RETROFIT AFTERTREATMENT SYSTEMS FOR DIESEL ENGINES

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ABSTRACT

The problem of abatement of diesel particulates emissions is considered now as one of the main challenges in the quest for better air quality. In order to succeed in this quest, and in view of the long service life of heavy-duty diesels, it could be beneficial to install retrofit exhaust aftertreatment devices in diesel vehicles. The paper reviews the leading retrofit technologies: diesel oxidation catalysts (DOC), diesel particulate filters (DPF), continuously regenerating trap (CRT), fuel borne catalysts (FBC), filter catalytic coating, catalytic particulate oxidizers (CPO) and continuous soot combustion catalysts (CSCC). These need to be carefully matched to the individual vehicle and usually require very low sulfur fuel. They are capable of significant reductions of PM and, to a lesser extent, NOx emissions.

Keywords: *Retrofit aftertreatment system, diesel oxidation catalyst, diesel particulate filter, catalytic particulate oxidizer.*

INTRODUCTION

While new legislation worldwide requires newly manufactured heavy-duty diesel engines to meet tough new emission standards, there have been no major regulatory actions to similarly clean up diesels that are in use today. Because of the long service life of heavy-duty diesels, cleaning up exhaust gases of in-use heavy-duty diesel vehicles would certainly lead to improvements in air quality on the short term.

Retrofit exhaust aftertreatment technologies have emerged in recent years and are increasingly being utilized. Such systems need to be carefully matched to the individual vehicle and may require very low sulfur fuel. When optimized, they are capable of very significant reductions of PM and, to a lesser extent, NOx emissions. The combination of retrofit controls, new engine designs, low-sulfur fuels and advanced lubricants would assist in minimizing urban air pollution by diesel exhaust, while providing the durability and efficiency required from heavy-duty vehicles.

2. AVAILABLE AFTERTREATMENT TECHNOLOGIES SUITABLE FOR RETROFITTING

Diesel aftertreatment technologies could be divided into two main groups: systems designed to mitigate PM emissions and systems aimed at reduction of NO_x emissions. Of course, their combination is possible too. Not all currently available aftertreatment technologies are suitable for retrofitting in-use vehicles. Some of them would require adaptations of the engine control strategy and other complicate and multiple changes to be introduced. Such complicated systems are not well suited for retrofitting, due to the associated costs and the potential loss of warranties from the vehicle manufacturer. Most of diesel aftertreatment technologies suitable for retrofitting are focused on reduction of PM emissions, and to less extent – NOx emissions.

Further review and analysis will be focused only on PM reduction technologies. Due to the fact that this survey is aimed at analysis of issues relevant for retrofit applications, and taking into account the available information - see, for example, state-of-the-art reviews of Johnson (2002, 2003), subjects such as optimization of filtration processes, catalyst coatings, etc, will not be covered by here. Technologies aimed at PM reduction that are suitable for retrofitting in-use heavy-duty diesel vehicles (HDDV), could be classified into three groups: diesel oxidation catalysts (DOC), diesel particulate filters (DPF), and catalytic particulate oxidizers (CPO). These systems are described in the following chapters.

2.1. Diesel oxidation catalysts

Diesel oxidation catalysts are similar in their design to the widely used gasoline three-way catalytic converters, but operate in an oxygen-rich environment. This results in their ability to stipulate oxidation reactions only. Therefore, DOCs assist in further oxidation of engine-out CO, HC and the soluble organic fraction (SOF) of particulates. The lower SOF reduces the particulate mass. At high exhaust gas temperatures, DOC may tend to produce sulfates with adverse effects on the PM mass emissions. The Main desirable chemical reactions that occur in a DOC are:

$$2CO + O_2 \longrightarrow 2CO_2$$
$$2HC + 2.5O_2 \longrightarrow 2CO_2 + H_2O$$
$$SOF + O2 \longrightarrow CO_2 + H_2O$$

Typically, diesel oxidation catalysts were found to provide PM reduction of up to 50%, Brown et al. (1997). Data were presented, which indicates that DOC reduces significantly also the particle number on at least 11 of 13 particulate size bands, Brown (1997). It is noted that DOC contributes also to significant reduction of polycyclic aromatic hydrocarbons (PAH) and other toxic hydrocarbon emissions, Khair and McKinnon (1999).

2.2. Diesel particulate filters

Diesel particulate filters, or particulate traps, are very efficient in filtering fine particulates. A DPF system usually contains a filter positioned in the exhaust stream and designed to collect a significant fraction of the particulate emissions, while allowing the exhaust gases to pass through the system. The main technological challenge concerning DPF is controlled regeneration of the filter (burning of the trapped particles), where the particle load has to be burned below temperatures critical for damaging the filter material. Without or with delayed regeneration, the filter becomes blocked, which rapidly increases the exhaust gas backpressure. To start the filter regeneration process, temperatures above 300°C are necessary in modern systems. Such temperature levels do not occur under all loads for today's HD engines. A rather low overload of only 3-4 grams per liter of filter volume causes a rise of the regeneration temperature in the order of 300-400°C. Such temperatures can damage the filter, Hausberger (2003).

The requirements for filter material in terms of high trapping efficiency, together with low hydraulic resistance, thermal stability and acceptable cost, are quite challenging. The following surface-rich structures are found to be suitable, Mayer et al. (2000), Mayer (2003): ceramic monolithic-porous cell filter or foam, highly alloyed porous sintered metals or metal foams, filament-structures like fleeces, winded yarn or textile webbing (knitted or wicker-work) of ceramic or metallic fibers.

Many procedures of regeneration have been developed, which may be classified as so called active (if regeneration is triggered by external energy supply) and passive (if soot burn reaction is started due to the exhaust gas temperature occurring during real-world driving). Active regeneration is usually based on using various fuel burners or electric heaters. Such systems are complex, very expensive, and therefore less attractive for retrofit purposes. Their main benefit is in the possibility to ensure DPF regeneration at any operation condition. Increasing the exhaust gas temperature by means of engine combustion controls is used frequently for original equipment (OE), but most often not applicable for retrofitting. Detailed descriptions of active regeneration procedures have been published, e.g. Johnson (2002), Mayer et al. (2000), Mayer (2003). In the present review, the analysis is focused on passive regeneration methods that receive growing attention in various retrofit applications. The most popular methods of passive regeneration are: fuel borne catalysts (fuel additives), catalytic coating of the filter or a pre-catalyst to increase the NO_2 fraction in the NO_x for soot burn facilitation and combinations of these methods. It is noted that omitting active regeneration may bee risky: filter regeneration could be not sufficient if the vehicle operation conditions do not result in reasonably high exhaust gas temperatures. Thus, at least a monitoring device for the exhaust gas backpressure is recommended if passive regeneration is used alone.

Fuel borne catalysts (FBC) have the ability to substantially lower the soot ignition temperature and increase its burn-off rate. They are mostly elements from the so-called transition metals. Some typical examples are cerium, iron, copper and platinum. Today additives allow filter regeneration at temperatures of about 300°C, Valentine et al. (2000). A principal disadvantage of FBC is that the oxides of the additive substances are deposited in the filter, thus gradually increasing the backpressure. Of course, use of FBC requires an on-board dosing system. Special care should be given to verify the absence of secondary emissions. An appropriate procedure is applied in Switzerland and called VERT Secondary Emissions Test, Mayer et al. (2002). The list of verified FBCs is published by SAEFL (2004). Table 1 presents these verified additives together with their maximal dosing rates.

Table1: Fuel borne catalysts verified by SAEFL (2004).

Manufacturer	Additive trade name	Effective element	Max. dosing rate, mg metal/kg fuel
OCTEL	Satacen	Fe	36
OCTEL	OCTIMAX	Fe + Sr	25
Rhodia	EOLYS	Ce	100
Rhodia	EOLYS-2	Ce + Fe	17
Clean Diesel Technologies	DFX-DPF	Ce + Pt	8

FBC could also be used alone or in combination with DOC, in order to achieve better PM reduction efficiency, US EPA Voluntary Retrofit Program (2005).

Use of NO_2 for soot burn facilitation was initially suggested by Johnson Matthey in 1989 and known worldwide as the continuously regenerating trap (CRT) technology, Cooper and Thoss (1989). This system is usually comprised of a platinum based oxidation catalyst installed upstream of a non-catalyzed wall flow particulate filter. The Pt catalyst stipulates oxidation of NO in the exhaust stream to form NO_2. Soot oxidizes with NO_2 at much lower temperatures than with O_2. The regeneration in CRT systems starts at approx. 300°C. This enables DPF regeneration under many typical operation conditions of a heavy-duty diesel engine. The main reactions are:

Catalyst: $2NO + O_2 = 2NO_2$

DPF: $C + NO_2 = CO_2 + 2NO$

$C + O_2 = CO_2$

High efficiency of CRT is quite well documented: it allows up to 99% PM removal, together with deep reduction of CO and HC emissions (Fig. 1), Chatterjee et al. (2002).

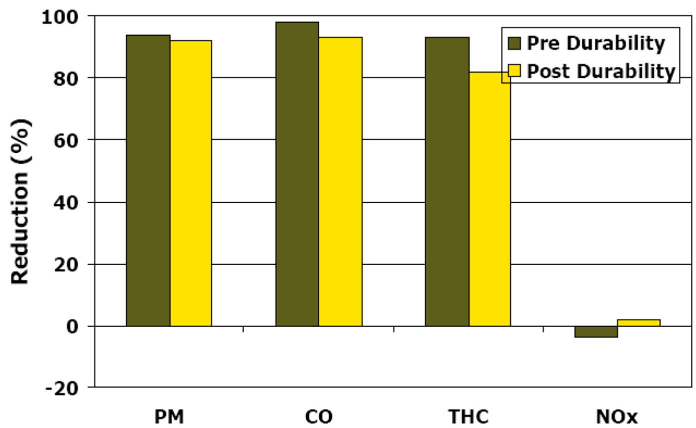

Figure 1: Emissions reduction comparison for New York bus under New York Bus Cycle, Chatterjee et al. (2002). "Pre-durability" – measurements carried out before field tests; "Post-durability" - measurements carried out after 9-12 months of bus operation.

Main drawbacks of CRT technology are sensitivity to sulfur content in a fuel, clogging tendency because of ash accumulation (these issues are discussed below) and the requirement for enough amount of NO_x in the exhaust gases - NO_x/PM ratio of at least 8 must be provided, as mentioned in the US EPA Voluntary Retrofit Program (2005). In the project PARTICULATES (5th EU-Framework program) extensive research on the formation of the nucleation mode was done, e.g. Thompson et al. (2004). It showed that the nucleation mode mainly consists of droplets without carboneous nucleus, which disappears if the exhaust gas is heated. Thompson et al. (2004) also found a pronounced number of nucleation particles for HD engines (EURO 2 and EURO 3) retrofitted with CRT systems. The nucleation mode was high using diesel fuel with both 38ppm and 8ppm sulfur content. Only with Swedish Class 1 diesel (3 ppm sulfur) the nucleation mode was suppressed. Fig. 2 shows that solid particle numbers were reduced by approx. 2 orders of magnitude by retrofit CRT, while the potential increase of nucleation particles can compensate for the lower number of solid particles in the total number emission. In general, low dilution of the exhaust gas and cold conditions increase the tendency for nucleation formation. Thus the nucleation may have much less relevance in real-world driving than in test bed measurements due to the much higher exhaust gas dilution compared to the test bed.

Filter catalytic coating. The application of a transition or precious metal coating applied to the surface of a filter reduces the ignition temperature necessary for oxidation of the particulates. No further measures are necessary, if the pertinent engine exhaust temperatures are attained sufficiently frequently and sufficiently long. This is a completely passive regeneration method, where regeneration occurs at a catalytically coated surface. Soot ignition temperatures are reduced to the values similar to or even lower than those obtained by using FBC, Johnson (2002), Mayer et al. (2000). Since ashes from fuel additives are avoided, such DPF systems can have longer maintenance intervals. Other demonstrated benefits of catalyzed filters over systems using FBC are their simplicity, reduced size and lower cost. The catalytic coating leads to deep conversion of CO and HC emissions together with high PM reduction efficiency (usually, around 90%).

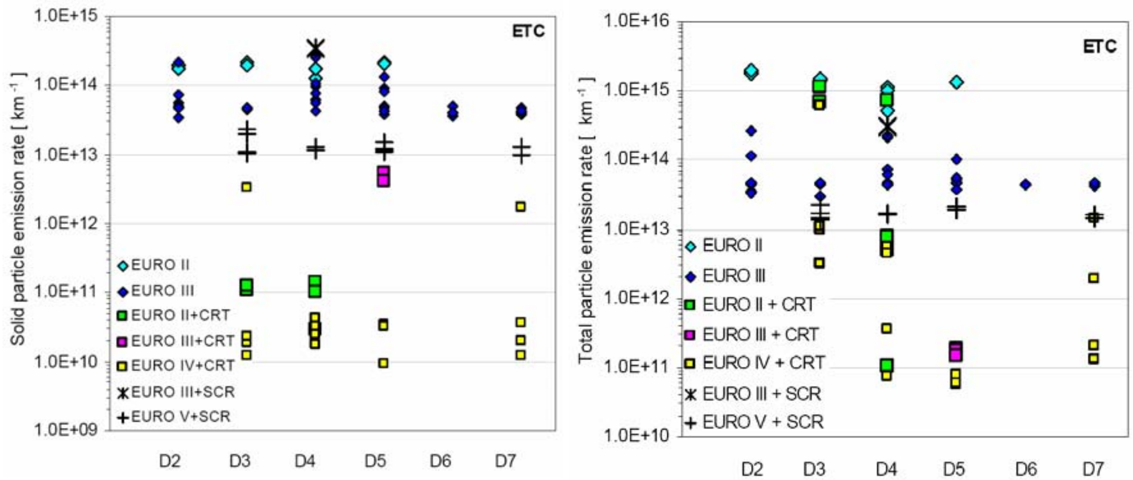

Figure 2: Total solid particle emissions (left) and total particle emissions including the nucleation mode (right) measured by Thompson et al. (2004), units in [#/kWh].

Combinations of DPF regeneration methods have recently gained rising popularity. A typical example of such a combination is the so-called catalyzed continuously regenerating trap (CCRT) developed by Johnson Matthey Inc., Chatterjee (2004), and verified by EPA and CARB in 2004 for retrofit applications, CARB (2005), US EPA Voluntary Retrofit Program (2005). The CCRT is a combination of CRT with a filter catalytic coating. According to Chatterjee (2004), its main advantages over CRT are the ability to successfully operate at low exhaust gas temperatures (200 – 250^oC) and lower NO_x/PM ratios.

2.3. Catalytic particulate oxidizers

Catalytic particulate oxidizers (CPO), continuous soot combustion catalysts (CSCC) or particulate catalysts (POC)[1] have been developed to overcome one of the main DPF drawbacks – increase in backpressure and possible filter plugging, together with achievement of higher PM reductions compared to DOC technology. CPO's can have a (limited) storage capacity for particles either on the surface of the catalyst, due to a special coating, or in a special storage medium. Like the catalytic coated DPF, the CPO's oxidize the particles if a sufficiently high exhaust gas temperature is reached. In contrast to DPF, the open structure of CPO can prevent plugging (Fig. 3). If the given storage capacity is exploited, e.g. due to long operation at low exhaust gas temperatures, the particles in the exhaust gas will simply pass the CPO but they shall not contribute to its overload. Thus the exhaust gas backpressure shall not exceed critical values even under worst conditions.

[1] Several different names exist now for similar systems. A key characteristic is an open structure where the exhaust gas is not flowing through a wall.

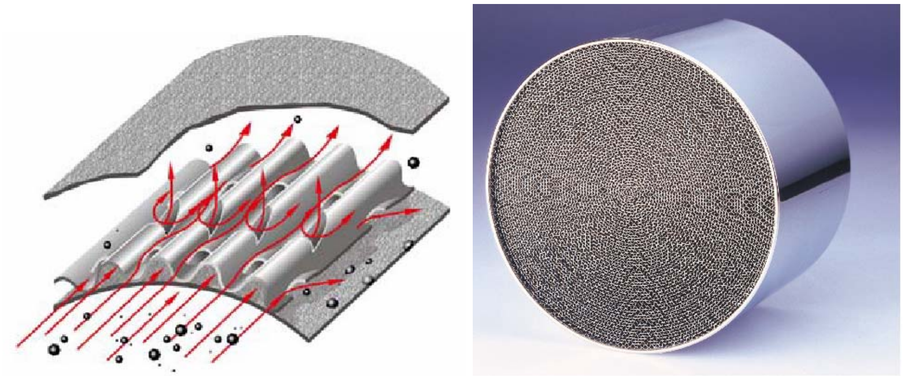

Figure 3: Operational scheme of particulate catalysts (Source Emitec).

Experience with particle catalysts as retrofit systems has been gained in the last five years. Tests in buses in Styria showed 40% to 70% reduction in particle mass (Fig. 4) and solid particle number emissions for Particle Oxidation Catalysts from Emitec, Pankl and Oberland.Mangold in real-world bus cycles, e.g. Hausberger (2003), Hausberger and Vuckovic (2005). Particulate number emissions in the nucleation mode were reduced typically by more than 90% by these systems. Durability tests over approx. 70.000 km were performed, which showed a steady or even increasing efficiency of these systems. Results from longer durability runs are not known to the authors.

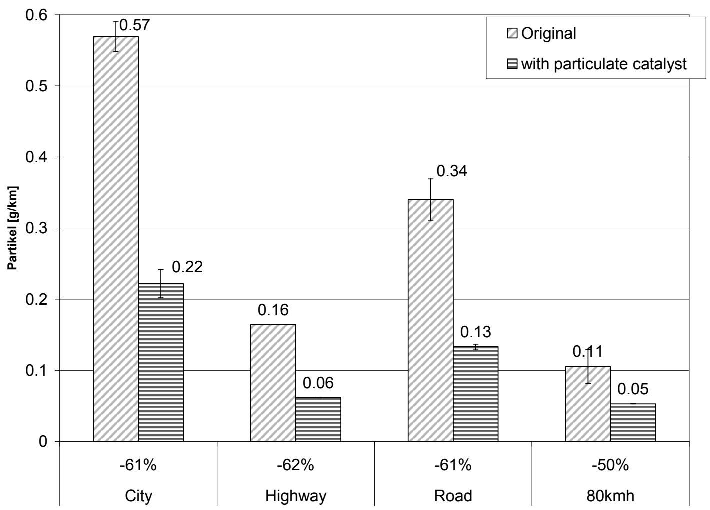

Figure 4: Particulate mass emissions: Setra S315H bus; different real-world bus cycles with & without a Pankl-POK particulate catalyst, Hausberger & Vuckovic (2005).

According to ETG (former Erland Nilson AB), the CPO can oxidize soot at much lower exhaust gas temperatures (≥200^{o}C) than conventional wall-flow monolith based filters. The system is maintenance-free, as ashes shall not accumulate in the filter. The CPO device was recently tested by the AVL MTC laboratory and was approved for use in the Swedish Environmental Zones retrofit program. Currently it is undergoing VERT verification procedure. Although the particulate reduction the CPO is less than of the DPF, the easier handling (according to the manufacturers no active regeneration is necessary) and usually lower cost make particle catalysts attractive for the retrofit market.

3. TECHNICAL ISSUES TO BE CONSIDERED IN RETROFITTING

When retrofitting diesel aftertreatment technologies to in-use vehicles, several factors should be considered. These include: the vehicle application and operation conditions, fuel quality, lubricant quality and vehicle maintenance, MECA (2002). They will influence the selection of an appropriate aftertreatment technology. The emission reduction target and system cost will also play an important role in technology selection.

3.1. Compatibility with vehicle operation conditions

Different aftertreatment technologies require different vehicle working conditions for their effective and durable operation. Not any aftertreatment system could be retrofitted to any vehicle. A careful selection process is a must for successful retrofit. Table 2 contains some compatibility conditions for few examples of various aftertreatment technologies that have been verified by US EPA, CARB or VERT, SAEFL (2004), CARB (2005), US EPA Voluntary Retrofit Program (2005).

Table 2: Compatibility conditions for various aftertreatment technologies.

Technology	Manufacturer	Exhaust gas temperature, ^{o}C	Engine-out NOx/PM ratio	Verified by
DOC+FBC	CDT	≥225 during 15% of the duty cycle	Not specified	EPA
DOC	Johnson Matthey	≥150 during the duty cycle	Not specified	EPA, CARB
DPF+FBC	CDT	≥225 during 20% of the duty cycle	Not specified	EPA
CRT	Johnson Matthey	≥275 during 40-50% of the duty cycle	Min – 8 Optimal ≥20	EPA, CARB, VERT
Catalyzed DPF	Lubrizol ECS	≥280 during 25% of the duty cycle	Not specified	EPA, CARB, VERT
CCRT	Johnson Matthey	≥210 during 40% of the duty cycle	Min – 8 Optimal ≥20	EPA, CARB
CPO	ETG	≥200	Not specified	Certified for Swedish Envir. Zones

Some conditions such as requirements that the engine should be well maintained and have oil consumption as prescribed by the manufacturer, are common for all vehicles that are considered for retrofit of any aftertreatment system. As can be seen from Table 2, CCRT and CPO technologies allow engine operation at lowest temperatures.

3.2. Fuel quality

Sulfur contained in automotive diesel fuels influences negatively performance and durability of aftertreatment systems, Zvirin et al. (2003). This influence is quite well documented and widely discussed. Therefore, most aftertreatment technologies require, for their successful operation, ultra-low sulfur diesel fuel (ULSDF) that contains no more than 50 ppm sulfur with a recommendation to use a fuel with no more than 10-15 ppm sulfur. 50 ppm sulfur fuel is prescribed today over the EU, 10 ppm sulfur fuel is becoming increasingly available over Europe and worldwide too. For those markets, where ULSDF is still unavailable, some aftertreatment technologies tolerant to sulfur content have been developed. Most of DOCs, as well as CPO, permit operation with fuels containing up to 350-500 ppm sulfur. This results from the fact that catalyst formulations have been developed recently that selectively oxidize SOF, while inhibit oxidation of the sulfur dioxide, MECA (2002).

3.3. Lubricant quality

The impact of lubricants on engine-out particulate emissions, and especially on deterioration in performance of DPFs, is much less studied than the impact of fuel quality, Froelund and Yilmaz (2002). The DPF is sensitive to organo-metallic ash derived from Calcium- and Magnesium-containing additives in the lubricant oil. These ashes melt at high temperatures (>1100°C) during regeneration and can react with the filter substrate and clog the filter permanently (glazing effect). Catalyst deactivation is possible as a result of lubricant oil Zinc- and Phosphorous containing ash, Froelund and Yilmaz (2002). It is clear now that ash management is an issue, and DPF manufacturers work on increase of ash storage capacity in their filters, as well as on methods of continuous ash removal using natural vehicle or engine vibrations, Johnson (2003). It is noted that some CPOs benefit from reduced permanent ash retention, ETG (2004). On the other hand, the best way to mitigate ash-in-DPF issue is to minimize ash generation. This can be done by reduction of both oil consumption and lubricant oil ash content.

Traditionally, ash content of heavy-duty diesel engine oil has been in the range of 1.2 up to about 2.0%. With the introduction of the OEM guidelines for Low Emission Diesel Lubricants, the maximum permissible amount of ash should be 1.0% for heavy-duty applications, Takeuchi et al (2003).

4. COST ISSUES

The Manufacturers of Emission Controls Association (MECA) has estimated costs for retrofit diesel aftertreatment technologies, MECA (2000, 2002). The information taken from their publications is given below for DOC and DPF technologies. Diesel oxidation catalysts are estimated to cost from $425 to more than $1,750 per DOC, depending on engine size, sales volume and whether the installation is muffler replacement or an in-line installation. In most cases, oxidation catalysts are easy to install. Installations typically take less than 2 hours. Diesel particulate filters are sold for about $3,000 to more than $11,000 each. The prices vary depending on the size of the engine, the number of vehicles being retrofitted, the amount of particulate matter emitted by the engine, the emission target to be achieved, the regeneration method and other factors. Filters that are sold as muffler replacements generally cost more that in-line filters. Detailed costs analysis has been performed in Switzerland for DPF, Mayer et al. (2000). Total retrofit costs were divided into purchase cost, installation costs, cost of monitoring systems and operating cost. A summary of their findings appears in Table 3.

Table 3: Summary of costs for DPF retrofitting in US Dollars, Mayer et al. (2000).

Cost component	Engine power 100 kW	Engine power 300 kW
Purchase	2,700	7,100
Installation	600	900
On-board monitoring	600	
Service	600 (every 100,000 km)	
Disposal	Manufacturer's responsibility	
Replacement	80% of purchase price	

It is clear from the data shown above that the successful market penetration of HDV retrofit programs would be achieved only if valuable incentives are suggested to vehicle operators.

5. SUMMARY

Diesel oxidation catalysts (DOC) were found to provide PM emission reduction of up to 50%. The efficiency increases with higher shares of the soluble fraction on PM and decreases at higher sulfur content of the fuel, together with high exhaust gas temperatures, due to formation of sulfates. Diesel oxidation catalysts have been one of the most popular control options for both on-road and off-road applications to date because of their low cost, maintenance-free service and negligible impact on vehicle fuel economy.

Diesel particulate filters, or particulate traps, are very efficient in filtering of particulates. VERT-certification carried out by the Swiss Environmental Agency revealed results of trapping by more than 99% of particulates emission. Diesel particulate filters retrofits have rapidly expanded during the last years, mainly due to their very high efficiency of PM reduction and recently achieved high level of durability and reliability in suitable applications. Retrofit of DPF usually leads to fuel economy penalty of between 1% and 3%. A main technical target is to enable sufficient regeneration of the DPF, i.e. a continuous or periodical oxidation of the trapped particles, to prevent overloading of the filter which would result in increased fuel penalties and potential damages.

The Catalytic Particulate Oxidizer (CPO or particulate catalyst) technology is characterized by open structures, which can prevent overloading with particles under insufficient thermal operating conditions. This feature is an advantage, especially for retrofit systems where active regeneration most often would need an external heater. Particulate catalysts can usually achieve particulate emission reductions lower than DPF but higher than DOC.

Most DOCs, as well as CPOs, permit operation with fuels containing up to 350-500 ppm sulfur. For successful operation of DPF, fuel with no more than 50 ppm sulfur is required. The DPF and maybe also some CPOs are sensitive to ashes from additives in the lubrication oil and in the fuel, since these ashes accumulate in the filter and increase the backpressure (e.g. organo-metallic ash). New specifications are being developed recently for engine lubrication oil with reduced ash content that will be compatible with aftertreatment devices.

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