A Perspective on the Potential Development of Environmentally Acceptable Light-duty Diesel Vehicles

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Between 1979 and 1985, an international technical focus was placed upon potential human health effects associated with exposure to diesel emissions. A substantial data base was developed on the composition of diesel emissions; the fate of these emissions in the atmosphere; and the effects of whole particles and their chemical constituents on microorganisms, cells, and animals. Since that time, a number of significant developments have been made in diesel engine technology that require a new look at the future acceptability of introducing significant numbers of light-duty diesel automobiles into the European and American markets. Significant engineering improvements have been made in engine design, catalysts, and traps. As a result, particle emissions and particle associated organic emissions have been reduced by about 10 and 30 times, respectively, during the past 10 years. Research studies to help assess the environmental acceptability of these fuel-efficient engines include the development of an emissions data base for current and advanced diesel engines, the effect of diesel emissions on urban ozone formation and atmospheric particle concentrations, the effect of fuel composition, e.g., lower sulfur and additives on emissions, animal inhalation toxicology studies, and fundamental molecular biology studies. — Environ Health Perspect 102(Suppl 4):25–30 (1994).

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Introduction

The presence of polycyclic aromatic hydrocarbons (PAH), compounds that are suspected to cause cancer, was reported first in vehicle exhaust in the 1950s and early 1960s (1–4). In the late 1970s and 1980s, there was a substantial international effort to chemically characterize the PAH in gasoline and diesel vehicle exhaust and to test the effect of these emissions on microorganisms, cells, and animals. This effort became concentrated on diesel vehicles because of much higher PAH and particulate emissions (4), which were found to cause cancer in some laboratory animals (5). This process culminated in the classification of diesel particulate as a probable carcinogen (4) and in regulations to reduce diesel particulate emissions in the United States (6).

Over the last 10 to 15 years, there has been an equally substantial effort to lower diesel emissions. It began with detailed mapping of diesel engine operation to define operating modes with low emissions and progressed to improved engine control to maintain the engine in those low emission modes (7). This trend continues today (8,9). More recently, the effort has resulted in modification of the engines, which has led to still lower emissions. The fuel injection process, for example, was modified for direct injection engines to distribute the fuel better so that it could be burned more completely. Together, such engine and engine control modifications have already lowered particulate emissions by about a factor of 3 (10). Additional modifications, which are planned for the near future, such as higher pressure fuel injection and individual control of each injector are expected to lower emissions further.

The effort to lower diesel emissions has also led to the introduction of exhaust treatment devices, namely catalysts and particle traps. Today, the catalysts that are used on most new gasoline vehicles are capable of removing roughly 95% of the hydrocarbon (HC) and nitrogen oxide (NOx) emissions and about 90% of the carbon monoxide (CO) emissions (11). Catalysts are capable of removing about 90% of the organic and CO emissions for diesel vehicles, but they cannot lower the NOx emissions because diesel exhaust has excess oxygen. However, the catalysts that have been used for the last few years on diesel vehicles are less active than those used on gasoline vehicles; they typically remove less than 50% of the organic emissions (12). More active catalysts cannot be used because the sulfur present at high levels in commercial diesel fuel would be converted to particulate sulfate. Sulfur levels in diesel fuel are being reduced, both in the United States and Europe.

While particle traps are effective and have been tested on diesel engines extensively for more than 10 years, their commercial introduction has been very slow because they are difficult to control. The problem is regularly burning the collected soot without restricting the engine exhaust flow or melting the trap. Solutions based on using catalytic metallic compounds in the fuel have been tested successfully (13,14), but not implemented fully, apparently because of control difficulties and concern over the emissions of metallic aerosol.

Using a full range of the emission control technology developed for diesel vehicles over the past 10 years, the particle emissions and the particle-associated organic emissions can be reduced by about 10 and 30 times, respectively. The success of particulate emission control efforts raises the issue of whether the environmental acceptability of diesel vehicles should be judged only on their potentially carcinogenic particulate emissions or whether it should be judged by its overall effect, including other problems such as urban ozone and global warming.

Public Health Risk from Diesel Particulate Matter

Diesel particulate matter (PM) contains many PAH and their chemical derivatives which are suspected carcinogens (4). Most
of these compounds have relatively low vapor pressure and condense along with other organic material on carbonaceous core particles before they leave the exhaust pipe. Together, the condensed organic material and the dry soot make up the PM at ambient temperatures. After collecting the PM, the PAH and derivatives can be extracted with solvents, so they are considered part of the soluble organic fraction (SOF).

In 1989, inhaled diesel exhaust was classified as probably carcinogenic (4). This classification means that, while there is a positive association between diesel exhaust and cancer in humans, chance or bias could be responsible. At the same time, it requires a positive association between cancer in animals and diesel exhaust that chance or bias cannot negate. A very brief summary of the health risk of inhaling diesel PM is included here for completeness. The interested reader is referred to an excellent review elsewhere (4).

"Definitive evidence of carcinogenicity in humans can only be provided by epidemiological studies" (4). There have been several epidemiological studies of workers exposed to high levels of diesel exhaust; they show associations between exposure and lung cancer, but the associations are too weak to be free of possible bias (4). One major problem stems from cigarette smoking, which causes the overwhelming majority of the lung cancers observed in such studies (4).

The largest of these studies was done for U.S. railroad workers who started railroad work between 1939 and 1949 (4,15–17). Using only exposure data from the 1980s, this study reconstructed the workers’ diesel exhaust exposure from their careers and classified them according to that exposure. Classes with the greatest exposure had roughly five times greater exposure than those with the lowest exposure, but they had 1.45 times higher risk of developing lung cancer. This higher risk was barely significant with the 95% confidence interval, which ranged from a relative risk of 1.11 to 1.89. Classes with intermediate exposures had 1.2 times higher risk with a 95% confidence interval of 1.1 to 1.3. The limited significance of this result may be confounded by different smoking habits of the various classes, which was not considered (4).

The risks for the railroad workers developing other cancers were also studied, but they were nearly independent of the diesel exposure classes. For example, classes with the largest exposure had 1.03 times greater risk of developing bladder cancer than those with the lowest exposure. Here the 95% confidence interval ranged from a relative risk of 0.88 to 1.19. Bladder problems could result from inhalation of diesel exhaust because particulate matter deposited in the lung is normally cleared from the lung and swallowed. In other studies, people with bladder cancer appeared to have been associated with diesel exhaust more regularly than those without (4).

After these human population studies, the case for a health risk from diesel exhaust inhalation rests on animal studies (4). While several animal species have been exposed to diesel exhaust, only the rat develops lung cancer. There is no evidence that the exposed animals develop bladder cancer (4). Many studies have shown unequivocally that the rat develops lung tumors after many hours of daily exposure for at least 24 months to very high levels of diesel PM, above about 4 mg/m³. The high level of PM overloads the natural self-clearing mechanism so that diesel particles build up in the lung. No tumors were found in animals exposed to filtered diesel exhaust from which the PM was removed.

Similar studies show that rats develop lung cancer after exposure to inert PM, which consists of insoluble particles such as titanium oxide or carbon black that is free of organic material. In these tests, the incidence of lung tumors in rats exposed to inert PM was equivalent to that in rats exposed to unfiltered diesel exhaust (18–20). Apparently, inhalation of the suspected carcinogenic PAH on the diesel PM has no additional effect. In other words, the risk of inhaling diesel exhaust was the same as inhaling similar levels of other particulate matter. These results suggest that overloading the lung clearance mechanism and not the PM may be responsible for the lung tumors in rats (21). Atmospheric particulate levels are typically below 100 μg/m³ in the United States, or more than forty times lower than the 4 mg/m³ used in the above animal tests.

Recent Developments in Diesel Engines

Over the last 15 years, there has been a substantial effort to lower diesel emissions. Early diesel vehicles were designed for optimum fuel economy and performance. They provided approximately 15 to 30% better volumetric fuel efficiency miles per gallon of fuel, than similar gasoline engines of comparable power level. No one could be certain that they could maintain this advantage with a diesel designed for low emissions. The efforts to lower emissions began with detailed mapping of the diesel engine operating modes that provide lower emissions (6). Careful control of the fuel injection and the addition of exhaust gas recirculation (EGR) were necessary to meet the emission standards. For the lowest emissions, injection parameters and EGR rate must change as precise functions of vehicle and engine speeds, accelerator pedal position, inlet air temperature and pressure, and engine temperature (22).

In the early 1980s, the fuel injection and EGR on most diesel passenger vehicles were controlled by mechanical systems that could drift out of tune. More precise control of fuel injection and EGR was achieved with the introduction of electronic control units in the late 1980s (23,24). These electronic control units were equipped with sensors that monitored the engine operation and helped maintain low emissions throughout the vehicle’s life. The trend toward greater electronic control of diesel engines will probably continue over the next 5 to 10 years (8).

The precision of fuel injection has been improved substantially by the development of new fuel detergents and new fuel injectors that reduce injector fouling (25,26). In the early 1980s, the flow area of the injectors typically was oversized by nearly 40% to allow better engine operation over the lifetime of the vehicle as the inevitable buildup of the carbonaceous deposits gradually reduced the injection rate (25). New ashless detergents (25,26) and new designs for injectors are available to reduce the fouling and lower emissions greatly. The detergent composition is often proprietary; however, one detergent is a high molecular weight polyisobutylene amine functional (27). Using properly sized injectors that remain clean, the HC and particulate emissions can be reduced by roughly 25% compared to fouled injectors.

In addition, modifications to the seal between piston and cylinder wall have reduced the emission of engine lubricating oil substantially (28). In the 1980s, lubrication oil usually contributed to the dominant portion of the SOF of the PM (29). Sufficient oil must flow between the piston and cylinder wall to prevent wear, but little oil should be drawn into the combustion process. A number of improvements such as chromium faced piston rings (28), optimized ring design (30), control of ring and cylinder bore corrosion, in-cylinder distortion, and controlled surface finishes (24), and optimized honing patterns, have reduced the oil contribution to the PM greatly.

Together these engine and engine-control modifications have lowered PM emissions.

26 Environmental Health Perspectives
by a factor of 3, from 246 to 82 mg/km, and the SOF by a factor of 7, from 123 to 16 mg/km (Table 1). The base emission levels, 246 mg/km with 50% SOF, appear to be representative of diesel engines in the late 1970s and early 1980s and were cited by International Agency for Research on Cancer in 1989 when it classified diesel PM as a probable carcinogen. Table 1 shows the PM and SOF emissions for several diesel vehicles including many, but not all, of the improvements indicated above. Thus, this downward trend is expected to continue.

Exhaust Treatment Systems

In addition to modifying the engine, the effort to lower emissions has led to the development of catalysts and particle traps. Early attempts to use catalysts on diesel engines were limited by the buildup of PM in the catalyst (31). As the soot built up, it blocked the diffusion of the organic gases and oxygen to the catalytic surfaces, greatly reducing the rate of oxidation. In extreme cases, the soot could restrict the flow of exhaust gases from the engine. Engine modifications that lowered the particulate emissions can eliminate this catalyst plugging problem.

Catalysts are capable of removing about 90% of the organic emissions of a diesel vehicle (12). They can oxidize gaseous organic compounds, both the HC and the SOF that is gaseous in the catalyst, but not the dry, carbonaceous soot. Catalysts must be above 250 to 300°C to oxidize organics effectively. Typically, cold engines must operate for a few minutes for the catalysts to reach this temperature. During this cold start period, the light molecular weight HC with high vapor pressures are emitted from the vehicle uncat-
alyzed. However, the cold catalyst cools the gases emitted from the engine rapidly, causing the heavier HC present in diesel exhaust to condense on its walls. As the catalyst warms up and becomes active, the condensed organic material is oxidized. The result is that very little of the gaseous organic material escapes unoxidized (12).

To effectively remove the organic compounds, the catalyst must become very active at the lowest possible temperature. Unfortunately, the sulfur in the diesel fuel causes two problems. The sulfur emitted from the engine as sulfur dioxide is converted to sulfur trioxide or, at lower temperatures and in the presence of water, sulfuric acid, or sulfate. The acid emissions can cause environmental problems and add to the particulate mass emitted. Some catalyst formulations can oxidize the organics selectively while hardly oxidizing the sulfur dioxide. These catalysts are used today. They typically can remove about 50% of the SOF (12). However, if sulfur were not present, substantially more active catalysts could be used. They can remove about 90% of the SOF (12). This has been recognized, and diesel fuel sulfur in the United States will be reduced from about 2800 ppm in 1991 to less than 500 ppm starting in October 1993. Even at this level, the sulfate can contribute about 25 mg/km (40 mg/mi), assuming 25% conversion of the fuel sulfur to sulfate by the catalyst and 12.7 km/1 (30 mi/gal) fuel economy (Figure 1). This is about 50% of the U.S. 50 mg/km (80 mg/mi) PM standard.

The second, more fundamental problem is that sulfur raises the temperature at which a catalyst becomes active. It does this because sulfur dioxide is absorbed strongly by the catalyst at low temperatures, preventing the absorption of oxygen, which is needed to catalyze the organics. The sulfur dioxide begins to desorb at about 230°C, depending on the catalytic material. The U.S. Auto/Oil program found that lowering the gasoline sulfur level from 466 to 49 ppm lowered the HC emissions of 1989 model cars by more than 15% (32). Because the exhaust temperatures of diesel vehicles are lower than that of gasoline vehicles, one expects a greater reduction in HC emissions if the sulfur level in diesel fuel were reduced from 500 to 50 ppm. Catalysts could reduce the SOF by as much as 95%. In addition, the sulfate emissions would be reduced to a few mg/km. Today, fuel sulfur limits the application of catalysts to diesel vehicles like lead limited their application to gasoline vehicles in the 1970s.

Diesel particulate traps are capable of removing more than 80% of the particulate mass emissions (14). Traps remove the SOF and the dry, carbonaceous soot, while catalysts remove only the SOF. However, traps are substantially more difficult to operate on the vehicle than catalysts, and this has slowed their introduction on light-duty vehicles greatly. The problem occurs as the particulate material builds up in the trap; this causes the exhaust back pressure to increase, and higher back pressure reduces fuel economy and performance (15). Therefore, the collected particulate must be burned periodically to regenerate the vehicle’s performance. The regeneration can occur naturally when the engine is used at high speed and loads. However, for many applications, natural regeneration is too infrequent to maintain good vehicle performance.

Table 1. Particulate matter and soluble organic fraction for diesel engines without traps and catalysts.

| Vehicle       | PM, mg/km | SOF, mg/km | SOF/PM, % | References |
|---------------|-----------|------------|-----------|------------|
| Circa 1980, IDI | 246       | 124        | 50        | (4)        |
| 1983 1.6-L IDI | 200–250   | NA         | NA        | (41)       |
| 1983 1.6-L IDI turbocharged | 150–200 | NA         | NA        | (41)       |
| 1986 3.0-L IDI turbocharged, 1900 kg | 231 | 18.5 | 8 | (34) |
| 1987 1.6-L IDI 1200 kg | 122 | 27.5 | 22.5 | (34) |
| 1989 1.8-L IDI turbocharged, 1360 kg | 82 | 16 | 20 | (24) |

Abbreviations: PM, particulate matter; SOF, soluble organic fraction; IDI, indirect injection; NA, not analyzed. a Engine displacement. b Inertia weight used in the emission test.

Figure 1. Sulfate particulate emissions.
If regeneration is delayed, some of the SOF collected by a trap at low temperature can be volatilized later when it becomes hotter. The diesel PM in a trap will not ignite until its temperature is about 500 to 550 °C (13), thus all of the trapped SOF that becomes gaseous below 500 °C can be emitted. In addition, if regeneration is delayed, the direct-acting TA98 mutagenicity of the trapped PM can be greater than that of the directly emitted PM (33). This appears to be due to the reaction of the PM with the exhaust gases flowing through the trap to form nitrated PAH. Together these problems could eliminate any benefit of reduced biological activity provided by a trap.

A promising regeneration method is the use of fuel catalysts such as organic compounds containing iron or copper (14). These compounds are burned in the engine and then are emitted as finely dispersed metal particles that are collected in the trap and intermixed with the diesel particulate. They act as catalysts by reducing the temperature at which the PM will burn so that regeneration occurs more frequently. Their presence reduces the PM ignition temperature to below 300 °C (14), greatly reducing problems with vaporization or nitration of the trapped SOF (34). Very low concentrations of fuel catalyst, about 0.05 gm/L of fuel, are sufficient for regeneration. Over 95% of the metal used as fuel catalyst is collected in the trap. Tests show that some metals, such as calcium and copper can be purged from the traps during the high temperatures of regeneration (34). Great care must be taken to avoid release of the metal particles because they could cause environmental problems.

Together, exhaust treatment and engine modifications have lowered PM emissions by a factor of 10, from 246 to 26 mg/km, and the SOF by a factor of 40, from 123 to 3 mg/km (Table 2). Again, the base emission levels for this comparison is 246 mg/km with 50% SOF. Table 2 shows the PM and SOF emissions for several diesel vehicles, including some, but not all, of the improvements indicated above. This downward trend is expected to continue.

### Biological Activity of Emissions from Current Diesel Vehicles

To our knowledge, the biological activity of the PM emitted from current light-duty diesel vehicles, especially that from catalysts and traps, has not been determined completely. The Ames assays that have been conducted show that the TA98 mutagenic activity is decreased by between 50 and 90% (34,35). The PM from current diesel vehicles equipped with traps has fewer revertants/km than that from gasoline vehicles made in the early 1980s (Table 3). Limited supporting data from heavy-duty diesel vehicles shows that traps decreased the TA98–S9 activity of the SOF by 54 to 94% (36) and that oxidation catalysts decreased the TA98–S9 activity by about 65% (37).

### Future Work

Since the late 1970s, the primary health concern for diesel vehicles has been the emissions of PM and the PAH contained on the PM. This concern led to particulate emissions standards and efforts to reduce these emissions. As shown above, the PM and SOF emissions have been reduced by factors of 10 and 30, respectively. There is the potential for additional reductions, especially for vehicles with catalysts using very low sulfur fuel or with traps using fuel catalysts or other regeneration aids. The emissions from such systems have not been characterized fully to help establish the potential health effects, so more work must be done in this area.

At the same time, the health effects of inhaling diesel PM emissions remain uncertain, especially for low-emission vehicles. In the mid 1980s, chronic inhalation exposure of rats to their maximum tolerated dose of diesel PM was shown to cause lung tumors (4). However, by the early 1990s, it became clear that similar chronic exposure to inert PM such as titanium oxide also caused tumors in rats (18–20). In general, it has been found that chronic, maximum tolerated exposure of "more than half of all chemicals tested (both natural and synthetic) are carcinogens in rodents, and a high percentage of these carcinogens are not mutagens" (38). Maximum tolerated doses appear to simulate cell division (38), and there is growing evidence that induced cell division has a dominant role in carcinogenesis (39). This suggests that tests at maximum tolerated exposure provide little

### Table 2. Particulate matter and soluble organic fraction emissions for diesel engines with traps and catalysts.

| Vehicle | PM, mg/km | SOF, mg/km | Mutagenicity TA98 – S9 rev/m | Mutagenicity TA98 + S9 rev/m | References |
|---------|-----------|------------|-----------------------------|-----------------------------|------------|
| 1986 3.0-L ´ IDI turbocharged 1900 kg ´ trap and fuel cat | 31         | 3.4        | 12.5                        | (34)          |
| 1987 1.6-L ´ IDI 1200 kg ´ trap and fuel cat | 26         | 12         | 46.5                        | (34)          |
| 1987 3.0-L ´ IDI turbocharged trap and fuel cat | 8.75       | NA         | NA                          | (42)          |
| 1992 2.5-L ´ IDI catalyst | 120        | 12         | 10                          | (35)          |

Abbreviations: PM, particulate matter; SOF, soluble organic fraction; IDI, indirect injection; NA, not analyzed. * Engine displacement. ° Inertia weight in the emission test.

### Table 3. Mutagenicity of the soluble organic fraction from various diesel vehicles.

| Vehicle | PM, mg/km | SOF, mg/km | Mutagenicity TA98 – S9 rev/m | Mutagenicity TA98 + S9 rev/m | References |
|---------|-----------|------------|-----------------------------|-----------------------------|------------|
| Circa 1980 IDI diesel | 246        | 123        | 595                         | 240                         | (4)        |
| Circa 1980 gasoline with catalyst | 62         | 10         | 61                          | 180                         | (4)        |
| 1986 3.0-L ´ IDI diesel | 232 ´       | 019 ´      | 480 ´                       | 486 ´                       | (34)       |
| 1986 3.0-L ´ IDI diesel with trap and fuel cat | 31 ´       | 3 ´        | 67 ´                        | 39 ´                        |           |
| % Reduction | 87%        | 84%        | 86%                         | 92%                         | (34)       |
| 1987 1.6-L ´ IDI diesel | 123 ´       | 29 ´       | 521 ´                       | 422 ´                       | (34)       |
| 1987 1.6-L ´ IDI diesel with trap and fuel cat | 26 ´       | 12 ´       | 108 ´                       | 71 ´                        | (34)       |
| % Reduction | 79%        | 57%        | 79%                         | 83%                         | (35)       |
| 1992 IDI diesel | 81 ´       | NA         | 14 ´                        | NA                          | (35)       |
| 1992 IDI diesel with catalyst | 75 ´       | 6 ´        |                             | 60%                        |             |

Abbreviations: PM, particulate matter; SOF, soluble organic fraction; IDI, indirect injection; NA, not analyzed. * Engine displacement. ° Engine out. ° Tail-pipe.
information about low dose risk. More needs to be done to define the health risk of inhaling atmospheric levels of diesel exhaust.

If, as suspected, the health risk of inhaling diesel exhaust can be reduced substantially by lowering their PM and SOF emissions, then a wider view of the environmental acceptability of light-duty diesel vehicles becomes appropriate. Such a view should also consider local and regional levels of ozone, carbon monoxide, toxic compounds and PM, and global climate change.

The impact of diesel and reformulated gasoline vehicles appear similar, and changes to current uncertainties could easily affect their relative environmental standing (40). Currently available data shows that diesel vehicles emit less nonmethane organic compounds, carbon monoxide, and carbon dioxide than gasoline vehicles but more nitrogen oxides and PM than gasoline vehicles with state-of-the-art control systems (40). More data on current and future diesel vehicles is needed to assess the effect of diesel emissions on urban ozone formation, atmospheric particle concentrations, and attendant health risk.

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