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Influence of Exhaust Aftertreatment Devices on Heavy-Duty Diesel Engine's Particulate Emissions

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This paper describes the impact of two exhaust aftertreatment devices, an oxidation catalytic converter and a diesel particulate filter, on the particulate emissions of a heavy-duty diesel engine, the emphasis being on diesel particulate composition and size distribution. The analysis showed that the catalytic converter did not alter the size distribution significantly, except for an increased production of nanosized particles below 30 n miles in mean diameter. On the other hand, the ceramic monolith wall-flow filter reduced the emitted particle number up to 100 times. The results again indicated that a diesel particulate filter can also increase the number of nanosized particles. Further investigations, however, revealed that the formation of nanosized particles downstream of the oxidation catalytic converters and diesel particulate filters, respectively, is biased by inadequate sampling conditions. As long as realistic sampling conditions are applied, both catalyst and filter reduce particle emissions over the entire size range.

Introduction

THE reduction of particulate emissions from diesel engines is one of the most challenging problems associated with exhaust pollution control, together with the control of NO_x emissions from any "lean burn" application. Diesel exhaust-gas aftertreatment devices hold out the prospect of substantially reducing particulate mass emissions from diesel engines. Results from current studies, however, imply that particulate mass does not properly mirror the variety of health effects associated with particulate emissions from any combustion process. There are indications that particle size or particle surface emissions might be even more important than particulate mass.[1] For this reason, in developing exhaust aftertreatment devices for diesel engines, it is important to study the impact of such devices on all aspects of the emitted particulate matter, i.e., particulate mass, particle size, and particle surface emissions.

Theoretical Considerations

As shown in Fig. 1, the total particulate matter (TPM) consists of an agglomerate of solid phase emissions, made up primarily of small carbon particles called SOL (solid carbides or soot); adsorbed to the SOL are hydrocarbons, which can be removed by an organic solvent, called SOF (soluble organic fraction), and sulfates (SULF), which can be removed by water.[3] Also associated with the TPM can be droplets of liquid, condensed hydrocarbons, and condensed sulfates.[2]

Generally, the TPM is composed mainly of three parts, the SOL, the SOF, and the SULF

$$\text{TPM} = \text{SOL} + \text{SOF} + \text{SULF} \tag{1}$$

The impact of diesel particulate matter on human health remains to be determined, as a wide variety of potential effects has been reported from laboratory animals and human studies. Nevertheless, it is beyond doubt that diesel particles are hazardous and could cause serious problems to humans and other living creatures.

The carcinogenic potential of exhaust gases from diesel engines is governed basically by two exhaust components, polycyclic aromatic hydrocarbons (PAH) and soot particles.

The carcinogenic effect of some of the PAH emitted by diesel engines has been known for more than 50 years. These carcinogenic PAH accumulate primarily along with SOF on the extremely fine soot particles that are present in the exhaust, and a certain percentage of them deposits with the soot particles in the lung after inhalation.

Recent research shows that the soot particle itself, or the carbon core of the soot particle, seems to be more significant for the carcinogenic potential of diesel exhaust emissions in the lung than PAH, only relatively small amounts of which are found in exhaust gas.[4]

The health effects of a particle are dependent on its extent and mode of entry into the body and on the body's ability to remove or neutralize the compounds present on the particle. Diesel particulate matter is small enough to result in deposition of the particle in the respiratory tract. It has been determined that 90% of diesel particulate mass is smaller than 1 μm. The bulk of the particulate greater than 0.3 μm in diameter will be removed by the upper respiratory tract, while the remaining small particles can enter and eventually get trapped in the bronchial passages and alveoli of the lungs. Recent studies show that particles smaller than 50 nm in diameter contribute about 7% of the total mass but, at the same time, about 20% of the total surface and about 70% of the total number emission (Fig. 2). Therefore, it is a matter of great importance to study the influence of diesel exhaust aftertreatment systems on the size distribution of diesel particulate matter.

Diesel exhaust aftertreatment devices can be divided into a group comprising filtering devices and a group comprising nonfiltering devices. For the group of filtering devices, the trap oxidizers or diesel particulate filters (DPFs), the most challenging problem is the regeneration of the loaded filters by burning off the accumulated particulate matter. For the other group, the oxidation catalytic converters (OCCs), the main problem lies in the hindering of sulfate formation.[5,6]

Experimental Conditions

Two exhaust aftertreatment devices, an OCC and a ceramic monolith wall-flow particulate filter with an electrical regeneration system, were installed, one at a time, on a heavy-duty 6.8-liter diesel engine (EURO II). Its specifications are shown in Table 1.

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Table 1 Engine characteristics (manufacturer's data)

Property	Specifications
Working characteristics	Direct injection (DI), turbocharged/intercooled
Displacement	6.8 liters
Cylinders	Six in-line
Compression ratio	16.5:1
Max. power	169 kW by 2400 min^{-1}
Max. torque	850 Nm by 1440 min^{-1}

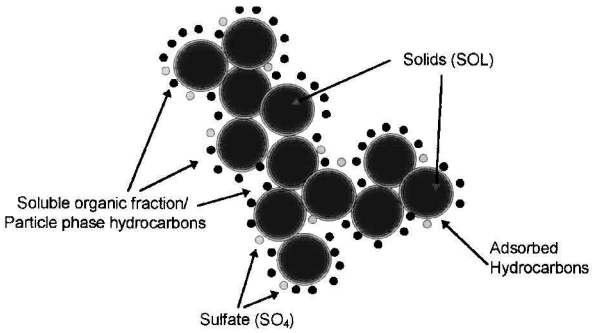

Fig. 1 Schematic of diesel particulate matter.[3]

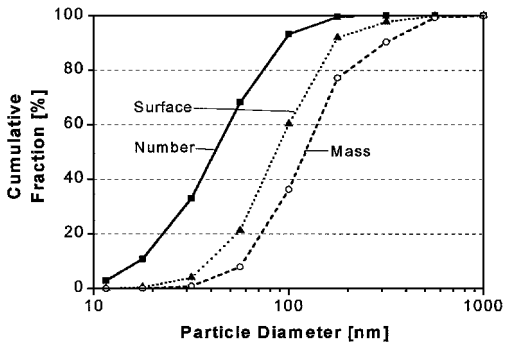

Fig. 2 Typical cumulative fraction of particle mass, surface, and number emissions.

Exhaust emissions were analyzed on four operating points (Table 2), which simulate the engine's overall behavior and are also present in the 13-mode step test, introduced by European Union (EU) directives 88/77/EEC[7] (European Economic Community) and 91/542/EEC.[8] In addition, each operating point was characterized by a standard tunnel dilution ratio (DR), since the DR can influence the interactions between the particles and change their size distribution by altering the tunnel temperature, whereas the total emitted mass remains unchanged. DR values are typically chosen in such a way that, for each point, the tunnel temperature does not exceed 52°C (as described by EU directives).

Particulate-analysis equipment included three particulate sampling filter lines, used for mass, chemical composition, and thermogravimetric analysis, three particulate size-distribution analysis devices [cascade impactor (IMP), electrical aerosol analyzer (EAA), and differential mobility analyzer (DMA)] and two other devices, the epiphaniometer (EPI), used for surface analysis, as well as the aethalometer (BC), used for black carbon measurements. Figure 3 shows the general test bench arrangements.

The diesel fuel's major properties are shown in Table 3.

Size-Distribution Analysis Equipment

The size distribution of diesel particulate matter was calculated using three measurement devices, the EAA, the DMA, and the IMP.

Table 2 Operating points and DR

Rotation speed, min^{-1}	Load, %	Torque, Nm	DR
Intermediate 1440	50	428	32
Intermediate 1440	100	855	22
Rated 2400	25	169	19
Rated 2400	100	660	12

Table 3 Fuel properties

Property	Analysis method	Value
Density (kg/m^3), 15°C/25°C	DIN 51/ISO 91	824.5/817.7
Sulfur content (wt%)	DIN 51 400 T.6	0.026
Carbon/hydrogen ratio	——	$C_{11.9}H_{23.0}$

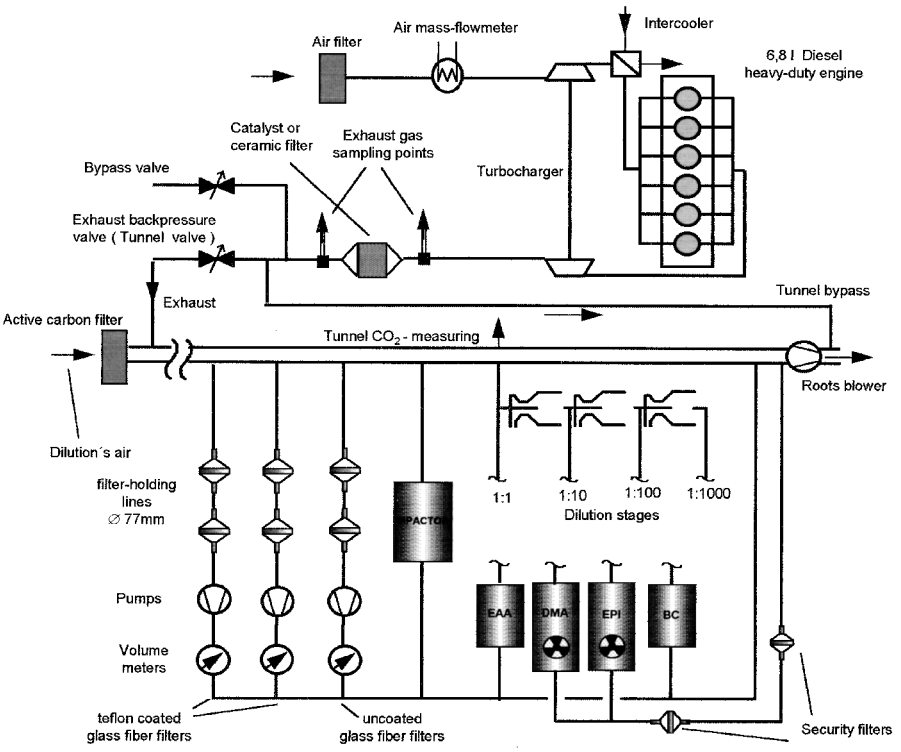

Fig. 3 Test bench arrangement.

Both the EAA and the DMA measure particles with diameters ranging from 10 to 1000 nm. However, the DMA covers better the smaller range of particles, up to 205 nm, whereas the EAA has a broader range, up to 1000 nm, and starts precisely at 15 nm. The IMP, on the other hand, covers the whole range of particle diameters, giving better results on high-diameter values.

The EAA and the DMA classify the particles according to their electrical mobility. The electrical mobility of a particle is a measure of its ability to move in an electric field, expressed as the field-induced velocity component per unit electric field intensity. By knowing the particle's elementary charge, it is possible to calculate its mean diameter from its electrical mobility and, consequently, determine the whole particle size distribution of the gas analyzed.[9,10]

According to the DMA's operating principle, an aerosol passes through an electric field after its particles have become electrically charged. Because of their different electrical mobilities, particles of the same size range can be collected by adjusting the electrode voltage to different values. Since the particles are too small for detection by optical means, they are increased by condensation, each one growing to a droplet, so that they can finally be counted.

The IMP's operating principle, on the other hand, is based on the fact that, when a moving aerosol changes direction, the particles, because of their inertia, continue moving in the same direction. By forcing an aerosol to flow through an orifice and impinge a flat surface where the gas stream undergoes an abrupt change of direction, it is possible to classify the particles into several size groups, using several stages in series, each having a different lower size limit for collection.

Results and Discussion

Emission Reduction

The catalyst's and the filter's efficiencies in reducing the gaseous as well as the particulate emissions are shown in Table 4, where the particulate matter is further separated into its components, SOL, SOF, and SULF. Differences in particulate matter and the sum of SOL, SOF, and SULF are attributed to the different measurement techniques used.

As anticipated, the OCC alters mainly the carbon monoxide and hydrocarbon emission levels, by reducing them up to 90%, in relation to the operating point. The nitrogen oxide emissions are generally not altered, but there is a slight reduction tendency, up to 5%, which can be attributed to the slight filter backpressure rise, caused by the use of the aftertreatment device. Increased backpressure generally causes the engine's internal exhaust-gas recirculation (EGR) to rise, which subsequently decreases the emitted NO_x.[11] In the case of particulate emissions, the catalyst does not influence their mass production.

On the other hand, the ceramic filter's behavior is quite different from that of the catalyst, as anticipated. The filter does not reduce significantly the carbon monoxide (CO) and hydrocarbon (HC) emissions. In fact, the HC emissions remain practically unchanged, whereas the CO emissions are slightly increased, especially on full load points. Nitrogen oxides are decreased up to 30%, in relation to the test point. The CO increase and simultaneous NO_x decrease are attributed to experimental conditions related to a significant backpressure ascent. The particulate emission decrease ranges from 85 to 95%. Unlike the OCC, the particulate ceramic monolith filter, operating as a typical shallow-bed trap, does not allow the particles to pass through, diminishing their production.

Particulate Composition

Particulate composition data are given by SOF and SULF analysis, which is made on Teflon-coated glass-fiber filters, and SOL, SOF, and SULF are related to the total particulate mass as shown in Eq. (1).

Data from Table 4 show that the oxidation catalyst does not generally affect the amount of solids present in the TPM. It seems, on the other hand, that the aftertreatment device tends to reduce the amount of SOF, but at the same time, it produces more SULF. Noble metal catalysts typically oxidize CO and HC, which is the desired reaction path, but at the same time they also oxidize SO_2 to SO_3, which, in combination with water, forms H_2SO_4. On rated speed, full load (2400 min^{-1}, 100%), however, this SULF increase is not observed, although the temperature is favorable; the space velocity is too high and does not give the SULF enough time to be built.[12]

The particulate filter, on the other hand, seems to alter significantly the SOL, reducing simultaneously both the SOF and the SULF. The SOL reduction is the main result of the filter's operation, as SOL production is minimized. The last conclusion is in accordance with the results of other analytical methods, suggesting that the solid carbides and the black carbon are reduced considerably.

Particulate Size Distribution

Diesel exhaust size-distribution analysis, as carried out with the DMA, EAA, and IMP, gives some interesting results. EAA and DMA measurements are almost-identical in the size range covered by both devices. Nevertheless, the DMA shows a high resolution and measurement accuracy in the size range from 15 to 205 nm in mean diameter, this range being considered more important toxicologically, and for this reason, its results are more meaningful. IMP mass measurements, on the other hand, are always very small, even for tests without the use of any exhaust aftertreatment device, producing big error factors, which do not allow us to consider them seriously. The IMP analysis verifies that the main bulk of the particulate matter is emitted in very small diameter ranges, which cannot be analyzed accurately by the specific device.[13]

Size-distribution analysis results, as carried out with the DMA, are given in Figs. 4–7. Measurement accuracy was calculated to be approximately ±20%, whereas experimental uncertainties are expected to be caused only from the measuring devices, since the engine's operation was very stable for all test repetitions.

The OCC does not seem to alter significantly the particulate size distribution. On medium speed, 50% load, and on rated speed, 25% load, the particle distribution is similar to that without aftertreatment, except for the ultrafine particle size range, where a slight particulate reduction is observed. This reduction could be explained

Table 4 Test emission levels

Rotation speed, min^{-1}	Load, %	Version	CO, g/kWh[a]	HC, g/kWh[a]	NO_x, g/kWh[a]	Particulate matter, g/kWh[b]	SOL, g/kWh[b]	SOF, g/kWh[b]	SULF, g/kWh[b]
1440	50	Baseline	0.56	0.14	7.65	0.09	0.059	0.011	0.024
		Catalyst	0.06	0.04	7.68	0.10	0.060	0.010	0.033
		Filter	0.58	0.13	5.90	0.01	0.000	0.004	0.003
	100	Baseline	0.81	0.06	5.87	0.10	0.050	0.027	0.020
		Catalyst	0.05	0.01	5.60	0.10	0.051	0.019	0.033
		Filter	1.14	0.04	4.05	0.01	0.001	0.002	0.002
2400	25	Baseline	1.55	0.45	4.30	0.43	0.280	0.130	0.019
		Catalyst	0.33	0.22	4.15	0.44	0.250	0.098	0.022
		Filter	1.93	0.45	3.94	0.05	0.010	0.032	0.007
	100	Baseline	0.58	0.08	3.60	0.10	0.074	0.014	0.014
		Catalyst	0.11	0.01	3.50	0.09	0.049	0.006	0.012
		Filter	0.73	0.07	3.11	0.01	0.008	0.006	0.003

[a]Experimental accuracy, ±5%. [b]Experimental accuracy, ±10%.

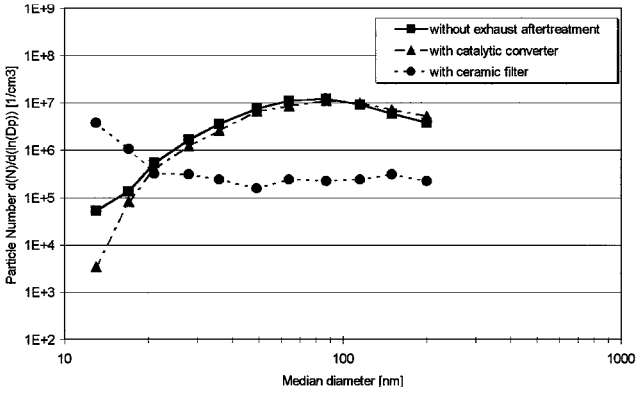

Fig. 4 Particulate size distribution: 1440 min^{-1}, 50% load.

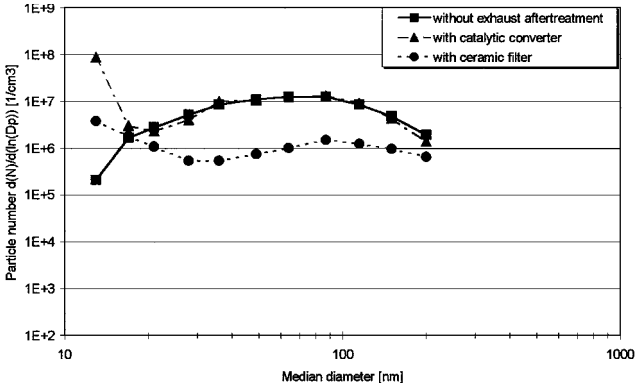

Fig. 5 Particulate size distribution: 1440 min^{-1}, 100% load.

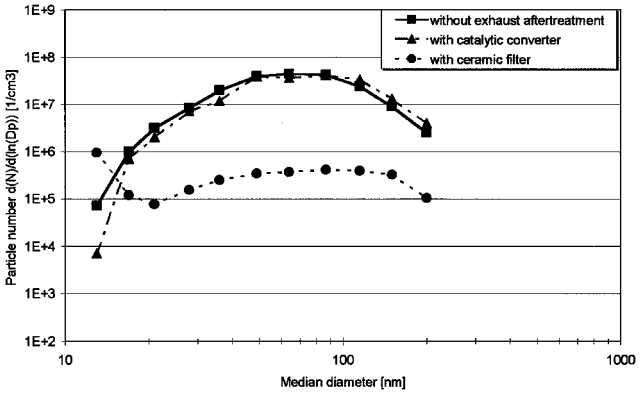

Fig. 6 Particulate size distribution: 2400 min^{-1}, 25% load.

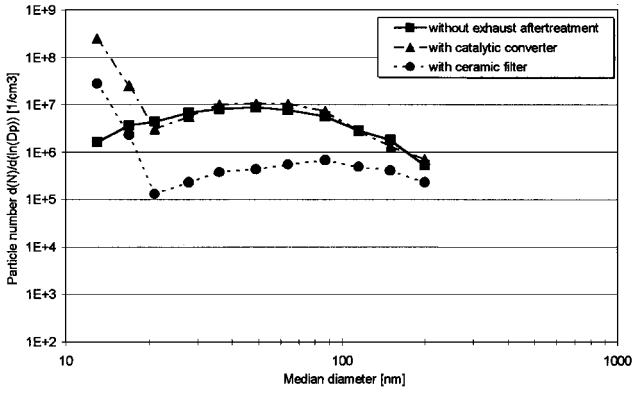

Fig. 7 Particulate size distribution: 2400 min^{-1}, 100% load.

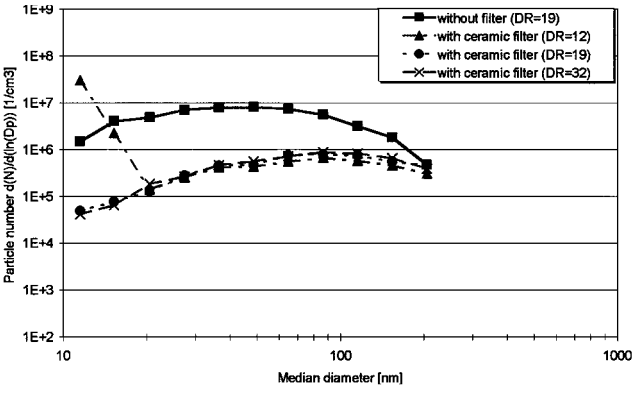

Fig. 8 Effect of DR on particle size distribution: 2400 min^{-1}, 100% load.

considering that particles of this size range are mainly condensed hydrocarbons (SOF), which have shown a reduction tendency according to SOF analysis.

On full-load test points, the size distribution shows no alteration in diameters above 30 nm, whereas in smaller diameters a very big increase, up to 100 times, in the particulate number is observed.

The ceramic filter, on the other hand, generally affects the particulate size distribution to a great extent. The number of particles with diameters above 30 nm is decreased on all test points from 10 to 100 times, due to the reduction of the emitted particulate matter, caused by the use of the specific aftertreatment device. An increase in the production of particles below 30 nm is observed once again, especially on full-load points, as well as on medium speed, 50% load.

It has been shown, however, that formation of nanosized particles downstream a ceramic monolith filter is provoked by inadequate measuring conditions (too low a DR), which result in spontaneous homogeneous condensation.[14] Figure 8 shows the effect of DR on particulate size distribution. A low DR results in increased nanoparticle production. The conditions resulting in particle formation downstream from a filter are far away from the dilution conditions occurring in the atmosphere under real driving conditions. The studies clearly indicated that particulate filters substantially reduce particle number emissions, in all size classes, as long as measuring conditions close to reality are applied.

It should also be noted that the reasons for the load and speed dependence of the particle size distribution have not been fully determined yet. Changes in load and engine speed affect numerous operation parameters such as peak pressure, residence time, and air/fuel ratio. At the time of the tests it was just known that such an effect exists and remains under investigation.

Conclusions

This research was carried out to study the influence of an OCC and a particulate filter on the particulate emissions of a heavy-duty diesel engine. The analysis focused on diesel particulate composition and size distribution.

The OCC altered mainly the engine's gaseous emissions, as it significantly reduced the HC and CO emissions, whereas the NO_x remained constant and the TPM showed a small increase tendency. This small particulate increase was the result of a SULF production increase, verified by composition analysis, which also showed stable SOL and SOF production levels. The catalyst did not alter the particulate size distribution, except for an increase in nanosized particles, which is related to the inadequate measuring conditions.

On the other hand, the ceramic monolith particulate filter did not affect the gaseous emissions significantly, and a slight CO increase and NO_x reduction were caused by the filter backpressure rise. The TPM was reduced significantly, as the filter's efficiency was between 85 and 95%. SOF and SULF analysis indicated that the particulate reduction was due mainly to extreme SOL reduction, whereas SOF and SULF remained stable. The particulate size

distribution was affected positively, as the number of emitted particles was considerably reduced. Formation of nanosized particles downstream from the ceramic monolith filter was provoked by inadequate measuring conditions (too low a DR), which resulted in spontaneous homogeneous condensation. Since atmospheric DRs are considerably higher, formation of nanosized particles is not expected to occur in the exhaust plume of a vehicle.

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