Standard Organization (ISO) guidelines for lifecycle assessment (LCA) recommend that allocation should be avoided wherever possible, but where this is not possible, the allocation should reflect quantitatively or qualitatively how environmental impact changes with product yield. Some authors have suggested options to refine the ISO methodology and the accuracy of the results (Ekvall and Finnveden 2001). Ultimately, however, it is left to the LCA practitioner to decide how to follow these recommendations. As a result, the literature contains several different estimates for the carbon intensity of gasoline and diesel production even for similar systems (Furuholt 1995).

The problems faced in solving the issue of allocation in multi-product systems are fairly well known, and they have been extensively discussed in the literature (Azapagic and Clift 1999; Ekvall 1999; Babusiaux 2003; Ekvall and Weidema 2004). Different accounting schemes have been proposed to assign emissions to the plant products typically based on mass, energy, or market value shares of products. More recently, linear programming (LP) models, which have a long tradition in the refining industry (Charnes et al. 1952; Griffin 1972; Palmer et al. 1984), have been extended to calculate CO_2 emissions, and to assign individual product contributions to the CO_2 emissions in refineries through a marginal approach (Azapagic and Clift 1999; Babusiaux 2003). These models follow a similar logic to that used in assigning costs to refinery products: global CO_2 emissions are allocated to products based on the incremental CO_2 emissions generated in manufacturing an additional volume of the products. The resulting product CO_2 intensities are sometimes, but not always, different from those estimated under traditional mass/energy allocation schemes. Neither type of method is superior; but each has its domain of validity and applicability.

Furuholt (1995) compared the energy consumption and pollutant emissions in the production and end use of regular gasoline, gasoline with MTBE, and diesel. Energy consumption and emissions were tracked through the production chain and emissions were allocated to products based on their energy content. The results were highly sensitive to the product specifications, and it was predicted that emissions from diesel production were significantly lower than those from production of gasoline as a consequence of "diesel's lower process energy requirement".

Wang and coworkers (Wang et al. 2004) compared the impact of different allocation rules applied at the process unit level in a US refinery. They used as an archetype refinery a detailed quantitative process-step model of petroleum refining developed in the late 1970s at Drexel University (Brown et al. 1996). The mass and energy balances at each process step of this archetype constitute the reference process-step model for petroleum refineries (Ozalp and Hyman 2007). Wang et al. (2004) compared the use of mass, energy content, and market value share of final and intermediate petroleum products as allocation weight factors at the process unit and the refinery levels. They defined product energy intensities for major refinery products (defined as the fraction of process energy invested in producing a particular product relative to its weight factor), and concluded that wherever possible, energy use allocation should be made at the lowest sub-process level (Wang et al. 2004). They found diesel production to be less energy intensive than gasoline production in each of the allocation weighting methods used (mass/energy/market value; refinery/process unit level) as predicted by Furuholt (Furuholt 1995).

Tehrani (Tehrani 2007) used an LP model to study the CO_2 emissions allocation problem for a European price-taking refinery operating in a cost-minimizing environment. It was assumed that the refiner's objective is to satisfy a petroleum production target at the minimum cost and subject to constraints of prevailing technology, commodity prices, input availabilities, oil product demand, capacity constraints, material balance, and product quality. Tehrani concluded that emissions could be allocated among products using "average allocation" coefficients containing two contributions, a direct one, which is its marginal CO_2 intensity, and an indirect contribution, which depends upon the production elasticity of unit processes and is calculated at the LP optimal solution ex-post. This approach was later used (Tehrani and Saint-Antonin 2007) to assess the impact of reducing sulfur in European automotive fuels on the refining emissions intensity of gasoline and diesel. It was shown that, contrary to prior results (Furuholt 1995; Wang et al. 2004), gasoline refining could be less emissions intensive than diesel refining.

Pierru (2007) used an alternative LP optimization function including operating costs and cost associated with the refinery's CO_2 emissions to calculate the marginal emissions (in accordance with economic theory) from the various refinery products. The study highlights the impact of constraints such as demand, refinery capacity, and raw material supply on the CO_2 emissions originated at refineries. It was concluded that contrary to traditional LCA studies, diesel has a higher marginal contribution to refinery emissions than gasoline.

The common features in the above studies, notwithstanding the different approaches, constraints, and results are: single-fixed refinery configuration, fixed unit throughput capacities and fixed crude diet.

The crude barrel is changing, as are fuel specifications, and these will lead to changes in refining emissions intensities. In this paper, we therefore focus on the consequences of varying the crude diet, the severity of conversion to gasoline or diesel, and the complexity of the refinery. The critical element is the hydrogen requirement, since its production and consumption is highly carbon intensive. A detailed analysis of the hydrogen flow through the refinery is carried out at each refinery unit, in order to establish the carbon footprint of products. Based on this work, we propose a more realistic way to estimate the energy and emissions intensities of refinery products.

2 Materials and methods

The refinery simulation model is a case study model used by Shell to select crude type, determine refinery products, and calculate refinery economics for major investment decisions. Shell has high confidence in its accuracy.

Yield representations reflect crude boiling curve, hydrogen content, aromaticity, sulfur, nitrogen, and other relevant parameters associated with the refinery crude diet. Several of those terms (boiling curve, hydrogen content, and aromaticity) are at least partially covariant with crude density (API gravity), but it is more accurate to handle them individually. Processing severity can be adjusted by distributing feeds differently within the refinery flow matrix, by changing reactor severity of individual processes, and by varying fractionator cut points. Energy consumption was determined by summing feed-rate-based consumption factors for each process unit (some of which are functions of that unit's severity). Feed gas and fuel gas energy for H_2 manufacture are included. Hydrogen balance is maintained throughout the model, meaning the hydrogen contained in all feeds equals the hydrogen contained in all products from each unit. Relatively few refinery models have that feature; meaning that their prediction of how much hydrogen is required from the hydrogen plant is less reliable. Since hydrogen plant size is critical to refinery CO_2 emissions, this is an important advantage for this study.

Specific process units included were: crude distillation, delayed coking, fluid catalytic cracking, hydrocracking, naphtha reforming, alkylation, hydrotreating (naphtha, distillates, fluid catalytic cracking (FCC) feed), hydrogen manufacture, sulfur recovery, and various other enabling process units typically included in a refinery (the refinery flow chart is available as Online Resource 1).

Product specifications were gasoline was US reformulated gasoline in a typical grade mix of regular to premium. Diesel was US ultra low sulfur diesel. Jet was Jet-A, and in cases where produced, residual was US Gulf Coast high sulfur Fuel Oil #6. Naphtha from the catalytic cracker was hydrotreated such that gasoline pool sulfur was 25 ppm. Jet smoke and diesel cetane number using a normal severity distillate hydrotreating unit were inside fuel specifications for all except two of the crudes analyzed. This was ignored because real refineries have some scope to blend streams to meet specifications, and if not, the refinery would run a blend of crude rather than neat crude. The three low value residual streams (Cat slurry, Fuel Oil #6 and Coker Coke) were summed into a single product class called residual/coke. To summarize, the product streams considered were liquefied petroleum gas (LPG), gasoline, distillate (including gasoil and kerosene), and residual/coke.

It was considered critical that the results from the allocation methods and the results from the model runs be consistent. In other words, if the refinery runs showed no difference in total refinery CO_2 emissions as the gasoline to diesel ratio was varied, then the CO_2 intensity of those two fuels should be the same.

3 Results

Three issues were studied explicitly: crude heaviness (fraction boiling >1,000°F/540°C), production ratio of gasoline to distillates, and whether the refinery processed its 1,000°F/540°C+vacuum resid in a delayed coker or blended it to Fuel Oil no. 6. Issues such as ratio of FCC to hydrocracking capacity, the type of benzene production controls employed, whether C_5/C_6 isomerization is employed, in cases with residue reduction, whether the residue reduction unit was a delayed coker, other type of coker, or other type of unit such as LC-Finer or resid hydrotreater, and any number of similar configurational issues could perturb the numerical results. Pair cases simulations (base Vs base + δ), where δ refers to a perturbation on the variable under analysis were run to assess the robustness of the results and to ensure that they did not have a material impact on the conclusion reached through the study

3.1 Matrix of cases

Crude heaviness was studied by selecting six crudes with quantity of vacuum bottoms (>550°C) ranging from 10% to 35% (lightest Brent, heaviest Maya). Production ratio of gasoline to distillate was varied by shifting from gasoline to distillate mode which means lowering FCC and HCU reactor severities, and changing cut points at crude unit, cat cracker, and hydrocracker. Cut points were shifted on both ends, lowering naphtha/distillate cut point and raising distillate to FCC feed cut point. Production of resid was changed by shutting down the coker, and sending coker feed to #6 oil blending instead. Case names of these conditions were captured in a four character code. The first character was either K or 6, representing a coker case or a case that produced #6 residual fuel oil. The second and third characters were C for crude, and a number, meaning the crude heaviness choices from 1 to 6. The final case was H or L meaning high or low severity to gasoline. So for example, KC3L was a coker case on crude 3, with low severity to gasoline. Or case 6C5H was a #6 fuel oil case on crude 5 with high severity to gasoline. In all, the refinery was run in four modes (high/low gasoline, with/without coker) with six different crudes to produce a matrix of 24 data points. For each case, refinery yields and fuel/CO_2 data were generated. Refinery yields data are available as Online Resource 2. The fuel/CO_2 data were split by process needs and H_2 generation needs.

One aspect of these runs was different from typical model running strategy. In most model studies, one must stay within capacity constraints of the various process units. But in this study, there are wide variations of crude heaviness, which would far exceed the acceptable flow rate variations for individual units in any given refinery. So individual process unit throughputs were allowed to vary as needed, such that each intermediate stream in the refinery headed to its normal consuming unit. Had that not been done, the results would have been strongly and inappropriately biased by internal constraints. This way, it was as though each case had a custom tailored refinery to allow ideal flows for that case.

3.2 Numerical results

Consider the results as being four blocks of data, with six cases in each block. The four blocks are with/without coker (i.e., high/low resid production), high/low conversion to gasoline, and within each of those four blocks, the six crudes of varying heaviness. These four blocks are shown in Fig. 1.

Comparing the left two with the right two blocks on Fig. 1 shows that adding the coker to eliminate the no. 6 fuel oil production clearly increases CO_2 emissions for all case pairs involving that switch. Not only does the coker consume energy in its own right, it upgrades a low hydrogen content product stream (no. 6 fuel oil). This in turn requires the refinery to run other cracking and hydrogen consuming units harder to boost the hydrogen content up from resid hydrogen levels (because resid is no longer being produced) to mogas/jet/diesel hydrogen levels (because those higher hydrogen content products are being produced instead of resid).

Changing the severity and cut points to vary the ratio of gasoline to distillate has very little effect in any of the cases in any of the case pairs where that change was made (see Fig. 1). At first, this might seem illogical because to go to lower boiling point gasoline, the level of cracking needed is harder, and that would seem to require more energy. The counter-balancing point is H_2 content. In gasoline production, aromatics are favored due to higher octane ratings and this is where the reformer's H_2 production comes into play. To make more gasoline, reformer feed rate increases and as reformers also produce H_2, the amount of H_2 that must be made in the CO_2 intensive H_2 plant decreases, and on balance, the overall CO_2 emissions do not change very much. In contrast, for jet and diesel production, paraffins are favored. In fact, despite its lower boiling point, H_2 content of gasoline is similar to jet and diesel.

What happens with crude heaviness depends on whether there is a coker (or other residue reduction unit). The left two blocks of Fig. 1 show that if there is a coker to eliminate resid, heavier crude needs a bigger coker, which consumes more energy, and demands more hydrogen consumption in downstream units, thus increasing CO_2 emissions (from running the hydrogen plant at a higher rate). The right two blocks of Fig. 1 show that without a coker, the refinery produces resid as a product, so CO_2 emissions do not change very much with crude heaviness. However, the heavier crude makes more resid in comparison to transportation fuel, and that is an indirect CO_2 penalty because more carbon intensive resid product fuels

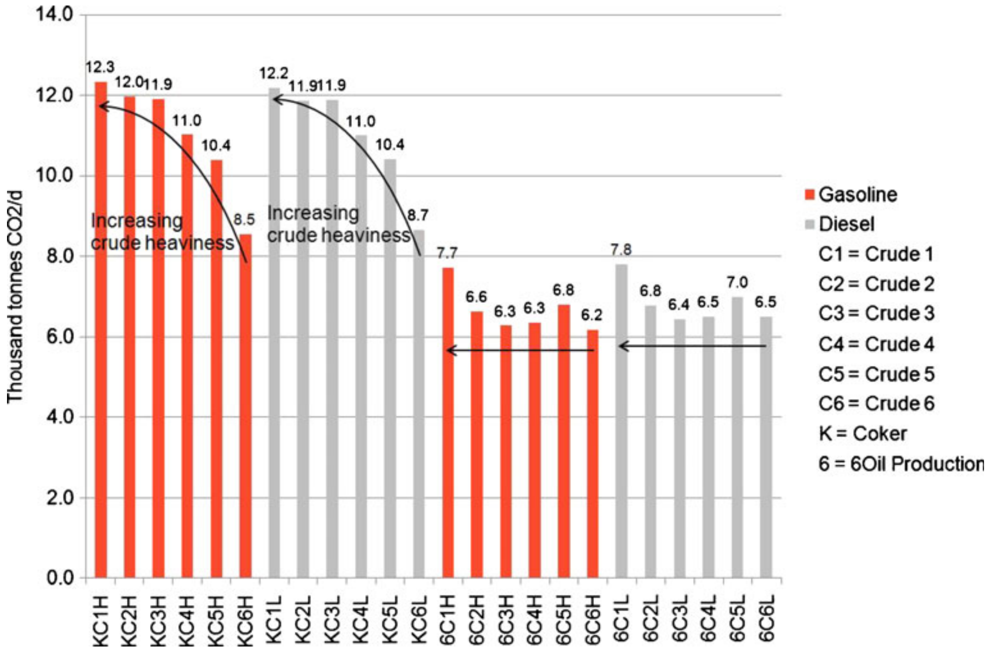

Fig. 1 Overall refinery CO_2 emissions

are being produced. Note that this issue of with/without coker, or higher/lower residual fuel production is sometimes referred to as refinery complexity. The coker (or other residue reduction unit) adds complexity not only because it is an added large process unit, but also because products from residue reduction units are low quality, which requires other units within the refinery to be larger and higher severity in order to upgrade them.

The fact that CO_2 emissions are practically independent of light product ratio shifts from gasoline to diesel shows that the CO_2 emissions at refinery level are not driven by the differential energy demands of these products, but by other factors: crude heaviness and whether the refinery has a coker to eliminate production of residual fuel. A third route to CO_2 emissions reductions is energy conservation; all routes can be influenced by external issues such as crude availability, product demands, and prices.

4 Discussion

It was shown in Section 3 that two operational routes significantly lowered total refinery CO_2 emissions. The production ratio of gasoline to diesel fuel was not one of those factors, because interaction of some non-obvious hydrogen issues equalizes the total refinery CO_2 emissions from production of gasoline and diesel fuel. The hydrogen balance at the refinery, together with the results from tracking products through process units in terms of the energy consumed during their production and their associated CO_2 emissions are described in the next sections. Both results are used to develop an allocation strategy consistent with refinery CO_2 emissions behavior.

4.1 Hydrogen balance

One of the most critical factors in refining is hydrogen balance. This is not just hydrogen balance in the sense of flows of elemental hydrogen gas as a processing stream but also the hydrogen content of feeds and products. Since crude oil is generally low in hydrogen content, and refined products (except for residual fuel and coke) are high in hydrogen content, refineries are forced to produce the additional H_2 that satisfies their needs in a process that its intrinsically highly CO_2 emissions intensive.

Carrying this hydrogen issue a bit further, if the crude has less hydrogen coming in (most common explanation being that it is heavier), or the products have more hydrogen going out (most common explanation being more transportation fuel with correspondingly less residual fuel), the refinery energy consumption will invariably be higher. While it is true that there are many possible routes and configurations of refineries (for example, cat cracking versus hydrocracking), all refineries by all routes are bound by this hydrogen balance issue. The exact configuration of a refinery can cause minor variations in energy/CO_2, but the simple difference in hydrogen content between crude coming in and products going out are by far, the controlling factor.

In a typical refinery, roughly half of the H_2 is produced as a by-product from the catalytic reformer (and in the few refineries that have them, from the olefins plant) (NETL 2008). Most allocation schemes allocate the energy and CO_2 from the "on purpose" H_2 plant properly, but they ignore the impact of the reformer H_2, and if applicable, from the H_2 produced at the olefins plant. Ignoring the reformer H_2 production means that the H_2 consuming units get a substantial part of their H_2 requirements as a CO_2-free stream, and also that the reformer is not credited for the large CO_2 avoidance associated with its H_2 production and the displaced H_2 from the "on purpose" H_2 plant.

Production of gaseous H_2 in "on purpose" H_2 plants can be typically characterized by a well to tank footprint of circa 108 gCO_2e/MJ (GREET 2008). By comparison, the gasoline footprint is around 90 gCO_2e/MJ in GREET. This highlights the importance of correctly accounting for CO_2 emissions in processes involving hydrogen production.

If one looks at what drives hydrogen content of crude, it is mostly the heaviness, i.e., how much boils above 1,000°F/540°C. There is a modest added effect for whether the crude is of naphthenic or paraffinic character, but heaviness is more important. One would expect that the heavier the crude, and thus the less hydrogen that the crude contains, the higher the energy requirement and CO_2 intensity of the refinery.

On the product side, gasoline, jet, and diesel have roughly equivalent hydrogen content: For the main transport fuels[1], the C/H ratio would range for gasoline (EN220) ~1.7–1.9, for diesel (EN590) ~1.7–1.9 and for jet A-1 (AFQRJOS[2]) ~1.7–1.9. The mass ratio (carbon to hydrogen) estimated for these fuels range between 6.3 and 6.9 m/m for all of them (see footnote 1). It might seem logical to think that gasoline should have more hydrogen than jet or diesel because it has a lower boiling temperature range, and hydrogen content is normally higher as boiling point gets lower. But actually, because quality issues force a bias toward aromatic species for gasoline to maintain its octane rating, while at the same time there is an opposite bias toward paraffinic content for jet and diesel to maintain their smoke point and cetane ratings things balance out in such a way that the main transportation fuels are similar in hydrogen content, and thus should be similar in their CO_2 emissions intensity.

LPG (generally C_3 and C_4 molecules) contains more hydrogen than gasoline, jet, and diesel, so should have higher CO_2 intensity. Some might think LPG should be low CO_2 intensity since much of it comes from simple fractionators. But LPG is not an "on-purpose" product, it is a byproduct. If more LPG were made by choosing catalysts that did more overcracking, the LPG would carry away more hydrogen in the product, requiring more refining and hydrogen manufacturing energy.

By contrast to high hydrogen LPG, residual fuel oil has very low hydrogen content. Resid can either be produced by the refinery as a product, or cracked in a resid cracking unit such as a coker. Coking is energy intensive, not only because of the coker itself, but also because the coker makes hydrogen deficient products which need extra hydrogen to be added in subsequent refining steps. Allowing the resid to go out as residual product rather than cracking it to lighter products saves large amounts of energy, thus making resid a very low energy product.

While not explicitly studied in the model runs described in this paper, other factors can influence refinery CO_2 emissions. One example has already been mentioned, namely, energy conservation which would lower CO_2 emissions. Others would include product specification changes such as lower sulfur or lower aromatics, which would raise CO_2 emissions. And finally, going to production ratios of products outside "normal ranges" could negate the conclusion that all of the light transportation fuels have "roughly equal" CO_2 emissions. If a refinery is forced to make more of a particular fuel than can be accommodated within "natural refinery flexibility" (such as very high diesel production, with very low gasoline production), CO_2 emissions would clearly increase. Variations in production ratios modeled in this paper were all within normal ranges of refinery flexibility, with an average swing between gasoline and diesel for high to low gasoline cases of around 4% on crude, and ranged between 2% and 6% depending on crude type and refinery configuration.

Subject to these caveats, we might expect that the refinery production of CO_2 (i.e., consumption of fuel, including the fuel needed to manufacture hydrogen) to produce gasoline, jet, and diesel should be roughly equal. Because refinery energy is mostly proportional to product versus feed hydrogen content, and the hydrogen content of gasoline, jet, and diesel products are similar. Using this same logic, LPG should be higher in CO_2 intensity and bunker-type residual fuel lower. CO_2 emission and energy consumption will be higher for heavier crudes than light, and slightly higher for naphthenic than for paraffinic crudes. Other factors should not influence refinery energy consumption as shown by the refinery model runs described in Section 3. Hydrogen content of the various feed and product streams is the main driver of refinery CO_2 intensity critically important in developing a proper allocation scheme.

4.2 Allocation approaches

Many allocation methods have concluded that refining to gasoline is much more energy intensive than distillate, which is inconsistent with the findings in the previous section, where varying gasoline/distillate ratio did not have much effect on CO_2 emissions. To understand why, a typical allocation approach was applied to the data from Section 3.

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[1] Shell Internal data

[2] Join Inspection Group, Products Specifications. Aviation Fuel Quality Requirements for Jointly Operated Systems (AFQRJOS). Issue 22–28 June 2007

The energy consumptions of the individual process units from the 24 runs in Section 3 were distributed into products according to process unit yields from those runs. For example, if a given unit consumed 10 units of energy, and its yields were 40% gasoline, 40% distillate 10% LPG, and 10% resid; its 10 units of energy would be allocated 4, 4, 1, 1 to those products. For the hydrogen plant, energy was distributed to the individual units according to the relative hydrogen consumption of that unit and from there by-product, as with the normal fuel. Using this approach, gasoline was approaching a factor of two times more energy intense than distillate. But this handles hydrogen incorrectly.

In the above scheme, the fuel and feed gas associated with the hydrogen plant is allocated to the hydrogen-consuming units on the basis of their relative hydrogen consumptions, and from there to products. However, only about half of the refinery's hydrogen comes from the hydrogen plant. The remaining half comes from the catalytic reformer, which is totally associated with gasoline production. Recall from Section 4.1 that gasoline is biased toward aromatics for quality purposes (i.e., octane rating), and the reformer is the process step that gives this bias. If the refinery makes less gasoline, it would have a smaller reformer, which would make less hydrogen, which would then require a larger hydrogen plant, which would consume more energy. So the reformer, from a refinery energy and CO_2 emissions standpoint, is an energy/CO_2 equalizing device, shifting energy/CO_2 from gasoline into distillates.

If the allocation scheme does not recognize this hydrogen-equalizing feature of catalytic reforming, it will conclude that gasoline has greater CO_2 and energy intensity than jet or diesel. But once the hydrogen production of the reformer is included in the allocation, the allocation will correctly show essentially equivalent energy intensity for gasoline, jet, and diesel. Note that this decision on how to allocate is not arbitrary. Without the reformer hydrogen correction, the allocation does not match actual refinery behavior, while with it, it does. So refinery reality, not arbitrary shifting, is being used to guide the allocation method.

There are various algebraic ways of including the reformer hydrogen production in the allocation scheme. The one chosen counts the energy equivalent of hydrogen as a credit/debit to each unit (credit to H_2 producing units, debit to consuming units), and does not count the hydrogen plant (because it is implicitly counted by debiting the consuming units for the energy equivalent of their hydrogen consumption). Using this technique, the consuming units pay the CO_2 penalty for all of their hydrogen, not just the fraction of hydrogen coming from the hydrogen plant. With this technique, the CO_2 intensity of gasoline versus distillate equals out, which agrees with the observed refinery behavior, which is that refinery energy consumption does not change as gasoline to distillate ratio changes. If gasoline was more energy intensive than distillate, that would not be true.

4.3 Allocation results

The behavior described in Section 4.2 is shown quantitatively in Figs. 2 and 3. Starting with Fig. 2, which has only the coker cases, the right hand side has the results from the simple allocation without hydrogen correction. It shows much greater CO_2 intensity for gasoline using that approach. The left side of the figure includes the hydrogen correction, and gasoline is similar to distillate in CO_2 intensity. There is a slope in both blocks, with heavier crudes showing more energy consumption. This is the same slope as was seen in the left two blocks of Fig. 1 (discussed in Section 3), and is caused by the fact that heavier crudes require more coking. Fig. 3 is similar to Fig. 2, except that it has the #6 oil cases rather than the coker cases. It shows most of the same trends, for the same reasons, as Fig. 2. The only differences are that there is essentially no bias for crude heaviness, and the overall levels are lower than in Fig. 2. These differences also link back to Fig. 1, where the #6 oil cases had similar CO_2 emissions regardless of crude heaviness, and had lower CO_2 emissions than the coker cases. The slight slope with regard to crude heaviness in Fig. 3 is caused by two things: (1) the highly paraffinic far right crude is slightly low, while the highly naphthenic far left crude is slightly high, and (2) there is an eye-catching slope in Fig. 3 with regard to LPG, but LPG is a small flow, explained by other factors (see next paragraph). So concentrating on the gasoline and distillate, Fig. 3 is essentially flat with regard to crude heaviness. But while CO_2 emissions are flat, there is an indirect, heavy crude CO_2 penalty in the Fig. 3 cases because with no coker, more carbon-rich resid product leaves the refinery as the crude gets heavier.

Looking at the corrected distributions, a few other observations can be made. First, resid product has very low CO_2 intensity as no energy has been spent cracking it or adding hydrogen to it. Second, LPG has very high CO_2 intensity. While a very small amount of LPG is contained in crude oil, and is thus produced with low CO_2 intensity through simple fractionation, most of it is produced by cracking in the high CO_2 intensity cracking units. Indeed, the LPG CO_2 intensity increases with heavier crude. As crude gets heavier, the cracking units get larger, so a larger proportion of LPG comes from cracking rather than simple fractionation. And if a refinery were forced to make even more LPG on purpose by over-cracking, the LPG energy intensity would go up even further. So LPG over and above the very small quantity contained in crude oil should not be regarded as a low energy intensity product.

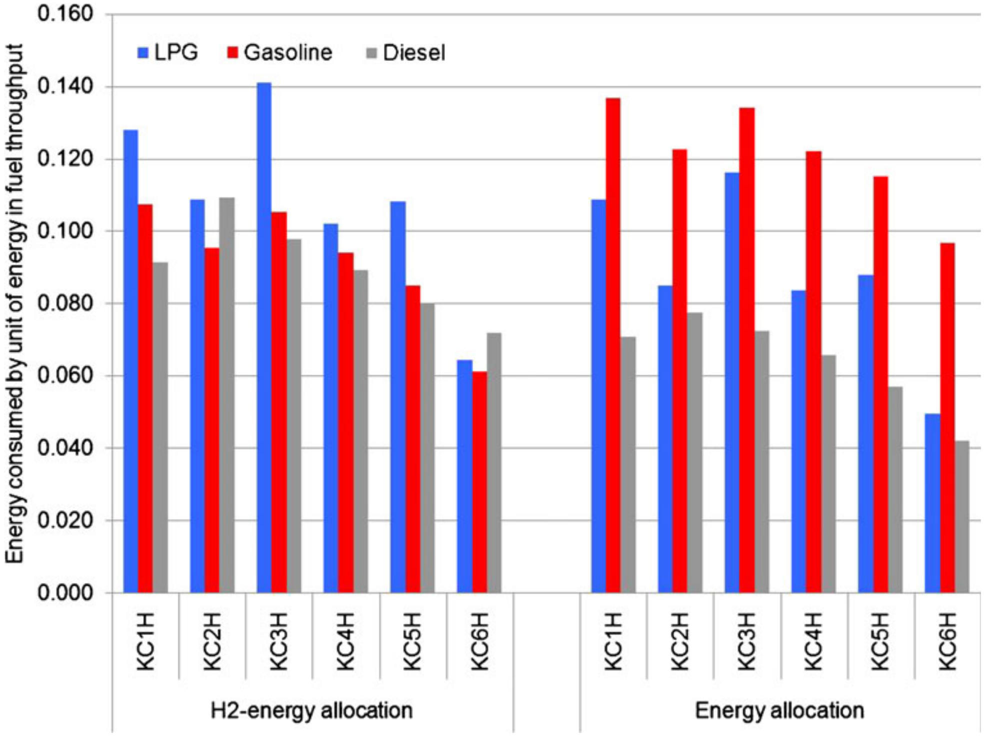

Fig. 2 Comparison between allocation methods for coker cases

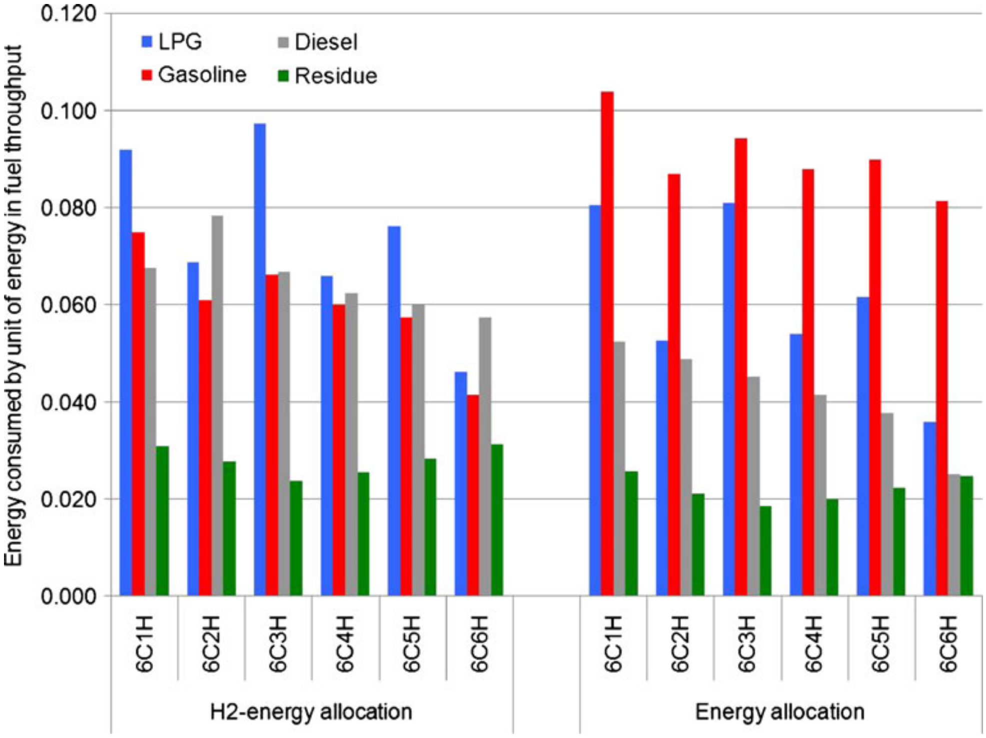

Fig. 3 Comparison between allocation methods for six oil cases

5 Conclusions

Total refinery CO_2 emissions are not strongly affected by ratio of gasoline to distillate product.

To agree with the above conclusion, an allocation scheme cannot conclude that gasoline is more CO_2 emissions intensive than distillate. To avoid that result, the allocation scheme must distribute energy into the various refinery products in a way that takes reformer hydrogen into account.

Refinery CO_2 emissions increase as it produces more transportation fuel and correspondingly less resid product. Operationally, this means that the refinery has a coker or other residue reduction unit, or said in another way, it is more complex.

In a complex refinery with a coker (or other residue reduction unit), making little or no residual fuel product, refinery CO_2 intensity is increased by running heavier crude. In a refinery that does not have a coker, and thus produces substantial quantities of residual fuel product, crude heaviness has little impact on total CO_2 emissions.

Refineries cannot vary LPG production by much, but if forced to make more LPG, total CO_2 emissions would increase. There is no way to make less LPG, it is minimized already.

While not studied explicitly in this paper, it should be self-evident that total refinery CO_2 emissions are also affected by degree of energy conservation excellence (i.e., capital equipment for energy conservation purposes) and by product specifications such as sulfur and aromatics.

6 Recommendations and perspectives

The conclusions on what impacts CO_2 intensity would seem to have obvious implications for regulatory methodologies. But there are a few added considerations that may not be immediately obvious from the conclusions themselves.

Allocation of refinery CO_2 emissions to individual products which does not stick to the technical reality is, by its very nature, rather arbitrary. This can be seen from the fact that using or not using the hydrogen corrections described in this paper has a dramatic impact on the allocation results. That arbitrariness should caution one against taking allocation results too literally. But if one insists on doing an allocation, at least it should be consistent with observed refinery behavior. The refinery behavior is that CO_2 emissions do not change very much with production ratio of gasoline to distillate. Thus, any allocation scheme which shows CO_2 intensities of gasoline and distillate are substantially different must be seen with caution, and special care should be put into understanding the handling of internal flows, the technical premises assumed, and how they align with the scope and goals of the LCA. Only with the understanding of the full context it is possible to conclude about the results and their implications.

The conclusion that CO_2 can be reduced by making more residual product in less complex refineries without cokers must be tempered with recognition that: (1) it would also lead to a carbon-rich stream (the resid) leaving the refinery; (2) refinery configurations and decision on make yield are driven many other external factors, for example, supply/demand balance of different products; and (3) well-to-wheels or life cycle effect should be considered in determining CO_2 reduction.

Similarly, the conclusion that CO_2 can be reduced by running lighter crude must be tempered with the realization that world crude demand is expected to continue to increase while world supply of light crude is limited [LBST 2007; EIA 2009]. Given that, it is likely that world demand for heavier crudes will continue to increase in the near future to meet consumer demand for transportation fuels.

Areas for further development This paper has not thoroughly handled jet versus diesel, grouping them instead as combined “distillate” fuel. If done simplistically, jet would show as being less energy intensive, because most jet comes via the crude unit and a low severity hydrotreater. But in similar fashion to LPG, if forced to make added jet, a refinery would need to include hydrocracked jet, and that is very energy intensive, often requiring a post-saturation step. Allocation methods could be developed to handle that complication, but that was thought to be beyond the scope of this paper. Instead, the simplifying step of combining jet and diesel into “distillate fuel” was adopted. However, this simplification does not undermine the conclusion that gasoline and diesel have similar overall refinery CO_2 emissions intensity. Simplistically, if jet is viewed as low CO_2 intensity, the algebra of the situation would force the intensity of diesel to be higher to balance. Thus, it does not offer a path back to the conclusion that gasoline is worse than diesel.

It is also acknowledged that precise refinery configuration or exact fuels specifications have not been studied in this study. Some runs were conducted to verify that those issues are far less important than the factors described herein, but it cannot be concluded that their effect is zero. In fact, the next phase of our work will be to study those issues more closely to determine which, if any, of such effects are non-trivial.

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Attachment 15

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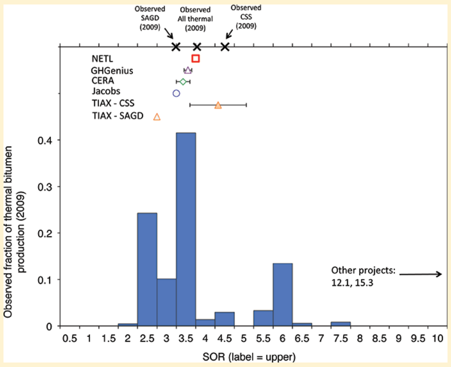
Variability and Uncertainty in Life Cycle Assessment Models for Greenhouse Gas Emissions from Canadian Oil Sands Production

Adam R. Brandt*

Department of Energy Resources Engineering, Stanford University, Stanford, California 94305-6105, United States

Ⓢ *Supporting Information*

ABSTRACT: Because of interest in greenhouse gas (GHG) emissions from transportation fuels production, a number of recent life cycle assessment (LCA) studies have calculated GHG emissions from oil sands extraction, upgrading, and refining pathways. The results from these studies vary considerably. This paper reviews factors affecting energy consumption and GHG emissions from oil sands extraction. It then uses publicly available data to analyze the assumptions made in the LCA models to better understand the causes of variability in emissions estimates. It is found that the variation in oil sands GHG estimates is due to a variety of causes. In approximate order of importance, these are scope of modeling and choice of projects analyzed (e.g., specific projects vs industry averages); differences in assumed energy intensities of extraction and upgrading; differences in the fuel mix assumptions; treatment of secondary noncombustion emissions sources, such as venting, flaring, and fugitive emissions; and treatment of ecological emissions sources, such as land-use change-associated emissions. The GHGenius model is recommended as the LCA model that is most congruent with reported industry average data. GHGenius also has the most comprehensive system boundaries. Last, remaining uncertainties and future research needs are discussed.



INTRODUCTION

As conventional oil production becomes constrained, transportation fuels are being produced from low-quality hydrocarbon resources, such as bitumen deposits and other unconventional fossil resources. These include oil sands, enhanced oil recovery, coal-to-liquids and gas-to-liquids synthetic fuels, and oil shale.

Production of crude bitumen from the oil sands was almost 1.5 M bbl/d in 2009.[1,2] Production of liquid products from oil sands, including raw bitumen and synthetic crude oil (SCO), reached 1.35 M bbl/d in 2009. This represents an increase from ≈600 k bbl/d in 2000.[3] Current plans for expansion of production suggest over 7000 k bbl/d of capacity in all stages of operation, construction, and planning.[2]

In general, liquid fuels produced from unconventional resources have higher energy consumption per unit of fuel produced than those produced from conventional petroleum deposits. This is due to the higher energy intensity of primary resource extraction and the energy requirements of hydrocarbon processing and upgrading. Greenhouse gas (GHG) regulations such as the California Low Carbon Fuel Standard (LCFS) and European Union Fuel Quality Directive seek to properly account for the GHG intensities of these new fuel sources.

This paper examines models of upstream GHG emissions from Alberta oil sands production. The goal of this work is to understand the validity and comparability of previously published life cycle assessment models of GHGs from oil-sands-derived fuels, and to compile a range of emissions factors for oil-sands-derived fuel streams. Assumptions and data inputs to models are compared with observed data. Recommendations are then made for the use of these LCA results and for future research needs.

OVERVIEW OF OIL SANDS PRODUCTION METHODS

Oil sands are a mixture of sand and other mineral matter (80–85%), water (5–10%), and bitumen (10–18%).[4] Bitumen is a dense, viscous mixture of high-molecular-weight hydrocarbons. Bitumen is either diluted or upgraded to SCO before shipment to refineries for processing into liquid fuels.

Oil sands extraction. Bitumen is produced through surface mining or in situ production processes. Surface mining requires removal of overburden and mining of the bitumen/sand mixture (ore). The ore is transported to processing facilities where it is mixed with hot water, screened, and separated into bitumen and tailings.[4] A variety of in situ techniques exist, the most commonly applied being steam-assisted gravity drainage (SAGD) and cyclic steam stimulation (CSS).

Mining-Based Bitumen Production. Overburden removal is typically performed with a truck-and-shovel operation.[5] Bitumen ore is mined with diesel or electric hydraulic shovels. Large haul trucks move the ore to crushing and slurrying

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centers for hydrotransport to extraction centers (diesel-powered using fuel generated on site as SCO). Some processing equipment is powered with electricity coproduced on site from natural gas, upgrading process gas, or coke.[6] Published estimates of mining energy consumption vary by an order of magnitude (0.3−3.6 GJ/m^3 of SCO).[6−8] Given that the high end of this range (3.6 GJ/m^3 SCO) represents some 10% of the energy content of the SCO, this is most likely an overestimate of mining energy inputs.

At the extraction facilities, bitumen froth (60%+ bitumen, remainder water) is separated from sand, requiring warm water and consuming ≈40% of the energy used to produce a barrel of SCO.[5] Within integrated mining operations, upgrader by-products such as process gas and coke provide heat and power for the separation process.[6] After primary separation, bitumen froth is treated to remove water and solids, using naphtha or parrafinic solvents. This produces clean bitumen ready for upgrading to synthetic crude oil. Energy costs for separation of the bitumen are estimated at 0.9 GJ/m^3.[8,9]

In Situ Bitumen Production. Bitumen and heavy oil in the oil sands region are generally produced in situ using thermal methods such as CSS and SAGD, although smaller amounts of cold (primary) production of extra-heavy oil does occur in the oil sands region.[5,10] A significant reduction in hydrocarbon viscosity with modest increases in temperature allows bitumen to flow to the well for production. Thermal in situ production is generally more energy-intensive than mining-based production.

GHG emissions from in situ production result primarily from fuels combusted for steam generation. A key indicator is the steam oil ratio (SOR), often measured as cubic meters of cold-water equivalent (CWE) steam injected per cubic meter of oil produced. SORs for commercial thermal in situ recovery projects generally range from 2 to 5, with the production-weighted industry average being 3.6 in 2009.[10] This represents the volume-weighted average of projects listed in Energy Resources Conservation Board data sets as "commercial-CSS" and "commercial-SAGD". Primary production of bitumen is not included because steam is not injected. SORs above 10 have been reported, but these represent transient effects at the outset of SAGD operations.[10] SORs have tended to improve over time with the maturation of SAGD technology.

The SOR is not the sole driver of in situ extraction emissions.[11] The amount of energy required to convert water to steam for injection depends on steam quality and pressure, the efficiency of steam generation, and heat recovery from produced fluids. Because of the requirement for 100% quality steam, the energy content of steam for SAGD projects is higher than that in heavy oil TEOR projects,[12] at ≈2.8 GJ/m^3.[11,13] Steam enthalpy varies little at relevant SAGD pressures, but the partitioning between sensible and latent heat changes across low- and high-pressure SAGD operating pressures.[11] To produce 100% quality steam, 80% quality steam is first produced in once-through steam generators (OTSGs), and condensate is returned to the boiler using vapor−liquid separators. This requires rejection of solute-laden water ("blowdown" water). Energy can be lost as a result of warm blowdown water. This energy requirement can be offset by the fact that produced fluids in a mature SAGD operation are hot, allowing heat recovery from the produced fluids stream. This produced fluid heat recovery has been suggested to equal some 10−30% of the heat content of the steam.[11] Literature estimates for steam energy requirements vary: Charpentier cites up to 2.8 GJ/m^3 of steam, whereas Butler cites ≈3.4 GJ/m^3 for 100% quality steam generation with heat recovery.[14,15] Electricity consumption for in situ production has been estimated as 190 MJ/m^3 bitumen (8.25 kWh/bbl bitumen) but will vary with SOR due to dependence on pumping and separation loads.[5]

Steam generation for in situ production is generally fueled with natural gas. An exception is the OPTI-Nexen Long Lake project, which consumes gasified bitumen residues,[16,17] increasing GHG emissions compared with natural-gas-fueled SAGD.[17,18]

Bitumen Upgrading. Because contaminants are concentrated in heavy hydrocarbon fractions, bitumen has a high sulfur and metals content. In addition, bitumen is carbon-rich, hydrogen-deficient, and contains a larger fraction of asphaltenes than conventional crude oil. Thus, bitumen requires more intensive upgrading and refining than conventional crude oil.

Raw bitumen will not flow through a pipeline at ambient temperatures so it is upgraded to SCO or diluted with a light hydrocarbon diluent (creating "dilbit", or "synbit" if synthetic crude oil is used as the diluent) before transport. Diluent can be either returned to the processing site or included with bitumen to the refinery stream.

Greenhouse gas emissions from upgrading have three causes:

1 Combustion of fuels for process heat, including process gas, natural gas, and petroleum coke.
2 Hydrogen production using steam reformation of natural gas or, less commonly, from gasification of coke or bitumen residues.
3 Combustion for electricity generation (whether in cogeneration or off-site for from purchased electricity).

Upgrading bitumen to SCO is performed in two stages. Primary upgrading separates the bitumen into fractions and reduces the density of the resulting SCO. Secondary upgrading treats resulting SCO fractions to remove impurities such as sulfur, nitrogen, and metals.

Primary upgrading adjusts the H/C ratio by adding hydrogen or rejecting carbon from bitumen feedstock. The most common upgrading processes rely on fluid or delayed coking to reject carbon.[4,19,20] Coking generates upgraded oils as well as coke and process gas;[5] for example, Suncor's delayed coking upgrading resulted in 85% SCO, 9% process gas, and 6% coke by heating value.[21] Natural gas or coproduced process gas is often used to drive coking, but in a fluid coker, a portion of the coke is combusted to fuel the coking process. In existing operations, coke disposition varies: in 2009, Suncor combusted 26% of produced coke and exported another 7% for offsite use, and the rest was stockpiled or landfilled. In contrast, the CNRL Horizon project stockpiled all produced coke.[21]

A competing primary upgrading method uses hydrogen addition for primary upgrading. The Shell Scotford upgrader[22] uses an ebullating-bed catalytic hydrotreating process. Treating bitumen with hydrogen addition results in larger volumes of SCO produced from a given bitumen stream and a high-quality product. It also requires larger volumes of H_2, with associated natural gas consumption and GHG emissions. The Scotford upgrader produced 82% of process outputs as SCO, 18% as process gas, and no coke (on an energy content basis).[21]

In secondary upgrading, the heavier fractions of primary upgrading processes (which contain the majority of the contaminants) are hydrotreated (i.e., treated through the addition of H_2 in the presence of heat, pressure, and a catalyst). Light refinery-ready SCO of 30−34°API, 0.1 wt %

sulfur, and 500 ppm nitrogen is a common product.[23] Heavy SCO products, such as Suncor Synthetic H, are also produced, but in smaller quantities (≈20°API and sulfur content of ≈3 wt %).[24] In chemical composition, dilbit looks similar to heavy synthetic blends.

Hydrogen consumption by hydrotreaters is often in excess of 3 times the stoichiometric requirement for heteroatom removal because of simultaneous hydrogenation of unsaturated hydrocarbons.[23] Hydrogen consumed in secondary upgrading is generally produced via steam methane reformation of natural gas, regardless of primary upgrading process.[6] Current exceptions include the OPTI-Nexen integrated SAGD to SCO project, which gasifies bitumen residues for H_2 production. Consumption of H_2 in upgrading processes ranges from 1.2 to 3.1 GJ/m^3 of bitumen upgraded.[25]

Nearly all of the bitumen produced from mining is upgraded, while most of the in situ-based production is shipped as a bitumen/diluent mixture to refineries.[5] There is no fundamental physical or chemical reason that in situ-produced bitumen cannot be upgraded.[17]

SCO and Bitumen Refining. Nonupgraded bitumen supplied to refineries requires intensive refining because of quality deficiencies. Refining of bitumen also produces a less desirable slate of outputs without extensive processing as a result of high asphaltenes content. Light SCO is a high-value product with low sulfur content compared with conventional oils of similar density, because light SCOs lack the typical "bottoms" of a conventional crude oil (i.e., residual products from distillation). This is because components that would form the bottom of the distillation output profile are destroyed during upgrading.

Refining energy consumption is well correlated with the specific gravity and contaminant loading (e.g., sulfur) of input crude oil.[26,27] This is due to need for additional coking or additional hydrogen consumption, both of which are energy-intensive.

Noncombustion Process Emissions. Other process emissions include emissions from venting, flaring, and fugitive emissions (hereafter, VFF emissions). Environment Canada reported emissions of ~3 g CO_2/MJ bitumen mined and in situ emissions of less than 1 g CO_2/MJ of bitumen produced.[28] Yeh et al.[29] found for mining operations that tailings ponds fugitive emissions had a wider range than fugitive emissions reported by Environment Canada, with a range of 0−8.7 g CO_2/MJ and a representative value of 2.3 g CO_2/MJ. It is not clear whether Environment Canada incorporates tailings pond emissions in these figures.

Land Use Change Associated Emissions. Land use change emissions are associated with biomass disturbance and oxidation due to land clearing, soil disturbance, and peat disturbance. These emissions are likely smaller than venting and fugitive emissions, with values ranging from 1.0 to 2.3 g CO_2/MJ of bitumen produced (representative value 1.4 g CO_2/MJ) for mining operations.[29] In a case that development is 100% on peatlands, land use emissions would increase by a factor of 3, suggesting that peat disturbance is a key driver of oil sands land use GHG emissions.[29] In situ operations have negligible land use emissions, ≈0.1 g CO_2 equiv/MJ of crude produced.

COMPARING PREVIOUS OIL SANDS LCA RESULTS

A number of LCAs of oil sands production have been performed, although none are yet comprehensive with detailed coverage of all oil sands production processes.[25,30,31] Over time, LCA studies have improved in quality and quantity of documentation, although gaps remain in the realm of publicly available models (see the Discussion and Recommendations section, below).

This paper reviews recent studies to determine the differences between study assumptions and to explore the uncertainty in resulting GHG emissions. The studies reviewed include

- **GREET**, the Greenhouse gases Regulated Emissions and Energy in Transportation model by Wang et al., Argonne National Laboratory;[32,33]
- **GHGenius**, the GHGenius model by O'Connor S&T[2] Consultants;.[34,35]
- **Jacobs**, a study by Keesom et al., Jacobs Consultancy;[25]
- **TIAX**, a study by Rosenfeld et al., TIAX LLC, and MathPro Inc.;[18]
- **NETL**, two studies by Gerdes and Skone, National Energy Technology Laboratory.[36,37]

A previous comprehensive comparison of oil sands GHG studies[6,19,38−41] was produced by Charpentier et al.[14] Other useful reviews are provided by Mui et al.[42,43] and by Hobbs et al., IHS-CERA Inc.[44] We will not attempt to recreate the analysis of these studies but in some cases use their results. One study reviewed but not included above is the Oil sands technology roadmap,[5] which is the source for GREET energy inputs to oil sands production.[45]

DIFFERENCES IN MODEL TREATMENT OF OIL SANDS PROCESSES

Determining the exact causes of differences between the results of reviewed models is impossible without access to original model calculations, but analysis of reported inputs and assumptions can give insight into reasons for divergence between estimates. These inputs can also suggest which model produces the most accurate estimates of project-specific or industry-wide emissions.

In all discussion below, energy content is reported on higher heating value basis (MJ or GJ HHV), and volumes are converted to cubic meters at standard conditions. Where required, volume- and mass-to-energy content conversions are made with fuel-specific compositions and relations between hydrocarbon density and chemical composition and heating values[46] (see the Supporting Information for calculation details).

System Boundaries and Study Scope. A main cause of variability between observed study results is the differences in broad methodological choices, such as study scope, system boundaries, and processes modeled (see Table 1).

A key difference between models is that some models assess emissions for an "average" oil-sands-derived fuel pathway, or generate industry averages (GREET, GHGenius, NETL), whereas others model emissions from specific oil sands projects (TIAX and Jacobs). This methodological difference overshadows many other sources of between-model variability.

The use of differing data sources of differing qualities is another major factor. As Charpentier et al. note, "the nature of the data used for the analysis varies significantly from theoretical literature values to project-specific material and energy balances".[14]

Another important difference is the study system boundary. Studies differ in the their treatment of indirect emissions (e.g.,

Table 1. Study Scope and System Boundaries by Reviewed Study

	scope of coverage	indirect emissions	embodied energy	venting, flaring, fugitives	land use
GREET	ind., pathway average	yes	no	yes	no
GHGenius	ind., pathway average	yes	no	yes	yes
Jacobs	process	NG + elec	no	no	no
TIAX	process	yes	no	yes	no
NETL	ind. average	yes	no	yes	no

emissions associated with producing natural gas consumed in upgrading operations), venting, flaring, and fugitive emissions as well as emissions from land use changes associated with oil sands mining. No study included emissions embodied in capital equipment (e.g., steel or cement upstream emissions).

Surface Mining. Emissions from mining are driven by the fuel consumed per unit of bitumen produced and the consumed fuel mix. In integrated operations, it is difficult to separate mining and upgrading inputs. Surface mining assumptions for each model are described below. The assumed fuel mixes and magnitudes of fuel consumption for mining and upgrading are shown by model in Figure 1. For comparison, industry reported fuel consumption (from regulatory data provided by the Alberta Energy Resources Conservation Board, or ERCB) are plotted in the right-most column.[47]

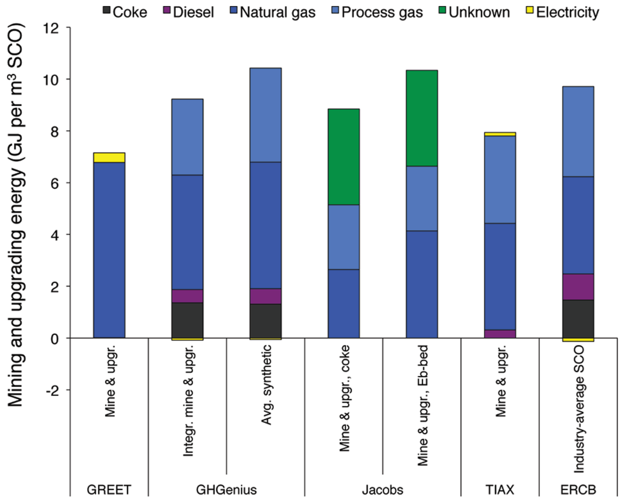

Figure 1. Fuel mix for mining and upgrading assumed by LCA models and industry average fuel mix (right). Fuel mix assumptions calculated from model inputs as described in text. Industry average fuel mix calculated from fuel consumption rates reported by ERCB for 2010 mining and upgrading operations.[47] See the Supporting Information for more detail on figure construction.

GREET. Estimates for diesel use are derived from Alberta Chamber of Resources data, which includes 340 MJ of electricity (94 kWh), 1573 MJ of natural gas, and 9 MJ diesel used/m^3 of bitumen mined.[45] This low diesel use is a possible difference between GREET results and those of other oil sands LCAs.

GREET assumes no coke consumption, which is at odds with empirical fuel mixes presented in Figure 1 and other reports.[6,19] In addition, although GREET figures are based on ACR fuel use data, GREET emissions are 15.9 g CO_2/MJ refined fuel delivered, whereas ACR emissions results are ≈19−22 g CO_2/MJ. (These figures are only approximate comparisons because ACR data are measured in kg CO_2/bbl of SCO produced, and conversion factors to energetic units are not provided in ACR.[5] SCO density and heating value were set to values for 31°API oil to allow comparison.) This is likely due to the omission of coke combustion in the GREET model. Charpentier previously noted these discrepancies, stating that "the energy balance in GREET appears to omit the diesel fuel used in mining and the coke used in upgrading".[14]

GHGenius. Data include emissions from off-site power and hydrogen production[35] as well as on-site cogeneration. Stand-alone mining operations consume 1.35 GJ diesel/m^3 of bitumen produced, 2.78 GJ natural gas, and coproduce 250 MJ of electricity for export. The weighted fuel mix in GHGenius for mining and upgrading to synthetic crude assumes 15% of energy content from coke,[34] closely in line with observed industry average mining fuel mix (see Figure 1).

Jacobs. The surface mining process model is not described in detail. It is stated that the energy for mining is "one-half of energy needed for SAGD at an SOR of 3." This represents an energy cost of ≈3.7 GJ/m^3 of bitumen of unknown fuel mix. Process model represents an integrated operation fueled with natural gas and using either ebullating-bed hydrogen-based upgrading or coking (no coke combustion). It is therefore similar to the CNRL Horizon oil sands project.

TIAX. The model represents the CNRL Horizon mining and upgrading project, which consumes natural gas and stockpiles coke generated during upgrading.[18] Total consumption for mining and upgrading is ≈8 GJ/m^3 of SCO.

NETL. The model uses emissions reported by Syncrude for integrated mining and upgrading operation,[37] as reported in Environment Canada facilities emission database.[48]

The TIAX and GREET models assume lower energy consumption than the industry average, whereas the Jacobs and GHGenius models are in line with observed consumption values. The GHGenius model has the most accurate fuel mix assumption for an industry average. Because Jacobs and TIAX model a specific project (e.g., CNRL Horizon) that is natural-gas-fueled, they do not replicate the industry average fuel mix.

This importance of fuel mix on emissions has implications for future emissions. Some argue that future projects will rely on coke as much as or more than current operations, because of decreasing availability of low-cost natural gas,[17,19] and others believe that unconventional gas resources will allow low gas prices in the long term.

One complication in comparing these studies is uneven modeling of cogeneration of electric power. This shortcoming is likely to be a secondary source of uncertainty. For example, Suncor exported some 4.1 PJ of electric power in 2009, compared with electricity consumption of 7.5 PJ and total energy consumption of 137.1 PJ,[21] suggesting that credits or debits due to cogeneration will likely be a secondary source of variation.

Upgrading Emissions. Upgrading emissions are driven by the energy consumed per unit of SCO produced plus the fuel mix used in upgrading. Study assumptions regarding upgrading include

GREET. Consumption of natural gas is ≈3.3 GJ/m^3 SCO produced.[45] No consumption of coke or process gas is recorded, which differs from reported fuel mixes by operators.[47]

Upgrading consumption values are low compared with other estimates (e.g., Jacobs).

GHGenius. Imputed upgrading consumption in integrated mining and upgrading is 5.1 GJ/m^3, whereas stand-alone upgrading is much more energy-intensive at ≈9.8 GJ/m^3 SCO.[35] Fuel mix is included in Figure 1.

Jacobs. Consumption is ≈5.7 GJ/m^3 SCO for coking, and 7.4 GJ/m^3 SCO for Eb-bed. Fuel mix includes both natural gas and process gas. The fuel mix is ≈50% each natural gas and process gas for the coking unit, 60% natural gas and 40% process gas in Eb-bed reactor,[25] with no consumption of coke.

TIAX. The study does not report upgrading consumption separately from mining or SAGD consumption. Integrated operations are modeled, and process flows are not delineated by mining and upgrading stages.[18]

NETL. A separate description of upgrading is not given in NETL studies.[36,37] Upgrading emissions are included in emissions from Syncrude integrating mining and upgrading operation, as described above.

Differences in emissions between Jacobs and GHGenius estimates are likely due to fuel mix differences, due to the similar energy consumption values. Given observed consumption of coke (see ERCB data in Figure 1), GHGenius estimates are more representative of industry-wide upgrading emissions. GHG-intensive upgrading using bitumen residues at OPTI-Nexen Long Lake project is neglected in all models except TIAX, but this is a relatively small operation, and therefore, this will not strongly affect model results in other models.

In Situ Production. Because of relatively homogeneous fuel mix consumed for in situ production, the primary determinants of emissions are the SOR and the energy consumed per unit of steam produced. In some studies, the product of these two terms—the energy consumed per volume of crude bitumen produced—is reported. Model assumptions include

GREET. Natural gas consumption is ~6.8 GJ/m^3 bitumen.[45] Because no SOR is reported, the energy consumed per cubic meter of steam cannot be calculated.

GHGenius. SORs of 3.2 and 3.4 assumed for SAGD and CSS, respectively.[14,49] Natural gas consumption is 9.6 and 10.2 GJ/m^3 of bitumen produced for CSS and SAGD, respectively.

Jacobs. Jacobs assumes SORs of 3.[25] Energy content of steam is 2.06 GJ/m^3 CWE steam, and efficiency is 85% (LHV basis), for total consumption of ≈8.1 GJ LHV/m^3 bitumen. Cogeneration of electric power provides an emissions offset in some cases.[25] Because SAGD net cogeneration exports are not reported in ERCB data sets, electricity exports cannot be verified using reported industry data.[22]

TIAX. Natural gas consumption rates are at the low end of the above cited range, 4.1 and 7.8 GJ/m^3 bitumen for Christina Lake (SAGD) and Cold Lake (CSS) respectively (without cogeneration).[18] The Christina Lake SAGD case has an SOR of 2.5 and a low implied energy consumption of 1.7 GJ/m^3 CWE of steam. These values are lower than the empirical values shown below, driving the low emissions from the TIAX natural gas case. Cases with cogeneration have somewhat higher effective steam energy requirements (see the Supporting Information). TIAX is the only report to consider integrated in situ production with bitumen residue or coke fueling. The TIAX case with asphaltenes residue gasification for steam generation (analogous to OPTI-Nexen Long Lake project) has a higher energy demand of 5.4 GJ/m^3 of steam generated, resulting in much higher emissions, as should be expected from the carbon intensity of asphaltene residue gasification.[18]

NETL. Emissions calculated for Imperial Oil Cold Lake project using CSS,[37] as reported in the Environment Canada facilities emission database.[48] In 2009, Cold Lake had an SOR of 3.5.[21]

The energy intensity of steam generation for the reviewed studies can be compared with calculated values from engineering fundamentals and values reported in the literature. These comparisons are shown in Figure 2. At top are fundamental computations of energy requirements, including the steam enthalpy at typical SAGD conditions (100% quality steam at 2000 kPa, or $h_g \approx 2.8$ GJ/m^3)[11] and the required energy consumption for steam generation, assuming no heat recovery from produced fluids. Also shown is a consumption band assuming 10−30% heat recovery from produced fluids. Next, estimates from the literature are presented, which are generally in line with fundamental values. Next, monthly energy intensities for 8 in situ projects are calculated from the reported literature. Last, assumptions for energy consumption in steam generation are shown for reviewed LCA models. A key result is that TIAX values are significantly lower than values from the literature. See the Supporting Information for figure construction details.

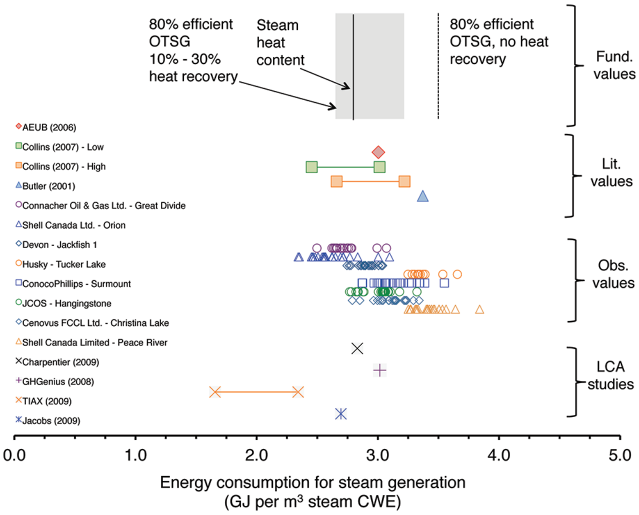

Figure 2. Assumed energy intensity of steam generation for studies and values from literature. Lines and shaded areas represent the energy content of the steam at typical SAGD conditions[11] (solid), the energy cost of obtaining this steam with an 80% efficient OTSG and complete heat recovery from blowdown water (dashed), and the energy cost with 80% efficient OTSG and heat recovery of 10−30% of the enthalpy of steam from warm produced fluids (shaded). Values are from the literature from various sources.[11,15,50]

In addition, the SORs assumed can be compared with SORs observed in practice, as in Figure 3. The SOR histogram shows SORs by fraction of industry output from reported data, as well as averages by process type (top axis). GHGenius and NETL report SORs in line with observed SORs, whereas the TIAX SAGD case is toward the low end of observed SORs.

Refining Emissions. Many LCA studies to date treat the refining of crude inputs (SCO and bitumen) in a simple fashion.[32,51] This is partly due to the absence of publicly available models of refinery operations and due to the fact that some models (such as GREET) have sought to produce a national average result, without modeling refining differences between individual crude oils.

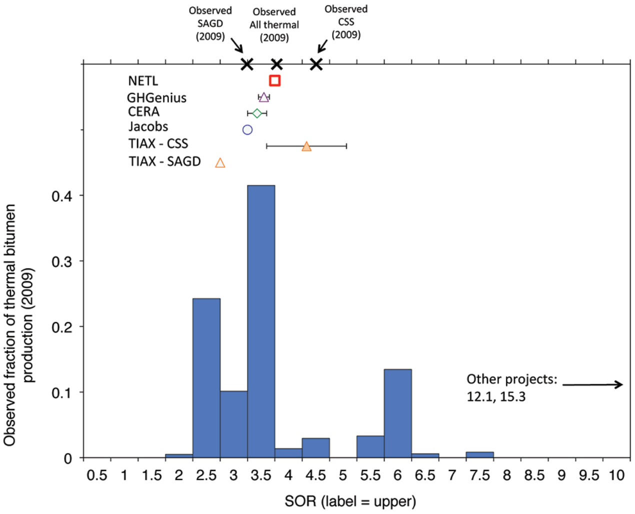

Figure 3. Assumed SORs for each model compared with observed SORs from ERCB data. Top marks represent production-weighted average for CSS and SAGD operations and 2009 full-year production volumes.

Refinery feedstock qualities differ by study, as shown in the Supporting Information. Some studies do not state explicitly the quality of refinery feedstock. SCO characteristics from studies align well with the reported characteristics of commercial SCO products. The resulting estimates of refining emissions as a function of crude specific gravity are plotted in Figure 4.

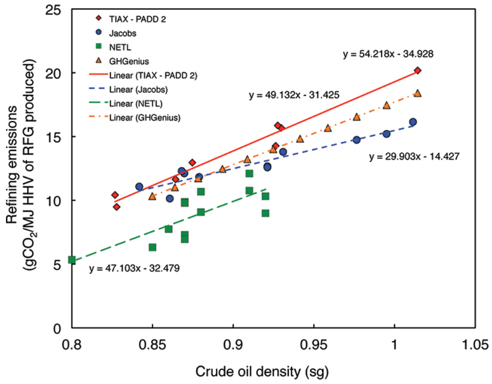

Figure 4. Refining emissions as a function of crude specific gravity for oil sands GHG emissions study. For TIAX, Jacobs, and NETL, the sulfur content varies with crude type. For GHGenius results, model version 3.20 was used with 2 wt % sulfur content for all crude oils.

GREET. The model calculates refinery emissions from processing oil-sands-derived streams as equivalent to processing conventional crude oil streams.[45,40] This assumption will not result in significant errors because GREET assumes bitumen is upgraded to SCO.[40]

GHGenius. The model relies (as of version 3.20) on a linear model of refinery emissions as a function of API gravity and sulfur, derived from Karras.[26] The relationship between sulfur and emissions is from Karras, and the slope of energy consumed as a function of density is set to one-half the Karras value.[34]

Jacobs. Detailed calculation of refinery inputs and outputs with refining simulation software. Results from the commercial refinery process model are presented in detail, with process throughputs and products breakdown provided for SCO, bitumen, and dilbit.[25] Detailed refining utilities consumption by subprocess is presented for Arab Medium crude, but not for oil sands pathways.[25]

TIAX. The model performs a detailed calculation of refinery inputs and outputs, using industry refinery modeling expertise, with extensive documentation. Model results include differential refining emissions based on the quality of the feedstock.[18]

NETL. The approach used by Gerdes et al.[36] is outlined in detail in Skone et al.[37] A novel approach is developed using US nationwide statistical data on refinery configurations, crude throughputs, crude qualities, and utilization factors for different crude processing stages (e.g., distillation utilized capacity vs fluid catalytic cracking utilized capacity). This approach is similar to that taken by Karras.[26] Heuristic models for the effect of crude density and sulfur content on refining intensity are developed.[36]

The Jacobs and TIAX models represent the most thorough efforts to date to model refinery emissions for refining oil-sands-derived fuels. The NETL model represents the most thorough treatment of the problem using public data. Given the relative similarity of refinery emissions model results, it is not clear that enough empirical data exists about refinery emissions to assess the relative merits of the different models. One concern in refinery modeling is that the different quality of SCO as compared with conventional oil will change refinery output slates, possibly indirectly affecting emissions in other sectors (see Discussion and Recommendations, below). In addition, a number of parameters not included in current simple refining models could be causing discrepancies between different model results (for example, Jacobs notes sensitivity to refinery configuration, which is not included in simpler models).

Other Process Emissions. Emissions from venting, fugitive emissions, and flaring (VFF) are unevenly addressed in the above studies. GREET does not include VFF emissions from bitumen extraction or upgrading.[40] GHGenius does include venting and flaring emissions.[34] Jacobs does not explicitly include VFF emissions from oil sands production.[25] TIAX does include VFF emissions, of 0.5 to 3.3 g CO_2 equiv/MJ[18] from regulatory documents related to the Horizon oil sands mine. NETL does include venting and flaring,[36] but does not describe method for estimating bitumen VFF emissions.

Land use emissions are considered only in the GHGenius model, which calculates soil and biomass disturbance per hectare and apportions this according to the type of operation (e.g., 100% disturbance on mined lands, no disturbance for SAGD).[35]

Resulting GHG Emissions Estimates. The resulting upstream GHG emissions estimates by study are shown in Figure 5. For simplicity, vehicular emissions (tank-to-wheel) emissions are given a nominal value of 70 g CO_2/MJ in all cases (TTW results are largely consistent across models and are not a focus of this study). A detailed breakdown of emissions for each data point is given in the Supporting Information.

General trends emerge among pathways as a result of the underlying fundamentals of process operation. In situ and upgrading projects have higher emissions, as should be expected from projects that combine energy-intensive extraction methods with energy-intensive upgrading.

Variability between estimates from a given study arise from varying process assumptions. For example, the four TIAX results for in situ-to-bitumen pathways differ in their

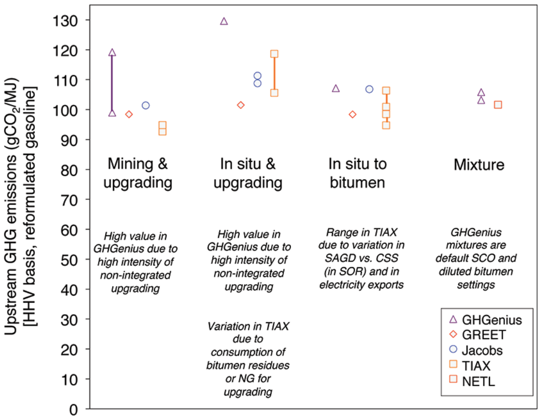

Figure 5. Full-fuel-cycle GHG emissions estimates for reformulated gasoline pathways by study. Nominal value of 70 g CO_2/MJ for combustion emissions is applied evenly across all studies. Details on construction of th estimates are given in the Supporting Information.

assumptions about the method of extraction used (i.e., SAGD vs CSS) and in whether they export cogenerated electric power. Clearly, emissions will vary between among implementations of similar pathways.

In general, GREET and TIAX model results are at the lower end of the emissions range. This should be expected from their assumptions about the energy intensity of extraction, as shown above for mining and in situ production.

In addition, in general, the GHGenius model tends to have somewhat higher emissions than other studies. A driver of these higher emissions is due to more careful accounting of energy consumption in GHGenius and due to industry-average fuel mixes that contain coke combustion. Some additional research is needed with respect to GHGenius stand-alone upgrading emissions, which are assigned a high emissions intensity. This does not strongly affect the overall results from GHGenius (as plotted in Figure 5 in the "mixture" column as default SCO and default bitumen pathways) because stand-alone upgrading is not a major pathway in current operations. In general, given the fidelity of GHGenius in replicating energy inputs to mining and in situ processes, GHGenius emissions estimates should not be considered overly pessimistic.

■ DISCUSSION AND RECOMMENDATIONS

Recommended Use of Model Results. The GHGenius model is recommended for use in generating industry-average GHG emissions values, such as those that might be required to assign default values in regulation. GHGenius contains the most accurate representation of observed energy consumption values for the industry as a whole, as seen in Figure 1 for surface mining and upgrading operations, in Figure 2 for steam energy content, and in Figure 3 for steam/oil ratios. It also includes emissions sources such as VFF and land use emissions that are not covered consistently by other models. In addition, its transparent and extensive documentation is a useful contribution to the literature and allows for fact checking of inputs.

Although the GREET model is publicly available and treats industry average pathways, its use for constructing industry-average emissions is not recommended because of less accurate energy intensity and fuel mix assumptions compared with GHGenius.

The Jacobs and TIAX models represent more detailed LCA studies of project-specific emissions. They provided important advances in refinery models compared with earlier studies. These estimates are useful for understanding specific pathways, but should not be considered representative of industry-wide emissions averages because of their focus on specific projects that may not be representative of general industry conditions.

Comparability of Studies. Figure 5 shows the considerable variation among model results for different processes and even significant variation within similar pathways. The key factor affecting the comparability of studies is whether study results are process-specific or pathway or industry-average emissions estimates. Process-specific emissions estimates and industry-average emissions estimates are useful in different contexts.

For regulatory purposes for determining the potential overall scale of differences in emissions among broad fuel types (e.g., conventional oil and oil sands), industry-wide production-weighted average emissions are more useful than process-specific assessments. For evaluating the GHG intensity of a given process or a given import stream, process-specific emissions estimates are required.

Other factors affecting the comparability of models include the study system boundaries. In the studied LCA models, study system boundaries are broadly commensurate (e.g., all are well-to-wheel LCA analyses), although smaller system boundary considerations were noted above, such as the inclusion or exclusion of land use emissions.

Uncertainties and Need for Future Work. A number of uncertainties remain in the area of oil sands GHG emissions. Treatment of cogenerated electric power varies among models. Given the CO_2 intensity of the Alberta grid, coproduction credits from cogenerated power could be provide emissions offsets. Important future research needs for electricity credits include variation with time, place, and characteristics of Alberta grid in relation to interconnected grids.

Treatment of refining is a difficulty in public-domain studies such as GREET and GHGenius because of a lack of access to industry-vetted refinery models. The Jacobs and TIAX refining models represent the most detailed work to date on refining emissions (although their models are not publicly available). The previous lack of data on refining emissions has been remedied somewhat recently, with increasing public access to correlations between emissions and crude density and sulfur content,[26] but additional work is needed. Importantly, refinery emissions vary with refinery configuration, the type of oil sands product refined (i.e., SCO, dilbit or synbit), and the refinery output slate.

Numerous coproduction issues arise that are not incorporated consistently in current studies. For example, the treatment of coproduced coke is a complex issue. This is noted in the Jacobs study but not treated elsewhere. At remote Alberta upgrading facilities, coproduced coke is generally stockpiled or burned on site to fuel operations. If bitumen is shipped to refineries as dilbit, this will result in coke generation near existing fuels markets, which could result in more coke being consumed, offsetting some coal consumption. Calculating the magnitude of credit or debit associated with such coproduction and displacement is nontrivial and requires understanding of the markets for solid fuels. Similar concerns arise with the treatment of diluent in dilbit pathways.

The interaction of oil sands products with existing fuel production systems and fuel demands is still poorly understood. For example, refinery outputs from refining a light SCO product will differ from outputs from a crude oil input of similar specific gravity and sulfur content (more middle distillate and less residual fuel from SCO). This could have ripple effects on other fuels markets and alter the energy requirements of producing a given refinery mix (e.g., EU refineries might not face as large an energy penalty associated with producing diesel-heavy refinery product slate).

The interaction of markets in LCA (as addressed in "consequential" LCA) is not studied in detail in any of the above models. Given a regulation that reduced the demand for oil sands products in North America (such as an expansion of the California LCFS to the national scale), there could be shifts in shipment of liquid fuels in the global fuels market (also known as crude shuffling). This shift of fuels could offset some of the desired reduction in emissions. The calculation of such impacts would require a combination of fuel market models with detailed LCA models. This is a difficult problem and likely subject to significant uncertainty.

Future work in oil sands GHG emissions should move toward modeling the emissions of specific process configurations. For example, models should be used to model emissions by project and compare those modeled emissions to reported emissions estimates. More vigorous calibration with available data (such as ERCB reported data sets) will help verify model accuracy. Much of the variability seen in the results above is driven by fundamental differences between different process operations (e.g., fuel mix or steam generation efficiency variation between project). Without more transparency and clarity about which processes are being modeled (and how representative they are of industry-wide operations), additional confusion will be introduced into assessing the environmental impacts of oil sands production.

■ ASSOCIATED CONTENT

Ⓢ Supporting Information

Additional results, including tabular results for important results figures. This material is available free of charge via the Internet at http://pubs.acs.org.

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: abrandt@stanford.edu.

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