

# Diesel Engines: Environmental Impact and Control

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## ABSTRACT

The diesel engine is the most efficient prime mover commonly available today. Diesel engines move a large portion of the world's goods, power much of the world's equipment, and generate electricity more economically than any other device in their size range. But the diesel is one of the largest contributors to environmental pollution problems worldwide, and will remain so, with large increases expected in vehicle population and vehicle miles traveled (VMT) causing ever-increasing global emissions. Diesel emissions contribute to the development of cancer; cardiovascular and respiratory health effects; pollution of air, water, and soil; soiling; reductions in visibility; and global climate change.

Where instituted, control programs have been effective in reducing diesel fleet emissions. Fuel changes, such as reduced sulfur and aromatics content, have resulted in immediate improvements across the entire diesel on- and off-road fleet, and promise more improvements with future control. In the United States, for example, 49-state (non-California) off-road diesel fuel sulfur content is 10 times higher than that of national on-road diesel fuel. Significantly reducing this sulfur content would reduce secondary particulate matter (PM) formation and allow the use of control technologies that have proven effective in the on-road arena. The use of essentially zero-sulfur fuels, such as natural gas, in heavy-duty applications is also expected to continue.

Technology changes, such as engine modifications, exhaust gas recirculation, and catalytic aftertreatment, take longer to fully implement, due to slow fleet turnover. However, they eventually result in significant emission reductions and will be continued on an ever-widening basis in the United States and worldwide. New technologies, such as hybrids and fuel cells, show significant promise in reducing emissions from sources currently dominated by diesel use. Lastly, the turnover of trucks and especially off-road equipment is slow; pollution control agencies need to address existing emissions with in-use programs, such as exhaust trap retrofits and smoke inspections. Such

a program is underway in California. These and other steps that can be continued and improved will allow the use of the diesel engine, with its superior fuel consumption, to continue to benefit society while greatly reducing its negative environmental and health impacts. The next ten years can and must become the "Decade of Clean Diesel."

## INTRODUCTION

This review summarizes and evaluates published information on diesel emissions, their effects on the environment and human health, and ways to minimize emissions and effects. It finds that diesels are an important part of the public and private transportation sector and their use will continue and grow into the future. Although large emission reductions have been achieved through fuel reformulation, engine redesign, and exhaust treatment, additional reductions are needed. These reductions are essential to reducing health risks, improving visibility, and minimizing water and soil contamination. They are practical and cost-effective. In the long term, fossil-fueled transportation systems may be replaced by renewable energies, probably using fuel cells for energy production.

The diesel engine is a major tool in the day-to-day life of modern society. It powers much of our land and sea transport, provides electrical power, and is used for many farming, construction, and industrial activities. The diesel does all this in a package that is robust and durable, with lower fuel consumption than any other prime mover. But diesel pollutes, and concern is growing over how much diesel pollution affects human health and well-being. However, technology advancements are now becoming available that may dramatically reduce diesel emissions and make 2001–2010 the "Decade of Clean Diesel."

Much of the history of the internal combustion engine involved a search for the best way to get more power for less fuel cost. For example, James Watt (1736–1819) successfully halved the coal consumption of Thomas Newcomen's (1663–1729) steam engine by modifying the cycle through the addition of a separate condenser.<sup>1</sup>

Similarly, Rudolf Diesel (1858–1913) used the developing science of thermodynamics and the ideas of Sadi Carnot (1837–1894) to develop his “economical heat motor,” which has subsequently evolved into the fuel-efficient diesel engine we know today.<sup>1</sup> But progress does not happen overnight. Diesel’s initial patent applications were filed in 1892,<sup>2</sup> but it took almost 8 years to develop the technology to the point of a producible and marketable engine.<sup>1</sup>

In its present form, the diesel engine has both advantages and disadvantages. To endure the diesel cycle’s high working pressures (1.5 times higher than those of gasoline engines of comparable power), heavier and more costly engine components are required. The diesel’s lean combustion characteristics produce less power per unit displacement than gasoline engines produce. Finally, the diesel’s diffusion flame combustion process is slower than the premixed combustion of a typical gasoline engine, thereby limiting diesel engines to lower maximum operating speeds.<sup>3</sup> These limitations, however, are balanced by the diesel’s benefits. At full load, the diesel engine uses only ~70% of the fuel that a comparable gasoline engine

consumes for the same power output. Under partial load conditions, the diesel’s advantage is even greater.<sup>3,4</sup> A heavy-duty diesel truck’s annual fuel cost is \$10,993, compared with \$15,977 for an equivalent gasoline-powered truck (if one were available).<sup>5</sup> The diesel engine’s lower fuel cost has made it the most attractive choice for vehicular and mechanical power in modern society.

Diesel engines power most of the commercial transportation of goods in the United States.<sup>5</sup> The engines are primarily used in trucks, freight locomotives, and marine vessels.<sup>6</sup> The number of diesel engines in use is large and growing. In the United States, the number of diesel trucks (including pickups, panels, minivans, sport utility vehicles, and station wagons) has increased by almost one-third, from 3,713,200 in 1992 to 4,913,300 in 1997.<sup>7</sup> Figure 1 shows how annual vehicle miles traveled (VMT) have increased during the same survey period in several diesel truck size categories.<sup>7,8</sup> The diesel is also used widely in other countries. Figure 2 shows the growth of diesel passenger car sales in Europe, with a prediction that one of every three cars sold there in 2001 will be

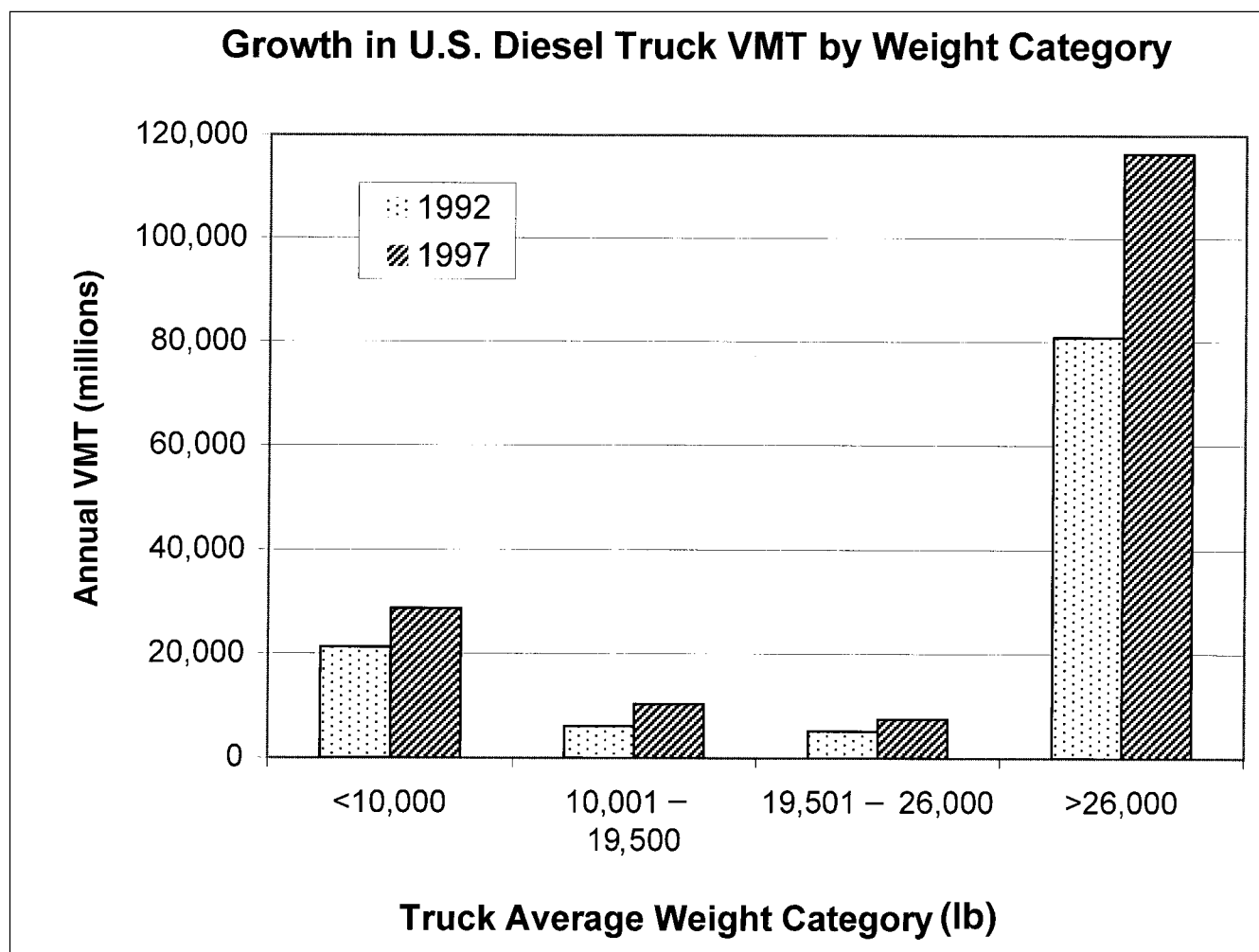


Figure 1. Annual VMT increased significantly in the early to mid-1980s in several diesel truck size categories in the U.S.<sup>7,8</sup>

diesel-powered.<sup>9,10</sup> The estimated contribution of the for-hire trucking industry to the 1997 U.S. economy was \$92 billion—more than 1% of the gross domestic product and almost \$18 billion more than the economic contribution from air transportation.<sup>6</sup> The manufacture of diesel engines and equipment, and the production and distribution of fuel, contributed an additional \$85 billion during that same year.<sup>5</sup>

Despite its many advantages, the diesel engine is inherently dirty. Particulate matter (PM) emissions from diesel combustion contribute to urban and regional hazes. Nitrogen oxides (NO<sub>x</sub>) and hydrocarbons (HCs) are precursors for O<sub>3</sub> and PM. Some components of HC and PM emissions have been identified as toxic substances with the potential for adverse health effects.<sup>11,12</sup> Since the 1970s, increasingly stringent emission standards have been imposed on diesel engine manufacturers. As a result, engine-specific NO<sub>x</sub> emissions have decreased ~80%, and new engines emit ~90% less PM than was emitted 1970. Visible smoke emissions have been reduced via state inspection programs that monitor and enforce manufacturer and operator compliance.

Figure 3 shows cost-effectiveness estimates for several typical California mobile-source control measures of the late 1980s and 1990s. With the exception of Phase 2

reformulated gasoline, these control measures (including several for diesel emissions) are all highly cost effective at about \$1/lb or less of ozone precursors (NO<sub>x</sub> and HC). Mobile source regulations have typically been considered cost-effective when such implementation costs are about \$6/lb or lower.<sup>13</sup> Because PM mass emission rates are lower than NO<sub>x</sub> or HC emission rates, PM cost-effectiveness values are higher. In California, the PM control cost effectiveness for new bus emission standards was about \$17.90/lb.<sup>14</sup> As experience is gained with PM emission controls, a better idea of what costs are reasonable and acceptable is evolving.

New engine emission standards have also resulted in diesel exhaust emission reductions. As shown in Figures 4a and 4b, off-road and heavy-duty on-road vehicles contribute a majority of the diesel NO<sub>x</sub> and PM emissions. Since the late 1980s, NO<sub>x</sub> emissions from off-road vehicles, which have few regulatory controls, have exceeded those from on-road vehicles.<sup>15</sup> Figure 4a shows NO<sub>x</sub> emissions for heavy-duty on-road vehicles decreasing in the late 1980s, but during the 1990s, the increasing numbers of diesel vehicles and mileage traveled countered reductions from individual vehicles. Figure 4b demonstrates a downward trend in PM emissions for heavy-duty on-road emissions, but off-road vehicle emissions leveled off after 1988.<sup>15</sup>

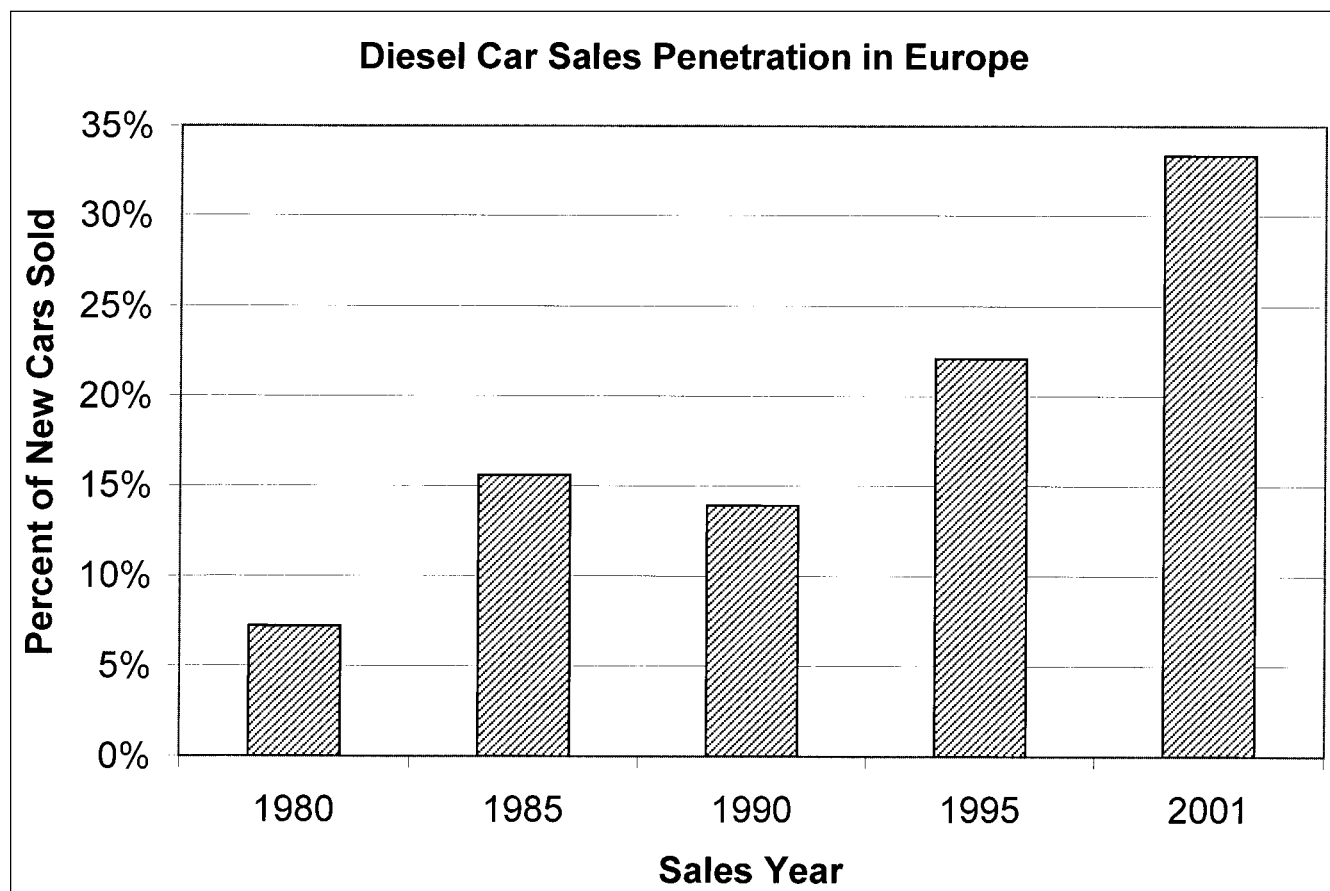


Figure 2. It is predicted that one of every three cars sold in Europe in 2001 will be diesel-powered.<sup>9,10</sup>

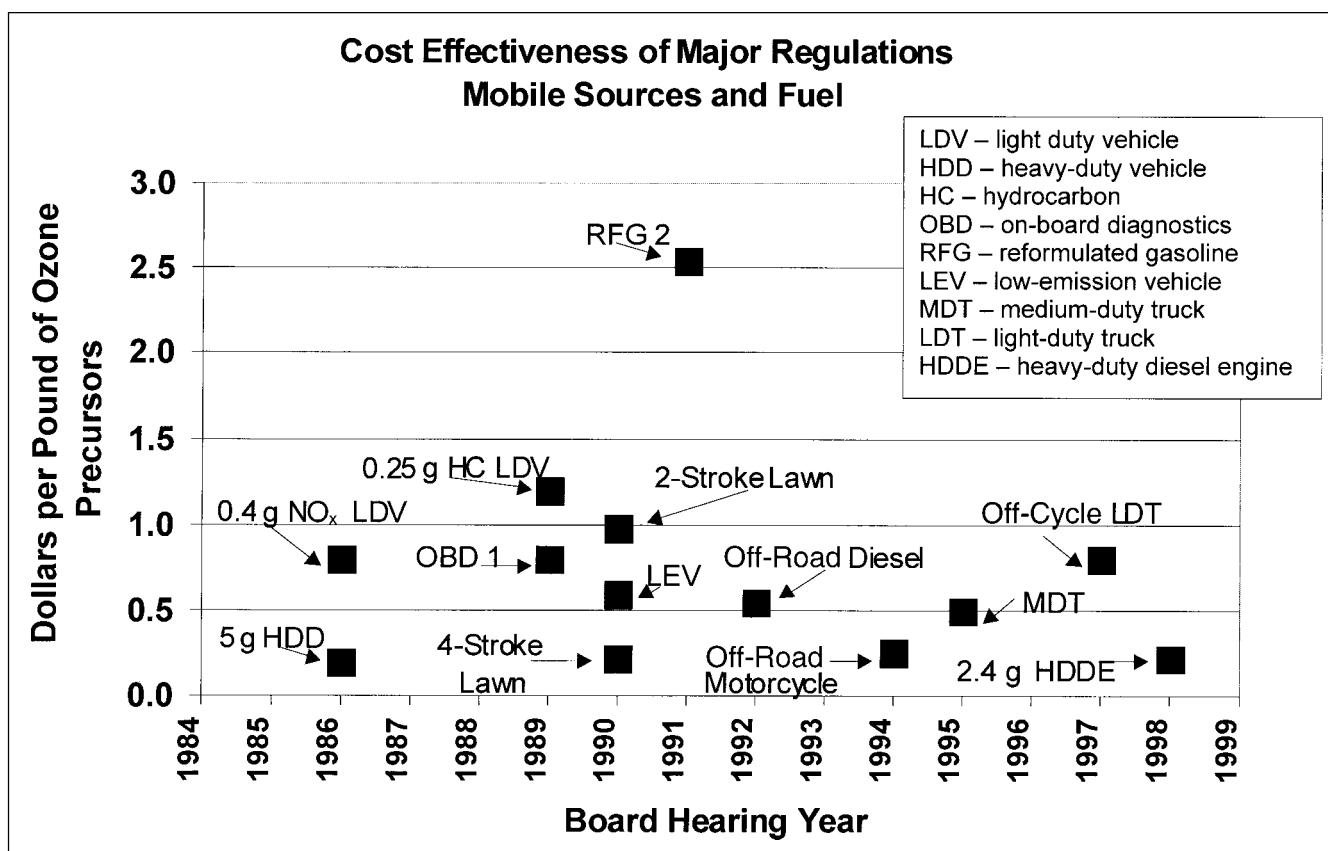


Figure 3. Cost-effectiveness values for various mobile source regulations approved by the California Air Resources Board in the late 1980s and 1990s show that most fall into a narrow and acceptable range.

Diesel's effects on air, water, and soil are multimedia, as illustrated in Figure 5, and are not limited to the effects of engine exhaust emissions. Diesel engine and vehicle manufacturing, as well as the production, storage, and distribution of diesel fuel, have adverse environmental consequences. Hydrocarbons are released from wells, refineries,

storage tanks, and pipelines into the atmosphere, ground, and groundwater. Runoff from leaks and spills enters surface water, while atmospheric deposition adds to ground and surface water contamination. Vehicle and engine production (including steel production and product transport) emit HCs and PM that also pollute air, water, and land.

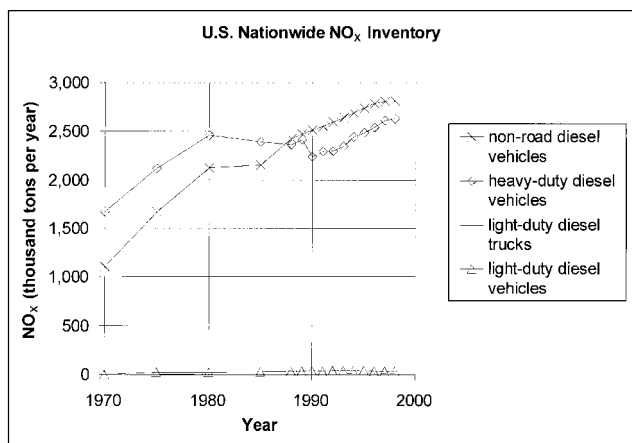


Figure 4a. The development and implementation of emission standards for new engines and vehicles is a primary means of reducing NO<sub>x</sub> from diesel exhaust emissions. The majority of diesel NO<sub>x</sub> emissions are from nonroad and larger on-road vehicles. Since the late 1980s, the amount of NO<sub>x</sub> emitted by nonroad vehicles, which have few regulatory controls, has exceeded on-road vehicle NO<sub>x</sub> emissions.<sup>15</sup>

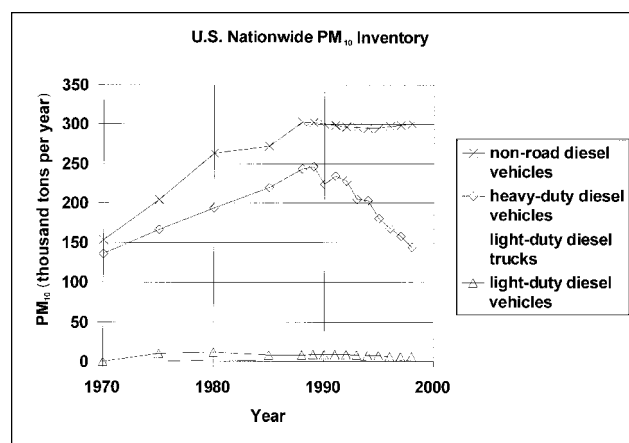


Figure 4b. The development and implementation of emission standards for new engines and vehicles is a primary means of reducing PM from diesel exhaust emissions. The majority of diesel PM emissions are from non-road and larger on-road vehicles. Large on-road diesel vehicle PM emissions show a downward trend, while the relatively uncontrolled nonroad vehicle emissions have leveled off, but at a relatively high value.<sup>15</sup>

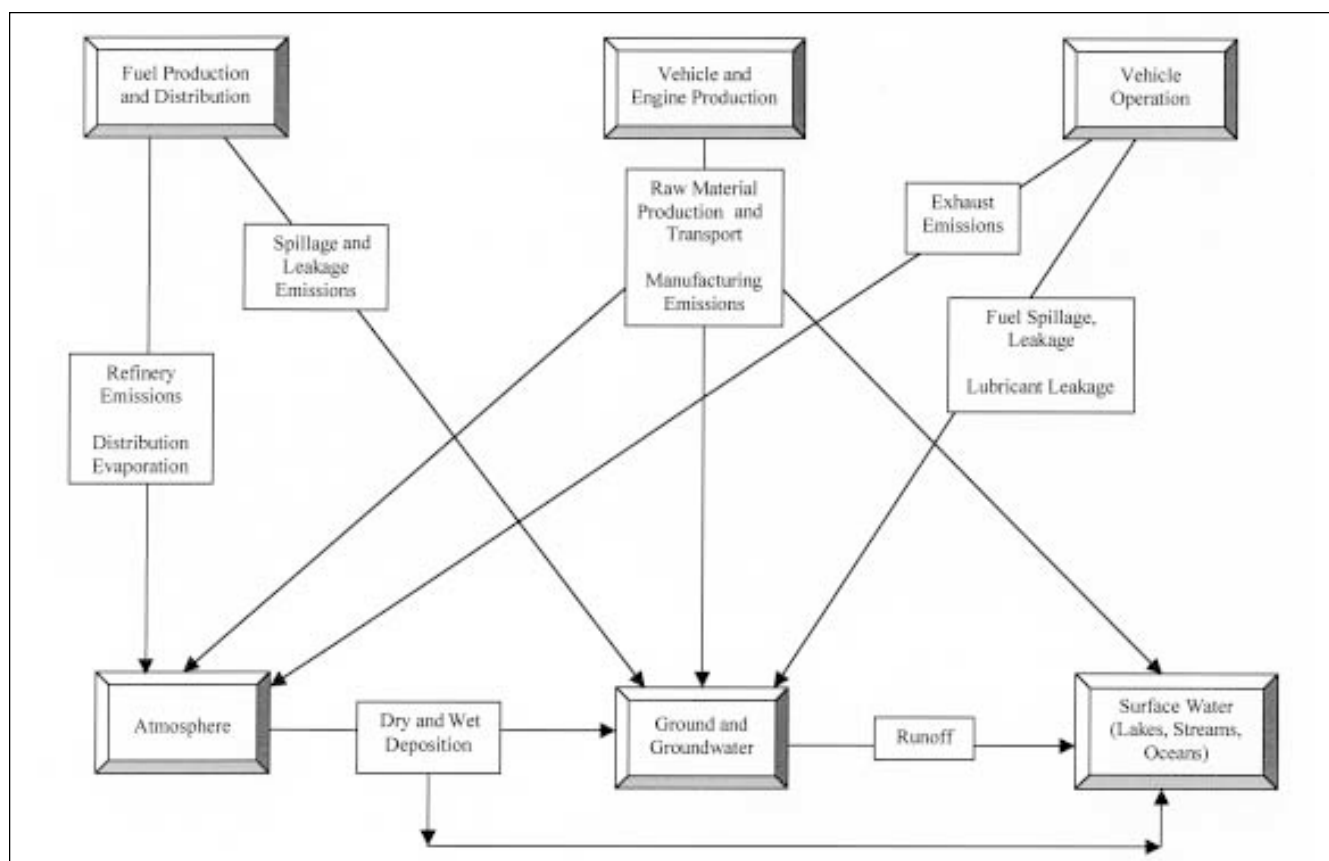


Figure 5. The environmental effects of diesel usage are highly multimedia and crossmedia in nature.

## EMISSIONS AND EXPOSURE FROM DIESEL ENGINES

Diesel exhaust consists of hundreds of gas-phase, semi-volatile, and particle-phase organic compounds that are produced through the combustion of fossil fuel.<sup>16-18</sup> A sample emissions profile from a chassis dynamometer study of a medium-duty diesel truck is shown in Table 1. Light-weight carbonyls make up the largest fraction of the gas-phase emissions of organic compounds, with acetaldehyde, formaldehyde, and acetone accounting for more than half of these emissions. Alkanes make up a much smaller fraction of gas-phase emissions, with low-molecular-weight alkanes ( $C_{10}$ ) and  $C_{10}$ – $C_{25}$  alkanes contributing roughly equally to the totals. A major portion of the high-molecular-weight semi-volatile organic compounds is unresolved by gas chromatography. This portion is composed predominantly of branched and cyclic HCs. The unresolved particle-phase organic compounds are similar to those detected in engine oil. Several polychlorinated dioxins and dibenzofurans have been found near detection limits, although the most toxic substances were not detected.<sup>17</sup> In comparison, diesel fuel consists of ~45% aliphatic HCs (predominantly  $C_{10}$ – $C_{25}$ ), ~25% cycloalkanes, <29% aromatic HCs [polycyclic aromatic hydrocarbons (PAHs) 2–5%], ~1% olefins, sulfur [100–300 parts per mil-

lion by weight (ppmw)], and nitrogen (40–500 ppmw).<sup>17</sup>

The exact composition of exhaust depends on operational parameters, such as speed, motor load, engine and vehicle type, fuel composition, ambient air temperature, and relative humidity. For example, increasing the cetane index (a measure of how readily the fuel autoignites) and lowering the aromatic content result in lower emissions of  $NO_x$ , HC, CO, and PM.<sup>20,21</sup> Lowering the sulfur content reduces  $SO_2$  and particulate sulfur emissions.<sup>22</sup> Ambient air temperature and the way in which diesel exhaust mixes in the atmosphere affect PM emission amounts and composition.<sup>23</sup>

Estimating ambient contributions of diesel exhaust is difficult because there are many other sources of the most common compounds in diesel exhaust.<sup>24</sup> Although some chemical markers have been identified, they are not commonly or easily measured. Black carbon [BC, also known as carbon black, soot, light-absorbing carbon, or elemental carbon (EC)] is used to estimate diesel particulate matter (DPM) because it constitutes a relatively high fraction of DPM,<sup>25,26</sup> and because PM emissions from diesel-powered vehicles are relatively high, compared with emissions from gasoline-powered vehicles.<sup>25,27</sup> Black carbon also originates from other sources, such as gasoline-powered vehicles, cooking, and vegetative burning.<sup>28</sup> Black carbon is defined by the measurement method, and different thermal evolution

protocols<sup>29-32</sup> yield different concentrations for the same samples.<sup>33</sup> Table 2 reports the results from several studies that estimate contributions from diesel exhaust or a combination of diesel and gasoline exhaust. Studies have shown that, depending on the vehicle fleet, DPM may be the primary component of motor vehicle PM.<sup>45</sup> Table 2 shows that DPM contributions to ambient PM ranged from 1 to 20 µg/m<sup>3</sup>, depending on monitoring location, sampling period, and method of estimating diesel source contributions.

To determine health consequences, ambient concentrations need to be related to human exposure. Because a typical person is indoors ~87% of the time, compared with ~5% outdoors and ~7% in vehicles,<sup>46,47</sup> indoor concentrations need to be accounted for in exposure assessment, but are poorly characterized for diesel exhaust. Studies of indoor diesel contributions have focused on occupational exposures, such as mines or warehouses, rather than on homes,

offices, or schools. Most residences, public buildings, and workplaces without internal sources of diesel exhaust are expected to have lower DPM concentrations than outdoor air has, owing to particle removal during infiltration and deposition. For example, in a recent DPM exposure assessment, the average indoor DPM contribution estimates were 2.0 ± 0.7 µg/m<sup>3</sup>, compared with an outdoor DPM concentration estimated at 3.0 ± 1.1 µg/m<sup>3</sup> (assuming no indoor DPM sources). The California Population Indoor Exposure Model (CPIEM)<sup>44</sup> was used for this study, and air exchange rates, particle penetration factors, and particle removal rates were included for a variety of building types. Using this same model, the estimated average annual exposures for California residents in 1995 and 2000 were 1.5 and 1.3 µg/m<sup>3</sup>, respectively. These calculations took into account population-weighted ambient concentrations (derived from receptor modeling studies and extrapolated to all air basins, using emission inventories) and activity patterns<sup>49,50</sup> in Cali-

**Table 1.** Example of mass balance and chemical composition from medium-duty diesel truck emissions. Values are from a chassis dynamometer study on the Federal Test Procedure urban driving cycle with hot start.<sup>16</sup>

Emission Rates (g/km)			
	CO <sub>2</sub> (935) <sup>a</sup> NO <sub>x</sub> (7.69) <sup>a</sup>		CO (1.76) <sup>a</sup> Fine particles (0.185 ±0.022)
Weight Percent of Fine Particle Mass			
	Organic carbon (19.7)		Elemental carbon (30.8)
	Nitrate (0.23)		Ammonium (0.73)
	Nitrite (0.01)		Sulfate (1.00)
	Aluminum (0.08 ±0.14)		Iron (0.05 ±0.01)
	Silicon (0.63 ±0.04)		Zinc (0.07 ±0.01)
	Phosphorus (0.01±0.06)		Cadmium (0.06 ±0.12)
	Indium (0.06 ±0.14)		Lead (0.01 ±0.04)
Emission Rates of Gas-Phase, Semi-Volatile, and Particle-Phase Organic Compounds and Elemental Carbon (mg/km)			
	C <sub>1</sub> -C <sub>10</sub> Organic Gases	C <sub>11</sub> -C <sub>30</sub> Semi-Volatile Organic Gases	Particle-Phase Organic Compounds
Resolved Organic Compounds	Alkanes (15.8)	Alkanes (4.6)	Alkanes (0.4)
	Olefins (17.3)	PAHs (1.8)	PAHs (1.0)
	Aromatics and cyclics (14.1)	Isoprenoids (2.2)	n-Alkanoic acids (0.4)
	Formaldehyde (22.3)	Aromatic acids (2.0)	Alkanedioic acids (0.3)
	Acetaldehyde (41.8)	Other compounds (1.5)	Other compounds (0.1)
	Propanal (14.0)		Elemental carbon (56)
	Acetone (22.0)		
	Crotonaldehyde (13.4)		
	Other carbonyls (55.4)		
Unresolved Organic Compounds	N/A	54.0	41.4

<sup>a</sup>Emission rate from EMFAC2000.<sup>19</sup>

fornia. They did not include in-vehicle or occupational exposures and, therefore, represent lower bounds.

Roadway studies show DPM contributions are higher than ambient measurements acquired at compliance monitoring sites. A study conducted near the Long Beach freeway reported DPM concentrations of 0.7–4  $\mu\text{g}/\text{m}^3$  higher than ambient.<sup>44</sup> At a Manhattan bus stop in New York City, with idling buses and heavy bus traffic, DPM contributions (as calculated from receptor modeling) ranged from 13 to 47  $\mu\text{g}/\text{m}^3$ .<sup>51</sup> In the Netherlands, BC measured near a busy roadway by filter light reflectance showed a 6.5- $\mu\text{g}/\text{m}^3$  increment over ambient.<sup>52</sup> Measurements at sidewalks in Harlem, New York City, reported BC concentrations that varied with truck traffic volumes from 1.5 to 6  $\mu\text{g}/\text{m}^3$ .<sup>53</sup>

Rodes et al.<sup>54</sup> measured BC inside vehicles, finding 0–10  $\mu\text{g}/\text{m}^3$  in Sacramento and 3–40  $\mu\text{g}/\text{m}^3$  in Los Angeles over 2-hr sample durations. BC concentrations were higher when following diesel vehicles. Fruin et al.<sup>55</sup> found average BC concentrations in Los Angeles of ~5  $\mu\text{g}/\text{m}^3$  with no vehicle in front; ~15  $\mu\text{g}/\text{m}^3$  when following a diesel-powered truck with a high, vertical exhaust pipe (i.e., tractor trailer rig) or a diesel passenger car; ~50  $\mu\text{g}/\text{m}^3$  when following a diesel-powered truck with low exhaust location; and ~130  $\mu\text{g}/\text{m}^3$  when following an urban transit bus making frequent stops (all runs were with windows closed). With an average of nearly 2 hr/day spent in vehicles,<sup>46</sup> in-vehicle DPM exposures can be a large fraction of human exposure.

Occupational DPM exposures can be high.<sup>57</sup> Mineworkers encounter some of the highest DPM

concentrations.<sup>58,59</sup> Estimated DPM exposures for mineworkers (based on respirable carbon or fine particle mass) range from 10 to 1280  $\mu\text{g}/\text{m}^3$ . Working near diesel-powered forklifts has also been associated with high exposures. Zaebs et al.<sup>26</sup> found average personal exposures of 31  $\mu\text{g}/\text{m}^3$  BC for those working around forklifts powered with diesel fuel, compared with an average of 3.5  $\mu\text{g}/\text{m}^3$  for those working around forklifts powered with other fuels. Groves and Cain<sup>60</sup> also found the highest BC exposures in the UK in areas where forklifts operate. In other job categories, average personal BC exposures were 27  $\mu\text{g}/\text{m}^3$  for mechanics and 5.2  $\mu\text{g}/\text{m}^3$  for truckers.<sup>26</sup> These levels far exceeded the measured highway BC concentration of 3.4  $\mu\text{g}/\text{m}^3$  and residential background concentrations of 1.1  $\mu\text{g}/\text{m}^3$ . However, most occupational exposure studies are relatively recent, and epidemiologic occupational studies have been hindered by the lack of good historical exposure estimates.<sup>56</sup>

#### HEALTH EFFECTS OF DIESEL EXHAUST

Adverse human health effects result from environmental exposure to diesel exhaust. These exposures have the potential to induce lung cancer in humans and animals and may cause acute and chronic noncancer adverse respiratory health effects.

#### Carcinogenicity

A consistent causal relationship between occupational diesel exhaust exposure and lung cancer was found in more than 30 human epidemiologic studies.<sup>61</sup> On average, long-term occupational exposures to diesel

**Table 2.** Summary of contributions to 24-hr average PM mass or PM carbon concentrations from diesel exhaust or a combination of diesel/gasoline exhaust.

Assessment Method	Location	Study Period	Emission Type	Estimated Contribution ( $\mu\text{g}/\text{m}^3$ )	Reference
CMB <sup>a</sup>	Tokyo	1987	DPM <sup>b</sup> PM <sub>7</sub>	24	Yoshizumi (1990) <sup>34</sup>
CMB	Boston, MA	1984–1985	MVPM <sup>c</sup>	2.5	Barnard (1999) <sup>35</sup>
CMB	Chicago, IL	June 1981–Jan 1982	MVPM	3.6	Barnard (1999) <sup>35</sup>
PCA <sup>d</sup>	Watertown, MA	June 1979–June 1981	MVPM <sub>2.5</sub>	2.5	Thurston and Spengler (1985) <sup>36</sup>
CMB	Phoenix, AZ	Sept 1989–Jan 1990	MVPM <sub>2.5</sub>	3–20	Chow et al. (1991) <sup>37</sup>
CMB	Phoenix, AZ	1994–1995		2.4–5.3	Maricopa (1999) <sup>38</sup>
CMB	Denver, CO	Dec 1996–Feb 1997	DPM PM <sub>2.5</sub>	1.1–2.4	Fujita et al. (1998) <sup>39</sup>
Grid modeling	Los Angeles Basin, CA	April 1998–Mar 1999	DPM PM <sub>10</sub>	3.0	SCAQMD (2000) <sup>40</sup>
Lagrangian model	Los Angeles Basin, CA	June 1982 and Dec 1982	DTC <sup>e</sup> PM <sub>2.1</sub>	1.3–5.2	Gray and Cass (1998) <sup>41</sup>
Trajectory model	Claremont, CA	Aug 1987	DPM PM <sub>2.5</sub>	2.4	Kleeman and Cass (1999) <sup>42</sup>
CMB	Los Angeles Basin, CA	1982	DPM PM <sub>2</sub>	4.4–11.6	Schauer et al. (1996) <sup>16</sup>
CMB	California	1990	DPM PM <sub>10</sub>	3.3	SAI (1994) <sup>43</sup>
CMB	California	1990	DPM PM <sub>10</sub>	3.0	CARB (1998a) <sup>44</sup>
Black carbon	Los Angeles Basin, CA	April 1998–Mar 1999	DPM PM <sub>10</sub>	3.4	SCAQMD (2000) <sup>40</sup>
Linear rollback	California	1995	DPM PM <sub>10</sub>	2.2	CARB (1998a) <sup>44</sup>

<sup>a</sup>CMB: chemical mass balance; <sup>b</sup>DPM: diesel particulate matter; <sup>c</sup>MVPM: motor vehicle exhaust particulate matter; <sup>d</sup>PCA: principal component analysis; <sup>e</sup>DTC: diesel total carbon.

exhaust were associated with an increase of ~40% in the relative risk of lung cancer.<sup>56,61</sup> Population-based case-control studies identified statistically significant increases in lung cancer risk for truck drivers,<sup>62-65</sup> railroad workers,<sup>64,66,67</sup> and heavy equipment operators.<sup>68</sup> These increases were consistent with self-reported diesel exhaust exposures.<sup>69</sup> These studies were adjusted for smoking. Industry-specific studies, both of case-control and cohort design, identified statistically elevated lung cancer risks for truck drivers,<sup>63,70-73</sup> professional drivers,<sup>74,75</sup> and railroad workers,<sup>66,67,76</sup> although only a few of these studies were adjusted for smoking.<sup>74-76</sup>

The Health Effects Institute (HEI) summarized 16 cohort and 19 case-control epidemiologic studies of occupational exposure to diesel emissions.<sup>77</sup> Control for smoking was identified in 15 studies. Six of the studies reported relative risk estimates less than 1, whereas 29 reported a relative risk greater than 1, indicating a positive association. These studies showed that long-term occupational exposure to diesel particulate exhaust is associated with a 20–50% increase in the relative risk of lung cancer. Bhatia et al.<sup>78</sup> found 21 of 23 studies reporting relative risks greater than 1. The pooled relative risk for these studies was 1.33 [95% confidence interval (CI): 1.24–1.44]. Lipsett and Campleman<sup>56</sup> reported a relative risk of 1.47 (95% CI: 1.29–1.67) from 39 independent estimates among 30 studies with adjustments for smoking. The U.S. Environmental Protection Agency (EPA)<sup>79</sup> reviewed 22 epidemiologic studies of workers exposed to diesel exhaust in various occupations. Increased lung cancer risk was observed in 8 of 10 cohort studies and 10 of 12 case-control studies for several industries, including railroad workers, truck drivers, heavy equipment operators, and professional drivers. The increased lung cancer relative risks generally ranged from 1.2 to 1.5, although a few studies showed relative risks as high as 2.6.

In Sweden, Gustavsson et al.<sup>80</sup> also found that lung cancer is positively associated with exposure to diesel exhaust. For the highest quartile of cumulative diesel exhaust exposure versus no exposure, the relative risk was 1.63 (95% CI: 1.14–2.33). Bruske-Hohlfeld et al.<sup>81</sup> pooled two large case-control studies on lung cancer in Germany (adjusted for smoking and asbestos exposure). Exposure to diesel exhaust resulted in an odds ratio of 1.43 (95% CI: 1.23–1.67) for lung cancer risk. The odds ratio applies only to case control studies, being the ratio of “odds” of occurrence of disease in the exposed and unexposed subgroups.

Several studies that accounted for at least one of the two principal confounders, smoking and exposure to asbestos, found significantly elevated risks, especially after longer-term exposures.<sup>62,64,67,71,82</sup> Additionally, these analyses demonstrated that the increases in lung cancer risk

remained after stratification by smoking or occupation, and risk was even higher in several instances.<sup>61,79</sup> Findings from studies of other types of cancer (e.g., bladder, testes, prostate, gastrointestinal tract, and lymphatic system) do not show as strong associations as those found for lung cancer.<sup>73,83-95</sup>

Diesel vapor and DPM extracts are genotoxic to bacterial and mammalian cell systems and can induce adverse chromosomal changes in animals. Genotoxicity refers to changes in DNA and is believed to be an important biological cause of lung cancer. It is typically measured using short-term tests, or bioassays, conducted in cell cultures (e.g., bacterial, plant, and mammalian cells) and in animals (e.g., mice and rats).<sup>79</sup> DNA adducts (representing genotoxic compounds bound chemically to DNA) increase in laboratory animals following inhalation exposure to diesel exhaust and have been found in mammalian cells following treatment with DPM extracts.

Elevated levels of DNA adducts have been associated with occupational exposure to diesel exhaust. The largest database for the genotoxicity of diesel exhaust is based on the Salmonella microsome (Ames) test for bacterial mutagenicity.<sup>96</sup> Enzymes (or microsomes) are used to metabolically activate certain compounds, such as PAHs, to make the compounds detectable as mutagens (i.e., compounds that cause specific types of changes in DNA). The bioassay has been used to chemically isolate the most potent genotoxic compounds in DPM, including nitro-substituted PAH (nitro-PAH). Nitrobenzanthrone, one of the most mutagenic compounds, was found in DPM,<sup>97</sup> and nitro-PAHs may serve as biomarkers of exposure to diesel exhaust.<sup>98</sup>

DPM has also been evaluated for genotoxicity in short-term tests. Using the Salmonella microsome assay, Sjorgen et al.<sup>99</sup> tested DPM from combusting fuels with a variety of chemical and physical characteristics. Physical fuel characteristics that were positively associated with genotoxicity included density and flash point, while the most important chemical variables associated with genotoxicity included PAHs, nitro-PAHs, particle-bound nitrate, and emitted PM mass.

In 1988, the National Institute of Occupational Safety and Health (NIOSH) first recommended that diesel exhaust be regarded as a potential carcinogen, based on animal and human evidence. In 1989, the International Agency for Research on Cancer (IARC) concluded that diesel engine exhaust was probably carcinogenic to humans (Group 2A). In 1990, based on IARC findings, the State of California identified diesel exhaust as a chemical “known to the State to cause cancer,” under the Safe Drinking Water and Toxic Enforcement Act of 1986 (Proposition 65). HEI<sup>77</sup> and the World Health



Organization (WHO)<sup>100</sup> found the epidemiologic data to be consistent in showing associations between exposure to diesel exhaust and lung cancer. The U.S. Department of Health and Human Services (DHHS) concluded that diesel exhaust is reasonably anticipated to be a carcinogen.<sup>101</sup> The EPA has proposed to identify diesel exhaust as a probable human carcinogen and further states that diesel exhaust is likely to be carcinogenic to humans by inhalation at any exposure condition.<sup>79</sup>

The California Environmental Protection Agency (Cal/EPA)<sup>61</sup> concluded that existing epidemiologic studies provide evidence consistent with a causal relationship between occupational diesel exhaust exposure and lung cancer. These findings are unlikely to be by chance and, with the possible exception of some studies that did not take smoking into account, the results are unlikely to be explained by confounding or bias. Similarly, HEI,<sup>77</sup> Bhatia et al.,<sup>78</sup> and Lipsett and Campleman<sup>56</sup> concluded that potential confounding by smoking could not explain the association between occupational diesel exposure and lung cancer. Other studies<sup>56,77-79</sup> also conclude that there is a causal association between lung cancer and diesel exhaust exposure. The weight of evidence for potential human carcinogenicity for diesel exhaust exposure is strong, even though inferences were involved in the assessment.<sup>79</sup>

### Cancer Risk Estimates

Harris<sup>102</sup> assessed the risk of exposure to diesel exhaust, using data from an earlier London Transport Worker Study by Waller.<sup>103</sup> He derived a maximum likelihood excess relative risk estimate of  $1.23 \times 10^{-4}$  per  $\mu\text{g}/\text{m}^3$ , with a 95% upper confidence limit of  $5 \times 10^{-4}$  per  $\mu\text{g}/\text{m}^3$  DPM per year. McClellan et al.<sup>104</sup> reported risk estimates based on the Garshick et al.<sup>76</sup> railroad worker case-control study. Using an upper and lower assumption of exposure concentration of the workers, McClellan et al.<sup>104</sup> predicted the number of excess cancer deaths per year in the United States to range from 950 to 3800. Using data from the

Garshick et al.<sup>76</sup> case-control study and the Garshick et al.<sup>67</sup> cohort study, Cal/EPA<sup>61</sup> estimated the risk of lung cancer in the general population due to diesel exhaust. Because of uncertainties in the actual workplace exposures, a variety of scenarios were used to bracket the exposures. Based on these exposure estimates, the range of resulting estimates of cancer risk is  $1.3 \times 10^{-4}$  to  $2.4 \times 10^{-3}$  per  $\mu\text{g}/\text{m}^3$  (the unit risk represents the 95% upper confidence limit of cancer risk per million people exposed per microgram of DPM in a cubic meter of air over a 70-year lifetime).

Larkin et al.<sup>82</sup> showed elevated risks of lung cancer in jobs with diesel exhaust exposure, after considering the differences in smoking rates between workers who were exposed to diesel exhaust and those who were not. Dawson and Alexeeff<sup>105</sup> reanalyzed the Garshick et al.<sup>67</sup> cohort data using a multi-stage model and found a positive relationship between exposures to diesel exhaust and increased lung cancer risk. Steenland et al.<sup>65</sup> estimated lung cancer risk of truck drivers on the basis of a case-control study of decedents in the Teamsters Union.<sup>106</sup> Starting with a set of 1990 exposure measurements, past exposures were estimated by data on diesel VMT and engine emission rates. The 1990 job category estimates came from an extensive industrial hygiene survey of EC exposures in the trucking industry by Zaebs et al.<sup>26</sup> Assuming the most likely emission scenario of 4.5 g/mi in 1970 and a 45-year exposure to  $5 \mu\text{g}/\text{m}^3$ , the estimated excess lung cancer risk was determined to be 1–2% above a background risk of 5%. Using the same database, Stayner et al.<sup>107</sup> presented an estimate of excess lifetime risk of  $4.5 \times 10^{-4}$  per  $\mu\text{g}/\text{m}^3$  of diesel exhaust for 45 years.

Several quantitative risk assessment models were used by NIOSH to predict lung cancer in miners exposed to diesel exhaust.<sup>107</sup> The toxicologically based unit risk estimates varied widely (from 2 to  $220 \times 10^{-6}$  per  $\mu\text{g}/\text{m}^3$ ); the epidemiologically based unit risk estimates were less variable and suggest higher risks (from 100 to  $920 \times 10^{-6}$  per  $\mu\text{g}/\text{m}^3$ ). All of the models suggest high relative risks

**Table 3.** Comparison of unit risk estimates for diesel exhaust.<sup>105</sup>

Study Population	Reference	Unit Risk [70 year-( $\mu\text{g}$ diesel particles/ $\text{m}^3$ )] <sup>-1</sup>
London transport workers <sup>103</sup>	Harris et al. (1983) <sup>102</sup>	$1.4 \times 10^{-3\text{ab}}$
U.S. railroad worker case-control study <sup>76</sup>	McClellan et al. (1989) <sup>108</sup>	
	Mauderly et al. (1992) <sup>109</sup>	$2.9 \times 10^{-4}$ and $1.2 \times 10^{-3\text{ac}}$
	Stayner et al. (1998) <sup>107</sup>	$7.1 \times 10^{-4}$ and $3.3 \times 10^{-3\text{c}}$
U.S. truck drivers <sup>26,106</sup>	Steenland et al. (1998) <sup>65</sup>	$4.0 \times 10^{-4}$ to $1.5 \times 10^{-3\text{c}}$ , $1.1 \times 10^{-3\text{ad}}$
U.S. railroad workers <sup>67,76,110,111</sup>	Cal/EPA (1998) <sup>61</sup>	$1.3 \times 10^{-4}$ to $2.4 \times 10^{-3\text{a}}$
U.S. railroad workers <sup>67,110,111</sup>	Dawson and Alexeeff (2000) <sup>105</sup>	$2.1 \times 10^{-4}$ to $5.5 \times 10^{-4\text{a}}$

<sup>a</sup>95% upper confidence limit; <sup>b</sup>This hazard is not statistically significant; <sup>c</sup>Range based on an upper and a lower concentration assumption; <sup>d</sup>Obtained from the exposure coefficient  $3.5 \times 10^{-4}$  with standard error  $1.55 \times 10^{-4}$  for the best value of exposure.

(i.e., 1/1000) for miners with long-term exposures greater than 1000  $\mu\text{g}/\text{m}^3$  of DPM. Table 3 compares the DPM cancer unit risk estimates from several epidemiologic studies. The 70-year unit risk varies over 30-fold, from  $\sim 1 \times 10^{-4}$  to  $3 \times 10^{-3}$  per  $\mu\text{g}/\text{m}^3$  of DPM exposure. Many of these variations are attributable to different estimates of exposure.

Although the totality of evidence demonstrating the carcinogenic effects from exposure to diesel exhaust is persuasive, many uncertainties exist. This is due to data and knowledge gaps about human exposures to diesel exhaust and the mechanisms by which diesel exhaust causes cancer in humans and animals. For example, it is unclear how the physical and chemical nature of past exposures to diesel exhaust compare with present-day exposures. Extrapolation of high occupational exposures to lower environmental exposures is unproven. Potential confounding and other biases, inadequate follow-up, specification of the dose-response model, estimation of exposure and dose, and unrecognized variability and susceptibility all contribute to the uncertainties of the existing studies and may result in either under- or overestimates of cancer risk.<sup>61</sup> The uncertainties in using the human data include the representativeness of railroad workers for the general population, the choice of the analytical model, and the lack of knowledge of the exposure history of the railroad workers, including possible exposure to unknown confounders. There is limited diesel-specific information that addresses variable susceptibility, with regard to carcinogenicity, within the general human population and vulnerable subgroups, including infants, children, and people with pre-existing health conditions. These relative risk estimates have been questioned. Crump et al.<sup>112</sup> reported relative risks that were positively or negatively related to the duration of exposure, depending on the way age was controlled. Crump<sup>113</sup> found no relationship between exposure to diesel exhaust and lung cancer mortality. Epidemiologic studies are criticized as limited in their application to environmental risk assessment.<sup>77,79,112,113</sup> Nevertheless, the human data have been used to quantify the cancer risk associated with diesel exhaust exposures.<sup>61,65,102,105,107</sup>

#### Noncancer Health Effects

More than 90% of diesel exhaust-derived PM is smaller than 1  $\mu\text{m}$  in diameter.<sup>108</sup> Most of the mass is in the 0.1–1.0  $\mu\text{m}$  “accumulation” size fraction, while most of the particles are in the <0.1  $\mu\text{m}$  “ultrafine” or “nanoparticle” fraction.<sup>114</sup> These small particles are respirable and penetrate deep into the lungs.<sup>115</sup> A given mass of very small particles contains a larger number of particles, with a correspondingly larger surface area, than an equivalent mass of larger particles. Consequently, a given

mass of ultrafine particles will impact a larger surface area of lung tissue than will an equal mass of larger particles,<sup>116</sup> leading to more extensive exposure of the lung tissues.

Recent epidemiologic studies<sup>115,117</sup> have associated mortality and respiratory morbidity with exposure to ambient concentrations of ultrafine particles, raising concern that diesel exhaust could contribute to or be the cause of the observed health effects. However, the contribution of diesel exhaust to the ultrafine particulate fraction was not analyzed in these studies. In California, the annual average ambient diesel exhaust particulate concentration in ambient air was about 2–3  $\mu\text{g}/\text{m}^3$  in 1990,<sup>61</sup> compared with annual statewide average  $\text{PM}_{2.5}$  concentrations ranging from  $\sim 4 \mu\text{g}/\text{m}^3$  in rural Lake County to 27  $\mu\text{g}/\text{m}^3$  in urbanized Bakersfield.<sup>118</sup>

There is controversy over whether health effects of DPM should be considered as distinct from those of generic urban PM. Animal toxicology studies indicate that all particles in a given size fraction are not equivalent; degree and type of toxicity vary with particle type. Murphy et al.<sup>119</sup> and van Zijverden et al.<sup>120</sup> compared inflammatory and tissue changes in rats exposed to DPM, BC, and silica particles. Each type of particle was found to induce a different pattern and degree of toxic effects. Nikula et al.,<sup>121</sup> however, reported that the non-neoplastic responses of rats to DPM and BC were similar, while Castranova et al.<sup>122</sup> found that inflammatory responses of rats to DPM and coal dust particles were not equivalent.

Evidence for associations between diesel exposure and noncancer deaths is equivocal. Edling and Axelson<sup>123</sup> reported a 4-fold increase in risk of dying from cardiovascular disease for men who had been employed by a bus company. This association was significant even when smoking was taken into account. When the study was later extended to include 694 bus garage employees over a 32-year period,<sup>124</sup> the results indicated no difference between the observed and expected number of deaths from cancer or cardiovascular disease in the cohort, or with reference to differing degrees of exposure.

Mauderly et al.<sup>109</sup> reported that mice exposed to 0.35, 3.5, or 7.1  $\text{mg}/\text{m}^3$  of diesel exhaust or filtered air for 7 hr/day, 5 days/week for 24 months showed no consistent exposure-related increase in mortality. While the groups exposed to the low and medium concentrations of diesel exhaust had a shorter median survival, the high exposure group had a median survival rate slightly longer than the filtered air controls. Nikula et al.<sup>121</sup> reported that survival of female Fischer 344 rats exposed to 2.5 or 6.5  $\text{mg}/\text{m}^3$  of diesel exhaust for 16 hr/day, 5 days/week was similar to that of control animals exposed to filtered air. On the other hand, male rats exposed to the same concentrations of

diesel exhaust showed reduced survival, although fractional survival was greater in the group exposed to the higher diesel exhaust concentration. Overall, these conflicting results suggest an equivocal relationship between diesel exhaust exposure and mortality.

There are few published studies on human subjects exposed to controlled concentrations of diesel exhaust. Rudell et al. exposed human subjects to unfiltered diesel exhaust<sup>125</sup> and to diesel exhaust with and without a ceramic particle trap on the source engine<sup>126</sup> for 1 hr while they exercised lightly. The trap reduced the particle number by ~25%. Subjects reported small increases in eye and nose irritation and an unpleasant smell in both studies. Neither study found changes in volume or flow measures of pulmonary function, although the later study<sup>126</sup> reported small increases in airway resistance. Responses to exposure with and without the trap were similar, suggesting that the effects observed may have been at least partly related to the gas phase of the exhaust. Pepelko et al.<sup>127</sup> found no significant changes in pulmonary function in male weanling cats exposed to 6.4 mg/m<sup>3</sup> DPM for 20 hr/day on 28 consecutive days.

Most available information on pulmonary function changes in humans exposed to diesel exhaust comes from studies of changes occurring over a work shift<sup>128,129</sup> or after working for years in an occupation involving likely exposure to diesel exhaust.<sup>130-135</sup> The small and often statistically nonsignificant changes observed cannot be related to specific exposure levels. Ames et al.<sup>133</sup> found that pulmonary function changes were larger in the group thought to have lower diesel exhaust exposure. Evidence for symptoms associated with diesel exhaust exposure, including eye and mucus membrane irritation, cough, phlegm, dyspnea, headache, light-headedness, dizziness, nausea, and odor annoyance, comes largely from epidemiologic studies of workers in industries where diesel-powered equipment was used.<sup>126,128,132,134,135</sup> Ames et al.<sup>129,133</sup> and Gamble et al.,<sup>130</sup> on the other hand, reported no association between occupational exposure to diesel exhaust and symptoms in coal miners.

These epidemiologic studies have a number of methodological shortcomings. Exposure was not directly measured, but was assigned according to job category or by the number of diesel-powered engines in the workplace. Statistical models did not adequately control for confounding variables, such as dusts generated by mining activities, blasting, and the gaseous fraction of diesel exhaust. None of these studies included adequate characterization of smoking history or appropriate control factors for smoking. Suitability of control groups used in some of the occupational studies (in terms of age, ethnicity, occupation, ambient air pollutant exposure, smoking

history, and employment history) is questionable. Nonoccupational exposures to vapors, dusts, or toxic substances were not included.

Recent epidemiologic studies examining exposures outside the occupational arena report statistically significant associations of asthma and allergic rhinitis in children living in areas with truck traffic on the street of residence.<sup>136-139</sup> Truck traffic volume was the surrogate for diesel exhaust exposure. Only Brunekreef et al.<sup>138</sup> obtained actual traffic counts for trucks and cars on roadways near the children's homes. The other three studies relied on children's reports about nearby truck traffic. The fraction of traffic-related emission exposure that could be attributed to diesel sources was not estimated. These studies (with the exception of Brunekreef et al.,<sup>138</sup> who performed ambient air monitoring inside and outside of schools) assumed that exposure estimated at a child's residence was representative of exposure to diesel exhaust, even though a large fraction of each week-day was spent in school. These studies also relied on self-reports of respiratory symptoms, allergy, and asthma obtained primarily from the children themselves. Overall, the results of these studies suggest associations between PM and gaseous pollutant emissions from traffic-related sources and various respiratory health endpoints, but the extent to which the adverse health impacts are due specifically to diesel exhaust exposure, as opposed to PM and gaseous pollutants from all sources, is uncertain.

Animal studies suggest that diesel exhaust induces cellular and biochemical responses indicative of pulmonary inflammation and immunological responses.<sup>140-142</sup> Studies on mice exposed to DPM concentrations up to 3 mg/m<sup>3</sup> show both inflammatory and immunological changes in lung tissues and in immune cells isolated by bronchoalveolar lavage.<sup>142-145</sup> Heinrich et al.<sup>146</sup> studied rats, mice, and hamsters that underwent controlled lifetime exposures to 4 mg/m<sup>3</sup> filtered and unfiltered DPM (19 hr/day, 5 days/week). After the hamsters and rats were exposed for 1 or 2 years, respectively, changes were observed in cellular and biochemical constituents in bronchoalveolar lavage fluid, suggestive of lung inflammation. Rats were more sensitive to the exposure than either mice or hamsters. Henderson et al.<sup>147</sup> exposed rats and mice to 0.35, 3.5, or 7 mg/m<sup>3</sup> diesel exhaust for 7 hr/day, 5 days/week, for up to 30 months (lifetime exposures in these animals). No cytological or biochemical changes were noted in bronchoalveolar lavage fluid or lung tissue after exposure to 0.35 mg/m<sup>3</sup> DPM. Rats and mice exposed to the two higher DPM doses had cellular and biochemical responses characteristic of chronic inflammation. Castranova et al.,<sup>122</sup> however, found little

effect on alveolar macrophage function in rats exposed to 2 mg/m<sup>3</sup> DPM for 7 hr/day, 5 days/week for 2 years.

Several toxicology studies confirm that diesel exhaust induces inflammatory mediator production<sup>120,148</sup> and airways hyperresponsiveness and remodeling tissue changes<sup>149</sup> with exposures of up to several weeks. Although these exposures were less than the lifetime studies cited above, the exposure doses were higher than would be typical for the average person over a similar time frame. Moreover, exposure was via routes other than inhalation, such as intratracheal instillation or in vitro exposure of lung tissue slices.

Studies have shown that DPM induced inflammatory and immunological responses characteristic of allergy and asthma within nasal airway tissues and cells obtained by nasal lavage.<sup>140,150,151</sup> When subjects were challenged with a nasal spray of both DPM and an allergen, inflammatory and immunological responses were enhanced, compared with responses to either DPM or the allergen alone. Similar responses occurred in human-derived isolated cells<sup>150</sup> exposed to DPM and ragweed allergen. Diaz-Sanchez et al.<sup>141</sup> induced immune responses with DPM and an allergen to which the subjects were not previously allergic. Van Zijverden et al.'s<sup>120</sup> results with mice were consistent with Diaz-Sanchez et al.'s observations,<sup>141</sup> supporting the hypothesis that DPM may encourage allergic responses, development of new allergies, and possibly asthma.

No cardiovascular or lung function changes were found following exposure of human subjects to resuspended DPM<sub>10</sub> (DPM measured as PM<sub>10</sub>) at a concentration of 200 µg/m<sup>3</sup> for 2 hr, although analysis of induced sputum samples at 4 hr post-exposure indicated cellular and biochemical evidence for inflammatory lung response.<sup>152</sup> Salvi et al.<sup>153,154</sup> exposed human subjects to DPM<sub>10</sub> at a concentration of 300 µg/m<sup>3</sup> for 1 hr. There were no significant changes in pulmonary function, but analysis of bronchoalveolar lavage fluid and bronchial biopsy samples indicated changes in cytology and various immune factors indicative of lung inflammation.

There are no data on lung cellular or tissue responses of human subjects exposed to diesel exhaust. However, noncancer effects from high-concentration, long-term (mainly lifetime) exposure studies in several animal species have shown changes in lung tissues indicative of inflammation and fibrosis.<sup>109,121,127,147</sup> Murphy et al.<sup>119</sup> reported that instillation of 1 mg of DPM into the lungs of rats induced no inflammatory effects. The only notable effect was PM accumulation in the lung parenchyma 12 weeks after instillation, suggesting that exposure was insufficient to induce effects. Murphy et al.<sup>119</sup> only exposed their animals for 12 weeks, in contrast to the

lifetime exposures in the studies above.

Studies on rats suggest that the mechanism for cellular or tissue effects is overload of the alveolar macrophage particle removal system.<sup>146,155</sup> The mechanism appears to begin with interaction of diesel particles with airway epithelial cells and phagocytosis by alveolar macrophages. As the number of particles in the lung increases, particle-laden alveolar macrophages aggregate in alveoli adjacent to terminal bronchioles. This is accompanied by increases in the number of Type II cells lining particle-laden alveoli, the presence of particles within alveolar and peribronchial interstitial tissues and associated lymph nodes, and depression of alveolar macrophage phagocytosis. The severity of observed effects appears to be related to the accumulated particle burden in the lungs.<sup>109</sup>

Other studies on rats<sup>122,147</sup> reveal that inflammatory changes may be initiated by macrophages engulfing DPM, as well as airway epithelial cells, that release pro-inflammatory factors, which, in turn, induce inflammation, cell injury, cell proliferation, hyperplasia, and fibrosis. Activated macrophages also release chemotactic factors that attract neutrophils and eosinophils that also release inflammatory mediators and contribute to the inflammatory process.<sup>142-145</sup> Comparison of the cellular responses of laboratory animals to gas- and particle-phase diesel exhaust, and to diesel exhaust with the particulate fraction filtered out, suggest that the observed effects are related to particulate diesel exhaust rather than to gaseous emissions.<sup>146</sup>

Evidence for noncancer human health effects resulting from exposure to ambient diesel exhaust is not as consistent or strong as that for lung cancer. However, animal studies indicate that diesel exhaust is capable of inducing a variety of adverse, noncancer health effects, including pulmonary fibrosis and inflammation. However, these effects have typically resulted from exposures considerably in excess of typical ambient levels. Results from animal studies suggest that there may be a threshold of exposure to diesel exhaust below which adverse structural and biochemical effects may not occur in the lung (e.g., Henderson et al.<sup>147</sup>), although there are no similar data available on human subjects. Diesel exhaust appears to act as an adjuvant in allergic responses and possibly asthma, although additional research is needed at lower concentration levels.

#### Mortality and Morbidity

Morbidity and mortality effects of diesel PM<sub>2.5</sub> exposure have been calculated for California, using concentration-response equations from several epidemiologic studies to demonstrate the range of effects. The

mortality estimates are likely to exclude cancer cases discussed earlier in the paper, but may include some premature deaths due to cancer, because the epidemiologic studies did not identify the cause of death. Table 4 summarizes some morbidity (illness) effects based on different studies. Table 5 shows the mortality effects of direct and indirect diesel PM<sub>2.5</sub> exposure. Corresponding 5th and 95th percentiles are also shown in these tables. The number of incidences estimated in the two tables correspond to concentration levels of 1.8 µg/m<sup>3</sup> of direct DPM and 0.81 µg/m<sup>3</sup> of indirect DPM for the year 2000. Table 4 consists of the chronic bronchitis portion and the hospital admissions for different health endpoints. Table 5 includes estimates based on studies that analyzed long-term exposure data and death cases during elevated PM exposure days. Long-term and short-term studies tend to estimate different levels of effect. The Pope et al. study<sup>160</sup> serves as an upper bound mean estimate of 3566 deaths, and the Samet et al. study<sup>157</sup> is the lower bound mean estimate of 665 premature deaths due to diesel PM<sub>2.5</sub> exposure in 2000.

## EFFECTS OF DIESEL ENGINE EXHAUST ON THE ENVIRONMENT

### Visibility Reduction

Visibility degradation (or haze) is caused by both primary particles and “secondary” SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, and organic carbon particles formed from atmospheric conversion of gaseous NO<sub>x</sub>, SO<sub>2</sub>, NH<sub>3</sub>, and HC emissions. It is measured as the light extinction coefficient, which is the natural logarithm of the fractional reduction of light transmission per unit distance, usually expressed as Mm<sup>-1</sup> (“inverse megameters”).<sup>164</sup> Light extinction consists of scattering and absorption by gases and particles.<sup>165</sup> Clean air scatters light with ~12 Mm<sup>-1</sup> at sea level, and NO<sub>2</sub> is the only gaseous pollutant that appreciably absorbs light. Particle light scattering depends on the particle size distribution, which can shift as hygroscopic chemical species absorb water with increasing humidity.<sup>166</sup> Light absorption is mostly due to BC from incomplete combustion.

Diesel exhaust PM is a combination of particles rich in BC and sized to very effectively scatter light, giving diesel exhaust an extinction efficiency nearly double that

**Table 4.** California annual morbidity (illness) effects in diesel PM<sub>2.5</sub> health effects studies and corresponding 5th and 95th percentiles.

Health Endpoint	PM <sub>2.5</sub> Concentration (µg/m <sup>3</sup> )	Incidence (cases/year)			Study
		5th Percentile	Mean	95th Percentile	
Chronic Bronchitis (age 27+)	1.8 <sup>a</sup>	−17	1791	3644	Abbey et al. (1995) <sup>156</sup>
	0.81 <sup>b</sup>	−8	801	1618	
		−25	2592	5262	
Hospital Admission					
COPD (ICD codes 490–492, 494–496) (Age 65+)	1.8 <sup>a</sup>	29	441	867	Samet et al. (2000) <sup>157</sup>
	0.81 <sup>b</sup>	13	198	388	
		42	638	1255	
Pneumonia (ICD codes 480–487) (Age 65+)	1.8 <sup>a</sup>	243	537	837	Samet et al. (2000) <sup>157</sup>
	0.81 <sup>b</sup>	109	241	376	
		353	778	1213	
Cardiovascular (ICD codes 390–429) (Age 65+)	1.8 <sup>a</sup>	1057	1297	1527	Samet et al. (2000) <sup>157</sup>
	0.81 <sup>b</sup>	475	583	686	
		1532	1881	2213	
Asthma (ICD codes 493) (Age 64–)	1.8 <sup>a</sup>	70	209	414	Sheppard et al. (1999) <sup>158</sup>
	0.81 <sup>b</sup>	32	94	186	
		102	303	599	
Asthma-related ER visits (Age 64–)	1.8 <sup>a</sup>	765	1155	1547	Schwartz (1993) <sup>159</sup>
	0.81 <sup>b</sup>	343	518	693	
		1108	1673	2240	

<sup>a</sup>From Cal/EPA (2000).<sup>61</sup> <sup>b</sup>Estimated conversion of diesel NO<sub>x</sub> emissions to PM<sub>2.5</sub> nitrate using 1999 California air quality data. Likely to be an underestimate because of NH<sub>4</sub>NO<sub>3</sub> loss.

of most other particle sources. Under certain operating conditions, diesel engines emit visible particle (smoke) plumes. These plumes are composed of directly emitted “primary” particles. Secondary aerosol, formed from  $\text{NO}_x$  and  $\text{SO}_2$  emitted by diesels, further magnifies their contribution to visibility degradation. The net result is that the visibility impacts of the existing diesel fleet, though variable in time and location, are larger than their proportional fraction of vehicle mileage, and significant visibility benefits accompanying future reductions in diesel  $\text{NO}_x$  and PM emissions can be anticipated.

Although diesels constitute only ~5% of on-road vehicles in the United States, they contribute from 10 to 75% of the optically active PM in urban areas, depending on surrounding source characteristics.<sup>167</sup> Currently applied

aerosol sampling and analysis techniques do not produce the size-resolved and chemically speciated data needed to definitively distinguish primary diesel particles from those of other combustion sources. As a result, site- and time-specific estimates of the diesel contribution to visibility impairment must be extrapolated from analyses of data from a few specialized experiments, occasional large-scale air quality field projects, limited modeling, or approximate analyses based on emission inventories or long-term  $\text{PM}_{2.5}$  data sets.

Horvath<sup>168,169</sup> estimated diesel contributions to light extinction in Vienna, Austria. Diesel fuel tagged with rare earth markers at the refinery was marketed, and aerosol samples were acquired at several sites around the city. The markers permitted direct determination of mass

**Table 5.** California annual mortality effects in diesel  $\text{PM}_{2.5}$  health effects studies and corresponding 5th and 95th percentiles.

	<b><math>\text{PM}_{2.5}</math> Concentration (<math>\mu\text{g}/\text{m}^3</math>)</b>	<b>5th Percentile</b>	<b>Mean</b>	<b>95th Percentile</b>	<b>Study</b>
<b>Long-Term Exposures Mortality</b>					
Direct diesel PM	1.8 <sup>a</sup>	1287	2457	3621	Pope et al. <sup>160</sup>
Indirect PM	0.81 <sup>b</sup>	580	1109	1636	
		<b>1867</b>	<b>3566</b>	<b>5257</b>	
Direct diesel PM	1.8 <sup>a</sup>		2221		Average of Pope et al. <sup>160</sup> and Krewski et al. <sup>161</sup>
Indirect PM	0.81 <sup>b</sup>		1002		
		<b>—</b>	<b>3223</b>	<b>—</b>	
Direct diesel PM	1.8 <sup>a</sup>	974	1985	2991	Krewski et al. <sup>161</sup>
Indirect PM	0.81 <sup>b</sup>	439	895	1350	
		<b>1413</b>	<b>2880</b>	<b>4341</b>	
<b>Short-Term Exposures Mortality</b>					
Direct diesel PM	1.8 <sup>a</sup>	394	1747	3064	Fairley—multi-pollutants <sup>162</sup>
Indirect PM	0.81 <sup>b</sup>	177	784	1373	
		<b>571</b>	<b>2531</b>	<b>4437</b>	
Direct diesel PM	1.8 <sup>a</sup>	507	1293	2081	Fairley—single pollutant <sup>162</sup>
Indirect PM	0.81 <sup>b</sup>	228	581	934	
		<b>735</b>	<b>1874</b>	<b>3015</b>	
Direct diesel PM	1.8 <sup>a</sup>	467	567	668	Schwartz et al. <sup>163</sup>
Indirect PM	0.81 <sup>b</sup>	210	255	300	
		<b>677</b>	<b>822</b>	<b>968</b>	
Direct diesel PM	1.8 <sup>a</sup>	113	459	806	Samet et al. <sup>157</sup>
Indirect PM	0.81 <sup>b</sup>	51	206	362	
		<b>164</b>	<b>665</b>	<b>1168</b>	

*Note:* Long-term study should be used alone rather than considering the total effect to be the sum of estimated short-term and long-term effects, because summing creates the possibility of double-counting a portion of PM-related mortality; <sup>a</sup>From Cal/EPA (2000); <sup>b</sup>Estimated conversion of diesel  $\text{NO}_x$  emissions to  $\text{PM}_{2.5}$  nitrate using 1999 California air quality data. Likely to be an underestimate because of  $\text{NH}_4\text{NO}_3$  loss.

concentration for DPM. Light scattering for major aerosol components was theoretically and empirically estimated, and optical absorption was measured directly on the filters. This approach allowed direct calculation of the optical effects of diesel emissions across the city. DPM contributed from 8% of light extinction in the urban core, where it was mixed with significant amounts of local sources, to 25% of light extinction in suburban areas, where influences from other sources were much weaker. Diesel emissions in this experiment contained ~14% BC. The diesel absorption coefficient was estimated to be  $5 \text{ m}^2/\text{g}$ ,<sup>170</sup> and the diesel scattering efficiency was  $7.7 \text{ m}^2/\text{g}$ , for a total extinction efficiency of  $\sim 13 \text{ m}^2/\text{g}$ . The older-technology diesel engines and fuels may not result in similar contributions to haze in contemporary U.S. cities.

### Global Climate Change

The Earth receives energy as visible light from the sun, and returns energy to space as invisible infrared radiation.<sup>171</sup> The greenhouse effect is a warming influence caused by the presence of gases and clouds in the air that are very efficient absorbers and radiators of the infrared radiation. The light-absorbing properties of diesel exhaust also affect the earth's radiation balance. The transportation sector is currently responsible for ~26% of greenhouse gas emissions in the United States and, due to increased demand for gasoline and diesel fuel, is expected to be one of the fastest growing sources of greenhouse gas emissions in the foreseeable future.<sup>172</sup> Diesels produce lower  $\text{CO}_2$  emissions than equivalent gasoline engines per unit of work, owing to their greater efficiency. Because  $\text{CO}_2$  is the principal greenhouse gas, concerns about global warming have made diesel engines appear to be an attractive alternative to gasoline power. However, DPM may modify cloud cover and rainfall (and thus alter the albedo of the earth-atmosphere system), more than offsetting the diesel engine's  $\text{CO}_2$  advantage.<sup>173</sup>

In contrast to greenhouse gases, which act primarily on the outgoing infrared radiation, PM influences both sides of the energy balance. Because PM scatters solar radiation, a substantial portion of that radiation is returned to space, thereby decreasing the net energy at the Earth's surface. However, BC from diesels and other emitters absorb solar radiation, thereby warming the atmosphere. In addition to this direct (scattering-absorption) effect, there is also an indirect aerosol effect. Tropospheric PM emissions affect the size distribution of cloud droplets, thus altering the radiative properties of clouds (increasing their reflectivities). Particulate matter may also inhibit rainfall by potentially altering the cloud lifetime.<sup>174,175</sup>

Particulate matter sulfate is estimated to exert a global average cooling effect of  $-0.67 \text{ W}/\text{m}^2$ .<sup>176,177</sup> It is

tempting to conclude that greenhouse gas warming and aerosol cooling could cancel each other and neither would, therefore, be a cause for concern. However, Hobbs<sup>178</sup> reported that air samples from the eastern United States showed a predominance of BC, consistent with warming rather than cooling. Black carbon aerosol causes a positive (warming) climate forcing that is very uncertain in magnitude, but it appears to be about  $0.5\text{--}1 \text{ W}/\text{m}^2$ .<sup>173</sup>

Black carbon particles are in several possible mixing states:<sup>179,180</sup> distinct from other aerosol particles (externally mixed), incorporated within them (internally mixed), or approximated as a BC core surrounded by a well-mixed shell. Thus far, it has been assumed that aerosols exist predominantly as an external mixture. However, Jacobson<sup>181</sup> simulated the evolution of the chemical composition of aerosols, finding that the mixing state and direct forcing of the BC component approach those of an internal mixture, largely due to particle coagulation and growth. This causes higher BC light absorption than previously thought, suggesting that the warming effect from BC may nearly balance the net cooling effect of other anthropogenic aerosol constituents. The magnitude of the direct radiative forcing from BC exceeds that due to  $\text{CH}_4$ . Black carbon may be the second most important component of global warming, after  $\text{CO}_2$ , in terms of direct radiative forcing.

Jacobson<sup>181</sup> suggests that BC emission reductions may be more beneficial than  $\text{CH}_4$  reductions. Aerosol radiative effects also depend on the solar angle, relative humidity, particle size and composition, cloud cover, and albedo of the underlying surface. When superimposed on each other, the spatial distribution of global greenhouse gases warming and aerosol cooling are so uneven that, even if the global-average forcings are of equal and opposite sign, the pattern of climate change predicted by general circulation models differs markedly when aerosol forcing is added to global greenhouse gas forcing.<sup>182</sup>

Nitrous oxide ( $\text{N}_2\text{O}$ , a greenhouse gas) is a significant contributor to atmospheric warming, and diesel and spark ignition vehicles are considered as potentially significant sources of anthropogenic  $\text{N}_2\text{O}$  emissions.<sup>183</sup> U.S. anthropogenic  $\text{N}_2\text{O}$  emissions were 1.0 million tons in 1997, ~47,000 tons above the 1990 levels. Nearly all the increase from 1990 levels can be attributed to emissions from motor vehicles.<sup>184</sup> Nitrous oxide is produced as a byproduct of NO reduction and CO/HC oxidation on noble metal catalysts in gasoline vehicle exhaust systems. Catalysts are also being considered as after-engine treatments for diesel exhaust systems. Odaka et al.<sup>185</sup> found that catalyst temperature zones and levels of  $\text{N}_2\text{O}$  formation vary greatly by catalyst composition. Catalysts with lower metal content showed lower  $\text{N}_2\text{O}$  emissions

under both fresh and aged conditions.  $N_2O$  emissions increased as catalysts aged, but at different rates for different catalysts. Weaver and Chan<sup>186</sup> found that both new and early three-way catalysts have the same  $N_2O$  emission properties because catalyst aging occurred relatively quickly. If the entire fleet of existing automobiles were equipped with present-day catalysts, their  $N_2O$  emissions could account for 6–32% of emission increases.<sup>187</sup>

### Water and Soil Pollution

Evaporative emissions of diesel fuel constituents are lower than those from gasoline, due to closed systems and low volatility.<sup>188</sup> Diesel fuel spills (accidents), leaks from pipelines and tanks, and roadway runoff are the most common forms of inadvertent release.<sup>100,189</sup> Adverse effects of these incidents vary considerably and depend on transformation and removal processes after the leakage or spill.<sup>190–194</sup> Light alkanes and monocyclic aromatic hydrocarbons, such as benzene, toluene, ethylbenzene, and xylenes, evaporate and biodegrade readily.<sup>194</sup> Heavy metals and PAHs are long-lived and, because of their high molecular weight and high lipid solubility, they do not readily evaporate. Instead, they tend to accumulate in sediments,<sup>190,195</sup> where they pose a chronic threat to aquatic organisms long after the acute initial effects of the spill have abated.

The effects of diesel fuel spills on aquatic organisms include mortality, reproductive impairment, depressed growth rates, and increased susceptibility to infectious and noninfectious diseases.<sup>196–201</sup> In January 1988, a tank filled with ~3.8 million gallons of diesel fuel collapsed, and 750,000 gallons escaped into the Ohio River in Pennsylvania. The spill killed more than 10,000 fish and 2000 ducks and resulted in ~4000 damage claims totaling more than \$17 million. Long-term damage to fish and wildlife and contamination of sediments and groundwater are still being evaluated.<sup>202–204</sup>

Another example involves a 240,000-gallon diesel fuel spill that occurred in January 2001 near the Galapagos Islands, off the coast of Ecuador.<sup>205</sup> Most of the species of lizards, insects, birds, and plants that inhabit these islands, the site of Darwin's investigations that led to his theory of evolution, cannot be found anywhere else in the world. The fuel spread over an area larger than the city of Los Angeles and killed dozens of animals and birds, including pelicans and seagulls. Extensive long-term impacts are expected as the fuel sinks to the ocean bottom, smothering algae and other sea plants that fish, marine iguanas, and birds like to eat. At least 2 years are needed to complete the cleanup. Other spills and their environmental impacts are summarized by WHO.<sup>100</sup>

Although fuel spills are highly visible, long-term leakage from tanks and pipelines is more widespread and potentially more dangerous. Leaks usually occur over many

months or years before they are noticed and repaired. Long-term effects include damage to fish and wildlife, enhanced levels of toxic compounds, and contamination of sediments and groundwater.<sup>206–208</sup> A large petroleum bulk storage tank began to leak in northern Virginia during the mid-1980s.<sup>209,210</sup> Approximately 200,000 gallons of diesel fuel and gasoline traveled 2500 feet below the surface in less than 5 years. Natural biodegradation will take as long as 2500 years, and bioremediation will require ~200 years, at a cost of over \$100 million. In San Luis Obispo County, CA, 200,000–480,000 barrels of diesel fuel leaked from Unocal's Guadalupe oil field over several decades, beginning in the 1970s.<sup>207</sup> Seventeen diluent plumes have been identified as the most serious, with 73 lesser plumes detected. Cleanup is expected to take at least a decade and cost more than \$70 million.

Atmospheric deposition of air pollutants released from diesel exhaust to ecosystems and their components, such as forests, water bodies, and soils, is another significant source of contamination.<sup>211</sup> Water and soil are contaminated indirectly by dry and wet deposition of diesel exhaust emitted to the atmosphere. Wet deposition dominates the transfer of airborne contaminants to the Earth's surface, but dry deposition may be important in arid areas where ambient concentrations are high and rainfall is limited. In urban areas, sedimentation of large particles is more important than wet deposition and dry gaseous and small particle deposition.<sup>212</sup>

Environmental effects of atmospheric deposition have been studied for a long time, but the mechanism is poorly understood.<sup>211,213</sup> Enhanced levels of atmospherically deposited nitric and sulfuric acid (acid rain) adversely affect the health of ecological systems, such as agricultural crops, large water bodies, and forests. Arimoto<sup>211</sup> concluded that deposition is the dominant source for a variety of chemical contaminants in the Great Lakes. Atmospheric deposition provides most of the dissolved inorganic and total nitrogen in the nutrient load of the lakes and is responsible for water nitrification and forest damage.<sup>214</sup> Atmospheric wet deposition to a high-elevation forest at Whiteface Mountain, NY, was 12 times more efficient at transferring nitrogen to the forest canopy than was dry deposition.<sup>215</sup>

Heavy metals, PAHs, and dioxins common to diesel exhaust can be transported long distances as gases or PM. They are often resistant to degradation and are found in relatively high concentrations in many rural and remote areas.<sup>212,216</sup> Simcik et al.<sup>217</sup> and Wik and Renberg<sup>218</sup> reported higher atmospheric loading of PAHs in lake sediments in Michigan and Sweden, respectively. Schroder et al.<sup>212</sup> reported that wet deposition accounted for 85% of the total deposition of toxic materials in a terrestrial ecosystem. PAHs and dioxins have also been detected in German forest canopies.<sup>216</sup>



### Effects on Materials

The deposition of airborne particles on the surfaces of buildings, tunnels, highway bridges, and culturally important articles (e.g., statuary) can cause damage and soiling, thus reducing the useful life and aesthetic appeal of such structures.<sup>219,220</sup> Diesel exhaust builds up in tunnels and underground garages,<sup>221</sup> where it is difficult to remove. Soot promotes metal corrosion.<sup>222</sup> Many metals form a film that protects against corrosion; however, high concentrations of anthropogenic pollutants lessen the effectiveness of the protective film.<sup>223</sup> The costs associated with soiling are significant. A 1982 study<sup>224</sup> estimated an annual cost of \$800,000,000 (range of \$220,000,000–\$2,700,000,000) for a hypothetical dieselization of 20% of the light-duty vehicles in California. These figures include only the cost to households. Consideration of costs associated with the soiling of public, commercial, and industrial buildings and their contents, and of motor vehicles, would further increase the projected costs.

### DIESEL EMISSION CONTROLS

#### Heavy-Duty Diesel Vehicle Smoke Inspection Programs

Federal certification standards for smoke opacity levels from on-road heavy-duty vehicles commenced in 1970. To attain these standards, fuel and corresponding turbo boost settings were changed to reduce visible smoke during steady-state operation. However, since the largest portion of smoke is emitted during transient operation (when a truck accelerates from a complete stop or a low speed), transient fuel controls were also added. The heavy-duty PM standard has been reduced from 0.60 grams per brake horsepower-hour (g/bhp-hr, emissions as a function of energy produced) in 1988 to 0.10 g/bhp-hr today, and will continue to be further reduced in the future.<sup>225</sup>

In the United States, eight states have diesel testing programs for opacity.<sup>226</sup> British Columbia, Canada, and Taiwan have implemented enforcement programs, and Australia is conducting research before implementing its program.<sup>227,228</sup> The Society of Automotive Engineers (SAE) provides a uniform method for opacity testing.<sup>229</sup> Some states use a rolling acceleration in place of, or in addition to, the SAE procedure. A few states apply a loaded test using a dynamometer.<sup>230</sup> Some states (including California) use random roadside inspections; others mandate annual testing at state-run or contracted inspection stations prior to vehicle registration. California's Periodic Smoke Inspection Program (PSIP) also requires self-inspection that is not linked to vehicle registration.<sup>226</sup> Vehicles that are not in compliance with state standards must be repaired and retested before being used.

Under California's Heavy-Duty Vehicle Inspection Program (HDVIP), vehicles are inspected using SAE J667

test protocols at fleet facilities, randomly selected roadside locations, and truck weight and safety inspection stations. Noncomplying vehicles are cited and incur civil penalties ranging from a Notice of Violation (no monetary fine; similar to a "fix-it ticket") to an \$1800 fine for two or more violations in one year. Owners must pass a post-repair opacity test, in addition to paying a fine. The California Air Resources Board (ARB) audits fleet records to ensure PSIP compliance. HDVIP and PSIP will remove ~5.3 tons/day of reactive organic gases, ~14.0 tons/day of NO<sub>x</sub>, and ~3.2 tons/day of PM<sub>10</sub> by 2010. Annual diesel fuel consumption should also be reduced by 19.2 million gallons, owing to increased operating efficiency.<sup>231</sup>

Opacity test failure rates in California declined from ~30% in 1990 to ~8% currently. Future in-use compliance programs will add measurements of NO<sub>x</sub>, HC, and PM emissions to the existing opacity tests. The United States and some other countries are developing dynamometer-based emission inspections for heavy-duty diesel-powered vehicles. A transportable dynamometer and emission measurement system that measures real-time diesel gas and particle emissions has been tested in Australia. Future in-use compliance tests will also evaluate owner malmaintenance/tampering, poorly designed emission control systems, and low durability.<sup>232</sup>

### Engine Technology

On-road heavy-duty diesel emission standards were first implemented in California in 1973, and in the rest of the United States in 1974. These emission standards were harmonized in 1988 to reduce the certification and testing burden on the engine manufacturers and to ensure that clean trucks operate throughout the 50 states. Federal and California NO<sub>x</sub> and PM emission standards were based on a transient engine test called the "Federal Test Procedure" (FTP).<sup>233,234</sup> Figures 6a and 6b show the U.S. and California standards, as well as the European Union (EU) emission standards for those respective years.<sup>235</sup> The U.S. and EU standards are based on different test cycles for compliance, so they are not directly comparable. The figures, however, illustrate a continuous trend toward cleaner engines in both the United States and Europe. With respect to the EU test cycle, compliance with the standard prior to the year 2000 was determined using a 13-mode steady-state engine cycle called ECE R-49. Beginning with 2000, this test cycle was replaced by two new cycles: a European Stationary Cycle (ESC) and a European Transient Cycle (ETC). The current NO<sub>x</sub> and PM emission standards for the EU are 3.6 g/bhp-hr and 0.07 g/bhp-hr, respectively, on the ESC and ETC. As of 2001, the federal/California NO<sub>x</sub> and PM emission standards are 4.0 g and 0.10 g/bhp-hr on the FTP, respectively. The current federal/California standards represent a

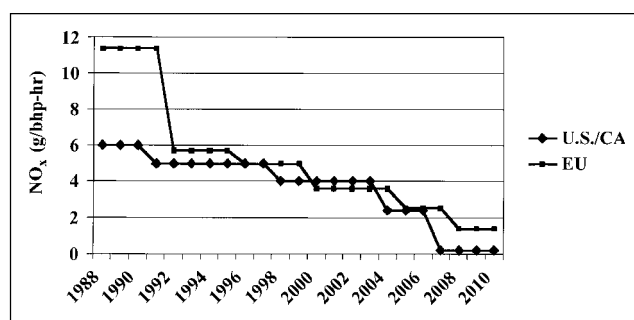


Figure 6a. Federal/California and European Union  $\text{NO}_x$  emission standards for heavy-duty diesel engines are being continually ratcheted downward.

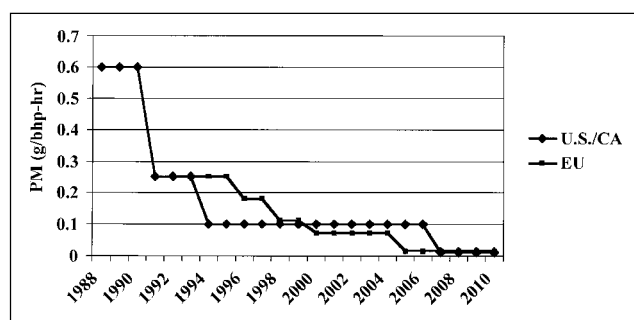


Figure 6b. Federal/California and European Union PM emission standards for heavy-duty diesel engines are being continually ratcheted downward.

more than 75% reduction in emissions, compared with precontrolled levels.

Off-road emission standards were first implemented in California in 1996. Federal and California regulations (shown in Table 6) are currently harmonized and have become increasingly stringent, based on engine horsepower and model year. The current emission standards for nonmethane hydrocarbons (NMHCs) plus  $\text{NO}_x$  and PM standards, called Tier 1, are approximately 15–50% lower than the uncontrolled levels.<sup>238</sup> Locomotive emissions are controlled under a separate U.S. regulation for newly manufactured and remanufactured locomotive engines that takes effect in 2001.<sup>239</sup> A Memorandum of Understanding, signed in July 1998 with the California railroads, EPA, and the ARB, provides for the early introduction of these cleaner locomotive units in California's South Coast Air Basin.<sup>240</sup> Commercial marine vessel standards begin in 2004 and are similar to the Tier 2 off-road and locomotive standards.<sup>241</sup>

Diesel engine emissions are currently controlled through improvements to the basic engine, rather than through the use of aftertreatment devices (other than diesel oxidation catalysts). Control techniques are usually limited by a  $\text{NO}_x$  and PM tradeoff, where strategies to reduce one pollutant may result in an increase to the other.

Table 6. Current and future U.S. and California emission standards for off-road compression-ignition engines (g/bhp-hr).<sup>236,237</sup>

Maximum Rated Power	Tier	Model Year	$\text{NO}_x$	THC	NMHC + $\text{NO}_x$	CO	PM
hp < 11	Tier 1	2000–2004	—	—	7.8	6.0	0.75
	Tier 2	2005 and later	—	—	5.6	6.0	0.60
11 < hp < 25	Tier 1	2000–2004	—	—	7.1	4.9	0.60
	Tier 2	2005 and later	—	—	5.6	4.9	0.60
25 < hp < 50	Tier 1	2000–2003	—	—	7.1	4.1	0.60
	Tier 2	2004 and later	—	—	5.6	4.1	0.45
50 < hp < 100	Tier 1	2000–2003	6.9	—	—	—	—
	Tier 2	2004–2007	—	—	5.6	3.7	0.30
	Tier 3	2008 and later	—	—	3.5	3.7	—
100 < hp < 175	Tier 1	2000–2002	6.9	—	—	—	—
	Tier 2	2003–2006	—	—	4.9	3.7	0.22
	Tier 3	2007 and later	—	—	3.0	3.7	—
175 < hp < 300	Tier 1	1996–2002	6.9	1.0	—	8.5	0.4
	Tier 2	2003–2005	—	—	4.9	2.6	0.15
	Tier 3	2006 and later	—	—	3.0	2.6	—
300 < hp < 600	Tier 1	1996–2000	6.9	1.0	—	8.5	0.4
	Tier 2	2001–2005	—	—	4.8	2.6	0.15
	Tier 3	2006 and later	—	—	3.0	2.6	—
600 < hp < 750	Tier 1	1996–2001	6.9	1.0	—	8.5	0.4
	Tier 2	2002–2005	—	—	4.8	2.6	0.15
	Tier 3	2006 and later	—	—	3.0	2.6	—
hp > 750	Tier 1	2000–2005	6.9	1.0	—	8.5	0.4
	Tier 2	2006 and later	—	—	4.8	2.6	0.15

Nitric oxide formation is directly related to combustion chamber temperature. Increased combustion temperatures result in higher NO<sub>x</sub> emissions. Nitric oxide reductions result from decreasing peak combustion temperatures and reducing the duration of high temperatures in the combustion chamber. Particulate matter, on the other hand, results from incomplete combustion of diesel fuel. Particulate matter emissions are reduced by an improvement in fuel combustion that results in higher combustion temperatures and increased NO<sub>x</sub>. Currently, diesel emissions are reduced by turbocharging, aftercooling, optimizing combustion chamber design, retarding injection timing, and high-pressure fuel injection.<sup>242-245</sup>

Turbochargers reduce both NO<sub>x</sub> and PM emissions by ~33%, compared with naturally aspirated engines.<sup>246</sup> The turbocharger boosts the pressure (and temperature) of the air entering the engine. This allows more fuel to be added to increase power output, while still inhibiting PM formation. Power to drive the turbocharger is extracted from the engine's exhaust stream. The turbocharger also increases engine power and fuel efficiency. Aftercooling with turbocharging yields even larger NO<sub>x</sub> and PM reductions by decreasing the temperature of the charged air after it has been heated by the turbocharger during compression. Aftercooling improves cylinder filling because higher-density cool air sinks faster than hotter air. Engine coolant circulating through a heat exchanger is sometimes used for aftercooling. However, aftercooling using an air-to-air heat exchanger is more effective because it attains lower temperatures. Both approaches are most effective when vehicle motion provides fresh air to cool the radiator or aftercooler.

Modifications to the shape of the combustion chamber, location of the injection swirl, crevice volumes, and compression ratios also optimize fuel efficiency and multipollutant reductions.<sup>243,245</sup> Improved understanding of diesel combustion and in-cylinder gas and particle formation is obtained via optical diagnostic techniques and computational models. Changes to combustion technology are continually being made to improve in-cylinder flow management, such as the geometrical design of the intake port and valve and increased swirl.<sup>247,248</sup>

Injection timing retard reduces the peak flame temperature, resulting in NO<sub>x</sub> reductions. However, timing retard typically lowers fuel efficiency, resulting in lower mileage and higher PM emissions. High pressure fuel injection can regain some of the efficiency loss by improving the atomization of the fuel spray and air utilization, resulting in more complete combustion.<sup>243,245</sup> Some fuel combustion efficiency has been traded for lower emissions to attain standards.

In the 1990s, oxidation catalysts were added to some

truck engines and many urban bus engines to reduce PM emissions. Flow-through oxidation catalysts effectively oxidize gaseous HC and the soluble organic fraction of PM. A recent test program showed that oxidation catalysts reduced transient FTP PM emissions by 23–29% and HC emissions by 52–88%, using a typical grade #2 diesel fuel (368 ppmw sulfur). Testing with a low sulfur diesel fuel (54 ppmw sulfur) resulted in an additional 13% reduction in PM.<sup>249</sup> Electronic computer control has also improved emissions. However, in some cases, these controls use special injection timing and other air and fuel management parameters to pass the FTP tests, while reverting to more polluting settings during on-road operation. A consent decree coupled with a large monetary settlement has stopped this deceptive practice.

### Fuels

Diesel fuel improvements have resulted in large reductions of SO<sub>2</sub> and PM emissions.<sup>250</sup> The guide standard for diesel fuel quality in the United States (ASTM D 975, Standard Specification for Diesel Fuel Oils) was originally adopted in 1948. Maximum sulfur contents of 0.5 and 1.0% by weight (i.e., 5000 and 10,000 ppmw, respectively) were established for Grades No. 1 and No. 2 diesel fuel oil, respectively, primarily to control engine wear and deposits.<sup>251</sup> ASTM D 975 specifies a minimum cetane number of 40 for both grades to ensure starting and smooth combustion. By the 1980s, the sulfur specification for Grade No. 2 decreased from 1.0 to 0.5% by weight.<sup>252</sup> In 1992, ASTM D 975 was updated to include a low sulfur fuel specification of 500 ppmw for Grades No. 1 and No. 2 diesel.<sup>253</sup> These changes paralleled U.S. standards for highway diesel fuel to reduce gaseous SO<sub>2</sub> and particulate sulfate emissions, which became effective in 1993.<sup>254,255</sup> In the South Coast Air Basin and Ventura County of California, a state standard for low sulfur fuel has been in effect for all on-road diesel fuel sold since 1985.<sup>256</sup>

Fuel sulfur content affects engine wear, deposit formation, and emissions. Fuel sulfur that is not deposited within the fuel system, engine, or exhaust system is emitted as sulfurous compounds, such as gaseous SO<sub>2</sub> and particulate sulfates. Sulfur compounds in engine exhaust also reduce the effectiveness of emission control equipment, especially the efficiency of catalysts. Cetane number requirements depend on engine design, size, nature of speed and load variations, starting, and atmospheric conditions. Too low a cetane number can result in poor combustion and high emissions under transient cycle operation.<sup>253</sup> The cetane number increases through the use of ignition improvement additives, such as 2-ethyl hexyl nitrate. Cetane number specifications for diesel fuels in Europe, Asia, and South Africa generally range from 45 to 50 ppmw.<sup>257</sup>

**Table 7.** Typical characteristics of equivalent alternative diesel fuel formulations.

Characteristic	Reference Fuel	Alternative Formulations <sup>a</sup>	Historical Average
Sulfur content (ppmw)	≤500	≤250	2800
Aromatic content (v %)	≤10	≤22	31
PAH content (wt. %)	≤1.4	≤4	NA
Cetane number	(natural) ≥48	≥54	NA

<sup>a</sup><http://www.arb.ca.gov/diesel/diesel.htm>, Certified Alternative Diesel Formulations.<sup>259,260</sup>

**Table 8.** Swedish diesel fuel classifications.

Requirements (partial)	Urban Diesel		Standard Grades			
	1	2	Summer	Winter 1	Winter 2	Winter 3
Sulfur (ppmw)	≤10	≤50	≤2000	≤2000	≤2000	≤2000
Aromatic (v %)	≤5	≤20	—	—	—	—
PAH (v %)	≤0.02	≤0.1	—	—	—	—
Density (kg/m <sup>3</sup> )	800–820	800–820	820–860	800–845	800–840	800–840
Cetane no.	≥50	≥47	≥49	≥47	≥48	≥45
Tax rate (\$/m <sup>3</sup> )	126	165	199	199	199	199

Aromatic compounds have high liquid densities. Monocyclic compounds have lower boiling points, and polycyclic compounds have higher boiling points. High aromaticity generally means high volumetric energy content, high combustion temperatures, poor combustion (due to low natural cetane number), and high emissions.<sup>258</sup>

To meet California standards,<sup>256</sup> diesel fuel must have a sulfur content less than 500 ppmw<sup>259</sup> and an average aromatic HC content (aromaticity) less than 10% by volume,<sup>260</sup> unless the fuel is produced under the regulation's flexibility provisions that accommodate small refiners and alternative formulations. Stationary engines, locomotives, and marine vessels are currently exempt from the California regulation. Producers can have diesel fuel certified by the ARB for use in California with a maximum aromaticity greater than 10%, if the alternative formulation is equivalent to the 10% aromatic fuel, as determined by engine emission tests.<sup>259,260</sup> Texas has also adopted the California diesel fuel regulations.<sup>261</sup> Table 7 shows typical characteristics of alternative formulations.

Sweden has established tax incentives for two urban diesel fuel classifications that are cleaner burning than Swedish standard grades.<sup>257</sup> Table 8 lists the requirements of the Swedish fuel classifications. Tables 9 and 10 show the diesel fuel sulfur levels in various countries that have incentives or plan to introduce incentives to accelerate the introduction of low sulfur fuels.

## EMISSION MODELING

For over 25 years, both EPA and ARB have maintained mathematical models to estimate the contribution of

on- and off-road mobile sources to the emissions inventory of the nation and state. According to California's most recent emissions inventory model, EMFAC2000, diesel-powered vehicles constitute only 3% of the population and contribute only 5% of the daily vehicle miles of travel (DVMT), yet 35% of the NO<sub>x</sub> and 56% of the vehicle exhaust particulate matter (PM<sub>ex</sub>) were attributable to these vehicles in California in the year 2000.

The United States has a lesser dependency on diesel fuel than do other countries, especially with respect to passenger cars. As can be seen in Table 11, the diesel fraction of the California fleet increases as a function of vehicle weight, ranging from 1% for passenger cars (PC) and light-duty trucks (LDT) to 10% of medium-duty vehicles (MDV) and 56% of heavy-duty trucks (HDT).

As with light-duty gasoline-powered vehicles, the emissions inventory for heavy-duty diesel vehicles is calculated as the product of an emissions rate, modeled

**Table 9.** Diesel specifications from fuel specifications of EU Directive 98/70/EC on petrol and diesel quality.<sup>262</sup>

Parameter	Limits from Jan 2000	Limits from Jan 2005
	(EURO3)	(EURO4)
Sulfur (ppmw)	≤350	≤50
PAH (wt%)	≤11	—
Density at 15 °C (kg/m <sup>3</sup> )	<845	800–845
Cetane no.	≥51	—
Distillation point 95% (°C)	<360	—

**Table 10.** Summary of diesel fuel regulations and incentive programs for selected countries.<sup>262-264</sup>

Country	Regulation or Incentive	Max S Limit	Conventional Fuel Limit (and typical content)	Introduced
United States	Highway diesel fuel sulfur control requirements	15 ppmw	3300 ppmw	June 1, 2006
EU	EURO2		500 ppmw (450)	Jan 1, 1997
	98/70/EC EURO3		350 ppmw	Jan 1, 2000
	98/70/EC EURO4		50 ppmw	Jan 1, 2005
Belgium	National incentive	50 ppmw	350 ppmw	Oct 1, 2001
Denmark <sup>a</sup>	National incentive	50 ppmw	500 ppmw	June 1999
Finland <sup>b</sup>	National incentive	50 ppmw	350 ppmw	2002
	Neste/Fortum initiative	10 ppmw		
Germany <sup>c</sup>	National incentive	50 ppmw	350 ppmw	Nov 1, 2001
		10 ppmw		Jan 2003
Netherlands	National incentive	50 ppmw	350 ppmw	Jan 2001
Sweden	National incentive <sup>d</sup>	10 ppmw	2000 ppmw	1991
	National incentive <sup>e</sup>	10 ppmw	350 ppmw	2001
		50 ppmw	350 ppmw	2001
Switzerland	National incentive	50/10 ppmw <sup>f</sup>	350 ppmw	2003
	Agrola initiative	10 ppmw <sup>g</sup>	350 ppmw	2000
	BP initiative	10 ppmw <sup>h</sup>	350 ppmw	2000
UK	National incentive	50 ppmw	500 ppmw	March 1999
	National incentive	50 ppmw	350 ppmw	March 7, 2001
Australia	National regulation	50 ppmw	1300 ppmw	Jan 2006
	BP initiative <sup>i</sup>	50 ppmw	500 ppmw	End 2000
Hong Kong <sup>j</sup>	"Ultra-low sulphur" national incentive	50 ppmw	500 ppmw	July 2000
Japan <sup>k</sup>	National regulatory proposal	50 ppmw	500 ppmw	Before 2005

<sup>a</sup>100% penetration by July 1999; <sup>b</sup>100% penetration; <sup>c</sup>From 2003, the incentive will shift from 50 ppm fuels to 10 ppm fuels; <sup>d</sup>City diesel; <sup>e</sup>Current incentive, last adjusted Jan 1, 2001; <sup>f</sup>Proposal before parliament; <sup>g</sup>Small market share; <sup>h</sup>Supply to public transport and army; <sup>i</sup>Capacity to supply 12% of national market; <sup>j</sup>Replaced regular diesel at all filling stations but high-sulfur fuel still used by bus fleets as tax-free; <sup>k</sup>Japan Air Quality Committee has recommended further reduction in the future.

**Table 11.** California's statewide on-road vehicle emissions for CY 2000 estimated using EMFAC2000 version 2.02.

Class <sup>a</sup>	Population	DVMT	HC	Tons/Day		
				CO	NO <sub>x</sub>	PM <sub>10</sub>
PC gas	14,258,518	463,490,000	762.48	6593.71	537.80	8.50
PC diesel	112,030	2,862,000	3.26	3.84	6.85	0.66
LDT gas	6,048,473	203,382,000	316.28	3509.51	359.87	5.68
LDT diesel	31,792	893,000	0.73	1.23	2.00	0.19
MDT gas	1,830,323	61,029,000	202.61	2374.67	209.04	1.92
MDT diesel	195,454	7,628,000	4.52	8.09	29.49	0.94
HDT gas	264,729	5,653,000	64.69	1020.82	77.82	0.12
HDT diesel	341,287	29,832,000	32.41	158.58	571.04	18.46
Diesel bus	13,955	1,478,000	5.55	94.93	35.91	0.55
Motorcycle	331,143	1,995,000	18.65	80.08	2.70	0.12
Total	23,427,704	778,242,000	1411.18	13,845.46	1832.52	37.14

<sup>a</sup>PC= passenger car; LDT= light-duty truck; MDT= medium-duty truck; HDT= heavy-duty truck.

**Table 12.** EMFAC2000-estimated emission rates for California heavy diesel trucks (>33,000 lb gross vehicle weight rating).

Model Year Group	Emissions @ 200,000 mi (g/mi)	
	NO <sub>x</sub>	PM
Pre-1975	28.76	2.30
1975–1976	27.43	2.17
1977–1979	27.43	2.19
1980–1983	27.45	2.21
1984–1986	20.40	1.42
1987–1990	17.09	1.00
1991–1993	16.57	0.69
1994–1997	19.90	0.52
1998	23.75	0.40
1999–2002	13.62	0.27
2003	6.82	0.32
2004+	6.82	0.32

as a linear equation and a measure of activity. Historically, the emissions rate is derived from emission tests of engines and is expressed in terms of mass per unit of work (g/bhp-hr). To derive an inventory, a correction factor must be used to obtain emissions in g/mi. Table 12 presents the latest estimates of NO<sub>x</sub> and PM emissions for the heaviest class of diesel trucks, based on EMFAC2000. It is difficult to assess the overall accuracy of the inventory estimates of emissions from diesel vehicles. Dreher and Harley<sup>265</sup> suggested that EMFAC7G, the predecessor to California's current estimation model, underestimates NO<sub>x</sub> emissions from on-road diesels by a factor of 2.3 and fine BC emissions by a factor of 4.5, compared with an assessment based on fuel use.

In a National Academy of Sciences report,<sup>266</sup> EPA was criticized for underestimating both NO<sub>x</sub> and PM emission rates for heavy-duty diesel vehicles in the national emissions inventory estimation model, MOBILE. It was recommended that EPA should design and undertake a large-scale testing program that will better assess real-world emissions from heavy-duty diesel vehicles. The testing program should include a broad range of engine technologies and ages and be based on driving cycles that accurately reflect real-world driving patterns.

There are many factors that may affect the accuracy of the inventory estimates of heavy-duty diesel emissions,

including uncertainty in the correction factors that convert grams per unit of work to grams per mile, the lack of real-world driving cycles, sparse data on vehicle activity, and characterization of emissions deterioration as the engine ages. Probably the most significant factor, however, is a lack of emission test data.

Because of the high cost of testing (historically, the engine is removed from the truck for testing on an engine dynamometer) and the difficulty of recruiting trucks that are used in daily commerce, gathering test data on the emissions characteristics of heavy-duty diesel trucks has lagged. While the California emission inventory for light-duty gasoline vehicles is based on data from tests performed on over 6000 vehicles, the heavy-duty diesel truck inventory is based on only 70 trucks,<sup>267</sup> as shown in Table 13. Because heavy-duty engines can be used in a host of different applications, the engines alone, rather than the complete vehicles, are tested for new engine certification. Emissions tests of heavy-duty engines yield an estimate expressed in terms of grams of pollutant per brake-horsepower hour (emissions per unit work) that must then be converted to gram per mile units. This conversion factor, which is essentially an estimate of fuel economy, is the second source of uncertainty.

The brake-specific fuel consumption estimates used for the development of these conversion factors have been derived from either data collected during the certification test or from surveys, such as the Truck Inventory and Use Survey (TIUS) conducted every 5 years by the Bureau of Census. Because heavy-duty diesel engines are only tested over a single cycle during certification, it is unlikely that the range of work that the engine might encounter in "real-world" operation is adequately reflected. For example, an engine destined for use in either a line-haul or a garbage truck would be subjected to the same certification procedure and, therefore, have identical brake-specific fuel consumption estimates. Furthermore, PM emissions are more strongly a function of the frequency and severity of the transients in the duty cycle than the fuel consumption of the vehicle. To paraphrase EPA documentation on this topic, "the best approach for determining conversion factors would be to develop in-use driving cycles and then test a statistically significant number of trucks and buses over those cycles to determine conversion factors for each

**Table 13.** California's heavy-duty diesel truck emission database (number of trucks tested by model year).

Year	66	81	82	83	84	85	86	87	88	89	90	91	92	93	94	95	96	97	98	99	Total
HHDT <sup>a</sup>	1	1	1	1	1	2			2	1	1	1		2	1	2	1	1	4		23
MHDT <sup>b</sup>						1		2	1	3	3		2	4	2	2	4	1	1	1	27
LHDT <sup>c</sup>			1		1	2	1	2	1	1		1	1	1	4	2	2				20
Total	1	1	2	1	2	5	1	4	4	5	4	2	3	7	7	6	7	2	5	1	70

<sup>a</sup>HHDT= heavy heavy-duty truck; <sup>b</sup>MHDT= medium heavy-duty truck; <sup>c</sup>LHDT= light heavy-duty truck.

pollutant and driving cycle.”<sup>268</sup> Rather than use this approach to refine the conversion factors, ARB and the environmental protection agencies of Hong Kong and Australia have opted to develop more representative heavy-duty test cycles and base inventory estimates directly on chassis tests of heavy-duty diesel vehicles.

In developing EMFAC2000, the practice of using engine test data with conversion factors to derive grams per mile emission rates was abandoned in favor of a chassis test-based inventory estimate. The lack of a uniform cycle, or set of cycles, on which to base the heavy-duty diesel inventory is the third area of uncertainty. A heavy-duty working group assembled by ARB concluded that the Urban Dynamometer Driving Schedule (UDDS) was the best cycle currently available on which to base the revised inventory.

The move to a chassis-based inventory reduced the difference between the emission test and fuel-based California  $\text{NO}_x$  inventory from a factor of 2.3 using EMFAC7G to a factor of 1.2 using EMFAC2000.<sup>265</sup> This modification did not affect PM emissions estimates significantly. However, when the chassis-derived emission factors for PM used in EMFAC2000 were compared with other chassis-derived emission rates, such as those collected by Parsons Australia Pty Ltd, the differences were found to be 15% or less for engines produced between 1990 and 1999.

Although the use of UDDS-generated chassis test data signifies a marked improvement in the inventory, few believe that the UDDS is adequate to capture the full range of heavy-duty diesel operation. The UDDS is just over 1000 sec in length and covers 5.55 mi at a speed of ~19 mph. Although the cycle was constructed from actual recorded driving patterns of heavy-duty trucks, the cycle lacks extended cruises, known to cause many larger heavy-duty diesel truck engines manufactured during the 1990s to default to a high  $\text{NO}_x$ -emitting, fuel-saving mode referred to as “off-cycle”  $\text{NO}_x$ . The UDDS also lacks hard accelerations known to increase PM emissions.

The fourth area of uncertainty in estimating the emissions inventory of heavy-duty diesel vehicles is activity. Since 1984, all gasoline-powered vehicles registered in California, with the exception of motorcycles, have been compelled to undergo periodic testing under the inspection and maintenance (I/M) program. Because odometer readings are taken at the time of testing, a comprehensive database of mileage accrual rates (the miles per vehicle per year driven as a function of vehicle age) is available for these vehicles. No equivalent source of information is available for diesel vehicles, as they are currently exempt from this type of inspection. This information has also been gathered in TIUS surveys; however, this source provides only a subjective assumption of heavy-duty truck travel within each state that is neither age- nor area-specific (below the state level). To address this issue, some have suggested the use of fuel sales rather than DVMT as the indicator of activity. However, this approach assumes that all fuel sold in a particular area will also be consumed in that area. The smaller the area of analysis, the larger this uncertainty becomes.

Another approach involves equipping heavy-duty vehicles with Global Positioning Systems (GPS) and data-logging devices capable of capturing highly resolved activity and location information on a second-by-second basis. Eighty-four heavy-duty diesel trucks were so instrumented and 2480 trips were recorded, covering over 60,000 miles of operation.<sup>269,270</sup> Preliminary analyses of these data reveals that the average vehicle speed is 37.5 mph, the average trip length is ~34 mi, and that these vehicles experienced extended periods of idle (~38% of their operating time).<sup>271</sup> Contrasting this information with the UDDS shows that the UDDS is too slow and too short to be representative of the average operation of the heavy-duty diesel fleet, and basing the inventory on this cycle is likely to lead to underestimation of  $\text{NO}_x$  emissions. Work is underway to use the instrumented vehicle information to develop more representative cycles for

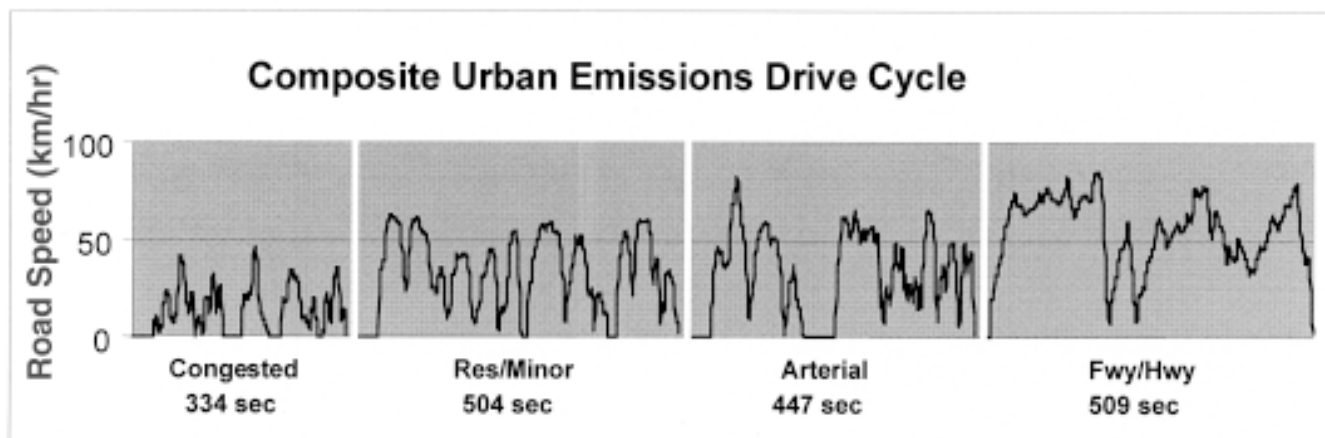


Figure 7. The CUEDC is intended to replicate several types of actual driving conditions. Res = residential, Fwy = freeway, Hwy = highway.

**Table 14.** Correlation of emissions on a short test to the CUECD.

Short Tests	Average NO <sub>x</sub> (g/sec)	Average HC (g/sec)	Correlation Coefficient (R <sup>2</sup> )		Average Opacity (%)	Maximum Opacity (%)	Rating 1 = Best 8 = Worst
			Average LLSP <sup>a</sup> (mg/sec)	Filter Mass (mg)			
AC5080 <sup>b</sup>	0.95	0.92	0.70	0.71	0.87	0.80	1
DT80	0.90	0.85	0.63	0.58	0.68	0.81	2
2 spd T <sup>c</sup>	0.62	0.72	0.30	—	0.40	0.68	3
DT80* <sup>d</sup>	0.80	0.74	0.35	—	0.15	0.21	4
Lug down <sup>e</sup>	0.60	0.68	0.22	—	0.26	0.68	5
2 spd P <sup>f</sup>	0.55	0.36	0.12	—	0.15	0.17	6
D550	0.64	0.53	0.18	0.23	0.03	0.23	7
Snap idle <sup>g</sup>	0.47	0.23	0.02	—	0.29	0.59	8

<sup>a</sup>LLSP is Laser Light Scattering Photometry—used to measure particles up to 10 µm; <sup>b</sup>AC5080 is the test cycle developed by ARB; <sup>c</sup>2 spd T is Two Speed Torque; <sup>d</sup>DT80\* is the last 10 sec of the DT80 test; <sup>e</sup>2 spd P is Two Speed Power; <sup>g</sup>Snap idle is used in California.

### heavy-duty truck testing.

A similar methodology utilized a number of vehicles that were instrumented during their normal driving, and the data were synthesized to form the average driving cycle for that area, called the Composite Urban Emissions Drive Cycle (CUEDC, as shown in Figure 7).<sup>272</sup> An effort was made during this study to develop short tests that could be performed in the field that would correlate well with laboratory tests over the CUEDC. The correlations of several short tests to the CUECD are shown in Table 14. The high correlations of the AC5080 and the DT80 cycles, both of which are less than 5 min in duration, suggest

that a relatively inexpensive field test could be developed and used to increase the heavy-duty diesel database, ultimately improving the emissions inventory.

The final area of uncertainty in the heavy-duty diesel emissions inventory is that of emissions deterioration, the increase in emissions as a function of engine usage. Deterioration in the diesel fleet is modeled as the product of the frequency of occurrence and the effect on emissions of 19 separate instances of tampering and mal-maintenance. Lacking a periodic inspection program for these vehicles, limited information is available to estimate the occurrence of these problems in the fleet, and quantifying their effects requires comparing baseline tests of vehicle with defects to their emission rates after repair. Such programs were performed by Radian Corporation in 1987<sup>273</sup> and the Colorado Institute for Fuels and Engine Research in 1998.<sup>274</sup> The results of these studies show that 10-fold increases in emissions associated with acts of tampering and mal-maintenance are frequent in the heavy-duty diesel fleet.

Off-road diesel-powered equipment is also a significant source of NO<sub>x</sub> and PM. Off-road diesel equipment emits roughly the same amount of NO<sub>x</sub> as on-road diesels emit, and nearly twice as much PM. The emissions inventory of off-road diesels is dominated by construction and agricultural equipment, as shown in Table 15. Compared with the on-road inventory, the off-road model is still in its infancy. Although no studies are known to quantify the uncertainty in the off-road diesel inventory, it can be assumed that all of the issues that pertain to the accuracy of the on-road inventory would also apply to the off-road model. In addition, while the on-road inventory concerns itself with 13 vehicle classes, the off-road model must estimate the emissions and activity patterns of equipment as diverse as generator sets and bulldozers.

**Table 15.** California emissions for off-road diesels for CY 2000 predicted using California's OFFROAD Model.

Category	Tons/Day	
	NO <sub>x</sub>	PM
Agricultural	149.43	9.70
Airport ground support	4.05	0.30
Commercial	27.26	2.04
Construction	331.67	21.12
Dredging	1.04	0.05
Drilling	11.87	0.64
Industrial	18.33	1.56
Lawn and garden	3.62	0.31
Logging	11.14	0.67
Military tactical support	1.42	0.08
Misc. portable equipment	0.13	0.01
Transportation refrigeration	25.68	2.60
Total off-road diesel	585.64	39.08
Total on-road diesel	645.29	20.80



As an innovative approach to gathering both activity and emissions information, the EPA is working to develop portable emissions monitoring systems. These systems, like the ROVER and West Virginia University's MEMS system, would be moved from engine to engine to develop an empirical inventory estimate. It is uncertain how this approach would be used to forecast future inventories. However, in order to improve both the on- and off-road diesel emissions inventory, an increased amount of testing and improved activity analyses for diesel vehicles must be made a priority. While both EPA and ARB have line-item budgets for the testing of light-duty gasoline vehicles, no such financial commitment is in place for diesels. To defer costs, it is suggested that the EPA, the ARB, and other interested parties, including other countries, cooperate in this area, as the proper characterization and control of diesel emissions are global priorities.

#### TECHNOLOGIES FOR THE FUTURE

##### Regulatory Requirements

By 2004, HC + NO<sub>x</sub> emissions on the transient FTP are required to be 50% lower than current standards.<sup>233,234</sup> EPA emission standards for 2007 will require ~90% emission reductions, compared with anticipated 2004 levels. These new on-road NO<sub>x</sub> and PM emission standards (shown in Figures 6a and 6b) are 0.2 and 0.01 g/bhp-hr, respectively.<sup>275</sup> A recent California regulation for transit buses sets a 0.01 g/bhp-hr PM standard beginning in October 2002, a 0.5 g/bhp-hr NO<sub>x</sub> standard for the 2004–2006 model years, and a 0.2 g/bhp-hr NO<sub>x</sub> standard for 2007 and subsequent model years.<sup>276</sup>

In 1998, seven heavy-duty diesel engine manufacturers signed consent decrees with EPA and ARB for violating certification regulations. Emission controls were automatically turned off or defeated during in-use highway driving. The consent decree included monetary penalties, completion of supplemental environmental projects, production of engines that meet the 2004 HC + NO<sub>x</sub> emission requirements 15 months earlier (in October 2002), and engine certification to supplemental test requirements (not-to-exceed and steady-state tests). Supplemental test procedures detect excessive modulation and turning off of emission controls during driving conditions not represented by the FTP, such as extended steady-state cruise conditions. In California, beginning in 2005, compliance with supplemental test procedures, as well as compliance with the FTP, will be required.

Future off-road emission standards are shown in Table 6. The introduction of the lower emission levels is based on power output and model year. Tier 2 HC + NO<sub>x</sub> and PM standards are about 20–60% lower than current Tier 1 standards. Tier 3 NO<sub>x</sub> standards are an additional

25% lower than Tier 2 standards.<sup>243–245</sup> Tier 3 PM standards are currently being developed because the appropriate levels could not be established at the time of the NO<sub>x</sub> standard, due to lack of a standard transient test procedure.

The 2000 California Diesel Risk Reduction Plan<sup>277</sup> outlines potential control measures that could be adopted to reduce exposure to toxic DPM. The plan calls for 14 control measures to be adopted by 2005, with full implementation before 2010. Highly efficient DPM aftertreatment devices, such as diesel particulate filters (DPF),<sup>278</sup> will be required on existing and new on- and off-road diesel engines.<sup>277</sup>

##### Future Engine Technology

As previously noted, existing emission standards have been met by modifying the diesel engine, rather than through the use of aftertreatment devices that remove pollutants from the exhaust stream. This is in contrast to gasoline-fueled passenger cars that have relied on reformulated fuel (unleaded gasoline) and aftertreatment devices (catalytic converters) to reduce emissions for more than 20 years. Fuel injection rate shaping, exhaust gas recirculation (EGR), and advanced combustion techniques can provide additional emissions reductions.<sup>242</sup> As combustion system refinements and EGR reach their limits, NO<sub>x</sub> and PM aftertreatment devices will be needed to comply with increasingly stringent emission standards.

Injection rate shaping tailors the fuel injection event to reduce peak flame temperatures without increasing fuel consumption. Injection rate shaping is possible because of electronic control and re-engineering of the fuel injectors. A pilot amount of fuel can be injected before the main injection event, or the main injection can be split into two or more events. Injection rate shaping has been shown to simultaneously reduce NO<sub>x</sub> by 20% and PM by 50% under certain operating conditions.<sup>279,280</sup> Fuel injection methods that achieve effective rate shaping include the common rail injector; the mechanically actuated, electronically controlled unit injector; and the hydraulically actuated, electronically controlled unit injector.<sup>242</sup>

EGR routes a portion of the exhaust gas into the engine air intake. It reduces NO<sub>x</sub> formation in the combustion chamber by diluting the air with inert exhaust gas that reduces peak flame temperatures when fuel is ignited. Laboratory studies have shown that EGR can reduce NO<sub>x</sub> by 40–50% at rated power, with no appreciable increase of PM emissions. Larger NO<sub>x</sub> reductions are found at other loads, with modest increases in PM emissions.<sup>9,281–284</sup> Precise control of the EGR rate is needed to minimize PM augmentation. Recirculated exhaust must be cooled for effective NO<sub>x</sub> reductions, but this cooling causes higher

PM emissions. Considering that cooling the inlet air, using an aftercooler, improves efficiency and reduces emissions, large amounts of uncooled EGR would heat the inlet charge air, partially offsetting effects on NO<sub>x</sub> emission reductions. However, with proper EGR design, these undesirable effects can be minimized.<sup>285</sup> Cooled EGR has been demonstrated to meet the future 2.4 g/bhp-hr HC + NO<sub>x</sub> standards with a 2% improvement in fuel economy; NO<sub>x</sub> emissions can be reduced by 50%.<sup>286</sup>

Conventional diesel engines inject fuel late in the compression stroke into hot, compressed air, resulting in autoignition. The rate of combustion is controlled by the rate at which fuel can mix with air because chemical reaction rates are much faster than mixing rates. NO<sub>x</sub> formation is high on the lean side of the flame, and PM formation is high on the rich side of the flame. In homogeneous charge compression ignition (HCCI) systems, fuel and air are premixed prior to introduction into the combustion chamber. Ignition occurs spontaneously throughout the mixture as a result of compression. This process produces ignition at a large number of sites throughout the combustion chamber, eliminating locally lean and rich zones that cause high NO<sub>x</sub> and PM. EGR helps regulate the conditions under which controlled combustion occurs.<sup>287,288</sup> Under low and medium loads, NO<sub>x</sub> reductions of 90–98% have been achieved.<sup>289</sup> The thermal efficiency of HCCI is comparable to that of conventional diesel combustion at partial loads. However, reduced efficiencies are observed for certain diesel engines, such as those using partial fumigation and direct injection. Challenges associated with HCCI include the control of combustion initiation and rate, effective fuel and air mixture preparation, and the achievement of stable HCCI under high loads and full power output.<sup>290</sup>

Aftertreatment systems require ultra-low sulfur fuel to be effective.<sup>291</sup> With the introduction of 15 ppmw sulfur diesel fuel in 2006, diesel engines equipped with aftertreatment devices and cooled EGR will be more than 90% cleaner than today's engines.<sup>275</sup> Sulfur levels in off-road diesel fuel will need to be similarly reduced to allow the transfer of on-road emission control technology to off-road engines. NO<sub>x</sub> aftertreatment devices include the lean NO<sub>x</sub> catalyst, the NO<sub>x</sub> adsorber, and selective catalytic reduction (SCR).

Lean NO<sub>x</sub> catalysts (active systems with diesel fuel as the reductant) have been shown to provide up to 30% NO<sub>x</sub> reduction under certain operating conditions. Fuel efficiency is reduced by 7% because some of the fuel is used as the reductant.<sup>292</sup> NO<sub>x</sub> adsorbers operate by storing NO<sub>x</sub> under typical diesel engine operations ("lean" conditions). Before the NO<sub>x</sub> adsorbent becomes fully saturated, engine operating conditions and fueling rates are

adjusted to produce a fuel-rich exhaust that reduces the stored NO<sub>x</sub> to nitrogen. NO<sub>x</sub> adsorbers have been demonstrated to reduce NO<sub>x</sub> emissions by more than 90% with ultra-low sulfur fuel for transient and steady-state conditions, but with reduced fuel economy.<sup>292–295</sup> NO<sub>x</sub> adsorbers' strong affinity for sulfur can deactivate the active catalyst sites and make the adsorbers less efficient over time. Improved NO<sub>x</sub> adsorber desulfurization systems, active catalyst layers that are more sulfur-resistant, and other methods are under development to maintain the NO<sub>x</sub> adsorber's high efficiency for the useful life of the engine.<sup>291,296</sup> One type of NO<sub>x</sub> adsorber includes an SO<sub>2</sub> sorbate catalyst upstream of the NO<sub>x</sub> adsorber to protect the NO<sub>x</sub> catalyst from sulfur poisoning. Testing showed NO<sub>x</sub> reductions >95% using Grade No. 2 diesel fuel (sulfur <500 ppmw).<sup>290</sup>

SCR has been used in stationary source applications for many years. It works by injecting NH<sub>3</sub> into the exhaust upstream of a catalyst to reduce NO<sub>x</sub> emissions. SCR can reduce NO<sub>x</sub> emissions by 70–85% for the transient FTP and by 65–99% for off-cycle tests.<sup>249,297–300</sup> Urea would probably be used to provide the reactant. The main challenges for SCR are controlling the rate of urea introduced to maximize NO<sub>x</sub> reductions without "ammonia slip" through the catalyst and ensuring that the urea is properly replenished throughout the vehicle life to ensure emission reductions.

The catalyzed DPF and the continuously regenerating DPF have been demonstrated to reduce particle emissions. In one program, using 54-ppmw sulfur fuel, the DPF reduced PM by 87% to a level of 0.008 g/bhp-hr.<sup>249,297</sup> Another program showed that heavy-duty trucks retrofitted with DPFs and fueled with ARCO's emission control-diesel fuel (EC-D, 7-ppmw sulfur) emitted 91–99% less PM, compared with trucks fueled by California diesel fuel (121-ppmw sulfur) and with no exhaust aftertreatment devices. These trucks have been operating reliably for more than 5 months, with an accumulation of ~50,000 mi per truck.<sup>301–303</sup> A continuously regenerating DPF reduced PM number counts by 1–2 orders of magnitude, as well as substantially reducing mass emissions.<sup>278</sup> In European field tests, DPFs have demonstrated highly efficient PM control and promising durability, when operated with ultra-low sulfur fuel.<sup>304</sup>

California's diesel risk reduction plan intends to reduce public exposure to DPM by retrofitting both on- and off-road diesel engines with high efficiency DPFs.<sup>277</sup> Worldwide, more than 10,000 buses and trucks have been equipped with passive high-efficiency DPFs, with some vehicles accumulating more than 300,000 miles.<sup>281</sup> Development and demonstration of DPFs for on- and off-road sources are underway in many countries, including

Sweden (Clean Cities Program), Switzerland,<sup>282,283</sup> Germany, Great Britain, Finland, France, South Korea, Taiwan, and the United States.<sup>9,281,284,304</sup>

In California, diesel transit buses are required to be retrofitted with PM aftertreatment devices demonstrating greater than 85% efficiency, beginning in 2003. Approximately 4500 diesel transit buses are expected to be retrofitted. In addition, a \$50 million program has been initiated to purchase new natural gas and diesel school buses and to retrofit existing diesel buses.<sup>285,289</sup>

Another alternative on the horizon is the hybrid electric engine system, which uses an electric drive, typically with a diesel internal combustion engine and a traction battery. Hybrids yield lower NO<sub>x</sub> and PM emissions and higher fuel economy. NO<sub>x</sub> and PM reductions of 25% have been shown with "off-the-shelf" diesel engines,<sup>305</sup> and larger reductions are possible with further optimization because the hybrid engine operates in a more tightly controlled range, compared with a conventional diesel engine.

### Fuels

Reducing the sulfur content of diesel fuels contributes directly to the reduction of SO<sub>2</sub> and PM emissions. Reducing fuel aromatic content reduces PM and NO<sub>x</sub> emissions. In California, due to required fuel sulfur reductions to date, SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> emissions have been reduced by ~95%. Total PM emissions (soluble organic compounds, BC, nitrates, sulfates, water, and ash) have been reduced by ~25% and NO<sub>x</sub> emissions have been reduced by ~7%, due to California's diesel fuel regulations.<sup>306</sup>

All of the advanced NO<sub>x</sub> and PM control technologies have the potential to produce particulate SO<sub>4</sub><sup>2-</sup>. EPA estimates that PM SO<sub>4</sub><sup>2-</sup> will exceed the total PM emission standard unless diesel fuel sulfur levels are at or below 15 ppmw.<sup>275</sup> Furthermore, fuel with sulfur levels higher than 15 ppmw significantly increases the chances of particulate filter failure and decreased efficiency of NO<sub>x</sub> after-engine devices. EPA's position is supported by the experience of Sweden and other European countries, where failure rates for PM control technology were higher with 50-ppmw sulfur fuel than with <10-ppmw sulfur fuel. EPA has not accepted the refining industry's limited evidence of the robustness of the PM control technology with fuel sulfur as high as 30-ppmw. DOE/EMA/MECA<sup>292</sup> determined that the desired emission reductions can only be realized with sulfur levels <15 ppmw. The EU's Conference of Ministers of Transport<sup>262</sup> concluded that "the successful achievement of 2008 EURO (NO<sub>x</sub>) standards for heavy-duty diesel vehicles is much more likely with the introduction of near zero sulphur fuels [<10 ppmw] than it is with the use of 50 ppmw sulphur diesel."

Beginning June 1, 2006, U.S. refiners must produce

highway diesel fuel that meets a maximum sulfur standard of 15 ppmw. All 2007 and later model year diesel-fueled vehicles must operate with this new low-sulfur diesel fuel.<sup>275</sup> The Texas Natural Resource Conservation Commission has adopted a maximum sulfur standard of 15 ppmw, applicable to all motor vehicle and off-road equipment diesel fuel sold in many counties, effective June 1, 2006.<sup>261</sup> California's Risk Reduction Plan calls for the adoption of a maximum sulfur standard of 15 ppmw for ARB Diesel in 2001. This standard would also be effective by June 1, 2006. The ARB Diesel applicability will be extended to stationary and other diesel engines with the adoption of airborne toxic control measures for nonvehicular sources.<sup>277</sup>

EU countries will limit sulfur in diesel fuel to 50 ppmw by 2005. The United Kingdom made a rapid conversion to a 50-ppmw sulfur cap for diesel fuel in 1999 by offering tax incentives to offset higher production costs. Some refinery production in that country is at levels well below 50 ppmw. Germany plans to introduce a 10-ppmw sulfur cap for diesel fuel by 2003, also with tax incentives, and is trying to revise the European Commission's 50-ppmw specification to 10 ppmw. With the help of a large incentive, Sweden introduced 10-ppmw sulfur fuel (Class I Swedish Diesel) into city areas in 1991. By 1999, more than 90% of the highway diesel fuel sold met the 10-ppmw sulfur maximum and other specifications (including a 5% by volume aromatics maximum) of the Class I Swedish Diesel specifications.<sup>275</sup>

Economic conditions in Asia and Africa will probably delay their adoption of ultra-low sulfur standards. A survey of 23 Asian and African countries showed little progress toward the North American and European 500-ppmw sulfur caps.<sup>307</sup> Hong Kong introduced a large tax incentive to promote 50-ppmw sulfur diesel over 500-ppmw diesel in July 2000, and Australia will introduce a tax incentive to reduce sulfur from the national average of 1300 ppmw.

In the United States, refiners should be able to meet the 15-ppmw specification with extensions of the same hydrotreating process they are using to meet the current 500-ppmw standard. Conventional hydrotreating combines hydrogen with the distillate at moderate pressures and temperatures and passes the mixture through a fixed bed of catalyst. Extensions of this process will likely involve a second stage of conventional hydrotreating with an additional reactor and hydrogen, sulfur recovery, and amine plant and sour-water scrubbing capacities. Two-stage hydrotreating is being used in Sweden to produce Swedish Class I diesel fuel, while United Kingdom refiners are meeting the 50-ppmw specification with one-stage hydrotreating. European diesel fuel is produced

from a lighter distillate fraction than is used for U.S. diesel fuel, and refining challenges will differ. Hydrotreating for desulfurization is more effective with lighter distillates and lower aromaticity.<sup>275</sup> The diesel fuels that are more difficult to desulfurize could be subjected to a more intense hydrotreating that might result in some improvements in aromatic content and cetane number. Severe hydrotreating reduces trace components containing nitrogen and oxygen that provide a natural lubricity in the fuel. This reduced lubricity can result in excessive engine wear, but it can be counteracted by blending in a very high lubricity biodiesel or a lubricating additive. Some loss in diesel volume may occur from reducing the sulfur content of diesel.

Early introduction of new and retrofitted fleet vehicles and equipment with sulfur-sensitive control technologies can be accommodated in California with the diesel fuel currently being produced. Of the 8–9 million gallons average ARB diesel production per day (based on 1999 consumption data),<sup>308</sup> more than 1 million gallons a day would meet a 15-ppmw sulfur standard. Approximately another million gallons a day can be produced upon demand. This fuel can be purchased from two or more California refiners.<sup>309</sup> To prevent blending and contamination in the distribution system, the fuel must be trucked from a refinery to the user's storage tank.

Reformulated and alternative diesel fuels have shown promise for achieving significant reductions in PM and NO<sub>x</sub> emissions. In addition to very low sulfur content, all of these fuels have relatively low density, with low aromatic and PAH contents. Alternative diesel fuels generally contain more than trace amounts of oxygenated fuel constituents or are emulsified with water. ARCO's EC-D fuel is a reformulated diesel fuel with higher aromatic and PAH contents than Swedish Class I diesel fuel. In emission tests of buses and trucks, EC-D fuel demonstrated emission reductions of PM, NO<sub>x</sub>, CO, and HC, relative to ARB diesel. Used in combination with catalyzed particulate filters, average emission reductions of more than 95% have been demonstrated for PM, along with 15–20% for NO<sub>x</sub> and 90% or more for CO and HC.<sup>258,301</sup>

Synthetic diesel fuel, with nearly zero sulfur and aromatic contents, is the cleanest burning of the reformulated diesel fuels. The fuel is produced by the gas-to-liquid chemical conversion process known as Fischer Tropsch (FT). First, sulfur is removed from natural gas. The natural gas is then reformed with air, producing a nitrogen-diluted synthesis gas containing mostly CO and hydrogen. A cobalt-based catalyst is used to reassemble the synthesis gas molecules into highly saturated synthetic oil and byproduct water. Laboratory engine and truck chassis dynamometer emission testing have demonstrated

average emission reductions of 26 and 24% for PM, 4 and 12% for NO<sub>x</sub>, 20 and 40% for HC, and 36 and 18% for CO, respectively, for FT diesel over ARB Diesel.<sup>258</sup>

Microemulsions of water or ethanol in diesel fuel have been shown to reduce both PM and NO<sub>x</sub> emissions through rapid vaporization of the emulsified droplets. These microexplosions break fuel droplets into smaller droplets, resulting in more complete vaporization and turbulent mixing and, consequently, more complete combustion of the fuel. The vaporization of the emulsified droplets also lowers peak combustion temperatures, thereby reducing NO<sub>x</sub> formation. Enhanced fuel atomization also reduces BC formation.

Lubrizol's PuriNO<sub>x</sub> is a microemulsion of ~20% water in diesel fuel, with less than 1% surfactants and other additives.<sup>310</sup> Following California's Interim Procedure for Verification of Emission Reductions for Alternative Diesel Fuels, emission reductions of 62.9% for PM and 14% for NO<sub>x</sub> were verified for a PuriNO<sub>x</sub> emulsion of water in a commercial California diesel fuel. Verification is based on the results of a series of laboratory engine emission tests, and the emission reductions are relative to the performance of a 10% aromatic ARB diesel reference fuel. The testing also showed no net increase in measured toxic emissions from PuriNO<sub>x</sub>.<sup>311,312</sup>

Biodiesel is a monoalkyl ester-based oxygenated fuel made from vegetable oil or animal fats. It contains 11% oxygen by weight and no sulfur or aromatic compounds. Otherwise, it has properties similar to petroleum-based diesel fuel and can be blended into conventional diesel fuel at any ratio. Neat biodiesel (B100) has been classified as an alternative fuel by the U.S. Department of Energy and the U.S. Department of Transportation. The use of B100 may reduce diesel engine emissions of PM by 30%, HC by 93%, and CO by 50% over conventional diesel fuel; however, its use tends to increase NO<sub>x</sub> emissions by 13%. Based on Ames mutagenicity studies, emissions from the use of B100 may provide a 90% reduction in cancer risk, compared to emissions from conventional diesel fuel. B20, a blend of 20% biodiesel with conventional diesel, is the most common biodiesel blend. B20 is useful for reducing PM, HC, and CO emissions and has a nearly neutral effect on NO<sub>x</sub> emissions.<sup>258</sup>

#### Alternatives to Diesel

Alternatives to diesel involve a different fuel and modifications to or substitutions for the compression ignition engine cycle. Methanol, natural gas (both compressed and liquefied), and hydrogen are considered alternative fuels.<sup>313</sup> Natural gas is a gaseous fuel composed primarily of CH<sub>4</sub>, with smaller amounts of ethane, propane, N, He, CO<sub>2</sub>, H<sub>2</sub>S, and water vapor. The gas is withdrawn from

wells, often in conjunction with oil production, and treated to remove water, certain HCs,  $H_2S$ , and other compounds. It is transported via pipeline and used to heat homes and offices, power electrical generators, and fuel stationary engines.<sup>314</sup> Natural gas has a higher hydrogen-to-carbon ratio than other hydrocarbon fuels, so its combustion typically produces less  $CO_2$  per unit of energy. However,  $CH_4$  itself is a potent greenhouse gas, so increased fugitive and other emissions of  $CH_4$ , resulting from increased use of natural gas, can offset the reduced exhaust  $CO_2$ .<sup>315</sup> Natural gas engine HC emissions are similar to the composition of the fuel.<sup>315</sup> Methane is relatively inert as a contributor to ozone formation, so it is often excluded from emission standards.<sup>315</sup>

Natural gas is not as convenient to use in vehicles as are liquid fuels. It must be either compressed to high pressures (compressed natural gas or CNG) or chilled and maintained at low temperatures for use as a liquid (liquefied natural gas or LNG). Current working pressures for CNG are about 2500–3500 psi, which are practical for cars, buses, and trucks. LNG must be cooled to less than  $-327.2^\circ F$  ( $-164^\circ C$ ) and is practical for locomotives. Although natural gas is available in most locations, due to the extensive pipeline network, preparing it for vehicle use requires compression or refrigeration equipment.<sup>316</sup>

Natural gas, due to its  $CH_4$  content, has a very high octane rating<sup>2</sup> and, therefore, does not readily ignite in diesel engines. Although Detroit Diesel Corporation (DDC) developed a direct-injection natural-gas-fueled version of its two-stroke diesel engine during the early 1990s,<sup>315</sup> most heavy-duty natural gas engines certified today use a spark-ignition, four-stroke cycle.<sup>317</sup> As an exception, Cummins and Westport Innovations intend to develop and market engines with high-pressure direct injection (HPDI) of natural gas. Natural gas is injected directly into the engine's combustion chamber with a small injection of diesel fuel as the ignition source. Since no throttle is needed and the diesel engine's high compression ratio is retained, these engines keep the characteristically high diesel fuel efficiency while operating on natural gas.<sup>318</sup> Westport Innovations has recently certified its HPDI system on a 14.9-L engine.<sup>319</sup>

Recent and ongoing tests of natural gas-fueled transit buses and comparable diesel-fueled buses demonstrate that natural gas engine emissions can be much lower than diesel engine emissions. One set of tests showed a natural gas bus emitted 8.7 g/mi  $NO_x$  and 0.1 g/mi PM, versus 20.6 g/mi  $NO_x$  and 0.38 g/mi PM for a comparable diesel bus. The natural gas bus consumed 17.8% more fuel, on an energy content basis, than the diesel bus consumed.<sup>320</sup> Some natural gas engines are not so clean. Analysis of transit bus emissions collected between 1992 and 1995 indicated statistically significant lower PM emissions, but

insignificant  $NO_x$  reductions, compared with comparable diesel buses.<sup>321</sup> An ARB staff survey of recently published test data shows that a heavy-duty natural gas engine can be categorized into one of three modes, depending on whether its  $NO_x$  emissions are low, intermediate, or high, relative to the sample population.<sup>322</sup> These data imply that natural gas-fueled engines have the capacity for greatly reduced emissions relative to diesel engines, but this cleanliness does not come automatically and requires careful engineering and maintenance to achieve.

Although the natural gas engine's PM emissions are lower than those of diesel engines, their composition, size, and possibly their effects on human health may be different. A test program<sup>323</sup> to quantify differences in natural gas versus diesel engines in terms of the number, size, and toxicity of PM emissions is ongoing in California, with results expected in the fall of 2001. Natural gas engines are becoming more widely used for transportation. The estimated number of heavy-duty CNG vehicles in the United States increased from 9874 in 1996 to 19,607 in 2000, and the number of heavy-duty LNG vehicles rose from 536 to 1564 during the same time period.<sup>324</sup> Use of CNG as a vehicle fuel has also increased, from 16,823,000 gasoline-equivalent gallons in 1992 to 104,501,000 in 2000. In the same time period, LNG consumption increased from 585,000 gasoline-equivalent gallons to 7,460,000.<sup>324</sup>

Liquefied petroleum gas (LPG), a mixture of propane/propylene, butane/butylene, and potentially higher HCs, is a byproduct of natural gas processing and crude oil refining.<sup>325</sup> The atmospheric boiling point of LPG is  $-43.73^\circ F$ <sup>315</sup> and it must be stored under moderate pressure ( $\sim 200$  psi)<sup>326</sup> to remain liquid. This pressure is significantly lower than that required for CNG storage. The volumetric energy content of LPG is about two-thirds that of diesel fuel, so it requires more volume to store enough fuel for comparable range. Although lower than natural gas, LPG's octane rating is still high,<sup>315</sup> making its use more suited to spark-ignition engines than to diesel engines. LPG consumption for vehicle use increased from  $\sim 208$  million gasoline-equivalent gallons in 1992 to almost 250 million in 2000. There were  $\sim 57,000$  heavy-duty LPG-fueled vehicles in the United States in 2000, up slightly from  $\sim 53,000$  in 1996.<sup>324</sup> LPG is also frequently used in stationary engines for applications that might otherwise make use of diesel fuels. Recent California certification of stationary engines using LPG shows low levels of  $HC + NO_x$  emissions relative to the certification standards.<sup>327-330</sup>

Methanol, sometimes called "wood alcohol," is made from natural gas through the use of steam reformation and a catalyst.<sup>313</sup> It has  $\sim 50\%$  of the volumetric energy

content of diesel fuel<sup>2</sup> and burns with a low luminosity flame, which is considered a potential safety hazard. Methanol combustion creates little PM. Its high latent heat of vaporization cools the intake charge, which reduces NO<sub>x</sub> formation temperatures, but also makes compression ignition more difficult. Its low adiabatic flame temperature also reduces NO<sub>x</sub> formation. The presence of oxygen in the fuel leads to the production of aldehydes (such as formaldehyde) during partial combustion,<sup>315</sup> which can be reduced with an oxidation catalyst.

In the early 1990s, DDC manufactured and sold a version of its two-stroke diesel engine that operated on methanol. The engine used glow plugs to ensure ignition during low load and starting conditions and high-flow injectors to compensate for the reduced energy content of the fuel. A catalytic converter reduced unburned methanol and formaldehyde emissions.<sup>315</sup> In the 1990s, the Los Angeles County Metropolitan Transit Authority (LACMTA, formerly the Los Angeles Regional Transit District) built up a fleet of ~330 methanol-fueled urban transit buses powered by this DDC engine. LACMTA's experience was discouraging, due to poor durability and the need for excessively frequent overhauls. LACMTA subsequently converted many of the methanol buses to use ethanol as a demonstration of that fuel.<sup>331</sup> As of August 2000, LACMTA had no methanol- or ethanol-fueled buses in service.<sup>332</sup>

Currently, no major U.S. heavy-duty engine manufacturer produces methanol-fueled engines as an alternative to its diesel-fueled product line. There were only ~200 heavy-duty methanol-fueled (pure methanol or M100) vehicles estimated to be in use in the United States in the year 2000, and those were all used by state and local governments.<sup>324</sup> Methanol fuel use has decreased from ~2.5 million gasoline-equivalent gallons in 1992 to only ~0.45 million in 2000.<sup>324</sup> Although not presently favored as a diesel fuel replacement, methanol and possibly ethanol may prove to be viable fuels for use with future fuel cell-powered vehicles.

Dimethyl ether (DME) is generally produced by dehydration of methanol, although other processes have been developed in recent years. Natural gas is the most common raw material for DME production, although DME can also be produced from coal. DME is nontoxic and is commonly used as an aerosol propellant. A gas at atmospheric conditions, DME requires handling and storage methods and equipment similar to LPG (i.e., low-pressure storage vessels). DME has a high cetane rating (nearly 60, compared with diesel fuel ratings in the low to mid-50s). This means that it can be readily used in diesel engines and yields fuel economies comparable to conventionally fueled diesel engines.<sup>333</sup> DME is not included on the U.S.

Department of Energy's list of alternative fuels,<sup>334</sup> perhaps because it is relatively new and its use is not widespread. DME has emission benefits, with PM emissions of 0.03 g/bhp-hr and NO<sub>x</sub> + HC emissions of less than 2.4 g/bhp-hr.<sup>333</sup>

NKK Corporation of Japan has built a pilot facility to produce 5 tons of DME per day in Japan using synthesis gas from coal.<sup>335</sup> BP Amoco, Indian Oil Corp. Ltd., and Gas Authority of India Ltd. have agreed to set up a \$500 million DME plant in the Middle East, scheduled to be in production by 2005.<sup>336</sup> Asian countries are primarily interested in DME as an economical alternative for transporting natural gas for use in power generation.<sup>337</sup>

### Fuel Cells for Heavy-Duty Transportation Applications

Fuel cells have the potential to replace diesel engines in truck, bus, and certain off-road applications. For example, an Allis-Chalmers farm tractor was demonstrated in 1959 utilizing alkaline fuel cells. Appleby<sup>338</sup> discusses various types of fuel cells for transportation. Typically, a fuel cell generates energy from hydrogen and oxygen; oxygen is freely available from the atmosphere, and hydrogen is generated from the electrolysis of water or by reducing HC-based fuels with a reforming mechanism.<sup>339,340</sup> In transportation applications, hydrogen can be carried on-board, reformed from methanol using a low-temperature reformer or reformed from higher HC fuels using high-temperature reformers.<sup>341</sup> Compared with internal combustion engines, emission benefits to human health, visibility, soil and water contamination, and the global radiation balance would be achieved by the use of hydrogen fuel cells for transportation.<sup>342,343</sup>

Transit buses will most likely be the first widespread commercial use of fuel cells. By 2003, fuel cells buses will be operated worldwide—in California, through the California Fuel Cell Partnership; in ten western European cities, through the Clean Urban Transport for Europe program; in Iceland, through the ECTOS project; and in five developing countries (Brazil, Mexico, China, India, and Egypt), in conjunction with the United Nations Development Fund/Global Environment Fund program.<sup>344,345</sup> California's transit bus regulations require large fleets that are predominantly diesel-fueled to demonstrate three zero-emission buses starting in July 2003.<sup>276</sup> Fuel cells are one of the three alternative technologies that qualify.

Buses are an ideal first application for fuel cells in transportation. They operate on set routes; maintenance is well-documented; the vehicles are driven under a wide range of conditions (e.g., temperature, topography); they have high visibility, so the public becomes personally acquainted with the new technology; and they are centrally

fueled, eliminating the need for a widespread fuel infrastructure. The first commercial fuel cell buses will not use an on-board reformer. Fuel cells, like most emerging technologies, are expensive for the first production units. The buses are publicly funded, so the initial cost is absorbed by society, but as the number of units produced increases, costs are expected to decrease.

Fuel cells are not yet cost-effective as the primary power source for diesel trucks. However, a fuel cell auxiliary power unit (APU) that complements the main diesel engine is much smaller, less expensive, and requires less fuel storage than a full-size fuel cell engine.<sup>341</sup> Given the large amount of time that diesel trucks spend idling (~6 hr/day for 303 days per year),<sup>346</sup> the fuel cell APU is a cost-efficient and environmentally benign means of meeting power needs for that portion of the driving cycle. Tests with a 1–5 kW fuel cell APU for diesel trucks show that the estimated savings per year include \$3127 for fuel (based on \$1.72/gallon), \$727 for preventative maintenance, and \$1454 for engine overhauls.<sup>341</sup> The reduced idling time also yields emission reductions of NO<sub>x</sub> (0.2–0.7 tons/year) and CO<sub>2</sub> (8–24 tons/year). For diesel engines, the elimination of DPM will lead to significant air quality benefits because the fuel cell emits only water vapor (if a reformer is used, emissions are likely to be near zero).

## CONCLUSIONS

Diesel engines are in widespread use throughout the world, and their numbers and worldwide diesel fuel consumption will probably grow for the foreseeable future. They are used for many on- and off-road applications, including passenger cars, line-haul trucks, buses, locomotives, farm and construction equipment, water pumps, and portable and peaking electrical generation. The engines are durable, lasting for many decades, and provide more energy per unit of fuel than gasoline engines provide. A large infrastructure of diesel fuel production and distribution exists that will take many decades to modify or replace.

Diesel engines and their fueling infrastructure adversely affect all aspects of the natural environment—land, water, and air. Diesel PM has been associated with lung cancer and short-term respiratory ailments, such as asthma, in occupational and general population epidemiologic studies. Small particles, which are both directly emitted from diesel engines and formed from gaseous emissions, can lead to premature death and major respiratory problems. Tumors and cell damage have been found in animals exposed to high DPM concentrations, although these concentrations are typically much larger than those found in normal atmospheres. The high BC content of diesel exhaust makes it an efficient light absorber,

contributing significantly to urban and regional haze, as well as to increases in the earth's radiation balance. Diesel particles soil bridges, tunnels, and other surfaces, with high associated clean-up costs. The NO<sub>x</sub> emissions from diesel (as well as gasoline) engines are important O<sub>3</sub> and PM precursors. Carbon monoxide and HC emissions from diesel engines are much lower than those from gasoline engines, but the HC compounds tend to be heavier and have a higher propensity to create secondary organic particles that can affect health and haze. Although CO<sub>2</sub> emissions are lower per unit of diesel fuel burned than with gasoline, they are still an important fraction of the total emissions of this greenhouse gas. Fuel spills and leaks contribute to the pollution load for land, water, and air; are dangerous; and are costly to clean up.

Significant progress has been made in reducing diesel emissions through improved engine design and fuel reformulation. These advances have often improved fuel economy, thereby offsetting some of the costs of new technology. Since 1980, up to 90% reductions in DPM and NO<sub>x</sub> emissions have been achieved with fuel injection rate shaping and combustion system refinements. However, current controls involve trade-offs between DPM and NO<sub>x</sub> emissions and may result in decreased fuel economy.

It is expected that high-efficiency aftertreatment devices will effectively reduce emissions even further, but these devices require fuels with sulfur contents <15 ppmw (possibly as low as 5 ppmw). NO<sub>x</sub> emissions can also be reduced using a lean NO<sub>x</sub> catalyst, NO<sub>x</sub> adsorber, or selective catalytic reduction. Diesel particle filters have shown more than 90% reduction in PM emissions and long life spans when used with low-sulfur fuels. Another promising new heavy-duty technology being demonstrated is a hybrid-electric engine system that can reduce DPM and NO<sub>x</sub> emissions by 25% or more, compared with the current diesel engine.

Outside of the United States, diesel engines represent a much higher proportion of the in-use vehicle fleet, particularly for light-duty vehicles. In the United States, increased diesel penetration has been proposed as one way to reduce CO<sub>2</sub> emissions and associated global climate change impacts from the transportation sector. However, DPM also negatively affects the global radiation balance, and a better understanding is needed of the comparative impacts of diesel, gasoline, and alternative fuels.

The most successful demonstrated alternative fuel for use in "diesel" engines is natural gas. Many newer urban transit buses now utilize CNG, and a limited fueling network for liquefied natural gas is being developed to support intrastate trucks. However, the lack of a widespread fueling infrastructure limits the application of alternative

fuels to interstate trucking and off-road engines. It is anticipated that the use of FT and DME will increase as the technologies to remotely manufacture these fuels become cost competitive and the infrastructure to support these fuels improves.

Fuel cells that convert hydrogen and oxygen to energy and water should begin to replace or complement diesel engines within this decade. Hydrogen can be economically generated from renewable sources, such as wind, solar, or geothermal sources. It can also be produced by reforming currently available HC fuels, such as gasoline, diesel, FT, natural gas, methanol, and ethanol. Fuel cell engines are currently practical for city buses that have a central fueling facility where hydrogen can be provided. Fuel-cell auxiliary power units with 1–5 kW capacity have been shown to reduce emissions and improve fuel economy, when used during idling of diesel engines in long-haul trucks. Fuel cell costs are expected to drop as manufacturing methods and sales volumes improve.

#### RECOMMENDATIONS

Environmental regulations are needed to stimulate further progress in reducing diesel and other vehicle emissions. Most of the technological advances that are enjoyed today would not exist if the diesel industry had not been challenged by more stringent emissions standards. Regulations should be based on good science that is practical to implement, inclusive of public and industry concerns, and can be concluded on a reasonable time schedule. Currently promulgated regulations for on-road vehicles will provide considerable improvements to air quality on urban, regional, and global scales. These regulations, which will require the use of exhaust aftertreatment, need to be extended to off-road diesel applications. This can only happen if ultra-low sulfur diesel fuel is available for off-road engines, as it will be for on-road engines in the United States beginning in 2006.

Additional monitoring of on-road emission performance is needed. Diesel certification tests are insufficient to understand how emissions change with variable driving conditions. They do not identify engines that are operating outside their range of specification. On-board, dynamometer, and remote sensing systems exist to take these measurements; roadside and periodic smoke inspection programs already exist in some areas; and approaches such as periodic inspection and maintenance programs (similar to “Smog Check” for light-duty vehicles) are being considered and developed. These and other similar technologies and methods all need to be incorporated into a comprehensive program to monitor actual emissions and

enforce emissions standards for individual vehicles.

Programs that introduce alternative fuels and potentially zero-emission technologies, such as fuel cell engines, as replacements for diesels need to be continued and enhanced. The cost-effectiveness of mass production, hydrogen production, and fuel distribution will not be realized until a critical mass of such vehicles exists. Practical problems with vehicle operation and maintenance will be identified and solved by these programs. This will increase public acceptance of hydrogen and other fuels that are perceived to be, but actually are not, more dangerous than the gasoline and diesel fuels in current use.

Better methods and data are needed to quantify the environmental trade-offs between diesel, gasoline, alternative fuels, and possible control technologies. This information will help ensure that decisions on future fuel strategies, fuel infrastructure investment, and regulation result in more environmentally benign modes of transport and power generation and protection for the health of future generations.

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