

A Study on the Emissions of Chemical Species from Heavy-Duty Diesel Engines and the Effects of Modern Aftertreatment Technology

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ABSTRACT

A comparative analysis was made on the emissions from a 2004 and a 2007 heavy-duty diesel engine to determine how new engine and emissions technologies have affected the chemical compounds found in the exhaust gases. Representative samples were collected from a source dilution sampling system and analyzed for both criteria and unregulated gaseous and particulate emissions. Results have shown that the 2007 regulations compliant engine and emissions technology not only reduced the specifically regulated exhaust pollutants, but also significantly reduced the majority of unregulated chemical species. It is believed that these reductions were achieved through the use of engine optimization, aftertreatment system integration, and ultra-low sulfur diesel fuel.

INTRODUCTION

In an effort to make diesel engines more friendly to the

environment during every day operation, engine manufacturers have begun to design and implement technology to achieve significant reduction of diesel emissions. Primarily, these emission control technologies

have been developed to reduce the amount of criteria pollutants, such as particulate matter (PM), nitrogen oxides (NO_x), carbon monoxide (CO), and hydrocarbons (HC). However, it is also important to understand what effects may occur for the emission levels of other chemical species which are of toxicological concern, such as certain polycyclic aromatic hydrocarbons (PAHs) and aldehydes.

Systematic approaches to the reduction of diesel emissions may include combustion optimization, exhaust gas recirculation (EGR), and aftertreatment systems. Each of these technologies was mainly designed to reduce the emission levels of the criteria pollutants, PM and NO_x. Additionally, these systems may have an effect on the levels of unregulated species due to a variety of physical processes or chemical reactions [1, 2].

Table 1. List of Mobile Source Air Toxics (MSATs) [3]

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

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Regulatory organizations, such as the U.S. Environmental Protection Agency (EPA) and the California Air Resources Board (CARB), have identified a greater 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) that contains a variety of compounds including fine particulate matter, aldehydes, and polycyclic organic matter (POM) (see Table 1) [3]. The POM component, found in Table 2, contains 16 species including seven PAHs which have been identified by the EPA as carcinogenic [3]. Additionally, a more comprehensive list of species has recently been developed as part of a program known as the Advanced Collaborative Emissions Study (ACES). Through the extensive work of several organizations including the U.S. Department of Energy, U.S. Environmental Protection Agency, the California Air Resources Board, the American Petroleum Institute, the Engine Manufacturers Association, the Health Effects Institute, and the Coordinating Research Council, the ACES program has identified more than 650 compounds which have been selected based upon established knowledge surrounding their toxicity and environmental impact.

Table 2. Polycyclic Organic Matter (POM) [3].

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, 4]. 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 also result from thermal alteration at high exhaust temperatures, whether found in-cylinder or during aftertreatment system regeneration [4, 5, 6, 7].

The goal of the present study was to perform a comparative analysis of the diesel engine exhaust products in order to gain a full understanding of the effects that the 2007 engine and emissions technology have in comparison with 2004 technology. The chemical species analyzed included the major PM constituents, EC, OC, ions, and metals, as well as a large variety of particulate and gaseous organic compounds.

EXPERIMENTAL SECTION

TEST CONFIGURATIONS - Two comparable Cummins heavy-duty diesel engines were tested to determine the effects of 2007 technology compared with 2004 technology. Each of the engines was equipped with the appropriate emissions technology necessary to meet the emission levels presented in Table 3.

Table 3. Test Engine Properties.

Property	2004 Engine	2007 Engine
Model	ISX 500	ISX 500
Rated Power	500 hp	500 hp
Displacement	15 L	15 L
Rated PM Level	0.1 g/bhp-hr	0.01 g/bhp-hr
Rated NO _x Level	2.5 g/bhp-hr	1.2 g/bhp-hr

Both the 2004 and 2007 ISX heavy-duty diesel engines were equipped with EGR systems to provide the necessary in-cylinder flame temperature and O₂ dilution ratio to help reduce NO_x emissions to levels below the regulatory limits. In addition, the 2007 ISX engine was equipped with a diesel particulate filter (DPF) and a crankcase ventilation (CV) coalescer to assist in the reduction of PM emissions.

Due to the low 2007 emission regulation for PM, a DPF system is utilized to reduce the PM emission levels by greater than 90%. The DPF consisted of a diesel oxidation catalyst (DOC), which converts NO to NO₂ to aid in the process of filter regeneration [1], and a catalyzed soot filter (CSF), which provides high efficiency 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 [8, 9].

Filter regeneration is a necessary process to maintain optimum diesel engine performance when a DPF is loaded. Filter regeneration is the process by which the soot deposits on the DPF are removed through combustion to reduce the pressure drop across the filter. While regeneration can occur passively through reactions involving O₂ already present in diesel exhaust or NO₂ produced from NO by an oxidation catalyst, it is often necessary to actively initiate the regeneration [1]. Active regeneration often involves hydrocarbon dosing for oxidation to generate sufficient heat, but can also be achieved through electrical heating of the filter elements [1].

New to the 2007 EPA regulations is the inclusion of engine crankcase emissions as part of the total tailpipe emissions. Crankcase emissions are primarily the result of leakage around the piston rings which force exhaust gases into the engine's crankcase. To reduce crankcase emissions, the 2007 engine included a CV coalescer to remove residual oil and water from the crankcase gases before they exit the engine.

During testing, each engine was lubricated with Valvoline Premium Blue 15W-40, which had a sulfated ash content of less than 1%. Additionally, the test engines were fueled with certified diesel fuel supplied by Chevron Phillips. The properties for each fuel were determined by Chevron Phillips and can be found in Table 4. Each of the fuels was certified to meet the appropriate standards for the corresponding engine model year.

Table 4. Fuel Properties for 2004 and 2007 Certified Diesel Fuel.

Property	2004 Fuel	2007 Fuel
API Gravity	36.2	36.2
Cetane Number	47	42
Sulfur Content (ppm)	308.5	9.2
PM Content (mg/L)	0.1	3.3
Aromatics (LV %)	29.5	32.0

TESTING AND SAMPLING PROCEDURE - After warming the engines 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 [10]. During sampling, the engines were 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 [3]. After each FTP cycle additional blank samples were collected during the remaining testing and post-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.

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 [11]. The system, shown in Figure 1, consisted of a primary dilution tunnel, secondary micro-diluter, residence time chamber (RTC), and multiple sampling trains capable of collecting both gaseous and particulate emissions. The sampling trains were connected to the sampling section of the RTC through isokinetic sampling probes, which were aligned with the flow direction to avoid interference as can be seen in Figure 2 [11]. 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 3. Further gaseous samples were collected using thermal desorption (TD) tubes, dinitrophenylhydrazine (DNPH) cartridges, and a high-volume sampling train with XAD cartridges as seen in Figure 4. The high-volume sampling train allowed for the quantification of chemical species that had ultra-low concentrations within the exhaust flow. Additionally, PTFE-coated aluminum PM_{2.5} cyclones were used upstream of the sampling media to pre-classify PM in the exhaust gases.

Chemical speciation of the collected samples was conducted by the Wisconsin State Laboratory of Hygiene and the Desert Research Institute. 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.

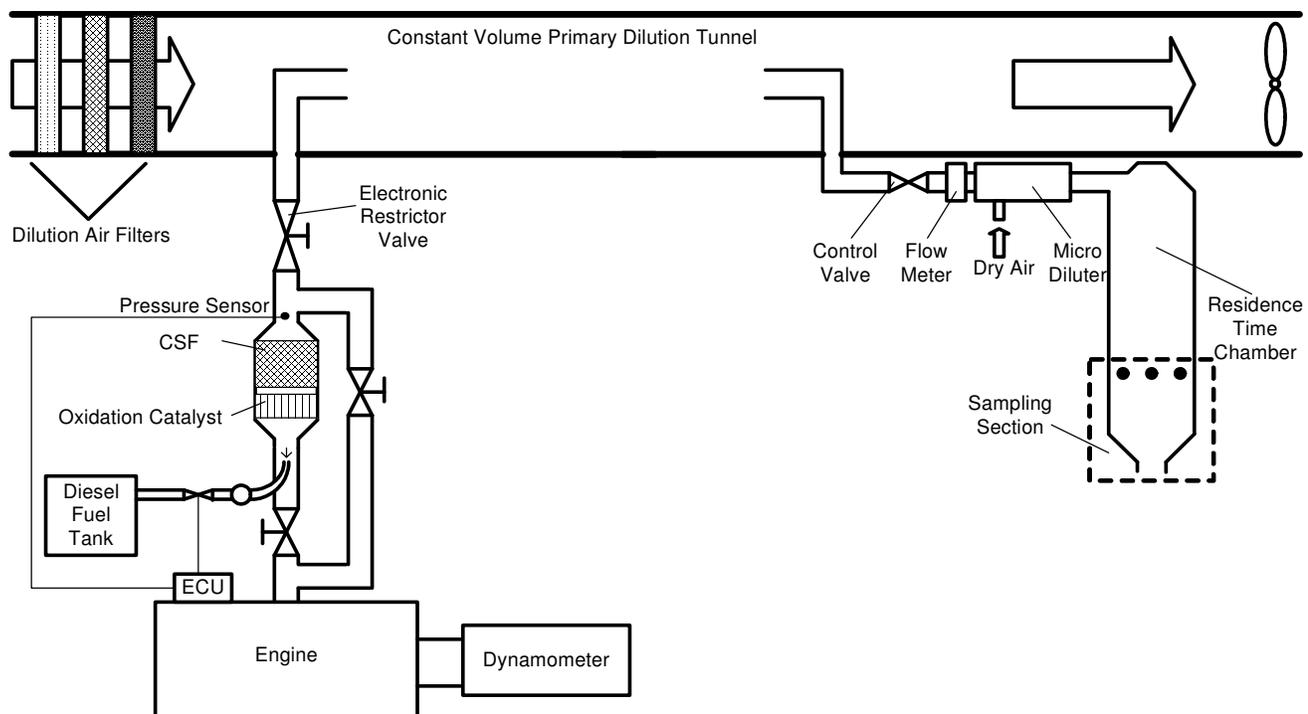


Figure 1. Source sampling system used for sample collection. (For the 2007 engine, crankcase emissions were vented into the exhaust downstream of the DPF and upstream of the electronic resistor valve.)

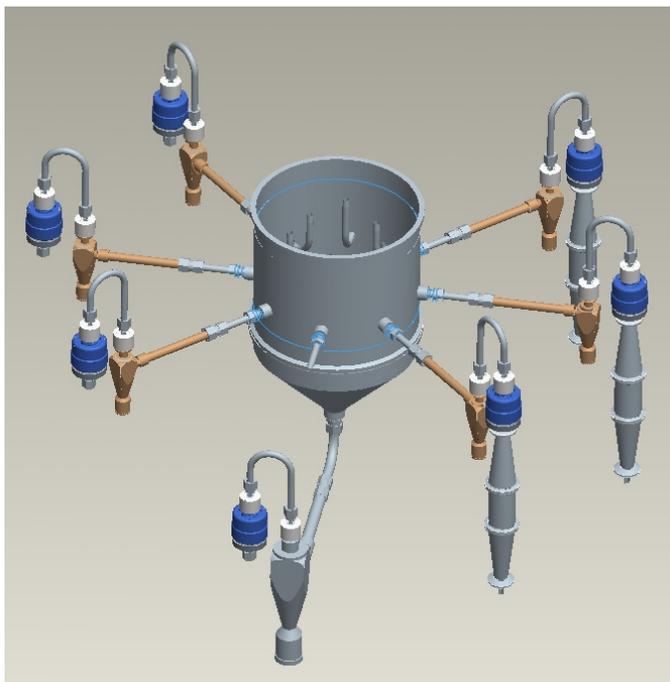


Figure 2. Sampling section of the residence time chamber.

Analysis of EC and OC samples in the particulate matter was performed using a thermal-optical method [12]. The filters were first heated in a helium atmosphere to evolve OC. The OC was then oxidized to CO₂ and reduced to methane before being measured by flame ionization detection. Pyrolytic conversion of OC to EC was continuously monitored by measuring the transmission of a laser through the filter. The final OC mass measurement was corrected for pyrolysis carbon by adding it to the evolved OC measurement and for non-adsorbed gaseous OC by subtracting the second series filter from the first [13]. Additionally, a factor of 1.2 was multiplied with the OC mass to account for the oxygen, hydrogen, and nitrogen components of the compounds, referred to as total organic matter (OM), which were absorbed onto the particulate EC emissions [14]. In the next stage of the procedure the sample was cooled and heated again, this time under a helium and oxygen environment to remove the remaining elemental carbon from the filter. The pyrolysis carbon was used to correct the total elemental carbon measured within the sample.

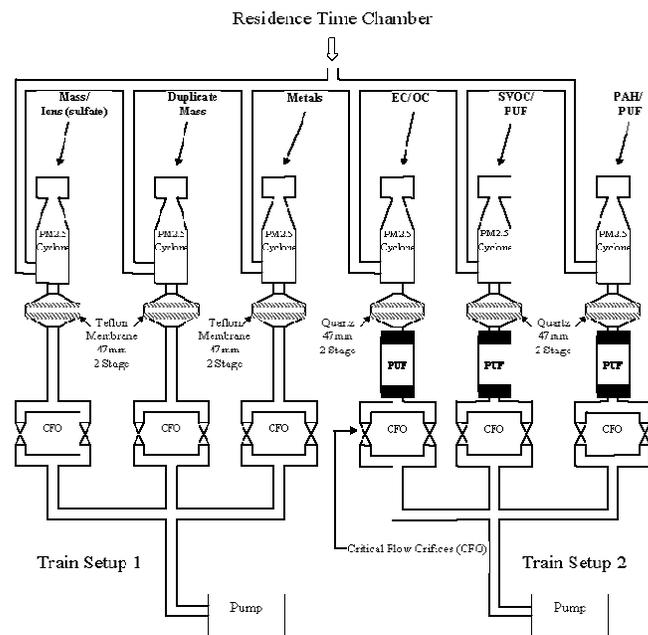


Figure 3. Sampling train diagram [11].

Further analysis of particle-phase compounds was performed on the baked quartz filters, PTFE impregnated glass fiber filters, and PTFE membrane filters. Inorganic ionic and trace metallic species were both collected using PTFE membrane filters. One set of filters that was analyzed for ionic compounds was first extracted in water and then analyzed by ion chromatography [15]. A second set of filters was prepared by extracting the filters in an acidic solution consisting of hydrofluoric, nitric, and hydrochloric acids. These filters were then analyzed for trace metallic compounds by high resolution inductively coupled plasma mass spectroscopy [15].

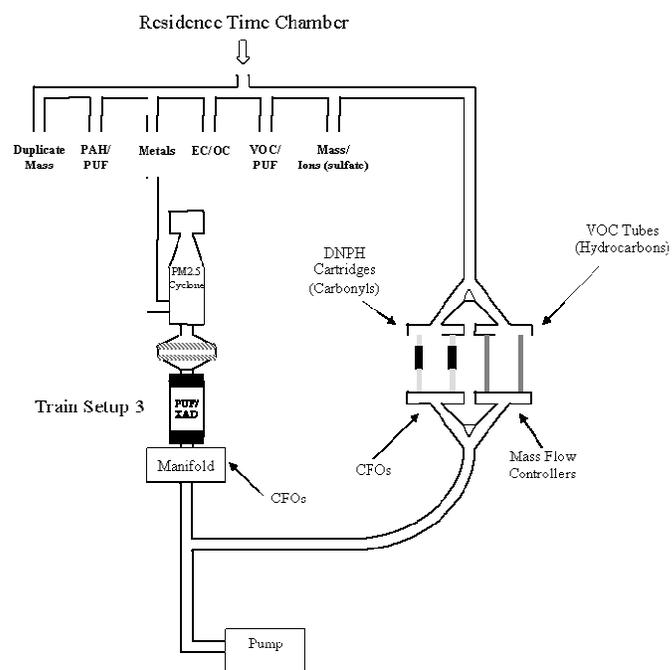


Figure 4. High flow sampling train [11].

Particulate organic species, collected on quartz and PTFE filters, were analyzed by gas chromatography mass spectrometry (GC/MS). The gaseous samples that were collected on TD tubes, XAD cartridges, PUF, and DNPH cartridges were analyzed for PAHs, alkanes, and aldehydes. TD tubes were analyzed using GC/MS according to a procedure based upon the National Institute of Occupational Safety and Health (NIOSH) 2549 method [16]. XAD cartridges were also analyzed by GC/MS equipped with an ion trap. PUF samples were analyzed together with the quartz filters used for particulate sample collection. Lastly, DNPH cartridges were analyzed using high performance liquid chromatography after being desorbed with acetonitrile [17].

RESULTS AND DISCUSSION

CRITERIA POLLUTANT EMISSIONS - Although not explicitly analyzed in the present study, the gaseous criteria emissions for each of the engine configurations were determined as part of the EPA certification testing program. The results of this testing showed that each engine family was designed to meet EPA regulations for each corresponding model year.

Previous studies have suggested that incomplete combustion of the soluble organic fraction (SOF) can result in increased emissions of HC and CO for engines without aftertreatment systems [9]. However, for the 2007 engine and emissions technology, a reduction of CO and HC emissions was achieved, most likely due to oxidation over the DOC to form CO₂ and H₂O [1, 8]. NO_x emissions were believed to be lowered through the use of an optimized in-cylinder flame temperature, which decreased the production of NO and NO₂ [2].

PM mass emissions were found to be significantly reduced with the use of 2007 heavy-duty diesel engine technology. Results show a filtration efficiency of ~99% for PM mass emissions when using the CSF, which has also been achieved in the past [18, 19, 20]. Although some increase in engine-out PM emissions was possible due to the inclusion of crankcase emissions and the lower in-cylinder flame temperature, those increases appear to be minor compared with the filtration efficiency of the CSF, which reduces tailpipe-out PM emissions primarily through particle filtration mechanisms such as interception and diffusion aided by thermophoresis and electrophoresis, followed by combustion of the PM as the filter undergoes regeneration [1].

The major contributors to the total particulate mass included EC, OM, sulfate ion, nitrate ion, ammonium ion, and trace metallic compound emissions. EC emissions, commonly known as soot, were found to be reduced significantly, by ~99.7%, with the use of new engine technology. EC is present in the particle-phase and is important to consider since the EC particles comprised

greater than 45% of the total PM mass emissions from the 2004 engine and often adsorbed organic compounds, such as organic carbon and PAHs [13].

Particulate OM, which was calculated through the quantification of OC emissions, consisted primarily of organic material adsorbed to the particle-phase EC. Therefore, the emission reduction of ~99.4% displayed by OM mass emissions was expected due to the correlation with particulate EC emissions. The other primary particulate emissions, including sulfate ion, ammonium ion, and trace metallic compounds, each were reduced by greater than 85% for 2007 technology versus 2004 technology, as seen in Figure 5. Error bars displayed in Figure 5 and in all succeeding charts represent standard error which is a measure of the variation of the sample mean. Some large errors may have been caused by ultra-low levels of certain species or difficulty in repeating regeneration patterns.

Further, significant reductions of metallic compounds across a CSF have also been reported in the past [19], and the emission levels for each of these compounds can be found in Figure 6, which shows that the majority were significantly reduced by the 2007 technology.

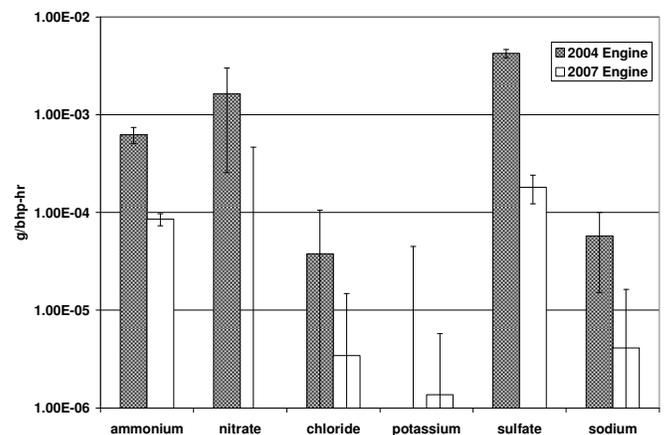


Figure 5. Inorganic ion emissions comparison.

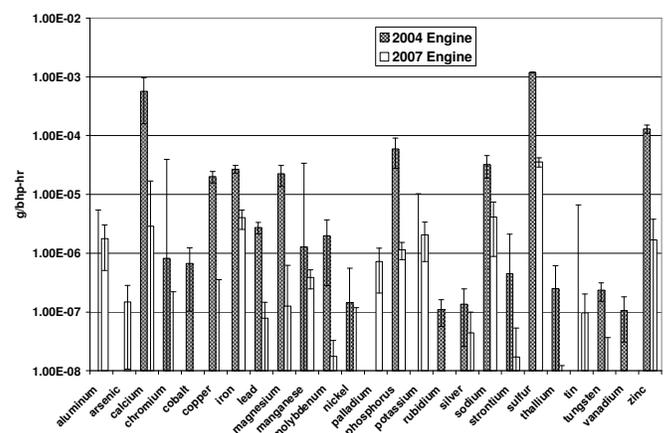


Figure 6. Metals emissions comparison

ORGANIC COMPOUNDS - A large variety of organic species were quantified in the present study. Particulate samples, collected on baked quartz filters, and gaseous samples, collected on PUF, XAD cartridges, TD tubes, and DNPH cartridges, were both analyzed for organic species. The samples were found to contain alkanes, aldehydes, biphenyls, and PAHs, with the largest portion found for aldehydes and PAHs. The full results of the organic species analysis are discussed in the following sections.

n-Alkanes and Biphenyls - Compounds classified as n-alkanes contained C₁₁ to C₂₄ species. These species have been observed in the emissions of diesel engines in the past and are thought to derive primarily from diesel fuel [4, 5, 6]. Total emission levels for the n-alkanes were found to be reduced by greater than 99% with the use of 2007 emission technology. Because of the low carbon number of the alkane compounds, their reduction is believed to be attributed to gas-phase oxidation over the DOC since adsorption onto the primary particles is not expected to be significant [5, 6, 21]. Additionally, emissions of branched alkanes and saturated cycloalkanes were observed from the test engines. These compounds, which ranged from C₁₈ to C₂₅, also displayed reductions of greater than 99%, and the emission levels were below the analysis detection limits of 1.0·10⁻⁸ g/bhp·hr from the 2007 engine setup. The emission levels of the aromatic compounds, C₁₂ biphenyl and the C₁₃ methylbiphenyls, also displayed significant reductions with the exception of 2-methylbiphenyl, which did display a slight increase as can be seen in Figure 7.

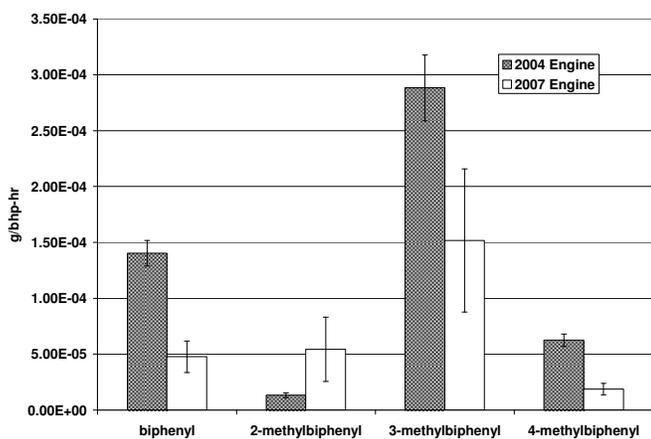


Figure 7. Emissions comparison for C₁₂ and C₁₃ biphenyls.

Polycyclic Aromatic Hydrocarbons and Aldehydes - The remaining low carbon number organic compounds consisted primarily of PAHs and aldehydes. PAHs and their derivatives, nitro-PAHs and oxygenated PAHs, included C₁₀ to C₂₄ hydrocarbon species also principally derived from diesel fuel. The emission rates of these compounds were typically reduced by greater than 85%, which most likely occurred through oxidation over the DOC and filtration through the CSF [1]. Additionally, the

ultra-low sulfur content of the 2007 fuel may have allowed for improved oxidation of the gaseous organic species since high fuel sulfur content can negatively affect the performance of the DOC [1].

Summarized in Figure 8, the greatest emission rates from the 2004 engine were formaldehyde, acetaldehyde, and naphthalene and its derivatives. However, each of these compounds was reduced by greater than 80% with the use of 2007 technology. Formaldehyde and acetaldehyde were each reduced to levels below the analysis detection limits. The substituted PAH compounds were affected by the 2007 technology by a greater amount than their unsubstituted complements. This observation may be attributed to thermal dealkylation, which may occur during the high exhaust temperatures of the active regeneration process [4]. The significant reduction of the PAHs and the aldehydes is particularly important due to the potential carcinogenic properties that they possess [22].

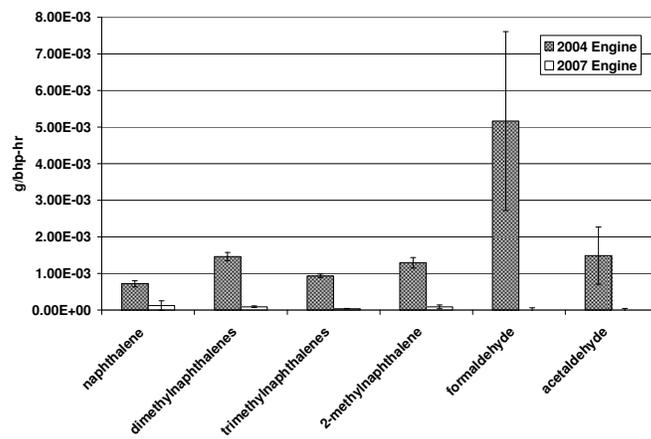


Figure 8. Emissions comparison of largest contributing organic species.

Hopanes and Steranes - The remaining organic species consisted of high molecular weight, C₂₇ to C₃₃, compounds which are typically considered to be derived from the engine lubrication oil [5, 6]. The high molecular weight compounds analyzed in the present study included hopanes and steranes, which are considered to be in the particle phase at diesel exhaust temperatures and have been used as diesel exhaust tracers in the past for air quality analysis [4]. Since these compounds are in the particle phase, it is unlikely that the DOC would have any effect on their emission rates unless exhaust temperatures were high enough for oxidation of these compounds to occur. Therefore, it is more likely that the reduction of these species, which was observed to be greater than 95%, occurred due to particle filtration mechanisms over the CSF [1].

As summarized in Figure 9, the largest contributors to total organic emissions included the aldehydes, ketones, and PAHs. Other organic species, including n-alkanes, branched alkanes, and aromatics, also displayed

significant emission rates from the 2004 engine. The summed total of most groups of organic compounds in Figure 9 displayed significant reduction with the use of 2007 emissions technology.

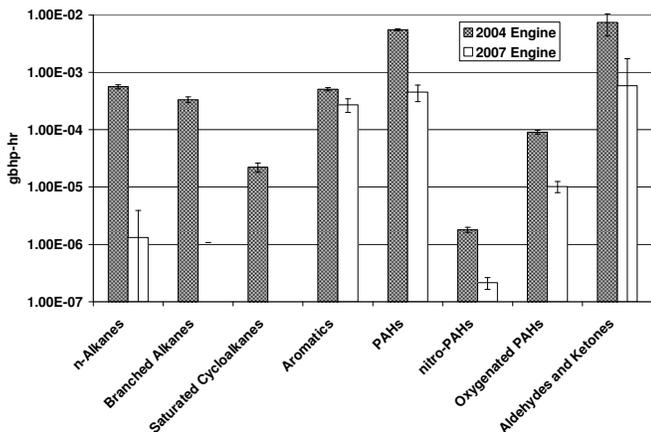


Figure 9. Organic species emissions summary.

CONCLUSIONS

The results of the present study yield the following conclusions:

- The systematic approach to 2007 engine emissions control, including engine optimization, aftertreatment integration, and fuel sulfur reduction, significantly decreased the emitted levels of a large variety of chemical compounds.
- Each of the test engines met its corresponding model-year emissions regulations, despite significant reductions of the allowable 2007 emission levels for PM and NO_x.
- The total PM mass emission was reduced by approximately 99% for the 2007 engine. The major PM constituents include EC, OC, inorganic ions, trace metals, and organics.
- The organic mass emissions contributed approximately 44% of the total PM mass emitted from the 2004 engine and 35% from the 2007 engine. This large portion renders it important to characterize organic mass when analyzing particulate mass emissions from heavy-duty diesel engines.
- Every category of organic species emissions displayed a significant reduction with the use of 2007 emissions technology. The largest contributing organic species to the 2004 engine emissions were formaldehyde, acetaldehyde, and naphthalene and its derivatives, all of which were significantly reduced in the 2007 emissions.

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DEFINITIONS, ACRONYMS, ABBREVIATIONS

ACES: Advanced Collaborative Emissions Study

CO: Carbon monoxide

CSF: Catalyzed soot filter

CV: Crankcase ventilation

DNPH: Dinitrophenylhydrazine

DOC: Diesel oxidation catalyst

DPF: Diesel particulate filter

EC: Elemental carbon

EGR: Exhaust gas recirculation

EPA: Environmental Protection Agency

GC/MS: Gas chromatography mass spectrometry

H₂O: Water

HC: Hydrocarbon

MSAT: Mobile source air toxic

NIOSH: National Institute of Occupational Safety and Health

NO: Nitric oxide

NO₂: Nitrogen dioxide

NO_x: Oxides of nitrogen

O₂: Oxygen

OC: Organic carbon

OM: Organic matter

PAH: Polycyclic aromatic hydrocarbon

PM: Particulate matter

POM: Polycyclic organic matter

PTFE: Polytetrafluoroethylene

PUF: Polyurethane foam

TD: Thermal desorption

RTC: Residence time chamber

XAD: Polymeric adsorbent

SOF: Soluble organic fraction