THE DIESEL PARADOX: WHY DIESELIZATION WILL LEAD TO CLEANER AIR

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GLOBAL CHALLENGES

There are challenges facing the U.S. and the world that are brought on by the growing demand for transporting people and goods. These include the growing consumption of petroleum, urban air pollution, and global climate change.

Transportation is almost entirely dependent on petroleum for its energy needs, currently consuming about 51 percent of the total world petroleum consumption. By the year 2020, the Energy Information Administration estimates [1] that transportation petroleum consumption worldwide could be as much as 58 percent of the projected world consumption of 106 million barrels per day. In industrialized nations, the number of vehicles is expected to be twice the current by 2050; in developing nations such as China and India the number of vehicles is expected to increase dramatically. In the next 50 years, the number of vehicles worldwide is projected to rise from 670 million to over 3.5 billion.

As the human population, number of vehicles, and vehicle miles traveled rise, urban air pollution in most of the world tends to worsen. Air pollution is known to contribute to cardiovascular and respiratory diseases such as asthma and emphysema.

Worldwide there is evidence of a trend in rising global average temperatures which many scientists attribute to human activities that are increasing atmospheric concentrations of gases such as carbon dioxide, methane (natural gas), and nitrous oxide. These are the preponderant "greenhouse" gases, so called because they absorb and hold heat within the earth's atmosphere. Specifically, burning of fossil fuels has increased atmospheric concentrations of carbon dioxide (CO_2) by about one-third over the past 150 years [2]. Worldwide, an additional 24 billion tons of CO_2 per year are now being generated. In the U.S., transportation accounts for one-third of CO_2 emissions [3]. Concern about the perceived threat of global warming has led to an agreement at an international conference in Kyoto, Japan in December 1997 to reduce greenhouse gas emissions to seven percent below 1990 levels by the 2008-2012 timeframe. The Kyoto Protocol has so far been signed by 55 developed nations. While the U.S. has yet to agree to this Protocol, there is a growing consensus that it is prudent for the U.S. to seek to reduce its emissions of greenhouse gases.

MEETING THE CHALLENGES

Improving transportation energy use efficiency is a part of a strategy for meeting these global challenges without adverse economic impacts. When the DOE Office of Heavy Vehicle Technologies (OHVT) was created in March 1996 to address the R&D needs of heavy vehicle customers, a strategy that was both real and viable had to be crafted because of the critical importance of heavy vehicles to economic activity. Trucks and other heavy vehicles are the mainstay for trade, commerce, and economic growth. The gross domestic product (GDP) of the U.S., and hence, the country's economic activity is strongly related to freight transport.

Heavy-duty trucks, rail, and inland marine vessels which are responsible for 99 percent of freight movement are virtually all diesel powered, and by industry consensus, are expected to remain so in the foreseeable future. The diesel engine is the engine-of-choice for heavy-duty freight transport where efficiency, durability, reliability, and low speed power requirements are important. Consequently, the diesel engine plays an important role in our economy.

The diesel engine is much more efficient than the spark-ignition gasoline engine, which accounts for its dominance in commercial transport where cost competitiveness is key to staying in business. Diesels produce less carbon dioxide for equivalent work. With regard to urban air pollution, interestingly, over the past 20 years, criteria pollutant emissions from each new generation of diesel engines have decreased (by 70 percent for nitrogen oxides and 95 percent for particulate matter) as the efficiency has increased from 37 percent to today's 44 percent (see Figures 1 and 2). In addition, emission standards originally agreed to in the Statement of Principles for 2004 will now have to be met in October 2002 by the diesel engine manufacturers as stipulated in the Consent Decree with the Environmental Protection Agency and the Department of Justice.

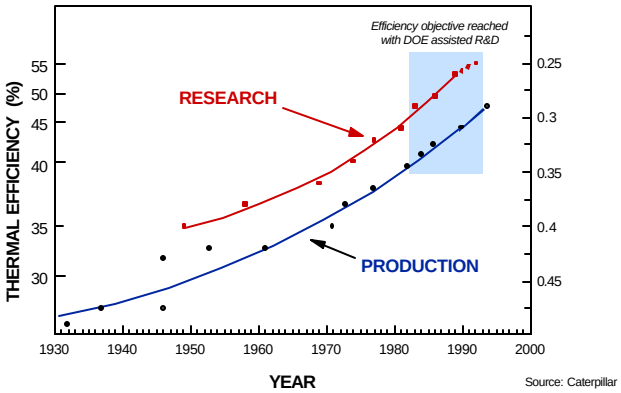


Figure 1. Increasing Diesel Efficiency

Through a series of workshops and meetings with its customers which included U.S. engine manufacturers, truck manufacturers, truck fleet owners/operators, fuel suppliers, and component suppliers, DOE/OHVT has crafted a strategy centered on the proven performance of the compression-ignition (Diesel cycle) engine. It is envisioned that a realistic approach to addressing transportation challenges is through the devolution of of an energy efficient, near-zero emissions heavy-duty diesel engine technology into all transportation vehicles. This includes not only heavy-duty trucks, but also light trucks such as the low fuel economy pickups, vans, and sport utility vehicles, and automobiles alike. Thus the "Dieselization" strategy.

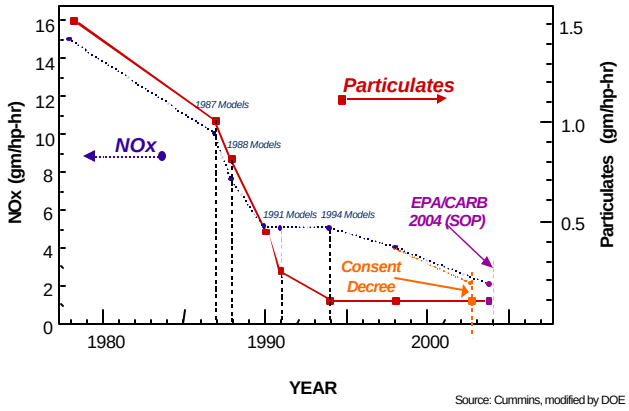


Figure 2. Diesel Engine Emission Trend

WHAT IS THE ALTERNATIVE TO THE DIESEL ENGINE?

The peak thermal efficiency of a number of energy conversion technologies that could lead to improved vehicle fuel economy are shown in Figure 3 for comparison. Of the two types of engines that are currently in production and widely used, the diesel is more efficient than the gasoline engine. Automakers are developing the gasoline direct injection engine which is expected to be more efficient than the current port fuel injected gasoline spark ignition engine. Gas turbines provide efficient air transport but attempts to adapt them to ground vehicles have been largely unsuccessful. For the future, if and when the "automotive" fuel cell becomes ready for production, and hydrogen becomes a widely available transportation fuel, there may be a strong competitor to the diesel engine. As shown

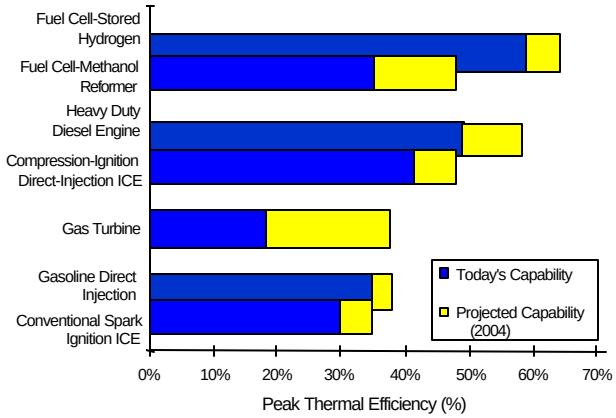


Figure 3. Comparison of Energy Conversion Efficiencies

in Figure 4, liquid hydrogen, has a relatively low energy density compared to hydrocarbon fuels. It is currently expensive to produce and difficult to distribute and store on-board a vehicle so as to provide a driving range that is comparable to that of diesel fuel. In the near- to mid-term, the diesel engine appears to be the most probable engine for improving transportation fuel efficiency.

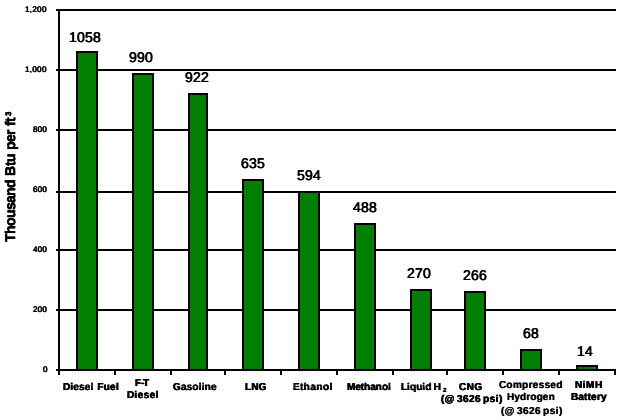


Figure 4. Energy Density of Fuels

CONCERN ABOUT DIESELS

The diesel engine has a reputation for emitting large quantities of oxides of nitrogen and soot. To this day the perception of the "smoky old diesel" persists even with the growing body of scientific and technical knowledge, and research results showing that diesel exhaust can achieve or even better the standards set for gasoline engines. As "dieselization" becomes more likely because of the growing concern about rising fuel costs and global climate change, and the absence of any other competitive transportation alternative to the less efficient gasoline engine, some environmental groups have raised concerns about increasing usage of diesel engines especially for light duty applications. Some direct quotes from these groups are:

> "Recent government-sponsored emissions tests of the newest, cleanest diesels have been unimpressive." [4]

> "At a minimum, regulators should close the historic loops that permit diesel cars to pollute more than those powered by gasoline. [5]

Many Groups Concerned About Diesels

- Union of Concerned Scientists
- Natural Resources Defense Council
- American Council for an Energy-Efficient Economy
- Environmental and Energy Study Institute
- Sierra Club
- American Lung Association
- Environmental Protection Agency
- California Air Resources Board
- South Coast Air Quality Management District
- Council on Environmental Quality

> "It's time to end the free ride for big rigs." [6]

In response to these concerns, the DOE Office of Heavy Vehicle Technologies has invited environmental groups to the Diesel Engine Emissions Reduction (DEER) workshops to facilitate dialog between them and the researchers whose work is showing that there are indeed technical approaches which can eliminate diesel emissions.

Diesel engines are currently considered to be the major source of particulate matter (PM) emissions. As measurement techniques become more sophisticated and more reliable data are developed, it is becoming apparent that the models being used to assign responsibility for the source of emissions may be unreliable.

In the recent Northern Front Range Air Quality Study (NFRAQS) [7] of the Denver, CO area, data obtained from actual vehicle emissions measurements were used to determine the proportion of PM2.5 emissions from various sources. Sophisticated analytical chemistry measurement techniques were used to differentiate PM2.5 emissions from various sources, for example, "smoking" gasoline engines, gasoline engine high emitters, well maintained gasoline engines, different diesel engines, and even cooking on barbecue grills. The data showed that 74 percent of the measured PM2.5 came from gasoline exhaust vs. 26 percent from diesel exhaust.

By comparison, the EPA Mobile 5 model was used to predict PM2.5 emissions inventory apportionment. The model predicted that 22 percent of PM2.5 emissions would come from gasoline exhaust vs. 78 percent from diesel exhaust. This contradicts the results of actual experimental measurements (see Figure 5) which suggests that the model may need some substantial revision to be considered to be reliable.

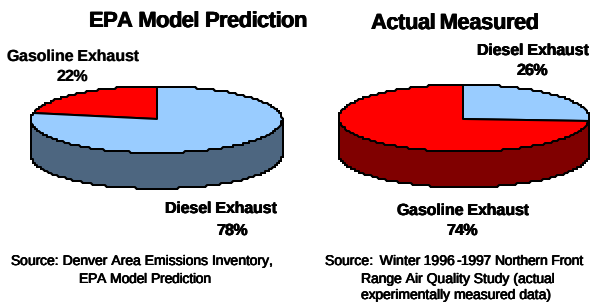


Figure 5. Big Difference Between EPA Predicted and Actual Measured PM Emission Inventories

These findings imply that the models may be inordinately overestimating the PM2.5 emission contributions from diesel engines. Such model predictions are often cited by those opposed to enabling "dieselization" of light duty vehicles.

It has been only been recently, in the last five years or so, that techniques for measuring ultrafine particulates from combustion sources have become more widely available. Particulate researchers in the United Kingdom, for example, have shown that PM from gasoline engines is normally distributed around 30 nanometers while diesel PM is normally distributed around 60 nanometers.

Health studies indicate that fine particulates may be highly toxic to the human lung at very low mass concentrations because of: a) large numbers per unit mass; b) high deposition efficiency in the lower respiratory tract; c) inability of the respiratory tract to clear itself of such particulates; and d) increased surface area available for interactions with cells [8]. These tendencies increase as the particles become smaller.

These factors imply that gasoline particulates, because they are smaller than diesel particulates, may be more readily embedded deeper into the lungs. By inference, therefore, gasoline particulates could pose an even greater health risk than diesel particulates since the greater number of our vehicles are gasoline fueled and the greater quantity of fuel consumed is gasoline. Unfortunately, "...no chronic inhalation bioassays have been carried out on gasoline emissions." Given this lack of toxicity data [9], it cannot be assumed that gasoline PM is benign, especially since it is known that there are carcinogens such as 1,3 butadeine on the particulates.

The small size of engine particulates presents difficulty in measuring and characterizing them let alone determining their biological toxicity and human health effects. With transmission electron microscopy (TEM) micrographs (see Figure 6) it is now possible to show the structural differences between gasoline and diesel PM.

At high magnification, the TEM micrographs [10] suggest that diesel particulates have a crystalline structure while gasoline particulates are amorphous and therefore, would have a greater

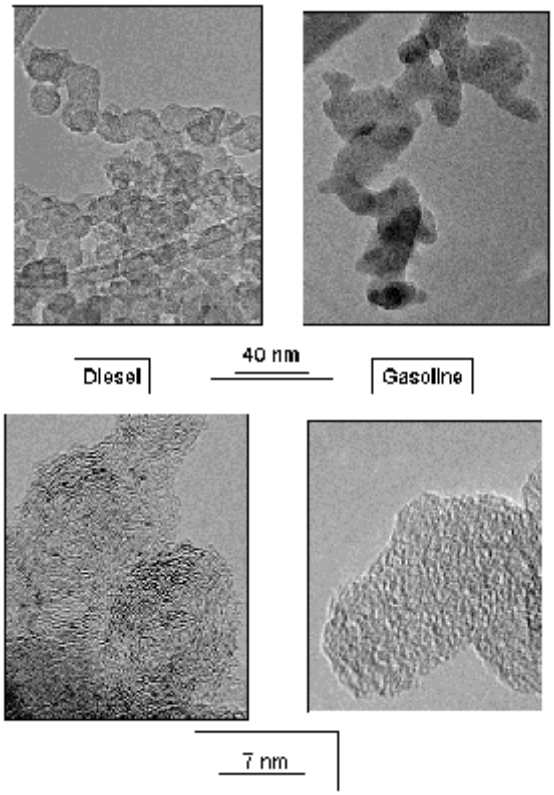


Figure 6. Comparison of Diesel and Gasoline Particulate Matter (at different magnification)

tendency to adsorb chemical species from the engine exhaust. Investigation is currently underway using ESCA/Auger electron microscopy to identify the chemical species on the surface of gasoline and diesel particulates.

Of the EPA list of some 188 hazardous air toxics only a few come from mobile sources. These are: acetaldehyde, benzene, biphenyl compounds, 1-3 butadiene, ethyl benzene, formaldehyde, methanol, methyl tert butyl ether (MTBE), naphthalene, polycyclic aromatics, styrene, toluene, and xylenes. Table 1 compares emissions of some of these toxics from diesel and gasoline vehicles. It is interesting to note that the levels of several of the most toxic compounds emanating from gasoline engines are higher (for example, benzene, 1-3 butadeine, formaldehyde) than from diesels.

Table 1. Comparison of Some Toxics from Diesel and Gasoline Exhaust

Toxics	Diesel Exhaust (Engine Out*) g/mi	Gasoline Exhaust (Engine Out*) g/mi	Gasoline Exhaust (Tailpipe**), g/mi
1,3 - butadiene	0.057	–nr--	0.087
acetaldehyde	–nr--	0.01974	0.00443
benezene	0.035	–nr--	0.365
formaldehyde	0.088	–nr--	0.121
methyl tert butyl ether	–nr--	0.0105	0.00142
toluene	–nr--	0.33836	0.03662

nr - not reported
* no exhaust aftertreatment device (i.e., catalytic converter)
** with catalytic converter

Sources: Gasoline FAQ (Industrial Research Ltd.)
EPA, "Locating and Estimating Air Emissions from Sources"

A comparison of the major components of gasoline and diesel fuel may also provide an indication of the fuel's propensity to produce toxic emissions. The typical composition of gasoline, No. 2 diesel fuel, and ARCO EC-Diesel is shown in Table 2. Of these components, paraffins are classified as being non-toxic, while olefins and aromatics are potentially toxic. Interestingly, aromatics are undesirable in diesel fuel because they lower the cetane number (i.e., reduce "compression ignition" quality of the fuel), but not in gasoline where they enhance octane quality. Hence, enhancing diesel fuel quality tends to remove aromatics, and therefore, lower toxicity.

Table 2. Composition of Gasoline and Diesel Fuel

Composition	Gasoline[1]	No.2 Diesel Fuel[1]	EC - Diesel[2] (ARCO)
Paraffins, vol. % (non-toxic)	67	71	90.7
Olefins, vol. % (potential toxics)	8	**	0.5
Aromatics, vol. % (potential toxics)	25	29	8.8
Performance Enhancers, weight %	*MTBE - 2 Benzene - 1*	*Amyl nitrate < 1*	

[1] 1999 Winter- Alliance of Automobile Manufacturers Survey.
[2] ARCO Press Release Package, October 1999.
** Olefins not measured, typically less than 5%

WHY DIESELIZATION WILL LEAD TO CLEANER AIR

It is our view that by conducting research to develop emissions control technologies for lean burn (diesel) engines, that implementation of these technologies will lead to cleaner air. Diesel engine emissions control technology development has lagged behind gasoline engine emissions control. Historically, EPA has focused on setting emissions standards for gasoline vehicles in order to reduce urban air pollution. Accordingly, technology development has focused on gasoline engine emissions control. The trends in gasoline and diesel light truck NOx emissions are shown in Figure 7. In the 1970s, gasoline "engine out" emissions were higher than diesel "engine out" emissions. Initial application of catalytic converters reduced gasoline NOx emissions to about the diesel engine out levels. The three-way catalyst system subsequently reduced gasoline NOx emissions to their current very low levels.

In the same timeframe that the catalytic converter has reduced gasoline engine emissions, diesel engine manufacturers have focused on reducing diesel engine-out emissions while optimizing engine efficiency. Diesel engine emission levels have been reduced (see Figure 2) thus far without

the benefit of exhaust aftertreatment. To say that diesel engine emissions cannot be made as clean as gasoline engine emissions, as some would claim, is, therefore, premature, since research on exhaust emissions aftertreatment for "lean burn" (diesel) engines is relatively recent.

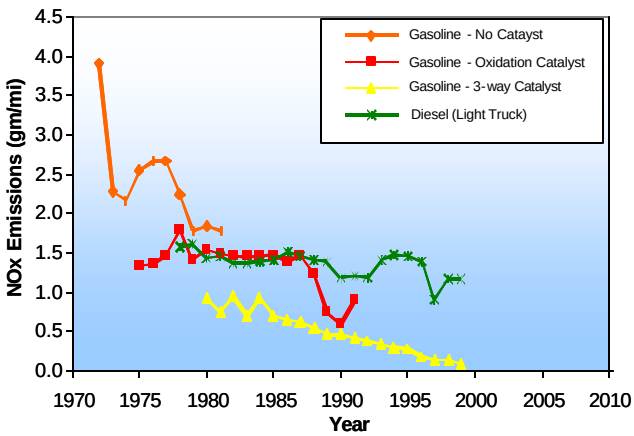
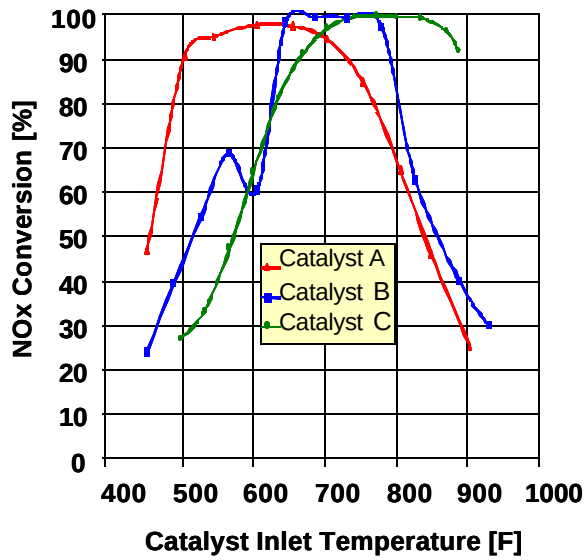


Figure 7. Light Truck NOx Emission Trends

One promising technology for NOx reduction is the NOx adsorber catalyst which has been shown to achieve NOx reduction efficiencies of up to 98 percent (see Figure 8). Using diesel fuel injected upstream of the catalyst as the reducing agent, a fuel penalty of 3 to 10 percent has been estimated. However, sulfur in the fuel poisons the current catalyst formulations and renders them ineffective after only a few hours of operation. Low sulfur diesel fuel or a practical sulfur trap upstream of the NOx adsorber (as discussed later) could make this an effective aftertreatment system.

More stringent emissions standards such as the California Air Resources Board low emission vehicles (CARB LEV II) standards and the Environmental Protection Agencies Tier 2 standards for light duty vehicles have been established. These emission levels are extremely challenging for diesels but they have lowered the bar for gasoline engines as well. To help gasoline vehicle emission control devices work effectively especially for particulate emissions, EPA issued the notice of proposed rulemaking on gasoline quality which limited the sulfur content of gasoline to 30 ppm. Lowering the sulfur content in gasoline reduces the formation of sulfate particulates thus enabling gasoline vehicles to achieve the Tier 2 particulate level of 0.01 g/mi.



Source: J. Stang, Cummins Engine Company

Figure 8. NOx Adsorber Catalyst

Diesel emission controls research and development have been spurred by these ever more stringent emissions standards, both for heavy-duty and light-duty vehicles. For its part, DOE has focused research and development on the control of diesel emissions, as this is the key enabling technology that will make the "dieselization" effort possible. A three-pronged systems approach (see Figure 9) is utilized to arrive at the most cost-effective and workable emissions control strategy for diesel engines for all types of vehicles. This approach involves: a) looking into the effects that fuel composition and properties have on engine performance and emissions; b) understanding the compression ignition lean burn combustion process itself as it occurs in the engine; and c) applying effective

Three-pronged systems approach appears necessary to meet very low emissions without sacrificing engine efficiency

Figure 9. Diesel Emissions Control Strategy

exhaust aftertreatment techniques to further clean up what comes from the engine. Implementation of these various technologies [11, 12, 13] in a vehicle is shown schematically in Figure 10.

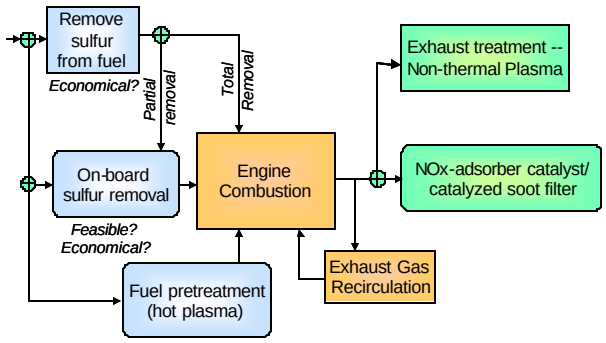


Figure 10. Schematic of Diesel Emissions Control

Sulfur in the fuel has been the primary concern in current diesel emissions control technologies [14], specifically in achieving the high efficiency of NOx and PM emissions reduction needed to meet future emissions standards. Three DOE projects are focused on evaluating the effects of sulfur content in the fuel on the ability of aftertreatment technologies to achieve the very low levels of emissions being established by EPA, namely: a) the Diesel Emission Control - Sulfur Effects (DECSE) Project for heavy duty engines; b) the Diesel Vehicle Emission Control - Sulfur Effects (DVECSE) Project for light-duty vehicles; and c) the evaluation of ARCO EC-Diesel (an experimental very low sulfur content petroleum-based diesel fuel) in California buses and trucks. Data from these projects have been used to provide supporting data justifying the need for very low sulfur levels in diesel fuel. EPA has taken into account these data in formulating its diesel emissions and fuel sulfur content regulations.

The benefit of using low sulfur diesel fuel such as the ARCO EC-Diesel is illustrated in Figure 11. By simply replacing conventional diesel with EC-Diesel fuel, particulate matter (PM) emissions from a tanker truck (55,000 lbs. when fully loaded with fuel being delivered to gas stations) were reduced from 0.807 g/mi to 0.562 g/mi. Use of a continuously regenerated particulate filter in conjunction with the EC-Diesel fuel enabled further reduction to 0.026 g/mi.

By comparison, a gasoline fueled car (3,300 lbs. with the driver and no passenger) complying with EPA Tier 2 emissions standards would be emitting 0.01 g/mile. Comparing vehicles on the basis of doing the same amount of work, 100 cars expend the same work (in ton-miles traveled) transporting just a driver around for the whole year as the tanker truck does delivering thousands of gallons of fuel to filling stations. These 100 Tier 2 emissions level-compliant cars emit 1.0 g/mi, almost 40 times as much as the tanker truck's 0.026 g/mi. Put another way, this huge truck emits only as much particulate matter as 10 Tier 2 compliant automobiles, a remarkable achievement.

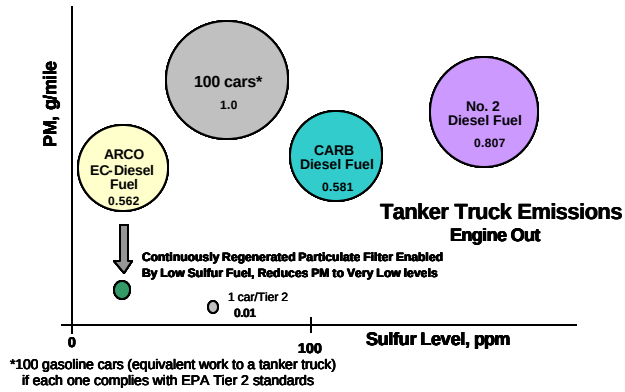


Figure 11. Low Sulfur Diesel Fuel Reduces PM Emissions from Current Tanker Trucks

In addition to lowering the sulfur content of diesel fuel during refining, other approaches include on-board sulfur removal (e.g., with the use of sulfur traps) and fuel pretreatment (e.g., with hot plasma). A primary consideration is to determine the practicality of such systems. Preliminary calculations indicate that a disposable 15 lb sulfur trap (using BaO as the active material taken to be 20 percent of the weight of the sulfur trapping material and assuming a 50 percent efficient adsorption process) would be good for 120,000 miles (in a 40 mpg light-duty vehicle) when using diesel fuel with 15 ppm sulfur. This sulfur trap would not need to be replaced for almost the lifetime of the vehicle.

As discussed briefly above, the diesel catalyzed soot filter (CSF) in conjunction with ultra low sulfur diesel fuel has been shown to be extremely

effective in eliminating ultrafine particulates from diesel exhaust. As shown in Figure 12 the particulate matter is completely filtered from the exhaust because the device is closed at the exhaust end. Catalytic combustion of the collected particulates occurs either continuously or periodically depending on the exhaust temperature as the catalyst "lights off". Greater than 90 percent reduction of particulate matter has been achieved.

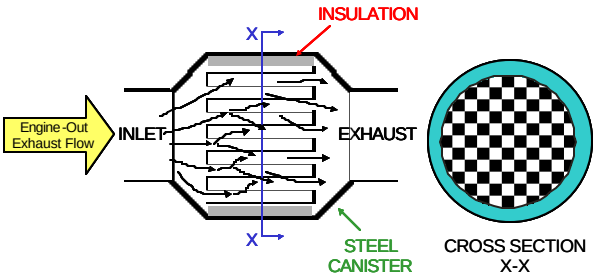


Figure 12. Catalyzed Diesel Particulate Filter

The prospect of having very low sulfur diesel fuel in the 2006 timeframe has stimulated increased commitment from catalyst manufacturers to bring the NOx adsorber and catalyzed diesel particulate filter technologies forward for diesel engine applications. Indeed, the newly approved stringent tailpipe emissions for large trucks and buses also direct refiners to produce virtually sulfur-free diesel fuel. By 2006, on average 15 parts per million of sulfur will be required of 80 percent of diesel fuel sold nationwide and, by 2010, of all diesel fuel.

Current port fuel injected gasoline engines with catalytic converters are "clean", i.e., they emit very low masses of PM, but emit very many ultrafine particulates. The primary difference between the catalyzed soot filter and the gasoline catalytic converter (see Figure 13) is in the physical design of the devices. The conventional catalytic converter is a flow through device while a catalyzed soot filter is a closed end device, which captures or "traps" all of the soot or particulate matter in the diesel exhaust.

Direct injection lean burn engines are more efficient than current port fuel injection gasoline engines. The direct injection diesel engine is the highest efficiency production engine today. The spark ignition direct injection gasoline engine has near-diesel efficiency at part load and its specific power at full load is better than the port fuel injection gasoline engine. The auto industry has been developing gasoline direct injection (GDI) engines for many years for the smaller light-duty vehicles, mostly automobiles. These GDI engines are expected to greatly improve the fuel economy of automobiles. Both the direct injection diesel and GDI engines have the same challenges which are the control of NOx in their lean burn operating regimes and control of particulates.

The diesel engine emission control strategies that are under development have potentially the same applicability to the direct injection gasoline engine. Specifically, the CSF will remove a greater number of ultrafine particulates than the gasoline catalytic converter. As transportation moves to lean burn engines for greater fuel economy and reasons of energy security and global climate change, the clean lean burn engines, both gasoline and diesel engines along with the new sulfur fuel they utilize will lead to cleaner air.

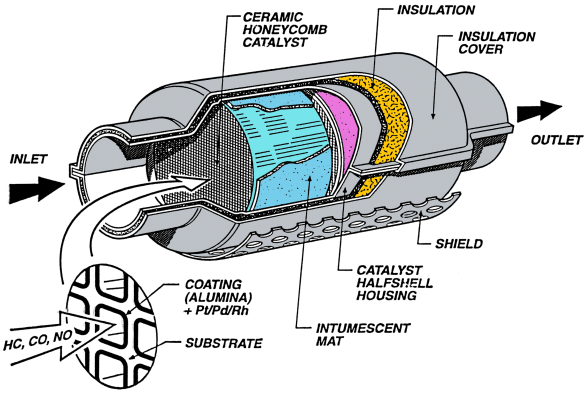


Figure 13. Automotive (Gasoline) Catalytic Converter (Three-Way Catalyst Design)

CONCLUSION

"Dieselization" is a real and viable strategy for reducing transportation energy use with the concomitant reduction in carbon dioxide emissions. Progress in diesel emission controls, together with ultra low sulfur fuel will enable these devices to work effectively, and will make "clean" diesel technology commercially viable in the near future. In addition, diesel engine emission control devices suitably modified will work as well with the gasoline direct injection engine which has

similar NOx and PM emission problems as the diesel since both are lean burn engines. As clean diesel and gasoline direct injection engines operating on low sulfur fuels replace the port fuel injected gasoline engines in automobiles and other light-duty vehicles the result will be cleaner air, since the emissions control technologies being developed for lean burn engines is more effective than the conventional catalytic converter. In retrospect, therefore, the efforts to promote dieselization ultimately will lead to cleaner air.

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