

Detailed Effects of a Diesel Particulate Filter on the Reduction of Chemical Species Emissions

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ABSTRACT

Diesel particulate filters are designed to reduce the mass emissions of diesel particulate matter and have been proven to be effective in this respect. Not much is known, however, about their effects on other unregulated chemical species. This study utilized source dilution sampling techniques to evaluate the effects of a catalyzed diesel particulate filter on a wide spectrum of chemical emissions from a heavy-duty diesel engine. The species analyzed included both criteria and unregulated compounds such as particulate matter (PM), carbon monoxide (CO), hydrocarbons (HC), inorganic ions, trace metallic compounds, elemental and organic carbon (EC and OC), polycyclic aromatic hydrocarbons (PAHs), and other organic compounds. Results showed a significant reduction for the emissions of PM mass, CO, HC, metals, EC, OC, and PAHs.

INTRODUCTION

For many years, diesel engine manufacturers have been working extensively to develop aftertreatment technologies that are capable of significantly reducing the emission levels of each of the primary pollutants which are considered to be a risk to the environment and public wellbeing. The majority of these aftertreatment technologies have been designed with the most consideration being given to compounds such as particulate matter (PM), oxides of nitrogen (NO_x), carbon monoxide (CO), and hydrocarbons (HC). However, it is

also important to understand what effects these technologies may have on the emission levels of other chemical species which are of toxicological concern, such as certain polycyclic aromatic hydrocarbons (PAHs), aldehydes, and the other species found in Table 1, due to a variety of physical processes or chemical reactions [1, 2].

One primary aftertreatment technology commonly used to reduce the PM emission levels of diesel exhaust is the diesel particulate filter (DPF). There are a variety of DPFs available. For example, the DPF could be made of composite fibrous materials which utilize fine fibers as a means of particle filtration. The DPF may also be an extruded wall-flow monolithic filter, which is commonly made of cordierite or silicon carbide material. The high filtration efficiency exhibited by wall-flow filters is achieved through cake formation on the filter surfaces [3]. As these cake layers collect particles, the overall filtration efficiency of the DPF improves at the cost of increased pressure drop across the filtration media. To reduce this pressure gradient across the filter it is often necessary to remove the cake layer through filter regeneration.

Filter regeneration is a necessary process to maintain continuous operation when a DPF is fully loaded. Regeneration is the process by which the soot deposits on the DPF are removed through oxidation to reduce the pressure drop across the filter. While regeneration can occur passively through reactions involving O₂ already

Table 1. List of Mobile Source Air Toxics (MSATs) [4]

Acetaldehyde	Diesel Particulate Matter + Diesel Exhaust Organic Gases (DPM + DEOG)	MTBE
Acrolein	Ethylbenzene	Naphthalene
Arsenic Compounds	Formaldehyde	Nickel Compounds
Benzene	n-Hexane	POM (See Table 2)
1,3-Butadiene	Lead Compounds	Styrene
Chromium Compounds	Manganese Compounds	Toluene
Dioxin/Furans	Mercury Compounds	Xylene

present in diesel exhaust at high temperatures, it is often necessary to actively initiate the regeneration [1]. Active regeneration may be achieved through hydrocarbon dosing or through electrical heating of the filter elements [1].

The DPF which was used in the present study consisted of a diesel oxidation catalyst (DOC) with dimensions of 26.7 cm (10.5") by 10.2 cm (4"), which converts NO to NO₂ to aid in the process of filter regeneration [1], and a catalyzed soot filter (CSF) with dimensions of 26.7 cm (10.5") by 22.9 cm (9"), which provides a high efficiency of PM filtration. Additionally, the catalyst coated DOC and CSF have the further benefit of reducing the emissions of gaseous CO, HC, and other organics through oxidation to form CO₂ and H₂O [5, 6].

Regulatory organizations, such as the U.S. Environmental Protection Agency (EPA) and the California Air Resources Board, have identified a wide range of compounds which may pose considerable risk to the environment and public health. The EPA has developed a list of Mobile Source Air Toxics (MSAT) which contains a variety of compounds including fine particulate matter, aldehydes, and polycyclic organic matter (POM) (see Table 1) [4]. The POM component, found in Table 2, contains 16 species including seven PAHs which have been identified by the EPA as carcinogenic [4].

Table 2. Polycyclic Organic Matter (POM) [4].

Acenaphthene	Chrysene
Acenaphthylene	Dibenz(a,h)anthracene
Anthracene	Fluoranthene
Benz(a)anthracene	Fluorene
Benzo(a)pyrene	Indeno(1,2,3-cd)pyrene
Benzo(b)fluoranthene	Naphthalene
Benzo(ghi)perylene	Phenanthrene
Benzo(k)fluoranthene	Pyrene

The compounds found in diesel engine exhaust are typically present in both the gas and particle phases depending upon the chemical composition of the particular species [1, 7]. Although a significant number of these compounds are considered to be derived from diesel fuel and lubrication oil, previous studies have shown that some species may also result from thermal alteration at high exhaust temperatures, whether found in-cylinder or during aftertreatment system regeneration [7, 8, 9, 10].

The goal of the present study was to perform a comprehensive analysis of the diesel engine exhaust products in order to gain a full understanding of the effects that DPF technology has upon these emissions. The chemical species analyzed included the criteria air pollutants, PM, CO, and HC, as well as a large variety of particulate and gaseous organic compounds.

EXPERIMENTAL SECTION

TEST ENGINE, LUBE OIL, AND FUEL - To determine the effects of DPF technology on the emissions of a wide spectrum of chemical species emissions, a heavy-duty diesel engine (435 HP), without exhaust gas recirculation (EGR), was tested both with and without the DPF aftertreatment system.

During testing, the engine was lubricated with Shell Rotella T 15W-40. Additionally, the test engine was fueled with certified diesel fuel supplied by Chevron Phillips. The fuel was #2 certified diesel which was determined to have a cetane number of 47, a sulfur content of 308.5 ppm, a particulate matter content of 0.1 mg/liter, and an aromatic composition of 29.5%. The experiments for the present study were conducted prior to the implementation of ultra low sulfur diesel (ULSD) in the U.S. Although the test engine and diesel fuel used in the present study did not correspond with current practices, this study did not concentrate on the effects of the engine or fuel content and thus the engine and fuel were used only as sources of chemical pollutants.

TESTING AND SAMPLING PROCEDURE - After warming the test engine for 2 hours at ISO mode 8 (50% load, intermediate speed), blank samples were collected to check for contamination from the dilution air and handling procedures [11]. During sampling, the engine was operated under the transient Federal Test Procedure (FTP) which simulates typical New York and Los Angeles freeway and non-freeway conditions over a 20 minute cycle [4]. After each FTP cycle, additional blank samples were collected during the remaining testing processes including background, storage, and transportation. Once testing was completed, all samples collected from the source sampling system as well as the blanks were stored and transported in temperature and humidity controlled containers to minimize the effects of external conditions.

The engine was first tested with the DPF in place, and the backpressure was measured and recorded with a data acquisition system. The backpressure was later matched by an electronic restrictor valve while the engine was being operated without the DPF. This ensured that the engine was operating under similar conditions for each testing configuration and reduced the amount of variation between them.

At least three separate samples were collected from the source sampling system for each test configuration to determine the sample variation. The source sampling system utilized a constant volume flow to minimize the effects of variable exhaust gas temperature [12]. As seen in Figure 1, the system consisted of a critical flow venturi-constant volume system (CFV-CVS) primary dilution tunnel, secondary micro-diluter, residence time chamber (RTC), and multiple sampling trains capable of collecting both gaseous and particulate emissions. The entire exhaust flow was introduced to the primary dilution tunnel where it was allowed to thoroughly mix

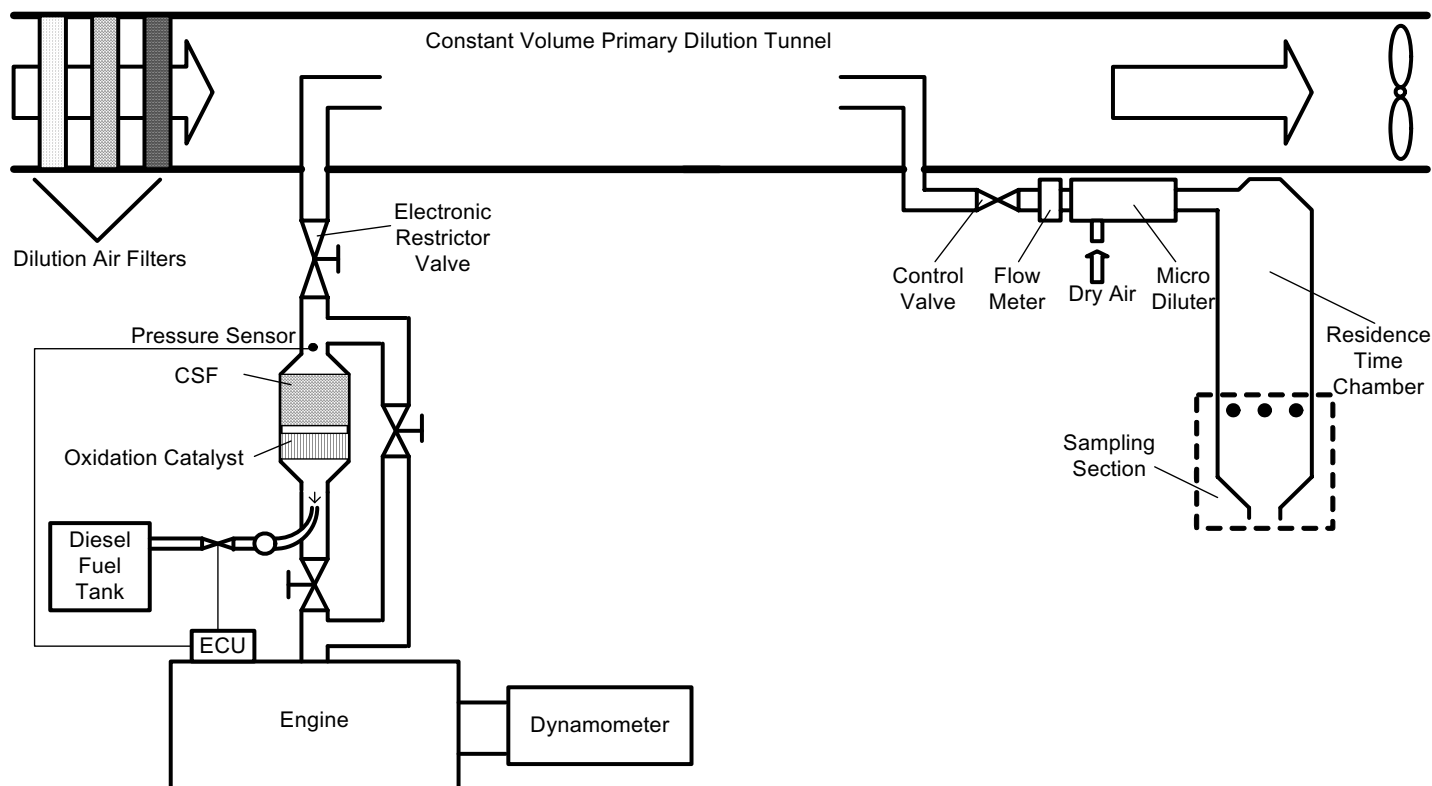


Figure 1. Source Sampling System Used for Sample Collection.

with the pre-cleaned dilution air. By introducing the entire exhaust flow in the primary dilution tunnel, the system is less prone to sampling error than systems which extract a partial sample of the exhaust flow for the primary dilution.

From the CFV-CVS tunnel, a partial sample was extracted by a secondary micro-diluter which was utilized to achieve the final dilution ratio and stabilize the samples. Mixing within the secondary micro-diluter was aided through the use of a static pressure chamber which allowed for uniform distribution of the dilution air.

A residence time chamber was incorporated as part of the source dilution sampling system to optimize the sample partitioning. The RTC allowed for the homogeneous and heterogeneous nucleation of particles to occur. It was determined that a residence time of 30 seconds allowed for the diffusion limited transport of gaseous organics onto existing particles. Additionally, the RTC enabled the exhaust flow to become uniformly distributed before samples were extracted.

The sampling trains were connected to the sampling section of the RTC through isokinetic sampling probes which were aligned with the flow direction and were sufficiently spaced to avoid interference [12]. Each of the sampling probes was made from thin walled stainless steel tubing and the edges of the tubing were sharpened to minimize particle loss due to partial sampling [13, 14].

The sampling trains utilized several types of media for the collection of exhaust samples. Particulate samples were collected with quartz and polytetrafluoroethylene (PTFE) filters while gaseous samples were collected using polyurethane foam (PUF) arranged as seen in Figure 2. Further, additional gaseous samples were collected using thermal desorption (TD) tubes and dinitrophenylhydrazine (DNPH) cartridges as seen in Figure 3. The high-volume sampling train, also seen in Figure 3, allowed for the quantification of chemical species which had ultra-low concentrations within the exhaust flow. Lastly, PTFE-coated aluminum PM_{2.5} cyclones were used upstream of the sampling media to pre-classify PM in the exhaust gases.

ANALYSIS PROCEDURES - Chemical speciation of the collected samples was conducted by the Wisconsin State Laboratory of Hygiene. Particulate emissions such as soot, trace metals, inorganic ions, and particle-phase organics were captured using quartz and PTFE membrane filters which were first weighed for total PM mass before being subjected to a more comprehensive chemical speciation. In order to obtain a mass measurement for the total particulate matter collected from the source dilution sampling system, two separate sets of 47 mm PTFE membrane filters were weighed in a draft and static free room with a robotic filter weighing apparatus which was certified to 2007 regulations and beyond.

Analysis of elemental and organic carbon (EC and OC) samples in the particulate matter was performed using a thermal-optical method as described by Birch and Cary