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Limited assessment of a selected particulate matter sensor technology in emission measurement applications on diesel vehicles

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LIMITED ASSESSMENT OF A SELECTED PARTICULATE MATTER
SENSOR TECHNOLOGY IN EMISSION MEASUREMENT
APPLICATIONS ON DIESEL VEHICLES

An Abstract of a Dissertation
Submitted
In Partial Fulfillment
of the Requirements for the Degree
Doctor of Industrial Technology

Approved:

Dr. Shahram VarzaVand, Committee Chair

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University of Northern Iowa

December 2012

ABSTRACT

The study assessed the ability of a novel particle sensor to provide adequate evaluation of real-time emissions from modern diesel engines and estimate the effectiveness of emission control devices. Emission data were obtained from vehicles in real-world field conditions under various test cycles. The datasets were used to determine whether there is an association between particulate matter (PM) measurements produced by particle sensor and the PM measurements from reference instrument. Exploratory analysis was combined with statistical techniques to investigate suitability of particle sensor to adequately measure PM mass concentrations in exhaust gases of modern diesel vehicles. Results of the study identified strong positive association between measurements from particle sensor and reference instrument. Study confirmed the suitability of the sensor in the field to measure PM emissions of diesel vehicles. These findings are useful to researchers and governmental agencies involved in regulation, control, and monitoring of diesel engine emissions.

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APPLICATIONS ON DIESEL VEHICLES

A Dissertation


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
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
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
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The content of this research reflects my views and does not necessarily reflect the official view of the California Air Resources Board. I am solely responsible for the facts and accuracy of the data presented herein. The California Air Resources Board did not technically review this report and takes no responsibility for its content. The mention of commercial products, their source, or their use in connection with material reported herein is not to be construed as actual or implied endorsement of such products.

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CHAPTER 1

INTRODUCTION

Introduction

Emissions from diesel vehicles are well known to significantly impact air quality and public health (Hsieh, Ming-Yang Wu, L.-C. Wang, Chang-Chien, & Yeh, 2011). Diesel exhaust emissions represent a complex mixture of gases and particles, including nitrogen oxides (NO_x), carbon monoxide (CO), carbon dioxide (CO₂), hydrocarbons (HC), sulfur dioxide (SO₂), and particulate matter [PM] (Faiz, Weaver, & Walsh, 1996; Westerholm & Egeback, 1994). Particulate matter is one of the most harmful emissions produced by diesel engines (Majewski & Khair, 2006; Ono-Ogasawara, Myojo, & Kobayashi, 2009).

Diesel particulates are mostly comprised of fine particles having diameters less than 2.5 μm, ultrafine with diameters less than 0.1 μm, and nanoparticles with diameter less than 0.05 μm (Hsieh et al., 2011; Majewski & Khair, 2006; Tsai et al., 2011). Because of its very small size, fine PM can penetrate deep into the lungs and cause adverse health effects, such as aggravation of respiratory and cardiovascular disease, acute respiratory symptoms, chronic bronchitis, aggravation of existing asthma, decreased lung function and even premature death. Numerous epidemiological studies demonstrated an association between ambient air levels of fine PM and elevated risks of lung cancer (Burtcher, 2001; Cohen & Pope, 1995; Diesel Epidemiology Working Group [DEWG], 2002; Lewne, Plato, & Gustavsson, 2007; Parent, Rousseau, Boffetta, Cohen, & Siemiatycki, 2007; Pope et al., 2002).

In 1998, diesel PM was identified as a toxic contaminant in the state of California due its carcinogenic potential and other adverse impacts on human health (California Air Resources Board [CARB], 2000a). Several occupational diesel exhaust studies support evidence of diesel exhaust carcinogenicity (Garshick et al., 2004; Laden, Hart, Eschenroeder, Smith, & Garshick, 2006; Lipsett & Campleman, 1999; Mine Safety and Health Administration; 2001; Olsson et al., 2011). Moreover, excessive amounts of particulate emissions reduce visibility and contribute to violations of the state and federal ambient air quality standards for particulate matter (Burtcher, 2001; Cohen & Pope, 1995; DEWG, 2002; Lewné et al., 2007; Parent et al., 2007).

NO_x represent another group of critical pollutants found in diesel exhaust. NO_x emissions are of specific concern because of their role in smog formation and PM pollution (Twigg, 2005). NO_x contribute to PM pollution by forming nitrates as a result of a secondary reaction in the atmosphere. The NO_x emissions form ozone when combined with various hydrocarbon (HC) emissions in the presence of sunlight. Consequently, NO_x and HC exhaust emissions from heavy-duty trucks and buses significantly contribute to violations of the state and federal ambient air quality standards for ozone (Faiz et al., 1996; Godish, 2004). Ozone causes a range of health problems related to breathing, including chest pain, coughing, and shortness of breath. Exposure to diesel PM and ozone is especially dangerous to children and the elderly. In addition, ozone, NO_x, and PM adversely affect the environment in various ways, including crop damage, acid rain, and visibility impairment (Beeson, Abbey, & Knutsen, 1998; C.-G.

Lee, Yuan, Chang, & Yuan, 2005; Office of Transportation and Air Quality [OTAQ], 2000; Srivastava, Agarwal, & Gupta, 2011).

In the Los Angeles basin for example, the atmosphere exhibits high levels of PM pollution due to contributions from highly congested traffic and other combustion sources (Arhami et al., 2009). According to the CARB estimates for the year 2000, the primary sources of PM emissions in the state of California are the 1,250,000 in-use diesel engines and vehicles, including on-road trucks and buses, portable and stationary engines, and off-road equipment. As a result, over 25,000 tons of diesel PM emissions are released annually in California (CARB, 2000a). Based on a research conducted in 2000 by the South Coast Air Quality Management District, cancer risk from toxic air pollution in Southern California averaged about 1,400 in a million with diesel particulates responsible for 71% of cancer risk (Coel, Irwin, Lieu, & Dabirian, 2000). Therefore, studies focused on diesel engine emissions represent a significant issue and are of great importance especially in the state of California.

Moreover, PM emission limits for diesel engines are becoming increasingly stringent requiring use of new exhaust after-treatment systems and advanced emission measurement technologies (K. K. Kelly et al., 2004; Majewski & Khair, 2006). This leads to a growing need for an assessment of PM testing equipment that would prove to be reliable and capable of measuring lower pollution levels.

Statement of the Problem

The problem of this study is to investigate the ability of a newly developed particle sensor to measure PM emissions in field testing environment in order to provide adequate evaluation of real-time emissions from modern diesel engines and assess effectiveness of emission control devices.

Statement of the Purpose

The objective of this study is to assess the suitability of a selected PM sensor to evaluate real-time emissions and investigate efficiency of exhaust after-treatment units. Specifically, the study examines the association between emission measurements produced by the sensor and the reference PM measurements from another instrument. Emissions are measured in exhaust gases of diesel vehicle in various real-time field settings, including snap-idle acceleration and driving conditions.

The particle sensor's ability to measure PM levels in raw exhaust gas is also investigated. This is especially important for field testing, since direct measurement of raw exhaust from the engine eliminates the need for additional dilution equipment, and therefore, significantly simplifies testing setup. Furthermore, modern low emission standards necessitate raw emission measurements. During these measurements, the instruments' response and sensitivity can be increased as a result of higher levels of PM emissions in raw exhaust sample compared to diluted gas.

The findings of this research would provide an insight on usability of the novel sensor to adequately measure PM emissions of mobile diesel engines in real-time field conditions. This addresses the need in reliable real-time sensors to monitor lower PM

levels in order to comply with new stringent diesel emission regulations. Additionally, adequate measurements of PM emissions on vehicles equipped with exhaust after-treatment devices, such as diesel particulate filters (DPFs), is useful to confirm proper operation of the engine and the DPF, as well as diagnose any operational problems. The results of this study may serve as the basis for establishment of future recommendations for engine emission measurement. Furthermore, the results may be useful to governmental agencies involved in regulation, control, and monitoring of diesel engine emissions.

Need of the Study/Justification

Many state, national, and international agencies have concluded that diesel exhaust is a probable lung carcinogen, including the International Agency for Research and Cancer in 1989, the World Health Organization (WHO) in 1996, and the National Toxicology Program in 2000 (DEWG, 2002). Additionally, constituents of diesel exhaust have been identified as toxic air contaminants under the CARB Toxic Air Contaminant Program (CARB, 2012b). The CARB estimates that “the majority of the potential airborne cancer risk in California” can be attributed to diesel emissions. In response to environmental and public health concerns, since the early 1990s, diesel emissions have been subject to more stringent emission standards worldwide (Faiz et al., 1996; Maricq, Xu, & Chase, 2006; Srivastava et al., 2011). A notable reduction of diesel exhaust emissions has been achieved during the last decades mainly due to the progress in emission-control technology, improved engine design, and cleaner fuels (Di Iorio, Mancaruso, & Vaglieco, 2012; Malik et al., 2011; Tsai et al., 2011).

However, with the growing use of diesel engines, their emissions continue to be of concern to federal and state air quality agencies. Despite an ongoing progress in air quality improvement, about 90 million tons of total emissions were released in the environment in the United States in 2010. These pollutants contributed to a number of existing air quality problems, including ozone and particles formation, deposition of acids, and visibility impairment (Environmental Protection Agency [EPA], 2012a). Experts estimate that despite the decrease of total particulate emission levels, the number of potentially more damaging fine particles have increased by as much as a factor of six (Gautam, Carder, Clark, & Lyons, 2002).

In recent years, the EPA and the CARB have promulgated regulations to further control diesel emissions. Stringent Tier 4 standards for non-road diesel engines introduced by the EPA will be phased in over the period of the years 2008-2015. These standards require a drastic 90% reduction of PM and NO_x emissions (DieselNet, 2004a). Similarly, the EPA's clean diesel regulations aimed to reduce emissions from on-road heavy-duty trucks became effective January 1, 2007, on a phased-in basis through 2010. By the year 2030, these rules project a 2.6 million ton reduction of smog-causing NO_x emissions and PM emissions reduction by 109,000 tons (OTAQ, 2000). With increasing demands on reduced engine-out emissions, the requirements in efficient exhaust after-treatment devices grow accordingly. In order to meet the 2007 heavy-duty on highway emission limits for PM, manufacturers are required to install DPFs on all new diesel-powered vehicles resulting, in some cases, in greater than a 90% reduction in PM (Khalek, Bougher, Merritt, & Zielinska, 2011; Malik et al., 2011). According to the 2011

data from the Manufacturers of Emission Control Association (MECA), since the year 2007 nearly two million heavy-duty and medium duty vehicles with installed DPFs have been sold in the United States to satisfy the EPA's heavy-duty highway rulemaking requirements (MECA, 2011).

Diesel PM filter operates by trapping (i.e., filtering) PM emissions from the exhaust stream as they get forced through the filter's body. Combustible and exhaust fractions of PM trapped by the DPF get periodically oxidized during the filter regeneration; while non-combustibles remain in the filter and must be regularly removed. Without proper regeneration or cleaning the pressure drop of the DPF increases to unacceptable levels. It can negatively affect engine performance and even cause DPF failure. Elevated back pressure levels are also accompanied by increased exhaust temperatures, fuel consumption, PM, CO, and HC emissions (DieselNet, 2005; Jääskeläinen, n.d.; Majewski, 2011; Mayer et al., 2000). Therefore, diagnostic measurements of the DPF-out and engine-out PM emissions on vehicles equipped with the DPFs are useful to ensure proper performance of the filter and the engine.

According to Title 40 of the U.S. Code of Federal Regulation, gravimetric method is currently used as a standard for diesel total PM mass emission measurements. However, classical gravimetric filtration technique is at its detection limit when it comes to measuring lower emissions of new engines as well as engines equipped with particulate filters. Accuracy of gravimetric results is affected by unstable nature of diesel emission samples with high fraction of volatile material. In this respect, the type of utilized filter media can have significant effect on sampling results of low diesel PM

levels. Additionally, gravimetric mass measurement is very labor intensive and unable to provide real-time results. Therefore, measurement of PM emissions of modern low emission vehicles using standard gravimetric procedure represents a challenging task and may no longer be adequate (Burtscher, 2001; Burtscher, Majewski, & Khalek, 2012; N. A. Kelly & Morgan, 2002; Khalek, 2005; Swanson, Kittelson, Pui, & Watts, 2010). Ultimately, real-time devices emerge as potentially valuable in estimating engine emissions and efficiency of emission control devices in accurate, fast, and cost-effective manner (Burtscher, 2001; K.K. Kelly et al., 2004).

Due to the existing need in instruments that would monitor PM exhaust emissions, it is important to investigate new PM sensor technologies in regard to its potential application to real-time diesel emission measurements. Further research is needed to investigate whether a selected portable PM sensor can serve as a robust instrument for real-time PM measurements under real-world conditions in the field environment. It would provide a better insight into possible potential applications of this sensor in regard to real-time field diesel emission testing. A critical assessment of PM sensor performance is vital in terms of understanding its usability to estimate PM engine emissions and to verify proper performance of the engine and emission control devices, such as DPFs.

Research Questions

The study is aimed to provide answers to the following proposed research questions:

1. Are there any trends in temporal distributions of emissions measurements produced by a novel particulate matter sensor compared to a reference PM measurement instrument?
2. Is there an association between outputs from a selected PM sensor and the reference device? If so, what is the strength of this relationship?
3. The study determines whether the particle sensor is suitable for DPF efficiency evaluation. Specifically, the study identifies how filtration efficiencies calculated using data from a selected PM sensor compare to those obtained from the reference instrument. This determines whether it is possible to use the novel PM sensor as a diagnostic tool for evaluation of the DPF operating condition.

Hypotheses

Study hypothesizes similarity in the ability of the particle sensor to measure PM mass concentrations compared to the mass monitor. The following hypotheses are generated in order to tests the significance of association between the instruments:

Null hypothesis H_0 : there is no significant association between PM measurements produced by the particulate sensor and the mass monitor.

Research hypothesis H_1 : there is a significant association between PM measurements produced by the particulate sensor and the mass monitor.

Assumptions

The study is based on several assumptions that are related to testing conditions and particulate matter emissions as described below.

1. The mass monitor measurements are assumed to be a reference due to instrument's satisfactory performance found in literature (Khalek, 2005, 2008; Lehmann, Niemela, & Mohr, 2004; Mamakos, Ntziachristos, & Samaras, 2006). Moreover, the mass monitor used in current study is compliant with manufacturer's requirements of calibration and maintenance.
2. Possible particle losses within the particulate matter testing equipment are assumed to be negligible since equipment transfer lines setup was done according to Code of Federal Regulations, specifically due to using "the heated transfer lines to minimize temperature differences between transfer lines and exhaust constituents" that were inert "with respect to PM and are electrically conductive on the inside surfaces" (40 C.F.R. § 1065, 2012).
3. Variation in ambient environmental conditions during testing is assumed to have insignificant effect on PM measurement results due to similarities in measured ambient characteristics that are presented in Appendix A.
4. Heated dilution system used during diluted exhaust sampling was able to effectively precondition gas exhaust sampling stream and remove volatile and semi-volatile compounds from the exhaust. Therefore, any organic materials are prevented from condensing on carbonaceous particles or from nucleating and forming new nuclei-mode particles.
5. PM losses of non-volatile particles after going through a heated dilution system are presumed to be minimal and "do not need to be corrected for separately" (Dekati, 2008). The latter and the former assumptions are made since operation

of heated dilution system satisfies requirements of European Particle Measurement Program for volatile particles removal (Dekati, 2008).

6. Variations of dilution rate of sample exhaust flow are considered to be marginal since the dilution system's "reduction factor is constant over time" with $\pm 5\%$ typical dilution ratio accuracy (Dekati, 2008). Therefore, dilution ratio accuracy of dilution system is assumed not to significantly affect the overall accuracy of PM measurement results.
7. The sampling probe configuration (e.g., sampling probe positioning and the attachment of sampling probes to the engine exhaust pipe) is assumed to be appropriate for diesel PM measurements. This configuration was implemented according to commonly accepted diesel testing procedures and was designed to "minimize the number of bends in transfer lines" and "maximize the radius of any unavoidable bends" (40 C.F.R. § 1065, 2012). The scientific justification of probe misalignment due to unavoidable bends was adopted from Besch et al. (2011). According to their calculations, since diesel PM is "in the size range of few nanometers to approximately 150 nanometers... it has been judged adequate to neglect any particle concentration corrections due to probe misalignment or anisokinetic sampling." It should also be noted that for submicron particles as those commonly found in diesel exhaust from modern vehicles, the isokinetic sampling is not as important as for larger particles from other sources (Kittelson, Arnold, & Watts, 1999) and inertial losses of submicron particles due to subisokinetic sampling are negligible (Canagaratna et al., 2004).

Limitations/Delimitations

Study is based on several limitations. Limitations that are taken into account in the conduct of the study are as follows:

1. The study is limited to an assessment of a single model of emission measurement device manufactured in the year 2011.
2. The focal point of the study is limited to particulate matter emissions.
3. The research is focused on measurement of emissions from diesel equipment.
4. Investigation is restricted to on-road diesel emissions measurement under environmental conditions typical for the state of California.
5. Study is limited to vehicle equipped with diesel engine with 2009 model year and the DPF.
6. Presented data does not include any sensitive information and confidential records. Specific models of particulate matter testing instruments and tested engine are omitted.

Methodology/Outline of Procedures to be Employed

Several procedures are used throughout the course of this study. It is intended that these items will be discussed in detail with appropriate explanations.

1. An assessment of a selected particulate matter sensor technology in diesel emission measurement application is conducted throughout the study.
2. This research investigates diesel vehicle emissions during real-time field testing.
3. PM emission measurements are performed on engines during several test cycles, such as snap-idle acceleration and urban driving.

4. Tests are conducted over multiple consecutive test runs under similar testing conditions.
5. Emission data needed to investigate proposed research questions and hypotheses are acquired using the following measurement tools: a selected PM sensor and a reference real-time PM emission measurement device. Measurement of PM concentrations was performed simultaneously using these instruments.
6. The particle sensor was investigated during diluted as well as raw exhaust PM measurements.
7. In order to evaluate the DPF efficiency, PM emissions were measured in the engine exhaust flow upstream and downstream of the DPF. Data were recorded during three consecutive runs, each consisting of six snap-acceleration events. First, instrument readings were simultaneously recorded during the measurements after the DPF. Then same procedure was repeated to obtain emission data before the DPF.

Statistical and Other Analysis of the Data

Assessment of a selected particulate matter sensor technology involved the evaluation of PM emissions using several descriptive and inferential statistical procedures preceded by general exploratory analysis described below. Data analysis consisted of several steps. The major components of the study design include the following:

1. Temporal PM distribution graphs were created as part of exploratory analysis using continuous data output. Visual comparison of distribution shapes further revealed existing trends and any discrepancies in instrument readings.

Differences in instrument response were identified based on PM distribution graphs and descriptive statistics results.

2. PM data obtained from the novel sensor were compared and correlated to measurements from the reference instrument. The continuous data output were correlated by means of simple linear regression. Strength, direction, and significance of relationship were identified (Aron, A., Aron, E., & Coups, 2005). Microsoft Excel software (Microsoft Inc., Redmond, Washington) was used to aid statistical analysis of the data.
3. The DPF filtration efficiency was calculated based on the PM sampling data, specifically PM mass concentrations before and after the filter. Results were used to conduct a comparison of the DPF efficiencies calculated using the PM sensor data against the DPF efficiencies computed using output from the reference instrument.

Definitions of Terms

The research involves several key definitions. Since various sources of literature might have different interpretation of the same concepts, current investigation adopted the following identification of terms.

Diesel engine represents a compression-ignition engine in which fuel is ignited by injecting it into highly compressed air as opposed to a spark-ignition engine (Beck, 2004; Majewski & Khair, 2006). It is “an embodiment of the internal combustion engine.” Its main function is to efficiently produce mechanical power from the chemical energy stored in fuel (Majewski & Khair, 2006). Regulatory definition of a diesel compression-

ignition language is expanded to include natural-gas fueled engine equipped with a sparkplug. “The definition focuses on the engine cycle, rather than the ignition mechanism, with the presence of a throttle as an indicator to distinguish between diesel-cycle and Otto-cycle operation. Regulating power by controlling the fuel supply in lieu of a throttle corresponds with lean combustion and diesel-cycle operation” (DieselNet, 2004a).

A diesel particulate filter (DPF) corresponds to a “ceramic device that collects the particulate matter in the exhaust stream. The high temperature of the exhaust heats the ceramic structure and allows the particles inside to break down (or oxidize) into less harmful components” (OTAQ, 2003).

Emission is “the discharge of pollutants into the atmosphere from stationary sources such as smokestacks, other vents, surface areas of commercial or industrial facilities, and mobile sources, for example, motor vehicles, locomotives and aircraft” (Statistics Division of the Department for Economic and Social Information and Policy Analysis, 1996).

Pollutants (pollution) are identified as “unwanted chemicals or other materials found in the air. Pollutants can harm health, the environment and property. Many air pollutants occur as gases or vapors, but some are very tiny solid particles: dust, smoke, or soot” (Beck, 2004).

Term "vehicle" is characterized as "collection of all categories of motor vehicles and motor vehicle engines. Specifically, it includes cars, motorcycles, light-duty trucks, heavy-duty vehicles, heavy-duty trucks, and heavy-duty engines" (EPA, 2012b).

Heavy-duty vehicle or heavy-duty truck is classified as "any motor vehicle (including buses) having a gross vehicle weight rating (GVWR) of greater than 8,500 pounds, or curb weight of more than 6,000 pounds, or that has a basic vehicle frontal area in excess of 45 square feet" (EPA, 2012b).

GVWR is defined as "the weight specified by the manufacturer as the loaded weight of a single vehicle. The loaded weight of the vehicle includes passengers, options, and cargo" (EPA, 2012b).

Non-road engines include "all internal combustion engines except motor vehicle (highway) engines, stationary engines (or engines that remain at one location for more than 12 months), engines used solely for competition, or engines used in aircraft". Additionally, all diesel powered engines utilized in agricultural operations in the State of California are also considered as non-road engines (EPA, 2012b).

Summary

This research is aimed to conduct an assessment of a selected particulate matter sensor technology in emission measurement applications on diesel vehicles. Several procedures were utilized throughout the study, such as identification of the problem and purpose, need/justification, research questions, assumptions, definition of terms, statistical methods, etc. Introduction and need of the study/justification represent the foundation of research that includes review of literature in this area of study and summary results of previous research.

The dissertation is comprised of an abstract and five chapters. The chapters include the introduction, literature review, materials and methods, results, and the

discussion and conclusion. Additional tables and figures are attached in appendices at the end of the dissertation. The first chapter, the introduction, is comprised of the former research proposal section. The literature review is summarized in the second chapter and further elaborates on the current need of selected research problem. Specifically, diesel engine fundamentals, major diesel emissions, their health and environmental effects, PM emission control, and measurement are discussed.

The materials and methods of the study are addressed in the subsequent chapter. The test setup, procedure, and data analysis techniques for PM emission evaluation are described in detail. Results of the assessment of a selected particulate matter sensor technology are presented in chapter four.

Finally, the last chapter presents discussion of major findings, summarizes the main conclusions of the dissertation, and provides further suggestions and recommendations regarding the usability of studied PM sensor in emission measurement applications on diesel vehicles.

CHAPTER 2

LITERATURE REVIEW

Diesel Engine Technology

Diesel Engine Fundamentals

The diesel engine represents a compression ignition engine, a category of internal combustion engines (Bauer, Dietsche, & Crepin, 1999; Heywood, 1988). Its main operational principle is based on utilization of chemical energy of the fuel in order to efficiently produce mechanical power. Similarly to other internal combustion engines, diesel engines use the conventional cylinder and piston system (Chambers, 1999; Pulkrabek, 2003). However, in contrast to spark ignition engines, such as gasoline engine, it does not use a spark plug for ignition. Instead, the heat of compression is used to initiate fuel ignition as it gets injected into the combustion chamber (Dobbins, 2007; Heywood, 1988; United States Department of Energy [USDE], 2012b). One combustion cycle of cylinder and piston arrangement consists of an intake, compression, expansion and exhaust strokes (Bauer et al., 1999; Bennett, 2010).

Diesel engine was invented by Dr. Rudolph Diesel in 1893. The theoretical mechanical efficiency of the second diesel engine model achieved a remarkable result of 75%. This was approximately seven times higher than the 10% efficiency of a steam engine, a prevailing power source of that time (Majewski & Khair, 2006). Since then diesel engine underwent further intensive improvements and was able to achieve a worldwide commercial success. In the United States, the majority of all heavy and medium-duty trucks utilize diesel engines. This is due to several advantages of diesel

engine, including its high efficiency, fuel economy, and durability (USDE, 2012b).

These are discussed in more detail later in the chapter.

Operational Characteristics

Operational principle of diesel engine is based on injection of a small amount of fuel into highly compressed air. After being injected into the combustion chamber, the fuel starts evaporating due to high temperatures of compressed air. Evaporated fuel subsequently begins mixing with the hot surrounding air. As a result, temperature of evaporated fuel starts rising until it reaches a fuel auto-ignition temperature limit. Ultimately, this causes burning of the fuel followed by release of chemical energy stored in that fuel (Bauer et al., 1999; Bennett, 2010; Dobbins, 2007).

Cylinder- and piston-type diesel internal combustion engines are categorized into two major types: two-stroke and four-stroke. Two-stroke engines are widely used in compact-scale engine applications due to their smaller size than that of four-stroke engines (Pulkrabek, 2003). Two-stroke engines also possess better specific power characteristics. Given the same power output, the specific power (power output for a given engine displacement) is higher in a two- than in a four-stroke engine designs (Gupta, 2006). Therefore, in order to enhance four-stroke engine's specific power output, modern engine designs oftentimes are equipped with devices that improve air-charging and fuel injection (Heywood, 1988). Particularly, turbochargers made significant contributions to latest enhancements of power and economy of diesel engines (Wharton, 2003).

Despite of having twice as many power strokes compared to their four-stroke counterparts, two-stroke engines do not generate twice their power mostly due to their shorter power stroke. The power stroke in two- and four-stroke engines occupies 90-95 and 120-140 crankshaft degrees, respectively. Additionally, two-stroke engines mostly have lower fuel efficiency than four-stroke designs, mainly due to low air scavenging and oftentimes small volumetric efficiency (Gupta, 2006).

The basic operation cycle in a two-stroke engine is comprised of two piston strokes that conclude one complete rotation of the crankshaft during one full combustion cycle, including intake, compression, expansion, and exhaust. In two-stroke engines, the air is introduced in the combustion chamber during the intake stroke of the combustion cycle immediately prior to compression. Consequently, the burned gases are exhausted into the atmosphere in the last portion of the expansion power stroke (Bennett, 2010; Gupta, 2006; Pulkrabek, 2003).

On the other hand, a combustion cycle of four-stroke engine designs is comprised of two crankshaft rotations, each consisting of two piston strokes. As the piston moves from its outermost top position (i.e., top-dead-center) toward its outermost bottom position (i.e., bottom-dead-center) during the intake stroke, the filtered air gets admitted into the cylinder. The inducted air is then being compressed during the subsequent compression stroke when the piston returns back to its initial position. During this stroke the air temperature inside of the cylinder raises above the fuel auto-ignition temperatures. At this time, the fuel is being injected into the cylinder where it starts burning. As a result, released heat energy creates additional pressure while gases in combustion

chamber start expanding. During the expansion stroke (or power stroke), this pressure is applied to the piston as it gets pushed back towards the bottom-dead-center position. Ultimately, the gases formed as a result of combustion process get expelled into the ambient air (Bauer et al., 1999; Heywood, 1988; Merker, Schwarz, & Teichmann, 2011).

Four-stroke engines are further subdivided into direct- and indirect-injected engines. Combustion chamber of indirect-injected engines (see Figure 1) is split into a main chamber and a smaller pre-chamber, latter generally incorporates a glow plug to help start-up the engine in cold weather conditions (HyperPhysics Project, 2012). Combustion begins in the pre-chamber after the fuel gets injected into it and comes in contact with hot surface of glow plug. Subsequently, gaseous turbulent mass flows through a narrow passage from a pre-chamber to the main chamber where it is further oxidized as it gets mixed with the remaining unburned fuel and partially combusted by-products. Combustion rate in indirect-injected engines greatly depends on the degree of turbulence (kinetic energy of the air) as opposed to fuel atomization. Therefore, high fuel injection pressure is not required to achieve proper mixing and combustion. This is also true during compression stroke as gaseous mass flows back to the pre-chamber (Bennett, 2010; Merker et al., 2011; Mollenhauer & Tschoke, 2010).

Indirect-injected engines have been traditionally used in various passenger vehicle applications. This is attributed to a lower noise rate of indirect-injected engine as well as to lower combustion temperatures and pressures that represent its major advantages. Major disadvantages of indirect-injected engines include increased cooling losses related

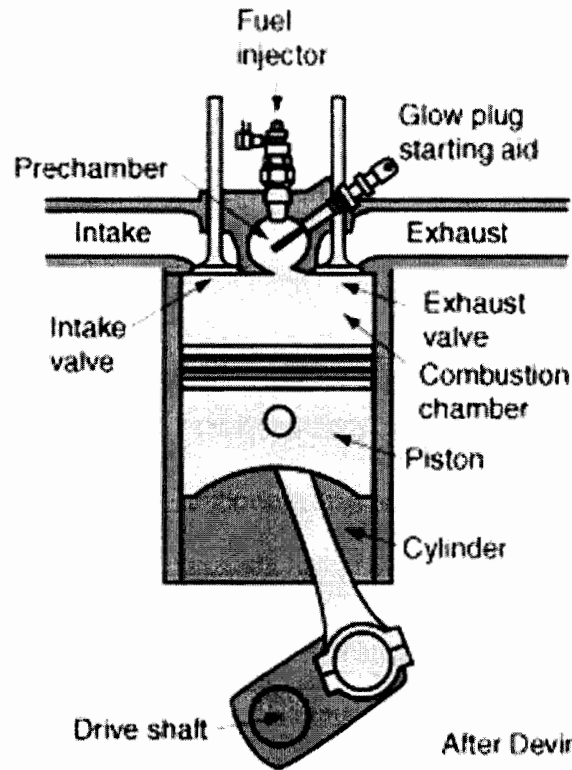


Figure 1. Diagram of indirect-injected diesel engine (HyperPhysics Project, 2012).

to somewhat high surface area exposed to the engine coolant, lower thermal efficiency, overheating of some critical engine components, and shorter oil lube life. Another disadvantage of indirect-injected engines is their overall lower efficiency related increased pumping losses due to the presence of the narrow passage connecting the main combustion chamber with pre-chamber (Bauer et al., 1999; Gupta, 2006; Majewski & Khair, 2006).

Direct injected engines designed to allow fuel to be introduced straight into the combustion chamber. Combustion efficiency of this engine design depends not only on kinetic energy of the air and fuel mixture, but also on injection pressure and injector

nozzle geometry. Absence of pre-chamber in direct-injected engines gives certain advantages to this type of engines compared to indirect-injected. These advantages include reduced pumping losses and up to 15% better fuel economy (Bauer et al., 1999; Bennett, 2010; Gupta, 2006).

Advantages and Disadvantages of Diesel Engines

The main advantages of diesel engines include fuel economy, high efficiency, durability, fuel safety, and low emissions of several major air pollutants. With oil prices continuously rising, the high fuel economy (one of the major advantages of diesel engines) is becoming increasingly important. Diesel engines deliver more power and fuel-efficiency compared to vehicles equipped with gasoline engines. On average, diesels consume 30-35% less fuel compared to their gasoline counterparts. Such high efficiency is partially attributed to higher compression ratios of diesel engines, which is twice as high as that of gasoline engines (Majewski & Khair, 2006; Heywood, 1988; USDE, 2012a).

High efficiency is also related to its inlet system configuration which lacks the throttle and allows ease of breathing. Unlike gasoline engines, instead of throttling inlet air the power output is controlled by varying the fuel rate. This helps reduce pumping losses, maintain good volumetric efficiency, and minimize the amount of air induction work done by engine to ensure efficient combustion (Gupta, 2006; Heywood, 1988).

Longer useful lives and better reliability is another advantage of diesel engines, especially important in industrial applications and trucking industry. However, even diesel engines installed in passenger vehicles frequently outlast the life period of their

vehicles. Generally, useful life of diesels may be three to four times longer compared to gasoline engines. Diesel engines are usually more robust to be able to withstand high operating pressure caused by the high compression ratio of the engine and the large amounts of produced torque. Additionally, more durable engine construction is impelled by more abrupt heat release occurring earlier in the combustion cycle. High durability is also related to the fact that diesels tend to have lower engine speeds than gasoline engines. Lower engine speeds reduce friction losses while improving the brake power and prolong engine service life (Gupta, 2006, Lloyd & Cackette, 2001; Majewski & Khair, 2006).

High durability of diesels allows maintaining emission levels within manufacturing emission certification requirements for a longer period of time. Additionally, diesel engines produce very low concentrations of HC and CO due to fuel combustion in lean regimes with excess air even at full load. Reduced HC emissions during fuel handling and transfer are also related to diesel fuel being less volatile. Other advantageous characteristics of diesel fuel include handling safety due to its lower ignition point (Majewski & Khair, 2006).

Major disadvantages of diesel engines include relatively high noise levels at idle, high levels of NO₂ (PM/NO₂ trade-off), need in expensive and robust transmissions, and high engine cost. Diesel noise is largely caused by characteristics of diesel combustion process, such as rapid pressure increase caused by sudden fuel ignition. Noise levels can be significantly reduced in engines with indirect injection. High torque at low engine speeds, while being useful for commercial applications, also creates need in more robust

and expensive transmissions (Gupta, 2006; Majewski & Khair, 2006; Mollenhauer & Tschoke, 2010).

High engine cost remains one of the most important issue of diesels mainly due to its highly precise fuel injection system needed to achieve proper degree of fuel atomization and mixing and therefore maintain required combustion efficiency. Utilization of advanced electronic injection systems in modern diesel engines helps reduce cost of their fuel injection equipment. Additionally, high price of fuel injection systems can be somewhat offset by enhanced performance and fuel efficiency of modern engines (Majewski & Khair, 2006; USDE, 2012b).

Low diesel exhaust temperatures minimize exhaust heat loss and increase diesel's efficiency. However, it also complicates operation of post-combustion emission control devices that usually require higher temperatures for efficient exhaust after-treatment. Therefore, the diesel's high NO_x emissions and related NO_x/PM trade-off represent major existing challenges for scientists and engine manufacturers (Gupta, 2006; USDE, 2012b).

Major Diesel Emissions

Diesel exhaust emissions come from “on-road” diesel engines (i.e., trucks, buses, and cars) as well as “non-road” diesel engines, including heavy-duty equipment, locomotives, marine vessels, and so on. Chemical composition of diesel engine emissions varies depending on engine types (e.g., heavy-, medium-, and light-duty), engine operating conditions (e.g., idle or acceleration modes), and fuel formulations (e.g., sulfur fuel content). Since most non-road engines are usually older than on-road engines,

their emission levels are usually higher than those from on-road engines (EPA, 2002; Lloyd & Cackette, 2001). Diesel emissions represent a complex combination of pollutants resulting from incomplete combustion of fuel mixture accompanied by various chemical reactions between its components. Major diesel pollutants are divided into regulated and unregulated. Generally, unregulated compounds are found in much lower concentrations in diesel exhaust compared to regulated pollutants. Main unregulated diesel emissions are polynuclear aromatic hydrocarbons (PAHs), soluble organic fraction of diesel particulates, aldehydes, sulfur dioxide, nitrous oxide (N₂O), dioxins, and metal oxides (Gupta, 2006; Khalek et al., 2011; Mullen, 2010).

Regulated diesel emissions consist of PM, NO_x, HC, and CO (Khalek et al., 2011; R. Zhu, Cheung, Huang, & X. Wang, 2011). Due to very lean fuel-air mixture diesel engines emit significantly lower quantities of HC and CO than spark-ignited gasoline engines (Gupta, 2006; Tsai et al., 2011). NO_x emissions from diesel and gasoline engines are relatively comparable to each other. While gasoline engines equipped with three-way catalysts produce lower NO_x emissions than diesel engines, NO_x released from diesel engines is usually lower compared to regular gasoline engines (Majewski & Khair, 2006). Diesel PM levels are significantly higher than those from gasoline engines and, therefore, represent a vital regulatory, environmental, and public health problem.

Carbon Monoxide and Carbon Dioxide Emissions

CO is one of regulated diesel emissions. It is an odorless, colorless, and tasteless gas slightly lighter than air. In high concentrations it can be toxic. CO can disrupt the process of oxygen delivery to bodily tissues as it combines with hemoglobin.

Additionally, exposures to CO can produce significant adverse effects to the heart and central nervous system. CO represents a product of incomplete combustion and is produced from the partial oxidation of carbon-containing compounds. CO is not chemically stable and gets easily combined with oxygen to form carbon dioxide and ozone in the atmosphere. Both anthropogenic (caused by human activities) and natural emissions contribute to production of carbon monoxide. The sources of carbon monoxide include technological processes, biogenic sources, biomass burning, oceans, and oxidation of methane gas and of non-methane hydrocarbons (Godish, 2004; Rodrigue, Comtois, & Slack, 2009).

The OTAQ estimates that input of mobile sources to carbon monoxide pollution is about 95% (OTAQ, 2012a; Godish, 2004). Rodrigue et al. (2009) identified that transportation is responsible for 70 – 90% of total carbon monoxide emissions. Operational characteristics of combustion in diesel cylinders result in low concentrations of CO in exhaust emissions. According to the OTAQ study, diesel vehicles contribute only four and five percent to total CO emissions from on-road and non-road, respectively (OTAQ, 2012a).

Carbon dioxide (CO₂) is unregulated component of diesel emissions. It is a greenhouse gas. CO₂ emissions are produced by natural phenomena as well as a result of fossil fuel combustion by anthropogenic sources. Total emissions of CO₂ from natural sources exceed those from man-made sources. Anthropogenic CO₂ emissions mainly come from power generation, heating, industrial activities, and transportation (Godish, 2004). Contribution of transportation to the total share of manmade CO₂ pollution is

accounted for about 30%. While diesel vehicles responsible for only 16% of that pollution; contribution from gasoline vehicles corresponds to about 66% (Rodrigue et al., 2009). According to experts, conversion from gasoline to modern diesel vehicles could potentially result in 25% reduction of transportation-related CO₂ emissions (Majewski & Khair, 2006).

Hydrocarbon Emissions

HCs correspond to a group of compounds that includes a large number of chemicals, such as gases, volatile liquids, semi-volatile substances, and solids. Diesel HC emissions are regulated in terms of total hydrocarbons (THC) or nonmethane hydrocarbons (NMHCs). Diesel exhaust contains combination of various HCs, including HCs with shorter carbon chains derived from diesel fuel and HCs containing heavier longer molecules from lubricating oil (Majewski & Khair, 2006). HCs possess distinctive irritating odor. Some HCs exhibit highly toxic properties and pose serious threat to human health. An exposure to these chemicals can cause difficulty breathing, lung damage, reduced cardiovascular functioning, and cancer. From environmental standpoint, HCs serve as precursors to ground-level ozone and are important in the formation of other photochemical oxidants. Indirectly HC contribute to smog formation (Chow, 1995; Godish, 2004; OTAQ, 2012b; Rodrigue et al., 2009).

Transportation sector is estimated to generate up to 50% of total HC input from manmade sources (Cunningham & Woodworth-Saigo, 1990; Rodrigue et al., 2009). Incomplete fuel combustion (70%), refueling operations (10%), and fuel evaporation mainly from storage units/gas tanks (20%) are the main sources of HC from

transportation. Diesel emissions account for 5 and 15% of HC from on-road and non-road mobile HC emissions, respectively (OTAQ, 2012b).

Due to lean fuel-to-air operating regimes, diesel engines produce extremely low HC emissions. Average levels of gaseous HC in diesel exhaust gas are around 20 to 300 ppm. Heavy-duty diesel vehicles generally produce more total HC emissions compared to light-duty vehicles. In terms of emission regulations, HCs are regulated as volatile gas-phase HC. The remaining particulate HC phase, liquid or adsorbed, is considered to belong to soluble organic portion of diesel PM. Utilization of catalysts in diesel emission control results in reduction of HC levels following its oxidation into CO₂ and water. However, mild oxidation conditions can lead to formation of harmful and toxic aldehydes or ketones (Majewski & Khair, 2006; National Center for Environmental Assessment EPA, 2002).

Nitrogen Oxides

The term NO_x is usually used to refer to a group of interconvertible gases that are produced during the fuel combustion or heating of the air to temperatures above 650 °C (1,200 °F) in the presence of oxygen. These gases can also be generated as a result of oxidation of nitrogen-containing compounds by bacteria in soil. The group of nitrogen oxides includes major gases, such as NO that can be further oxidized to NO₂ (Cunningham & Woodworth-Saigo, 1990). NO_x are of major environmental concern. They are very persistent in the atmosphere, can be transported over long distances from emission source, and cause a variety of health issues and environmental impacts. NO_x and HCs in the atmosphere create smog in presence of sunlight. Another negative effect

of NO_x is acidification (nitric acid NHO₃) as a result of their chemical reaction with water. Nitrogen dioxides and nitrous oxides also contribute to particulate matter formation. High concentrations of nitrous oxides lead to stratospheric ozone depletion, and cause global warming by means of ultraviolet light absorption (Godish, 2004; Lloyd & Cackette, 2001; OTAQ, 2012c).

Transportation contributes about 45-50% to total emissions of NO_x. Major part (95%) of anthropogenic emissions comes from fuel combustion in transportation and electric power generation. Examples of human-caused emissions include gasoline- and diesel-powered vehicles, industrial boilers, municipal incinerators, and fossil fuel-fired electric generating stations (Cunningham & Woodworth-Saigo, 1990; Godish, 2004; Rodrigue et al., 2009). According to the OTAQ (2012c) estimates, diesel vehicles represent the major contributors to national emission of nitrogen oxides from on-road and non-road mobile sources and input as much as 42 and 49%, respectively.

NO is a colorless and odorless gas. It is produced in heated and pressurized diesel combustion chamber as a result of chemical reaction between nitrogen and oxygen. The proportion of NO in diesel exhaust varies from 95% in older engines to 85% in newer turbocharged engines (Majewski & Khair, 2006). After being exhausted into the atmosphere, NO can be further oxidized into NO₂, a reddish-brown toxic gas with a characteristic irritating odor. It possesses strong oxidation properties and, therefore, it is extremely reactive and highly toxic (Godish, 2004).

N₂O is not included in diesel NO_x classification according to regulatory definition. Therefore, N₂O is currently one of unregulated diesel emissions. It is

commonly known as a laughing gas. At room temperatures, it is a colorless non-flammable gas with a somewhat sweet odor and taste. At high temperatures, N_2O acts as a strong oxidizer. As a result, N_2O is a greenhouse gas and air pollutant (Godish, 2004). However, diesel engines usually produce very low N_2O emissions levels of about 3 ppm (Majewski & Khair, 2006).

NO_x represents the second crucial and strictly regulated diesel exhaust pollutant after particulates due to its significant contribution to smog formation. Diesel NO_x and PM emissions are interdependent resulting in NO_x -PM trade-off, when reduction of NO_x is accompanied by increase of PM levels, and vice versa (Desantes, Bermúdez, Pastor, & Fuentes, 2006; Lahde et al., 2010). Emissions from diesel-powered engines account for about one-third of the total NO_x emissions in the United States and one-quarter of the total PM emissions from mobile sources and represent a significant air quality concern (Krishnan & Tarabulski, 2005).

Diesel Particulates

Diesel PM is considered to be one of the most harmful emissions and the main focal point of regulatory and emission control efforts. It is comprised of extremely complex aerosol system, including agglomerated particles of elemental carbon with various physical and chemical properties due to adsorption of other chemical compounds present in the exhaust gas. Ambient PM can be classified into particles with aerodynamic diameter less than $10\ \mu m$ (PM₁₀), particles with diameters less than $2.5\ \mu m$ (PM_{2.5}), ultrafine particles with diameters less than $0.1\ \mu m$, and nanoparticles with diameter less than 50 nm (Cunningham & Woodworth-Saigo, 1990; Godish, 2004; F. J. Kelly &

Fussell, 2012). Primary diesel PM emissions get released directly from diesel engines, whereas secondary diesel PM gets formed as a result of secondary reactions of the gaseous compounds present in the exhaust and in the atmosphere (Chow, 1995; EPA, 2002; F. J. Kelly & Fussell, 2012).

Main sources of particulate matter pollution are anthropogenic emissions from transportation, fuel combustion in stationary sources, power generation, industrial processes, and solid waste disposal (Godish, 2004; F. J. Kelly & Fussell, 2012). About 25-28% of total emissions of particulates come from transportation. Diesel vehicles and engines are major polluters and generate more than half of the mobile source particulate emissions. Diesel engines of non-road and on-road vehicles contribute 72 and 57% of PM pollution, respectively (OTAQ, 2012e; Rodrigue et al., 2009).

Size characterization of diesel PM. Based on their physical properties, PM diesel emissions are divided into two different modes: nuclei and accumulation modes. The coarse particles are not directly emitted by diesel engines; however they are produced by means of deposition and re-entrainment of PM from the sampling system, engine exhaust system, and engine walls. Particles in nuclei mode are between 0.007 and 0.04 μm in diameter and correspond to nanoparticles. The maximum nuclei mode levels are in the range between 10 and 20 nm. Accumulation mode includes particles with diameter from 0.04 to 1 μm and has maximum particle concentration between 0.1 and 0.2 μm (see Figure 2). Accumulation mode contains fine particulates, ultrafine PM, and a portion of nanoparticles. In regard to total PM mass, nuclei amount to only few percent. However,

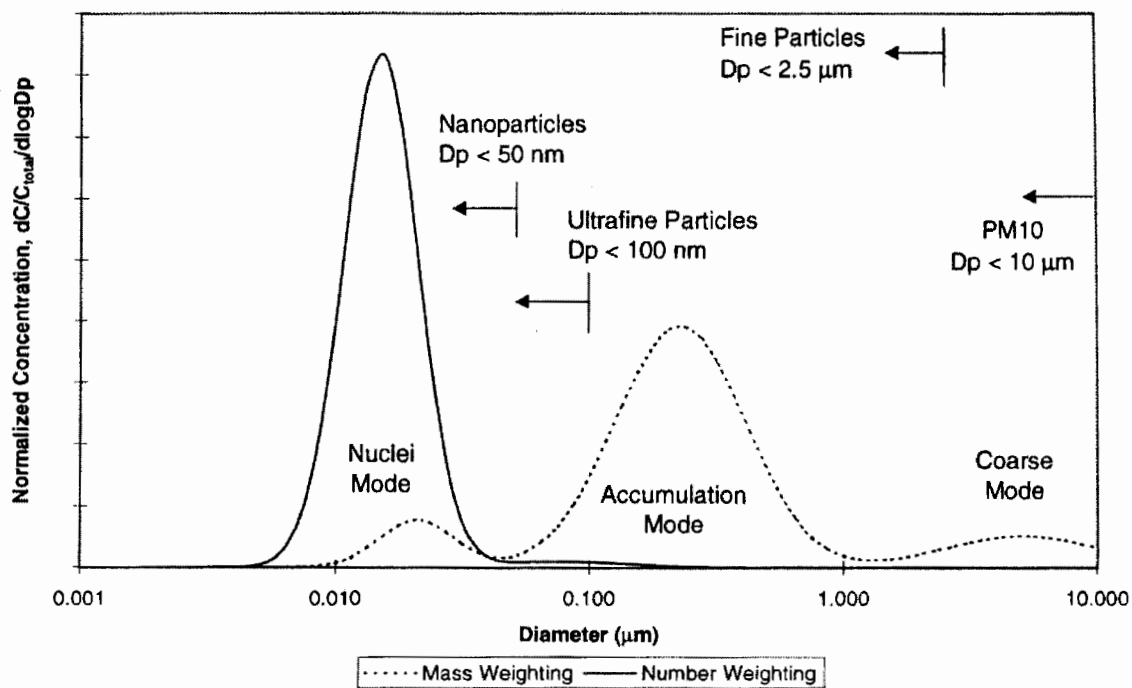


Figure 2. Engine exhaust PM size distribution (adopted from Kittelson, 1998).

because of their very small size they have extremely large particle number, which is about 90% of the total diesel PM number (Desantes et al., 2006; Kittelson, 1998; Majewski & Khair, 2006; National Center for Environmental Assessment EPA, 2002). This further underscores the findings of many scientists linking exposure to fine and ultrafine PM to adverse health effects (Arhami et al., 2009; Avino, Casciardi, Fanizza, & Manigrasso, 2011; S.-C. Chen et al., 2010; Tobias et al., 2001).

Particles of nuclei mode are mostly volatile unstable components, such as condensates of hydrocarbon and hydrated sulfuric acid condensates, with only minor portion of solid compounds generally from lube oil additives (i.e., carbon and metallic ash). As exhaust gas temperature gets lower during its dilution and mixing with

surrounding air, nuclei mode gets created from various gaseous precursors. Due to nuclei being highly dependent on dilution rate and mixing residence time, different PM sampling methods may yield different results due to variations in sampling conditions (Kittelson, 2001; Mathis et al., 2004; Samaras, Vouitsis, & Ntziachristos, 2008).

Accumulation mode particles include a mixture of solid carbon with condensed heavy HCs. Several other compounds can also be present in accumulation mode, such as metallic ash, sulfur, and various cylinder wear metals. Major creation mechanism of this mode is agglomeration of primary carbon particles and other solid material by collision with following adsorption of gases and condensation of vapors (Desantes et al., 2006; Díaz, Reed, & Fu, n.d.; Ma, Jung, & Kittelson, 2008; Majewski & Khair, 2006; Ziemann, Hiromu, & McMurry, 2002).

The total PM mass is comprised of the following fractions: solid fraction (SOL), soluble organic fraction (SOF), and sulfate particulates. Solid fraction includes elemental carbon and ash. Soluble organic fraction is further subdivided into organic material from engine lubricating oil and from fuel, whereas sulfate particulates consist of sulfuric acid and water (Agarwal, Gupta, & Kothari, 2010; Kittelson, 1998; National Center for Environmental Assessment EPA, 2002). According to Kittelson (1998), a typical particle composition for a heavy-duty diesel engine consists of 41% carbon, 25% unburned lube oil SOF, 14% sulfate particulates and water fraction, 13% ash and other, and 7% unburned fuel SOF. Figure 3 created using data from Kittelson (1998) study is presented below.

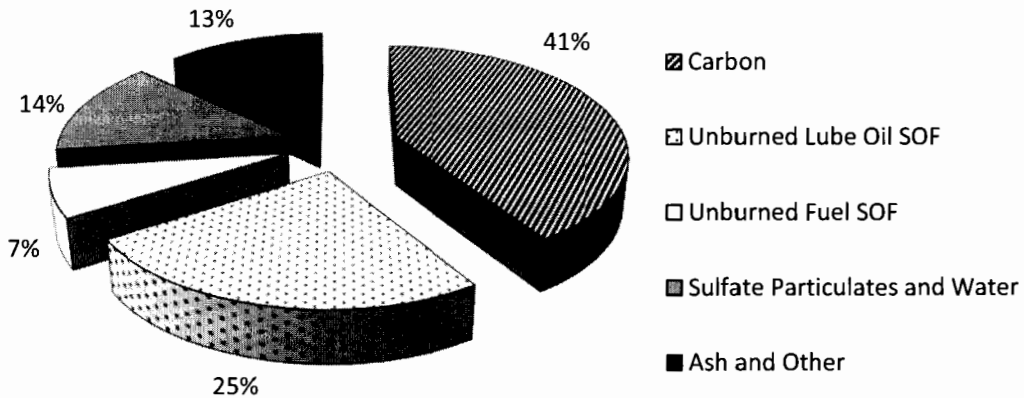


Figure 3. Typical composition of diesel PM tested in a heavy-duty transient cycle (based on data from Kittelson, 1998).

Solid PM fraction. Major portion of diesel PM is carbon SOL fraction (both nuclei and agglomerates) which is created as a result of combustion process in regions with locally rich air-fuel ratios. While most of carbon SOL gets further oxidized, the remaining solid residue undergoes additional agglomeration as it gets exhausted into the atmosphere (Avino et al., 2011; Di Iorio et al., 2012; Kittelson, 1998). Solid carbon is responsible for black smoke emissions that are typical for older diesel engines (Gupta, 2006). Primary nuclei particles have spherical shape and represent graphite crystallites consisting of several carbon atoms. In turn, hundreds of nuclei mode particles get combined into an accumulation mode particle with clustered structure lacking a well-defined diameter (Case & Hofeldt, 1996; Godish, 2004).

Ash portion of SOL particulate fraction is generated from lubricating oil additives and from engine wear during the power stroke of the diesel engine cylinder. Initially ash is comprised from nuclei mode particles that can later combine into agglomerates. Additionally, minor quantities of inorganic ash are produced from metal compounds in fuel and lube oil. Typical composition of diesel exhaust ash from lubricating oil additives includes sulfates, phosphates, calcium oxides, zinc oxides, and magnesium oxides. Furthermore, various metal oxide impurities are introduced into lube oil from engine wear, such as oxides of iron, copper, chromium, and aluminum. Finally, exhaust system corrosion can also contribute to ash generation by delivering the iron, chromium, nickel, and aluminum oxides (Agarwal et al., 2010; Majewski & Khair, 2006).

Soluble organic compounds. Soluble organic compounds are produced from condensation or adsorption of heavy hydrocarbons on carbon particles' surfaces during dilution and mixing of exhaust gases with air. Condensation of hydrocarbons resulting in the subsequent formation of volatile nuclei occurs when the amount of carbon particles is not sufficient for adsorption of all available hydrocarbons (Burtcher, 2001; Desantes et al., 2006; Di Iorio et al., 2012; Samaras et al., 2008). Usually SOFs are present in hot diesel exhaust in the form of vapors. Levels of SOF emissions depend on engine type and on its operating conditions. Higher SOF emissions are generally produced by two-stroke engines compared to their four-stroke counterparts. For each given engine, SOF content is normally at its peak at low engine loads. Particulates emitted from engines with low SOF portion compared to total PM mass are defined as dry particulates. Correspondingly, high SOF particles are identified as wet particulates. Share of SOF can

range from less than 10% to more than 50% of the total PM in dry and wet particulates, respectively (Du et al., 2007; Majewski & Khair, 2006; National Center for Environmental Assessment EPA, 2002; Sharma, Agarwal, & Bharathi, 2005).

Sulfate particulates. Sulfur in the diesel fuel serves as a precursor for sulfate particulate fraction. As a result of oxidation process, the main portion of sulfur gets oxidized to sulfur dioxide, an unregulated diesel compound. Its concentrations increase in direct proportion to increasing fuel sulfur level. The remaining sulfur from diesel fuel gets transformed into sulfur trioxide which can subsequently be converted into sulfuric acid in the presence of water. The proportion of sulfur dioxide and sulfur trioxide in diesel exhaust is usually around 95 to 5%, respectively. Reactions of sulfuric acid with water occurring during dilution process produce sulfate particulates as a result of hetero-nucleation. Nucleation process of sulfuric acid and water depends on vapor pressure of both sulfuric acid and water which, in turn, are influenced by fuel sulfur level, conversion rate of fuel sulfur to sulfur trioxide, air-to-fuel ratio, and dilution tunnel temperature and relative humidity. Diesel engines running on fuel with reduced sulfur content of 500 ppm release approximately 0.01 g/bhp-hr of sulfate particulate emissions. Some catalytic after-treatment technologies required in modern diesel vehicles, such as oxidation catalysts, produce a considerable portion of SO₃ emissions as they oxidize SO₂ during the course of their normal operation (Majewski & Khair, 2006; National Center for Environmental Assessment EPA, 2002; Samaras et al., 2008).

Sulfate particulates can be present separate from carbon particles. They also can exist in accumulation mode attached to carbon particles and mixed with soluble organic

substances. Moreover, solid nonvolatile sulfate salts, such as calcium sulfate, can be formed from sulfuric acid based on available quantities of metal-based compounds. Sulfate particulates with higher salt content bind less water compared to those with lower salt content due to being more hydrophilic. When using gravimetric filtering method during PM emission analysis, the amount of water could be calculated based on relative humidity and temperature conditions of the sampling filter maintained during its preconditioning (Du et al., 2007; Majewski & Khair, 2006; National Center for Environmental Assessment EPA, 2002; Samaras et al., 2008).

Polycyclic aromatic hydrocarbons and dioxins. Polycyclic aromatic hydrocarbons (PAHs) can be found in SOF fraction of PM and represent aromatic hydrocarbons with a complex structure consisting of up to six benzene rings. PAHs can also be comprised of cyclopentane and heterogeneous rings with atoms of sulfur (PASH) or nitrogen (PANH) that are generated in the engine at high concentrations of sulfur and nitrogen oxides, respectively. Many experts defined PAHs as toxic, mutagenic, and oftentimes carcinogenic compounds PAHs oftentimes account for a fraction of a percent of the total PM emissions (Agarwal et al., 2010; Dobbins, 2007; Hsieh et al., 2011; Rajput & Lakhani, 2009; Shi et al., 2010).

PAHS can be found in gas and particulate portions of diesel exhaust. Studies conducted by Rajput and Lakhani (2009) identified that the high molecular weight PAHs are dominant in exhaust emissions compared to low molecular weight PAHs in diesel fuel. Organic fraction of PM contains the most harmful species with four or more rings. PAHs come from various sources, including diesel fuel, pyro-synthesis occurring in the

engine cylinder, and high NO_x concentrations resulting in generation of PAHs nitro-derivatives (National Center for Environmental Assessment EPA, 2002; Scheepers & Bos, 1992). In diesel engines, PAHs are produced during the pyrosynthesis process occurring in the engine's cylinder and constitute about 1% or less of total PM (Rajput & Lakhani, 2009).

Dioxins represent a group of 210 chemical compounds (congeners) that share similar chemical structures and biological characteristics (Michigan Department of Environmental Quality, 2005). There are three major families in this group: the chlorinated dibenzo-p-dioxins (CDDs), chlorinated dibenzofurans (CDFs), and polychlorinated biphenyls [PCBs] (Hsieh et al., 2011; Office of Environmental Health Hazard Assessment, 2001). As many as 17 dioxins and furans in this group are considered toxic (Schechter, Birnbaum, Ryan, & Constable, 2006). The most toxic chemical in the group is 2,3,7,8-tetrachlorodibenzo-para-dioxin [2,3,7,8-TCDD] (Action Pennsylvania, 2012). The United States Department of Health and Human Services, the National Institute for Occupational Safety and the Health and the WHO's International Agency for Research on Cancer, as well as the EPA, identified 2,3,7,8-TCDD as a potent human carcinogen (Bertazzi et al., 2001; EPA, 2012e; Michigan Department of Environmental Quality, 2005, National Research Council of the National Academies, 2006; WHO, 1998). The main sources of dioxins include combustion of chlorine containing municipal and hazardous waste, chemical and pesticide manufacture, metal production, forest fires, and volcanic eruptions (Agency for Toxic Substances and Disease Registry, 1999; Office of Environmental Health Hazard Assessment, 2001).

Diesel fuel catalytic additives, such as copper, may increase the amount of released dioxins by orders of magnitude (Laroo, Schenk, Sanchez, & McDonald, 2011). Study by Heeb et al. (2007) found an increase of PCDD-F emissions up to three orders of magnitude when using copper-catalyzed DPFs.

PM emission trends of modern diesel engines. Diesel engine technology, design, and fuel formulations have evolved over the last decades to reduce air pollution emissions. In order to comply with increasingly stringent U.S. emission standards, modern post-2007 diesel engine models and most older retrofitted diesel engines are using ultra-low-sulfur diesel fuel and are equipped with technologically advanced emission controls, such as oxidation catalysts, wall-flow diesel particulate filters, and various electronic controls. As a result, PM in diesel exhausts of newer engines (especially of engine models 2007 year and later) is quantitatively and qualitatively very different from that found in traditional diesel exhaust. PM emissions of post-2007 engines have greater resemblance to those from engines using compressed natural gas or gasoline (Hesterberg et al., 2011). The concentrations of PM and associated compounds in modern diesel engines are more than hundred-fold lower than levels found in older diesel engines (Wall & McDonald, 2012).

However, newer diesel engines can generate higher numbers of ultrafine particles while still reducing PM mass emissions (Tobias et al., 2001). According to results of the study conducted by Bagley, Baumgard, Gratz, Johnson, & Leddy (1996) on older 1991 and newer 1988 year model engines, the utilization of particulate trap was able to substantially reduce the weight of particles emitted and subsequently lowered the

emissions of organic chemicals associated with PM and the overall mutagenicity of diesel exhaust. They concluded that low-sulfur fuel in combination with particulate trap and catalytic converter reduces PM levels without significantly increasing emissions of other pollutants. When newer model engine was operating without emission control devices, the research findings indicated a 30- to 60-fold increase in the number of small primary particles compared to older model engine leading to a greater total PM number. On the other hand, integrated data comparison of particle number emissions of different engines obtained from a number of studies indicate that 1999-2000 engine models generate fewer nanoparticles than 1991 model engines used in research of Bagley et al. (1998).

Other studies also report an increase of particle numbers in exhausts of newer diesel engines. However, most findings observe smaller increase of particle numbers than showed by Bagley et al. (1998). For example, results of comparison study conducted by Mayer et al. (1998) for newer and older engines revealed up to a sixfold increase in total particulate number in emissions of newer engines. On average the increase ranged between 15 and 50%. Research conducted by Khalek et al. (2011) on heavy-duty diesel engines 2007 model year equipped with high-efficiency catalyzed DPFs revealed a 90% increase in volatile nanoparticles during DPF active regeneration with measured particle numbers similar to those from previously tested 2004 model engine. However, for combined periods with and without active DPF regeneration, 2007 model engines demonstrated lower particle number emissions compared to results from older engine.

Additionally, research revealed that the biggest portion of PM emissions from the 2007 model engines mainly included volatile nuclei mode in the sub-30-nm size range. As seen in Figure 4, composition of PM emissions consisted of 53% sulfates, 30% organic carbon, 13% elemental carbon, and 4% metals and other elements. This represented a major difference compared to a common PM composition of pre-2007 engines with levels of elemental carbon of up to 90%. Furthermore, elemental carbon rates usually vary in pre-2007 engine emissions depending on operation mode. Contrary, engines equipped with high-efficiency DPFs are expected to generate stable small levels of elemental carbon regardless of engine operation (Khalek et al., 2011).

Reduction of the solid PM fraction in emissions of modern vehicles equipped with DPFs accompanied by increased particulate sulfate fraction identified by Khalek et al. (2011) is consistent with findings of other researchers (Hesterberg et al., 2011; Kleeman, Schauer, & Cass, 2000; Liu et al., 2009). Additionally, modern diesel engines produce lower lower PAH emissions, volatile organic compound, and aldehyde emissions (Hesterberg et al., 2011; Khalek et al., 2011). Currently, a variety of older and newer diesel engines is used in mobile sector applications. Therefore it is important to understand a broad spectrum of health and environmental effects of diesel PM emissions.

Health and Environmental Effects of Diesel PM Emissions

Rapid growth and development of transportation industry, including diesel vehicles sector, have resulted in significant increase of emissions, especially in the last decades. Historically, heavy-duty trucks have represented the majority of total utilized

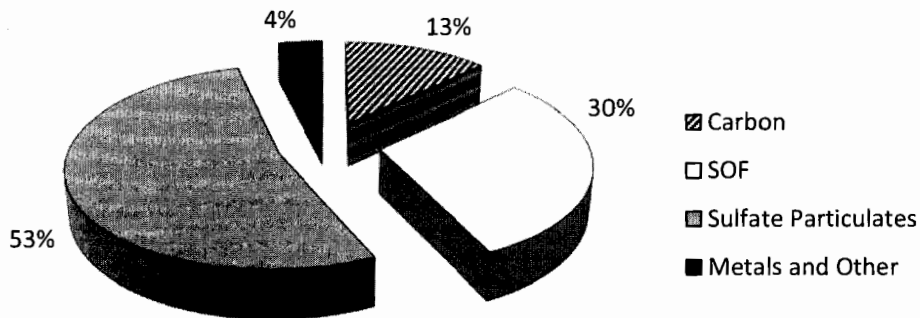


Figure 4. Diesel PM emissions from 2007 year model engines in a combined cycle 16-hour cycle consisting of heavy-duty transient cycle segments mixed with CARB 5-Modes cycle (based on data from Khalek et al., 2011).

diesel vehicles based on sales numbers and mileage (EPA, 2002). Data from the OTAQ and the National Highway Traffic Safety Administration (NHTSA) show that of all freight moved in year 2007 in the United States 71% by tonnage and 87% by value was transported using diesel vehicles, specifically heavy-duty trucks. According to the EPA, the total sales projections for diesel heavy-duty trucks are estimated to increase and account for up to 1,526,871 units per year in the year 2014 (OTAQ & NHTSA, 2011).

Diesel-equipped vehicles produce much greater gram per mile particulate emissions compared to those from gasoline vehicles. Additionally, diesel vehicles oftentimes have greater number of traveled miles. As a result, despite of relatively smaller number of diesel engines, their contribution to the national emissions inventory is higher compared other types of vehicles (EPA, 2002; X. Zhu, Durbin, Norbeck, & Cocker, 2004).

Diesel vehicles have been attracting immense interest due to being recognized as significant contributors to the atmospheric inventory of a number of pollutants, including fine PM with various adsorbed toxic compounds and other chemicals that act as respiratory irritants and inflammatory agents (EPA, 2002; Schneider & Hill, 2005). From an environmental standpoint, aerosol particles play a crucial role in the atmosphere influencing global climate change and resulting in visibility reduction (Godish, 2004). Exposure to exhaust from diesel engines is viewed as a serious threat to the environment and public health.

Health Effects

Exposure to diesel exhaust represents serious health hazard to humans and leads to numerous acute and chronic health effects. A variety of health effects caused by diesel exhaust can be explained by its complex composition comprised of hundreds of compounds in gas and particle form. Many constituents of diesel exhaust are individually known to be mutagenic and carcinogenic (Díaz et al., n.d.; EPA, 2002; National Toxicology Program, 2011).

PM phase of diesel exhaust, including solid inorganic carbon and the associated organic material, poses the most serious threat to human wellbeing (WHO, 1996). Therefore, oftentimes exposure to particulate phase of diesel exhaust is used as a surrogate measure of exposure to the whole diesel exhaust (EPA, 1999b; EPA, 2002; WHO, 1996). Majority of toxic organic compounds are concentrated on the surfaces of fine diesel particles. Large surface area of fine PM acts as an excellent medium for adsorbing toxic organics. Additionally, fine particles due to their small size can penetrate

deep into the lungs and enter the bloodstream (EPA, 2002; National Toxicology Program, 2011; Nel, Xia, Mädler, & Li, 2006; Schneider & Hill, 2005).

Exposure to fine PM has been associated with the following health impacts: respiratory symptoms (i.e., enhanced allergic responses, irritation of respiratory organs, lightheadedness, nausea, coughing, worsening of existing respiratory conditions, chronic bronchitis, asthma attacks, and reduced lung function), various cardiovascular problems (e.g., heart attacks), as well as premature mortality (DieselNet, 2004b; EPA, 1999b; Schneider & Hill, 2005). Data from Schneider and Hill (2005) study specifies that each year over 400,000 and 27,000 of nation's population suffers from asthma and heart attacks, respectively. Consequently, this leads to thousands of hospitalizations, emergency room visits, and lost work days. Modeling study conducted by the Clean Air Task Force based on 1999 National Emissions Inventory concluded that cancer risk from diesel exhaust is seven and a half times higher than the combined total cancer risk from all other air toxics in the nation. According to their estimates, lung cancer from exposure to diesel exhaust results in about 3,000 each year (Schneider & Hill, 2005).

The National Institute for Occupational Safety and Health (NIOSH) was the first to address the serious health effects posed by exposure to diesel exhaust. It confirmed carcinogenic evidence of diesel exhaust based on results of animal studies. Because of limited human evidence of diesel carcinogenicity, diesel exhaust was identified as potential occupational carcinogen (NIOSH, 1988). In 1989 and 1996, diesel motor exhaust was classified as probable carcinogen to humans (group 2A) by the IARC and by the WHO, respectively (IARC, 1989; WHO, 1996).

As a result of a national-scale air toxics assessment conducted by the EPA in 1999, a variety of diesel exhaust compounds have been listed as hazardous air pollutants. Diesel exhaust was characterized by the EPA as a mobile source air toxic due to its various carcinogenic and non-carcinogenic health impacts (EPA, 1999a). Chronic reference concentration value of $5 \mu\text{g}/\text{m}^3$ was identified by the EPA for diesel PM (EPA, 1999c). Additionally, particulate emissions from diesel-fueled engines were identified as air toxic and were included in the CARB's toxic air contaminant identification list in 1998 (CARB, 2012b). In the year 2000, the National Toxicology Program administered by the Department of Health and Public Services listed diesel exhaust PM as reasonably anticipated to be a human carcinogen (National Toxicology Program, 2011). The CARB listed more than 40 components of the diesel engine exhaust as toxic air contaminants (CARB, 2000a). Findings of international and national agencies regarding diesel toxic health impacts are presented in Table 1.

Occupational diesel PM exposure. Over the past years, various scientists examined diesel exhaust exposure in both ambient and occupational environments. Generally, higher PM exposure levels have been found in occupational settings compared to ambient exposures (DieselNet, 1999; National Toxicology Program, 2011; Olsson et al., 2011). Majority of occupational investigations focused on carcinogenic properties of diesel exhaust, specifically of diesel PM concentrations. Epidemiological studies involving groups of individuals exposed to diesel PM levels in occupational settings have

Table 1. Diesel carcinogenetic findings based on studies conducted by international and national environmental and health agencies (CARB, 2000a; EPA, 1999a, 1999c; IARC, 1989; National Toxicology Program, 2000; NIOSH, 1988; WHO, 1996).

Year	Agency	Findings
1988	National Institute for Occupational Safety and Health	Potential occupational carcinogen based on confirmatory animal and limited human evidence
1989	International Agency for Research on Cancer	Probable carcinogen (group 2A) based on limited evidence in humans and sufficient evidence in experimental animals
1996	World Health Organization	Probable carcinogen (group 2A) based on limited evidence in humans and sufficient evidence in experimental animals
1998	California Air Resources Board	Toxic air contaminant based upon the cancer risk to the public and the potential short-term and long-term respiratory effects of diesel exhaust
1999	Environmental Protection Agency	Mobile source air toxic due to various carcinogenic and non- carcinogenic health impacts: <ul style="list-style-type: none"> - acute (short-term episodic exposure) and chronic (long-term) exposure can pose hazards to humans - probable human carcinogen, or is likely to be carcinogenic in humans by inhalation at environmental or higher exposure conditions
2000	National Toxicology Program of the Department of Health and Public Services	Reasonably anticipated to be a human carcinogen based on animal and occupational human studies

shown increases in relative risks of lung cancer and respiratory disease (Boffetta, Jourenkova, & Gustavsson, 1997; Lipsett & Campleman, 1999; Sauvain, Vu Duc, & Guillemin, 2003). The following occupational groups have been identified to have an increased occupational lung cancer risk from exposure to diesel exhaust: miners, trucking company workers, bus garage employees, dock workers, and other groups involved in servicing or handling diesel-powered equipment (Cohen & Higgins, 1995; EPA, 2002). Apportionments studies found that diesel exhaust constitutes 78–98% of the total fine PM mass and greater than 90% of the fine particle carbon (McDonald, Zielinska, Sagebiel, McDaniel, & Mousset-Jones, 2003).

A cohort mortality study conducted by Attfield et al. (2012) on mining industry workers exposed to diesel exhaust found a further evidence of increased mortality risk from lung cancer. Additionally, the findings identified higher lung cancer mortality risk associated with diesel exhaust exposure among ever-underground workers compared to lower risk found in surface-only workers. Results of this study are consistent with conclusions of occupational exposure assessment performed by Pronk, Coble, and Stewart (2009). According to Pronk et al. (2009), exposure to diesel exhaust PM in underground mining and surface mining operations ranged from 148 to 637 $\mu\text{g}/\text{m}^3$ and from 13 to 23 $\mu\text{g}/\text{m}^3$, respectively. Somewhat lower average levels have been reported by Stewart et al. (2010) during 1998-2001 historical assessment. They estimated that the average exposures can range from 40 to 384 $\mu\text{g}/\text{m}^3$ for the underground workers and from 2 to 6 $\mu\text{g}/\text{m}^3$ for the surface workers. However, fine PM mass levels in underground mines can even exceed 1 mg/m^3 (McDonald, Zielinska, Sagebiel, & McDaniel, 2002).

Research on trucking industry workers exposures to diesel exhaust PM conducted by Zaebs et al. (1991) found that exposures of dock workers (especially diesel forklift trucks workers) and mechanics were significantly higher than background exposures. Exposures in these two groups were also significantly higher than those in the local and road drivers. Average diesel PM exposures of truck drivers ranged from 3 to 5 $\mu\text{g}/\text{m}^3$ which is consistent with 2 to 3 $\mu\text{g}/\text{m}^3$ more recent findings reported by Attfield et al. (2012). Based on relatively minor variations in exposure levels of highway truck drivers (3.8 $\mu\text{g}/\text{m}^3$) with highway background concentrations of 2.5 $\mu\text{g}/\text{m}^3$, Zaebs et al. (1991) concluded that the highway environment, rather than the trucker's vehicle, has greater contribution to diesel PM exposure. It has been suggested in that diesel truck drivers also have an increased lung cancer risk compared to workers in other industries.

Studies by Sauvain et al. (2003) showed an increase in exposures to elemental carbon and PAHs fractions of diesel PM by three to six times and ten times, respectively, in a bus depot, a truck repair workshop, during winter compared to summer. This trend has been attributed to decreased ventilation during the winter season. Several studies found evidence of increasing lung cancer risk with increasing employment time in trucking industry. These findings are consistent for older and recent studies (Garshick et al., 2008; Steenland, Silverman, & Hornung, 1990). Boffetta et al. (2007), Garshik et al. (2008), and Lipsett and Campleman (1999) specified that lung cancer risk not only rises with increasing years of work but is also greater in workers regularly exposed to diesel PM exhaust in direct proportion with increasing exposure duration. Results of this study support Zaebs et al. (1991) findings and indicate that surrounding vehicles and

background air pollution represent the main sources of truck drivers' exposure to diesel PM; whereas smaller exposure is attributable to emissions from driver's own vehicle. A retrospective assessment conducted by Davis, Hart, Laden, Garshick, and Smith (2011) identified significant variability in estimated historical exposures of trucking-related workers to elemental carbon in diesel exhaust with highest exposure levels (over $40 \mu\text{g}/\text{m}^3$) in the 1970s. For the lowest exposed categories in the 1990s, the average estimated diesel PM exposures during a typical work shift ranged from less than $1 \mu\text{g}/\text{m}^3$.

Ambient diesel PM exposure. Health impacts of ambient PM levels remain a concern on local and regional scale. PM emissions are primarily generated by automotive traffic, especially by diesel-powered engines (X. Zhu et al., 2004). Typically, people in industrialized and heavily populated urban areas with high traffic density have relatively high diesel PM exposure levels (DieselNet, 2004b; X. Zhu et al., 2004). Ambient exposure to diesel PM generally varies from 0.2 to $4 \mu\text{g}/\text{m}^3$; while average exposure levels are usually around $1.8 \mu\text{g}/\text{m}^3$ (Majewski & Khair, 2006).

Transportation sector provides significant contribution to atmospheric levels of fine PM pollution in Los Angeles area. Local source apportionment studies characterize that transportation sources contribute on average from 28% to 68% to total ambient levels of fine PM (Held, Ying, Kleeman, Schauer, & Fraser, 2005; Schauer, Fraser, Cass, & Simoneit, 2002; Valavanidis, Fiotakis, & Vlachogianni, 2008). Mobile sources from Los Angeles area also contribute 43% of total fine PM emissions in areas located downwind. In the state of California, total share of diesel PM emissions accounts for about 55% to 94% of the overall fine PM pollution generated from mobile sector (Held et al., 2005;

Schauer et al., 2002; Zhao & Hopke, 2004). Generally, in urban centers diesel PM_{2.5} can constitute as much as 36% of the total ambient PM inventory (EPA, 2002; Schneider & Hill, 2005).

Atmospheric particles, especially fine PM, have been receiving significant attention because of their potential for causing adverse health effects indicated by numerous toxicological investigations (Betts, 2011; S.-C. Chen et al., 2010; Li et al., 2003; Vinzents et al., 2005). Studies of the past decades were the first to identify that fine particles exhibit more toxicity compared to coarse PM in proportion to decrease in PM size (Ferin, Oberdörster, Soderholm, & Gelein, 1991; Oberdorster, Ferin, & Lehnert, 1994). Results from toxicological research of Valavanidis et al. (2008) and De Kok, Driee, Hogervorst, and Briede (2006) further suggested that oxidative stress, inflammation, and adverse cellular effects (i.e., cytotoxicity, mutagenicity, DNA damage, and stimulation of pro-inflammatory cytokine production) could be responsible for the PM of smaller size having higher toxicity compared to coarse particles. However, while fine PM have been more strongly associated with mortality and morbidity than coarse particles; the number of hospital admissions due to respiratory problems have been more strongly related with exposure to coarse fraction of PM (Brunekreef & Forsberg, 2005).

Significant epidemiological evidence supports an association between exposure to ambient air pollution and increased mortality and morbidity, including a wide range of adverse respiratory, cardiovascular, toxic, and cancerous effects (Castranova et al., 2001; Delfino, Sioutas, & Malik, 2005). The study conducted by Pope et al. (2002) provided evidence of association between elevated fine PM pollution in ambient air and increased

risk of cardiopulmonary and lung cancer mortality. Pope et al. (2002) estimated that each $10 \mu\text{g}/\text{m}^3$ increase in long-term fine PM levels resulted in a four, six, and eight percent increase in cardiopulmonary and lung cancer mortality risk. Increased lung cancer risk was also found to be associated with elevated long-term ambient concentrations of PM₁₀ and SO₂ (Beeson et al., 1998).

According to Cohen et al. (2005) study, ambient PM pollution on a global scale accounts for about four percent of adult cardiopulmonary disease mortality; about five percent of combined lung cancer, trachea, and bronchus mortality; and about one percent of children mortality from acute respiratory infection. This results in to 800,000 (1.2%) premature deaths and 6.4 million (0.5%) lost life years. These findings are consistent with previous epidemiologic studies suggesting that air pollution from combustion sources, including diesel internal combustion engines, significantly contributes to the lung cancer incidences among the general public (Cohen & Pope, 1995).

Several epidemiological studies explored the mechanisms of ambient ultrafine PM and related adverse respiratory, cardiovascular, and immune system effects. Results of study conducted by Brown, Zeman, and Bennett (2002) showed a high deposition efficiency of fine PM in the pulmonary region of healthy subjects. Patients with asthma or chronic obstructive lung diseases were observed to have an increased PM deposition rate compared to healthy patients (Brown et al., 2002; Chalupa, Morrow, Oberdörster, Utell, & Frampton, 2004). Nemmar, Hoylaerts, Hoet, and Nemery (2004) hypothesized that this effect could be explained by ability of fine PM to enter the systematic circulation and impose direct effects of on myocardium or coronary vasculature. Research on

mechanisms of diesel PM impacts on immune system identified that increased rate of respiratory allergies may be due to increased of the lung susceptibility to infections as a result of depressed antimicrobial potential of alveolar macrophages (Castranova et al., 2001; Diaz-Sanchez, 1997; Diaz-Sanchez, Dotson, Takenaka, & Saxon, 1994).

While non-carcinogenic fine PM exposures have been known to increase the risk of asthma and chronic bronchitis, cause airway inflammation, and lead to allergies, scientists examined additional threats of PM exposure on children. Numerous medical studies have documented that ambient PM is associated with adverse health impacts in children, such as crib death in infants, reduced lung development, deficits in lung function, and other respiratory effects (Pekkanen, Timonen, Ruuskanen, Reponen, & Mirme, 1997; Ritz, Wilhelm, & Zhao, 2006; Woodruff, Parker, & Schoendorf, 2006).

Environmental Effects

Environmental impacts of diesel exhaust represent a significant concern. Components of diesel exhaust get accumulated and dispersed in the atmosphere and other environmental media. Diesel particulates backscatter the direct sunlight and thus reduce the amount of sunlight reaching the earth (Davidson, Phalen, & Solomon, 2005; Lohmann & Feichter, 2004; Majewski & Khair, 2006). In urban areas the effects of cloudiness and fog resulting from this phenomenon are more evident than in rural areas. PM pollution represents a main cause of poor visibility and haze on local and global scale. Both coarse and fine fractions of diesel PM contribute to haze and visibility impairment. In general, visibility decreases as concentration of PM in the atmosphere increases. As a result, visual range can be reduced by as much as 70% from natural

levels (EPA, 2011a; Malm, 1999). Larger coarse particles settle more easily, faster, and in closer proximity to its generation source than fine PM. Remaining fine airborne aerosols, including particles and droplets, are likely to remain suspended in the ambient atmosphere for extended periods. Increased wind speeds significantly decreases the ultrafine particles levels. Oftentimes, they fine PM is transported over long distances and settles far from its original emission source (Godish, 2004; Sabbagh-Kupelwieser, Horvath & Szymanski, 2010).

Because of their size and composition, diesel PM can create other undesirable effects, such as rapid accumulation and dust deposition on buildings. Many historical monuments and architectural structures can be affected by settling of aerosols and soiling. The soiling represents general dirtiness of the environment that necessitates more frequent cleaning. Examples include more frequent cleaning of clothes, washing of automobiles, and repainting of structures. Ambient PM pollution is also responsible for the damage of vegetation, including both commercial crops and vegetation in natural areas. Particulates that settle on plants can disrupt normal functional processes within vegetation and cause other undesirable effects (Degobert, 1992; Godish, 2004).

In addition to causing damage to property and ecosystems as a result of deposition and soiling, PM can cause negative environmental effects as a result of acidification. After its initial release from emission source (i.e., diesel engines), primary aerosols undergo a series of chemical reactions in the atmosphere resulting in formation of secondary aerosols. Secondary aerosols from NO_x and SO₂ emissions significantly contribute to the acidification, nitrification, and eutrophication of water bodies, such as

lakes and streams, and harm plants and animals. Secondary acidic pollutants (e.g., sulfurous, sulfuric, and nitric acids) precipitate with rainfall, snow, or fog. Acidification of water bodies can result in reduction of fish species, changes in plankton, and alteration of water composition, such as increased levels of dissolved heavy-metals. Acidic pollutants can also get deposited on vegetation, soils, and manmade structures. This can result in crop and forestry losses, lead to biodiversity impairment of ecosystems, and cause deterioration of property (Degobert, 1992; F. J. Kelly & Fussell, 2012; Likens, Driscoll, & Buso, 1996; Majewski & Khair, 2006).

Diesel PM also contributes to photochemical smog, especially in the urban centers (Majewski & Khair, 2006). Smog gets created in the atmosphere in the presence of sunlight as a result of complex reactions involving fine particulates (e.g., HC portion of diesel SOF particulates and secondary nitrate particulates), nitrogen oxides, sulfur oxides, hydrocarbons, and carbon monoxide. Secondary chemical reactions involving photochemical smog constituents can also facilitate formation of ground-level ozone, an oxidizing compound with extreme chemical ability. Some studies suggest that the rate of ozone production increases with increasing light intensity on calm, clear, and dry days (S. B. Lee, Bae, Y. M. Lee, Moon, & Choi, 2010; Mauzerall, Sultan, Kima, & Bradford, 2005; Shin, Cho, Han, Kim, & Kim, 2012; X. Wang, Manning, Feng, & Zhu, 2007).

High levels of smog are characterized by an irritating brownish-grey haze that significantly affects visibility, environment, and causes adverse health impacts, such as eye irritation, coughing, choking, difficulty breathing, and others. The oxidants formed in the atmosphere due to photo chemically induced reactions are also damaging to

vegetation and inhibit its growth. This can affect environment on local, regional, and global scales. Local effects can potentially reduce vegetation cover, diversity, and ecosystem stability resulting from local photochemical smog events. Regional impacts occur via exposure to photochemical oxidants as they get transported over the long-range distances from their release source (EPA, 2010a; Godish, 2004; Harrison, 1996).

According to recent studies, black carbon (e.g., soot portion of diesel PM) has the potential to cause global warming by absorbing sunlight and heating the atmosphere and thus directly contributing to positive climate forcing. Black carbon also has an indirect radiative forcing through its effects on clouds. It can also get deposited on snow and ice, absorb direct sunlight, cause reduction of surface reflectiveness, and result in ambient temperature increase. Contribution of black carbon to global warming may be more than of any other greenhouse gases after CO₂ (Davidson et al., 2005; Houghton et al., 2001; Jacobson, 2002; Lohmann & Feichter, 2004; McConnell et al., 2007; Ramanathan & Carmichael, 2008; University of California San Diego, 2008). However, experts agree that diesel engines produce significantly less amounts of CO₂ and therefore offer a significant greenhouse reduction benefit compared to gasoline engines. According to their estimates, diesel greenhouse emissions are about 13% lower than those from gasoline vehicles (General Motors Corporation, 2001; Walsh, 1998).

Reduction of harmful effects of diesel PM on health and environment is possible only through a combined effort of legislators, engine manufacturers, and scientists. Stringent diesel emission standards play an important role and represent a driving force of emission reduction efforts. These are described in the following section.

Diesel PM Emission Regulations and Control

The widespread use of diesel equipment, increased emissions levels, and growing awareness of a wide range of health and environmental effects of diesel exhaust has necessitated a development and an ongoing strengthening of emission regulations. As a result of these efforts, starting from the 1950s, a variety of the air quality policies have been adopted on federal and state level resulting in significant advancements in emission control and air quality improvement.

PM Emission Regulations for Diesel Engines

Major development of federal and state emission regulations were authorized under the Clean Air Act of 1970 (EPA, 2011c; Sullivan, 2005). First federal air quality standards for PM were established by the EPA in 1971 to control total suspended particles. In the 1987, additional standards were adopted for PM₁₀, an inhalable portion of particulates with less than 10 microns in diameter. Consequently in view of numerous health studies on harmful effects of fine PM, in 1997 the new standards were adopted for PM_{2.5}, a portion of fine particles with aerodynamic diameter less than 2.5 microns (CARB, 2010a). As these standards get periodically reviewed and updated, the non-attainment areas are required to prepare the State Implementation Plans in order to establish strategies to meet new standards by the mandatory deadlines and avoid non-compliance sanctions (EPA, 2012d; Sullivan, 2005).

In addition to the EPA's national air quality standards that regulate ambient pollution levels in the atmosphere, regulative efforts also focus on engine emission standards. Diesel emission standards regulate emissions from the new engines and/or

vehicles and from in-use diesel engines. Additionally, occupational health regulations specify maximum allowable workplace exposures. Currently, occupational exposure limit for particulate fraction of diesel exhaust has been set by the Occupational Safety and Health Administration (OSHA) and Mine Safety and Health Administration for underground mining (OSHA, n.d.; OTAQ, 2009a, 2011c, 2012d).

Emission standards for new engines. On federal level these standards are adopted by the EPA. On state level, the State of California is authorized to implement its own emission regulations; while other states can decide to either adopt federal or California emission standards. Emission standards for new engines define the maximum amount of exhaust gas pollutants allowed to be emitted from a specific engine. Additionally, new emission standards need to identify the required emission warranty to ensure equipment meets emission standards over certain period of time or mileage. Engine manufacturer has to comply with established standards and ensure that each new engine is emission certified (DieselNet, 2010a; Faiz et al., 1996).

Compliance with emission standards usually is determined based on emissions as measured on dynamometer over standardized engine or vehicle test cycles that simulate certain operating conditions in a repeatable manner. Emissions can be recorded over steady-state cycles with engine operating during a series of modes under stabilized load and speed. Transient cycles are also used to measure new engine emissions and involves a particular driving pattern with segments of accelerations, decelerations, and changes of speed and load (DieselNet, 2010a; Majewski & Khair, 2006). Light- and medium-duty vehicles are usually certified on chassis dynamometer. Their emissions are expressed in

grams per unit of traveled distance, such as grams per mile. Emissions from heavy-duty engines have to be tested on engine dynamometer. These are stated in either grams per kilowatt-hour or grams per brake-horsepower-hour and represent emissions relative to the amount of mechanical work performed by the engine (DieselNet, 2006; Faiz et al., 1996).

Emission standards for new engines are divided into two major groups: on-road and non-road standards. Based on their gross weight classification, vehicles can be subject of either light- or heavy-duty legislation. Emissions from cars and light-duty on-road trucks have been controlled by federal Tier 1 standards, phased-in between 1994 and 1997, and Tier 2 standards, implemented between 2004 and 2009. Tier 1 PM diesel emission standards for 10 years of engine useful life controlled emissions from vehicles with less than 5,750 lbs and more than 5,750 lbs loaded weight at the levels of 0.1 and 0.12 g/mi, respectively (DieselNet, 2006, 2007a; EPA, 2010b). In order to comply with these regulations, engine manufacturers had to significantly enhance engine design and improve electronic fuel injection controls. Many of the emission control technologies from on-road engines were also applied to off-road engines (Kittelsson et al., 1999; Kliesch & Langer, 2003). Subsequent Tier 2 limits were more stringent compared to Tier 1 and imposed same emission limits to all weight categories, including heavier vehicles up to 8500 lbs. Vehicle manufacturers could certify their vehicles according to any of the available certification levels (bins) with PM limits ranging from zero to 0.02 g/mi. These standards also regulated NO_x, non-methane HC, CO, and formaldehyde (DieselNet, 2006, 2007a; EPA, 2011e; Kliesch & Langer, 2003).

The state of California emission standards for cars and light duty trucks are more stringent compared to federal rules and include: Tier 1/Low Emission Vehicle (LEV) phased-in through model years 2003, LEV II implemented through 2004-2010, and LEV III to be phased-in for model years 2014-2022. According to these standards, light-duty vehicles and trucks less than 8,500 lbs GVW had to comply with a full useful life PM standard of 0.01 g/mi. Certification to such low emission levels could be achieved only when using advanced exhaust gas after-treatment devices. This forced manufacturers to install various emission control technologies (e.g., DPFs and NO_x oxidation catalysts) on new vehicles (DieselNet, 2010b; Faiz et al., 1996).

The most significant emission control efforts for heavy-duty on road engines were 2007 emission standards requiring all diesel engines 2007 and later to comply with very stringent regulations by 2010 on a phased-in percent basis. According to the phase-in schedule, diesel manufacturers had to certify 50% vehicles from 2007 to 2009 and 100% vehicles in 2010. These regulations were first adopted by the EPA. The state of California signed similar regulations in the following year. These rules mandated more than 90% reduction in PM levels and 50% reduction of NO_x and HC over the previous 2004 emission standards. Figure 5 shows comparison of 2007 and older PM/NO_x emission standards for heavy-duty engines. Similar to light-duty requirements, these standards necessitated wide development and implementation of the DPFs and other exhaust after-treatment technologies (DieselNet, 2007b; OTAQ, 2009b).

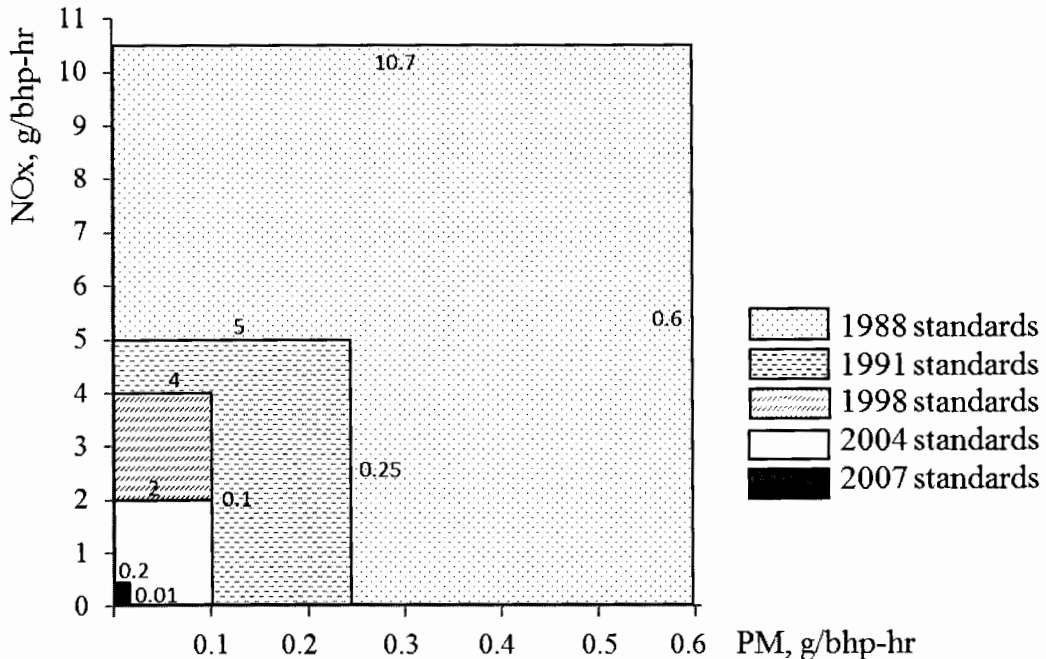


Figure 5. Emissions requirements for new on-road heavy duty diesel vehicles (created using data from DieselNet, 2007b).

Additionally, these regulations required a 97% reduction of sulfur levels in diesel fuel to allow the employment of pollution-control devices. The sulfur content had to be reduced from 500 to 15 ppm. The EPA estimated that implementation of 2007 standards will lower PM and NOx levels by 110,000 and 2.6 million tons each year. As a result, as many as 8,300 premature deaths, 5,500 cases of chronic bronchitis, 17,600 cases of acute bronchitis in children, 360,000 asthma attacks, and 386,000 cases of respiratory symptoms in asthmatic children will be prevented annually. Moreover, this regulation will prevent 1.5 million lost work days, 7,100 hospital visits and 2,400 emergency room visits for asthma (OTAQ, 2000, 2009b).

Engine emissions from non-road engines have also been reduced with adoption of Tier 4 standards that are to be phased-in from 2008 to 2015 and apply on federal as well as state level. Earlier Tier 1, 2, and 3 standards phased-in from 1996-2000, 2001-2006, and 2006-2008, respectively, could mostly be achieved through advanced engine design. In some cases oxidation catalysts are used to provide additional exhaust gas after-treatment. On the other hand, Tier 4 standards require significantly higher emission reductions of PM and NO_x levels by about 90%. Therefore, similarly to 2007 year and later heavy-duty on-road engines, emission reduction to non-road Tier 4 levels could only be accomplished using exhaust gas after-treatment technologies. To enable use of exhaust control devices, the sulfur level had to be also limited from unregulated levels of 3,000 ppm to 500 ppm and 7-15 ppm effective July 2007 and June 2010, respectively. According to the EPA estimates, Tier 4 compliant engines could reduce annual emissions by 129,000 tons of PM and 738,000 tons of NO_x. By 2030, this could annually prevent 12,000 premature deaths (DieselNet, 2004b; EPA, 2011b; OTAQ, 2011b).

Emission control for in-use engines. Implementation of voluntary or obligatory emission control retrofit programs helps reduce diesel emissions from older in-use vehicles to acceptable levels. Participation in voluntary retrofit programs usually involves various incentives, such as the State Implementation Plan emission credits, tax incentives, and grants. These programs involve installation of approved retrofit devices, such as the DPFs and catalytic converters on existing vehicles (CARB, 2010b; OTAQ, 2011a). The DPFs can trap from 60 to 90% of PM in the vehicle exhaust and must be used with ultra-low sulfur diesel fuel. Diesel oxidation catalysts use chemical oxidative

reactions to reduce exhaust emission levels. On average, utilization of oxidation catalysts results in 20, 50, and 40% reduction in PM, HC, and CO levels, respectively (EPA, 2012c; OTAQ, 2003).

Examples of retrofit programs include the Voluntary Diesel Retrofit Program (VDRP) established by the EPA and the state of California diesel retrofit program. Being a voluntary program, the VDRP provides the State Implementation Plan emission credits to participating states and approves the following heavy-duty diesel retrofits: the DPFs, catalytic converters, engine upgrades/replacements, and clean fuels and/or fuel additives. The state of California diesel retrofit voluntary program is administered by the CARB as part of its risk reduction plan for diesel PM. Therefore, it is mostly focused on retrofitting diesel vehicles with the DPFs. Major goal of this program is retrofitting of about 90% of all existing mobile and stationary in-use vehicles with the DPFs (CARB, 2000b; DieselNet, 2000; Majewski & Khair, 2006).

Emissions from in-use vehicles are also controlled by other means, such as through inspection and maintenance (I/M) programs. These programs are oftentimes maintained at the state level. Diesel I/M programs include smoke opacity measurement according to a specified test method protocol. Compliance with emission limits is enforced through vehicle registration. Additionally, I/M programs may assist in enforcement of on-board diagnostics (OBD) regulations (DieselNet, 2010c; EPA, 2011d; Faiz et al., 1996). The OBD system includes sensors, diagnostic software, and computer. Its operational principle is based on monitoring vehicle performance characteristics, detecting operational problems, illuminating a malfunction warning light to alert the

driver, and storing malfunction codes in vehicle's computer for later retrieval (DieselNet, 2007c; Equipment and Tool Institute, 2012).

The OBD requirements were first adopted by the state of California and later by the EPA. Currently, all diesel fueled passenger cars and trucks with model year 1997 and newer in the state of California are required to meet OBD requirements. For heavy-duty engines heavier than 14,000 lb, basic engine manufacturer diagnostic system is required for engines with model year 2007 and later. By the year 2013 these systems will be fully phased-out by advanced OBD system requirements on all heavy-duty engines in the state of California (DieselNet, 2009a, 2010c). Federal OBD regulations are mostly similar to those from the state of California with only few differences, such as more stringent limits adopted in the state in the year 2016 for all engines equipped with the DPFs and other exhaust after-treatment systems (DieselNet, 2009a). The OBD rules are especially important since they impose additional requirements on exhaust pollution sensors and specifically in regard to monitoring of the DPFs. Therefore, the OBD regulations create an additional need in real-time sensors that could continuously monitor PM levels and DPF filtration efficiencies on DPF equipped vehicles.

Emission Control Technologies - Diesel Particulate Filters

As engine emission standards are becoming more and more stringent over the years, the effective reduction of exhaust emissions to regulated limits becomes possible only when using new emission control technologies, such as the DPFs. The DPFs are exhaust after-treatment devices that are installed in the vehicle exhaust system and collect PM in the exhaust gas (National Clean Diesel Campaign, 2010a, 2010b; OTAQ, 2003).

The DPFs are enclosed in a steel housing and can be set up as a standalone unit or placed inside a muffler (Majewski & Khair, 2006). The DPFs are usually made of porous ceramic or metal material and physically capture diesel PM by means of particle deposition (MECA, 2007; National Clean Diesel Campaign, 2010a).

As collected particles get accumulated inside of the DPF, they create a pressure drop and may clog the filter. To ensure effective filtration efficiency and proper operation of the DPF itself and the engine, PM must be reduced to ash during the DPF regeneration. Noncombustible materials, such as ash, remain after filter regeneration and must be subsequently removed during periodic maintenance and filter cleaning. Filter maintenance can include removal of the DPF and washing it in specially designated cleaning station or reversal of the DPF flow direction. This procedure should be repeated every six or 12 months depending on soot loading and backpressure monitoring data (Barone, Storey, & Domingo, 2010; Kliesch & Langer, 2003; National Clean Diesel Campaign, 2010a; Sappok, Parks, & Prikhodko, 2010).

Regeneration can be conducted periodically or continuously depending on filter design and temperatures. Continuous “passive” regeneration is performed when filter operates at high exhaust gas temperatures that are sufficient to initiate an ongoing combustion of collected PM in the DPF. Passive regeneration occurs during the normal engine operation and does not require any additional heat source. Instead, passive systems oftentimes are catalyzed. In this case, addition of catalyst lowers the oxidation temperature that needs to be maintained during regeneration. On the other hand, “active” filters require periodical increase of filter temperature using an external source of heat,

such as an electric heating element or fuel injection into the filter followed by its subsequent ignition (CARB, 2012a; MECA, 2007; National Clean Diesel Campaign, 2010a; Reul-Chen, Ross, Steele, & Winer, 2005).

Common filter configurations. The most common DPF configuration type is a wall-flow monolith with honeycomb structure comprised of numerous adjacent channels placed parallel to the filter's axis and alternatively plugged at each end of the filter. Exhaust gas gets filtered as it passes through the porous walls of adjacent channels acting as a filter media. Wall-flow monolith filters are usually made of the following ceramic materials: cordierite or silicon carbide. Cordierite represents a synthetic ceramic material with good mechanical strength, resistance to thermal cycling, and high temperature resistance around 1,200 °C. Silicon carbide can resist up to 600 °C higher temperatures compared to cordierite, however it is also more expensive and has higher thermal expansion properties (Majewski, 2011; MECA, 2007, 2009; Merkel et al., 2003; Wu, Kuznetsov, & Jasper, 2011).

Particle filtration efficiencies of wall-flow monoliths are over 90%, which is highest compared to other filter types. This could be explained by combination of involved filtration mechanisms, including depth and surface (cake) filtration. Depth filtration is characterized by inertial and diffusional deposition of dust particles within the filter walls. As particles get accumulated on the surface of inner channels they also act as additional filtration media. Wall-flow filters can be used as catalyzed filters in passive systems. Additionally, they are quite compact in size. However, these filters are susceptible to thermal damage as a result of clogging due to relatively high increase in

pressure drop with increasing soot load (Majewski, 2011; MECA, 2007, 2009; Sappok et al., 2010; Wu et al., 2011).

Other filter materials used in DPFs are ceramic fibers, ceramic foams, metal fiber filters, sintered metals, and others. Ceramic fibers can have various designs, such as continuous fibers layered around a metal tube or thin fiber sheets combined to make filter cartridges of up to 90% filtration efficiency. Regeneration of fiber filters involves introduction of fuel additives, fuel burners, and electric heaters. This type of filters cannot be catalyzed. This represents a serious disadvantage since higher temperatures are required during regeneration of fiber filters compared to oxidized passive systems. Ceramic foam filters are comprised of a rigid ceramic structure of thermally treated polyurethane foam filled with ceramic paste. These filters can be catalyzed; however, they can achieve relatively low filtration efficiency of 60 to 70%. Somewhat lower efficiency between 50 and 70% is demonstrated by metal fiber filters. The filtration efficiency of this filter type varies depending on filter loading and gets decreased after reaching its peak due to re-entrainment of previously collected particles (Klouda, Fletcher, Gillen, & Verkouteren, 2011; K. M. Lee, Y. S. Lee, & Jo, 2006; Majewski & Khair, 2006; MECA, 2007, 2009).

Filter performance. Typical efficiency of the DPFs that have been verified by the EPA and the CARB ranges from 85 to 90% or more (National Clean Diesel Campaign, 2010a; MECA, 2009). Determination of the DPF performance is based on its filtration efficiency which is calculated as mass ratio of the collected PM to the PM entering the filter (Malik et al., 2011). The DPF efficiency depends on filtration mechanisms

involved in filtration process. The highest filtration rate occurs when a combination of filtration mechanisms are present, such as in wall-flow filters discussed earlier. Surface (cake) filtration mechanisms alone result in higher PM filtration compared to depth filtration. However, these mechanisms are also characterized by higher pressure drop. Surface filtration usually does not occur alone in the DPFs, rather it takes place at higher soot loads in combination with depth filtration after PM holding capacity of a deep-bed gets saturated to its limit. The depth filtration, on the other hand, is accompanied by somewhat lower pressure drop and PM collection efficiencies. Additionally, the PM blow-off can occur at high exhaust gas rates and or rapid flow accelerations. This occurrence takes place when PM collected by depth filtration gets separated from the deep-bed filter and re-entrained by the exhaust gas flow (Gallant et al., 2005; Karin & Hanamura, 2010; Kim, J. Wang, Shin, Scheckman, & Pui, 2009; Soldati, Campolo, & Sbrizzai, 2010).

The DPFs collect the solid fraction of diesel PM, such as ash and inorganic carbon fraction, in the most effective manner. The organic liquid portion of PM can be collected with varying degree of efficiency depending on a specific filter and its operational conditions. The sulfate particulate levels after the DPF either remain the same or increase. The latter occurs in the process of catalytic oxidation of sulfur dioxide emissions in catalytic filters. Moreover, under high DPF temperatures a significant portion of liquid PM, including organic fraction as well as sulfates, exists in the gas phase and cannot be trapped by the filter. Additional ultrafine particles can also be formed downstream of the DPF and get released into the atmosphere with exhaust gases. These

particles mostly exist in liquid nuclei nanoparticle mode due to little soot surface area of exhaust PM available for their further adsorption and condensation. PM sampling and measurement conditions can also affect diesel PM formation and transition (Barrios, Domínguez-Sáez, Rubio, & Pujadas, 2011; Burtscher, 2005; Canagaratna et al., 2010; Heikkilä et al., 2009; Kittelson & Watts, 2002; Kittelson, Watts, & Johnson, 2006). Most common PM measurement technologies and their characteristics are discussed in the following section.

Diesel PM Measurement Technologies

Based on different measurement methods, diesel PM can be characterized by various properties, such as particle mass, size distribution, particle count, surface area, and so on. Conventionally, PM emission standards have been mass based. Therefore, measurement of diesel particulate mass emissions for regulatory and compliance purposes is performed using gravimetric method. Over the past decades, increasingly stringent diesel emission standards and subsequent advances in emission control technologies produced new requirements and challenges in PM measurement at low emission levels (Burtscher, 2001; Khalek, 2008; Swanson et al., 2010). This created a need in development of new advanced real-time emission measurement techniques and technologies. The major emission measurement technologies and their characteristics are discussed as follows.

Gravimetric Mass Emissions Measurement

Gravimetric method is based upon collection and measurement of deposited PM mass. Sampling filters are used to collect PM emitted from diesel engines. The filters

are pre-conditioned and measured under controlled temperature and relative humidity before and after each test. For the duration of the test, a defined volume of vehicle exhaust gas is continuously drawn through filters. Ultimately, the initial filter weight before the test is compared to the final filter weight after the test. Material accumulated on filters is subsequently evaluated using analytical procedures (40 C.F.R. § 1065, 2012; CARB, 2008; Chow, 1995; National Exposure Research Laboratory, 1998). PM mass concentration is calculated as a function of collected PM mass in a known total volume of sampled air (40 C.F.R. § 1065, 2012; Chow, 1995; Price & Lacey, 2003).

Even though this PM measurement method is widely used, it has some major shortcomings. First of all, by its definition, gravimetric technique calculates the total emission mass and therefore, it cannot be used to measure emissions during vehicle transient operating conditions. They are unable to provide real-time data crucial for understanding the time-dependent performance of emission control devices, such as the DPFs, and the vehicle engines. As a result, other methods must be used to assess real-time (second-by-second) or near-real-time (minutes or less) variations of vehicle exhaust emissions. Additionally, gravimetric results require subsequent labor intensive and time consuming analysis of collected samples. Oftentimes, sample analysis is accompanied by long time delays (Maricq, Chase, & Xu, 2001; Montajir, Kusaka, Bamba, & Adachi, 2005).

Other disadvantages of gravimetric method are due to measurement challenges at low PM emission levels as seen in diesel engines compliant with 2007 emission standards (CARB, 2008; Khalek, 2006, 2008). For example, when dealing with low PM emissions,

the gravimetric method is at its detection limits and requires greater accuracy and sensitivity (Maricq & Maldonado, 2010; Nakhawa, 2012; Swanson et al., 2010). Studies conducted by Khalek (2007) identified laboratory challenges of PM emission measurements at or below 2007 standard levels. Specifically, emission levels of 10 percent of the 2007 standard measured by Khalek (2007) from DPF-equipped vehicle were nearly the same magnitude as levels measured from blank filters.

During sampling and analysis, the deposited and emitted particles may also undergo transformation resulting in changes of their properties. For example, particle mass overestimation can result from collection of particle “artifacts” on filter due to adsorption of semi-volatile species either on gravimetric filter itself or on collected on filter particulates (Kirchstetter, Corrigan, & Novakov, 2001; Park, Cao, Kittelson, & McMurry, 2003; Swanson et al., 2010). Furthermore, the amount of PM deposited on filter is affected by changes in sampling efficiency of semi-volatile particles, filter face velocity, sampling, and dilution factors (Khalek, 2005, 2008; Kirchstetter et al., 2001; Vecchi, Valli, Fermob, D’Alessandro, Piazzalunga, & Bernardoni, 2009). For example, long sampling time may lead to desorption of volatile gas component of the composite particles and result in negative filter “artifacts.” However, long sampling time is recommended to minimize positive “artifacts.” Maintaining high filter face velocity of up to 100 cm/sec will also reduce positive “artifacts” related to gas phase adsorption on the filter (Khalek, 2005, 2008).

In effort to improve PM mass emission quantification for low PM emitting engines, the EPA revised requirements for gravimetric PM measurement procedures,

such as temperature, dilution tunnel, filter media, filter handling, and weighing chamber specifications. However, recent diesel emission studies performed using new protocol still report variability in PM measurement results and suggest that additional effort is required to understand and minimize these phenomena (Giechaskiel, Dilara, & Andersson, 2008; Khalek, 2008; Liu et al., 2009; Swanson et al., 2010).

In-Situ Measurement Techniques

The modern trend in PM measurement is towards in-situ real-time techniques that are able to determine instantaneous variations in PM emissions as opposed to total filter PM mass. In-situ methods of PM emissions measurement usually involve continuous real-time or near real-time PM analysis that is performed in the aerosol phase as it moves through a designated measuring area of in-situ instrument, thus minimizing changes to the particle properties. This technique possesses a unique advantage over conventional gravimetric procedure since it enables instantaneous measurement of PM emissions and their properties, such as particle sizes and size distributions. This is especially useful for transient testing, instantaneous emission monitoring, real-time diagnostic of DPF-equipped vehicles and performance of their emission control devices (Burtscher & Majewski, 2012; Maricq & Maldonado, 2010). Certain in-situ devices can be coupled with gravimetric filters for subsequent gravimetric analysis. Example of this type of equipment is portable emission measurement systems (PEMS) that include various on-board emission analyzers to measure emissions in the field (Sensors, 2009).

Measurement of exhaust PM emissions oftentimes involve exhaust gas preconditioning prior to sampling. Sample conditioning is usually performed using a

specific dilution system or a constant volume sampling (CVS) tunnel. However, several systems are capable of measuring undiluted hot raw exhaust directly from the exhaust pipe. Generally, the final PM measurement results produced by in-situ instruments are usually comprised of measurements of both solid particles (e.g., carbonaceous matter, ash, and metals) as well as liquid particles (e.g., volatile organic compounds, sulfuric acid and/or water condensates). Therefore, sampling conditions significantly affect sampling and measurement results (Burtscher, 2001; Maricq, Chase, Podsiadlik, & Vogt, 1999; Maricq & Maldonado, 2010). Removal of volatile nucleation particles and measurement of remaining carbonaceous fraction has been suggested by the Europe's Particle Measurement Programme (PMP) protocol in attempt to minimize variability in PM measurements (Johnson et al., 2009; Zervas et al., 2005).

Contrary to gravimetric technique based on direct measurement of PM mass, when using in-situ methods the results are usually calculated from indirect measurement of various PM properties, such as physical, optical, aerodynamic, and electrical mobility. Because different in-situ instruments utilize various measurement approaches, a correlation between these instruments oftentimes involves significant uncertainty (Kittelson, Watts, & Arnold, 1998; McMurry, 2000; Vouitsis, Ntziachristos, & Samaras, 2005). Several major types of in-situ devices most commonly used for real-time or near real-time diesel PM measurement are presented below.

Number concentration measurement devices. The condensation particle counters (CPC) or the condensation nucleus counters (CNC) are widely used to measure the total aerosol number concentration. These instruments determine number concentrations of

diesel particles larger than a minimum detectable size. Detection limits of ultra-fine particle CPCs can be as low as 3 nm or less. After particles undergo condensation and grow in size they are detected using optical detection methods (Franklin, Bika, Watts, & Kittelson, 2010; McMurry, 2000; Mordas et al., 2008; Swanson et al., 2010).

The CPCs are sensitive enough for detecting low PM concentrations. However, the instruments' detection efficiency is influenced by particle chemical composition (Mordas et al., 2008). The CPCs are mostly used in laboratory settings due to dependence of their readings on temperature. The range of operating temperature is related to strict temperature tolerance requirements inside of CPC device in order to maintain super-saturation conditions. The CPCs readings are also affected by device positioning and vibration, thus creating additional difficulties for field measurement (Burtscher, 2001; McMurry, 2000). The CPCs are often used along with other devices, for example, with diesel particle sizing instruments, such as electrical mobility classifiers.

Diesel particle sizing instruments. PM sizing techniques can be divided into three general categories: methods using aerodynamic detection principles, electrical mobility analyzers, and diffusion charging devices (Burtscher & Majewski, 2012; McMurry, 2000). Particle sizing instruments identify the dimensional distributions of the diesel PM. Generally, the measurement results are reported in the form of particle mass or particle number distributions (Maricq & Maldonado, 2010; University of Essex, 2009).

Instruments that are based on aerodynamic principle belong to a class of cascade impactors. These devices categorize aerosols according to their size ranges by means of inertial impaction. The particles are separated based on difference in their momentum as

gas sample passes through a number of orifices of varying diameter (International Union of Pure and Applied Chemistry; 2012). Cascade impactors may be used together with other PM measurement devices in order to expand their operational particle size range (Lodge & Chan, 1988).

Electronic low pressure impactor (ELPI) is an example of cascade impactor used in vehicle emission testing that charges particles prior to entering the impaction stage. ELPI generally measures particles in 30 nm to 10 μm size range (Burtscher, 2001; Kittelson et al., 1998). Some literature reports ELPIs with extended particle size ranges down to 16.7 nm (Yli-Ojanperä, Kannosto, Marjamäki, & Keskinen, 2010) and 6 nm (Burtscher & Majewski, 2012). Given the particle charge and density, the ELPI can determine the PM mass concentration in the vehicle exhaust (University of Essex, 2009). A comparison of particle mass obtained from ELPI to gravimetric mass measurements shows that EPLI overestimates results of up to two times the gravimetric values (Khalek, 2000; Tsukamoto, Goto, & Odaka, 2000). Additionally, some researchers report difficulties in emission measurements from low-particle emitting vehicles (Zervas et al., 2005). Measurement of ultra-low PM concentrations using ELPI represents a challenging task since low pressure can cause evaporation of volatile particles (Kittelson et al., 1998; Burtscher, 2001). Additionally, ELPI is unable to produce high resolution second-by-second measurements. Instead it generates near real-time data with 2 to 15 seconds time resolution (Burtscher & Majewski, 2012; Kittelson et al., 1998).

Electrical mobility analyzers represent a second type of PM sizing instruments. They classify charged particles according to their electrical mobility, the ability to move

through sampling gas volume in response to applied electric field. Mobility diameter of particulates is then defined based on their mobility measurement. Particles of the same size possess the same electrical mobility and, therefore, get sorted according to their size distribution. Electrical mobility depends on gas properties, particle charge, and the geometric particle size. However, electrical mobility of particles is independent of their density and other properties (D. R. Chen, Li, & M. D. Cheng, 2007; Flagan, 2008; McMurry, 2000). Oftentimes, the analyzer is combined with a particle detector, such as condensation particle counter or aerosol spectrometer, in order to measure PM size distributions (Flagan, 2008). Electrical mobility analyzers correspond to a broad class of instruments that includes differential mobility particle sizers (DMPS), scanning mobility particle sizers (SMPS), engine exhaust particle sizers (EEPS), fast particulate spectrometers, nano-differential mobility analyzers, and others (Burtscher & Majewski, 2012).

DMPS utilizes a bipolar charging to charge PM particles as they enter the mobility analyzer. Electrical size mobility intervals are changed based on voltage that is applied in varying stages. Only particles of certain mobility can pass through each discrete voltage step (Durbin, Norbeck, Cocker, & Younglove, 2004; Flagan, 2008; Kittelson et al., 1998). Major disadvantage of DMPS is long characterization time of each particle sizing run of about 10 to 60 or more minutes which makes it unsuitable for transient real-time measurements. SMPS represent a modified variation of the latter device. Separation of PM by size is performed in the electrostatic classifier. In SMPS, the collection rod voltage is continuously varied resulting in reduced delay in the

measurement and improved determination speed of an entire particle size distribution. Using this instrument it is possible to significantly reduce the measurement time without considerably sacrificing size resolution (University of Essex, 2009; S. C. Wang & Flagan, 1990). As a result, the measurement time is reduced to 20-60 seconds with minor accuracy reduction (Burtscher & Majewski, 2012; S. C. Wang & Flagan, 1990). Kittelson et al. (1998) found that SMPS data obtained in scanning mode over two to four minute scan have much less distortion compared to one minute scans. When used in single size mode in transient testing, the instrument is capable to produce results with resolution of few seconds. However, this arrangement is rather costly and requires more time since several tests are needed to obtain one size distribution.

In order to enable real-time transient testing using electrical mobility analyzers, several analyzers models have been developed, such as EEPS and fast particulate spectrometers. These devices simultaneously measure particle concentrations of several size ranges. Each particle size is determined using separate electrometers. As a result, EEPS and fast particle spectrometers are able to produce particulate size distribution in 32 and 43 size channels, respectively, with 0.1 second time resolution (Burtscher & Majewski, 2012; J. Wang et al., 2006; Zervas & Dorlhène, 2006). Symonds, Reavell, Olfert, Campbell, and Swift (2007) found that data from fast particle spectrometer DMS500 obtained during diesel emission testing over a variety of engine conditions were good agreement with gravimetric method. However, DMS500 mass concentrations were below the gravimetric values.

Diesel emissions measurement studies conducted by Nakhawa (2012) revealed that EEPS can produce reliable real-time results. However, this testing demonstrated dependence of sensitivity for each size level on the minimum concentration and required an increase in minimum concentration as the particle size decreased. EEPS was shown to produce useful data during vehicle transient (Barrios et al., 2011; J. Wang et al., 2006) and background tunnel testing (Yao, Lau, Fang, & Chan, 2005). Transient test results from EEPS correlated well with ELPI and CPC upstream of the DPF (Zervas & Dorlhène, 2006). However, when measuring low exhaust emissions downstream of the DPF, the EEPS data were below their detection limits. While Barrios et al. (2011) defined this instrument to be suitable for transient nanometric emission testing, especially in laboratory conditions. EEPS was found to be sensitive to vibrations, high temperatures above 42 °C (107 °F), and high particle concentrations complicating the field testing requirements.

Third type of PM sizing instruments corresponds to diffusion batteries. They categorize particles based on their diffusion properties and determine their mobility and size from measured diffusion coefficients. Diffusion batteries prove especially beneficial in size distribution measurement of particles smaller than 300 nm (Burtscher, 2001; Burtscher & Majewski, 2012). Some disadvantages of this measurement method include slow sampling time and steady-state testing conditions requirement. When several diffusion batteries are arranged parallel to each other during testing, it is possible to obtain a near real-time instrument response. However, this design is more expensive and much less suitable for small size applications. Another variation of diffusion method is

electrical diffusion battery. This device determines size distribution of 5 to 300 nm charged particles by measuring the resulting electrical current. Electrical diffusion batteries allow for near-real time PM measurement while minimizing evaporation of volatile material. However, these improved scan times in two latter variations of diffusion battery designs are only possible at the cost of reduced particle size resolution (Burtcher, Scherrer, & Siegmann, 2000; Fierz, Scherrer, & Burtcher, 2002).

Dekati Mass Monitor (DMM) represents a real-time particle mass measurement device that combines several particle detection principles, including mobility analysis and aerodynamic sizing. Mobility and aerodynamic size information is obtained from the charger and impactor, respectively. First, the particles are charged in a diffusion charger region of the instrument's particle mobility analyzer. The smallest particles are then deflected to the charger mobility electrode using a static electrical field and measured using electrometer. Afterwards, the particles are being passed from the particle mobility size analyzer to a multi-stage inertial impactor for subsequent particle size classification. An electrical current is generated by particles at each of six impactor collection plates and measured by a multi-channel electrometer. Ultimately, the effective density is calculated based on mobility and aerodynamic size data and used together with measured current values to determine the particle mass and number concentrations (Dekati, 2010; Mamakos et al., 2006).

Evaluative study of PM measurement instruments conducted by Khalek (2005, 2008) revealed that DMM data correlated well with gravimetric data and were highly sensitive to low PM concentrations. Specifically, DMM, EEPs, and SMPS instruments

compared with CVS filter-based PM system yielded a linear regression with a correlation coefficient of better than .95 when tested with DPF bypass exhaust configuration. DMM measurements also showed more repeatability and less sensitivity to dilution parameters compared to those obtained using gravimetric method. These results are consistent with data from other studies. Studies conducted by Mamakos et al. (2006) employing DMM in steady state and transient tests found high correlation of DMM and filter-based method with a .95 correlation coefficient. Researchers found that DMM overestimated mass measurement values were generally 38% higher than gravimetric values.

Lehmann et al. (2004) studied a prototype DMM model in five different transient cycles and identified its ability to follow the transient cycle and detect individual PM peaks with a good second-by-second time response and repeatability. Measurement values from DMM and gravimetric methods were in a good agreement at transient test cycles with correlation coefficient of more than .95 for total PM mass per unit volume. According to Lehmann et al. (2004), DMM measurements exceeded gravimetric values with about 20% overestimation. When measuring low emission levels, Lehmann et al. (2004) found that filter-based method produced significantly higher mass values compared to the DMM possibly due to filter gas-phase hydrocarbon adsorption. However despite these differences, the DMM was deemed to produce more accurate results than gravimetric method. The DMM also showed good correlation to other mass-based instruments (Lehmann et al., 2004).

Optical methods. Optical methods are oftentimes used in exhaust gas measurement due to its suitability for fast transient vehicle testing (Burtscher, 2001).

Particle photometer DustTrak represents one example of PM mass measurement instruments that use optical detection methods (Maricq & Maldonado, 2010). The DustTrak corresponds to a single channel photometer that is capable to provide real-time aerosol mass readings. Its operating principle is based on light scattering technique. The sample flow gets illuminated by a sheet of light from a laser diode. Particles in the gas stream scatter and focus significant portion of light on a photo detector. The PM mass concentration is then calculated based on the voltage across the photo detector (TSI Incorporated, 2008; TSI Incorporated, 2011).

The response of the DustTrak depends on the size distribution, refractive index, shape, and density of the measured aerosol. Therefore, calibration of the instrument with the measured aerosol has positive effect on the accuracy of results (Stephenson, Lutte, & Spear, 2003; TSI Incorporated, 2011; X. Wang, Chancellor et al., 2009). The DustTrak is factory calibrated with Arizona road dust and requires custom calibration for precise measurements when using different aerosol of interest, such as diesel exhaust. The DustTrak photometer exhibits the highest sensitivity to fine particles while underestimating masses of ultra-fine and large particles. This phenomenon is due to photometric signal being the strongest when registering particle diameters close to the laser wavelength of 655 nm (X. Wang, Chancellor et al., 2009).

Occupational exposure studies conducted by Stephenson et al. (2003) suggested that DustTrak could be utilized to obtain accurate estimation of diesel PM concentrations. Several experiments on diesel PM emissions involving the DustTrak found it to be best suited for fast response mass measurements compared to other measurement devices used

in those studies, such as inertial mass measurement and optical PM detection methods. During the duration of test cycles the DustTrak maintained measurement capabilities without significant deterioration of the signal or need in cleaning showing its suitability for diesel testing environments (Moosmüller et al., 2001a, 2001b). Experiments revealed good correlation of the DustTrak with gravimetric method with correlation coefficients of .87 and .98 for vehicle models newer and older than the year 1998, respectively (Moosmüller et al., 2001a). The studies confirmed the dependence of instrument's calibration on vehicle type and need for individual vehicle-dependent calibration. It has been suggested to use the DustTrak in combination with other instruments to minimize calibration issues. Overall, test results indicated that DustTrak produced data with good sample resolution, signal-to-noise ratio, and low interference from exhaust gas properties. Additional notable benefits of DustTrak included its appealing compact design and inexpensive cost (Moosmüller et al., 2001a, 2001b). The major PM measurement instruments used in diesel emissions testing and their characteristics are summarized in Table 2.

Table 2. Major types of real-time and near real-time diesel PM measurement instruments (Barrios et al., 2011; BMI Aerosol Solutions, 2012; Burtscher, 2001; Burtscher et al., 2000; Burtscher & Majewski, 2012; Dekati, 2010; Fierz et al., 2002; Franklin et al., 2010; Khalek, 2005, 2008; Kittelson et al., 1998; Lehmann et al., 2004; Mamakos et al., 2006; Maricq & Maldonado, 2010; McMurry, 2000; Moio, Niemelä, & Tuomenoja, n.d.; Moosmüller et al., 2001a, 2001b; Mordas et al., 2008; Nakhawa, 2012; Stephenson et al., 2003; Swanson et al., 2010; Symonds et al., 2007; TSI Incorporated, 2008; University of Essex, 2009; J. Wang et al., 2006; S. C. Wang & Flagan, 1990; X. Wang, Chancellor et al., 2009).

Measurement device	Size range, nm	Measured property	Response time	Comments
Condensation particle counter (CPC)	5-3,000	Number	1 sec	Able to detect low PM concentrations. However, detection efficiency is influenced by particle chemical composition. Measurements are affected by device positioning and vibration.
Electrical low pressure impactor (ELPI)	6-10,000	Size distribution based on aerodynamic principle	2-15 sec	Volatile particles may evaporate due to low pressures when measuring low concentrations. Particle mass can be calculated from measured PM properties.
Scanning mobility particle sizer (SMPS)	10-700	Size distribution based on mobility analysis	2-4 min	Can be used in transient testing in single channel size mode with certain accuracy reduction. Concentrations and size must be kept constant during scan. Expensive method and requires several tests to obtain one complete size distribution. Particle mass can be calculated from measured PM properties.

table continues

Table 2 continued.

Measurement device	Size range, nm	Measured property	Response time	Comments
Engine exhaust particle sizer (EEPS)	5.6-560	Size distribution based on mobility analysis	0.1 sec	Suitable for transient low emission testing. Particle mass can be calculated from measured PM properties. Sensitive to vibrations, high temperatures above 42 °C, and high particle concentrations complicating the field testing requirements.
Fast aerosol spectrometer DMS5000	5-2,500	Size distribution based on mobility analysis	0.1 sec	Can be used in transient testing. Particle mass can be calculated from measured PM properties. Data in good agreement but below the gravimetric values.
Diffusion battery (DB)	10-500	Size distribution based on particle diffusion	Several minutes	Slow sampling time and steady-state testing conditions requirement. Possible to obtain a near real-time instrument response using more expensive multiple diffusion batteries design which is less suitable for small size applications. Particle mass can be calculated from measured PM properties.

table continues

Table 2 continued.

Measurement device	Size range, nm	Measured property	Response time	Comments
Electrical Diffusion Battery (EDB), Diffusion size classifier	5-300	Size distribution based on particle diffusion	1 sec	Suitable for transient testing. Can measure ultra-fine PM with accuracy and high time-resolution. Minor dependency of results on the average particle size and the sampling rate is observed. Particle mass can be calculated from measured PM properties.
DMM	5-1,200	Mass based on mobility analysis and impaction with electrical detection	1 sec	Can be used in transient testing. Design is suitable for laboratory conditions. DMM measurements correlate well with gravimetric data and are more repeatable and less sensitive to dilution parameters.
Particle photometer (DustTrak)	100-10,000	Optical (mass based on light scattering)	1 sec	Capable to provide real-time aerosol mass readings in broad size range with good correlation to gravimetric method. Compact design and inexpensive cost. Need for individual vehicle-dependent calibration.

CHAPTER 3

MATERIALS AND METHODS

The following chapter outlines the methodology used throughout the study. In particular, the description of the particle sensor under investigation, experimental procedure, testing equipment, set-up configuration, test cycles, and other research specifics are described below in detail.

Particle Sensor Description

The particle sensor, a major component of PM measurement unit evaluated during experiments, represents a real-time device for continuous exhaust measurements. According to the manufacturer's technical specifications, it is designed to measure the PM mass concentration in the particle size range of 23 nanometers to 2.5 micrometers. The particle sensor is capable of measuring concentration range from $1 \mu\text{g}/\text{m}^3$ to $250 \text{mg}/\text{m}^3$. As shown in Figure 6, the particle sensor represents a flow-through device with sample inlet, sample outlet, inlet for ionizing and sheath air, and electronics unit. Metal housing was added in order to enclose the sensor and incorporate additional accessories. As a result, the unit includes a particle sensor, an absolute filter for incoming sheath air, and temperature and flow control modules.

Diffusion particle charging and electrical detection of charged particles are the major operating principles used by the particle sensor. The PM measurement involves charging of pressurized air as it enters the sensor's sheath air inlet. The charging is done by unipolar corona discharge. Subsequently, the charged pressurized air gets mixed with

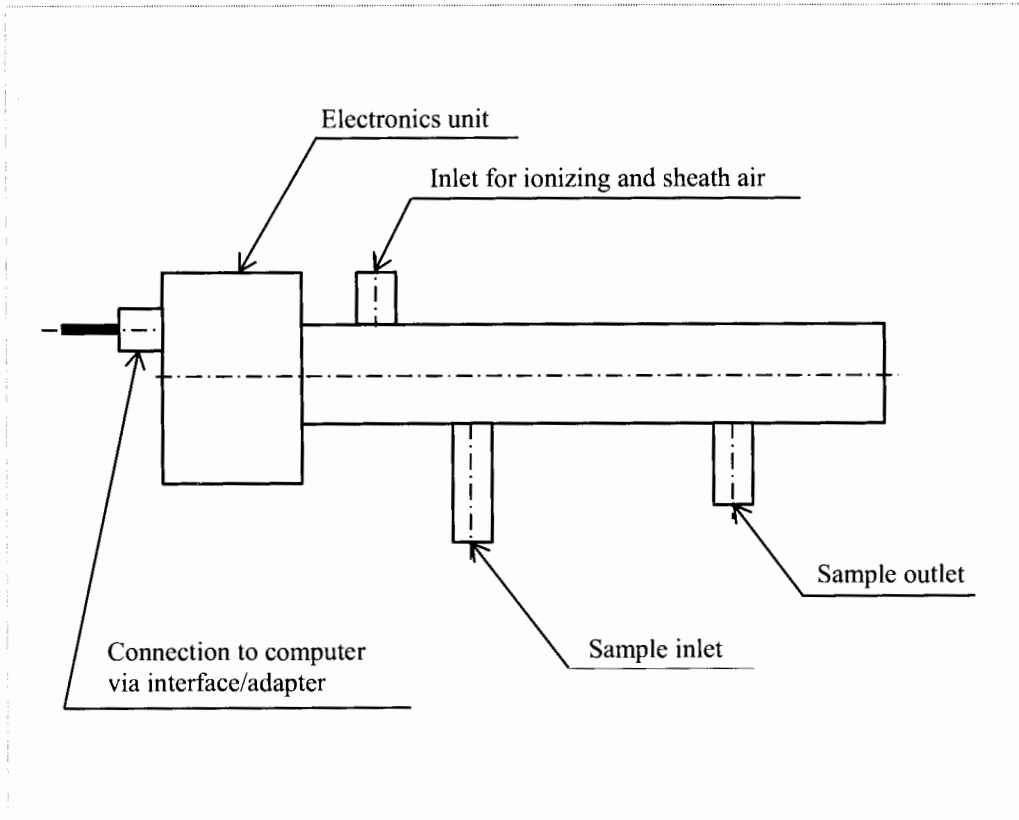


Figure 6. The particle sensor schematic.

exhaust sample gas as they enter the sensor's inner mixing chamber. The exhaust particles get charged in mixing chamber as they get mixed with corona produced ions carried by pressurized air.

Excess ions are captured by an electrostatic precipitator located after the mixing chamber. Electrical current is produced by charged particles flowing from the sensor. It is measured by a built-in electrometer and is proportional to the PM mass concentration (Besch et al., 2011). The reference instrument used for data comparison was Dekati Mass Monitor. Description of its operational principle and summary of recent research has been presented in a previous chapter. According to research findings, this particular

instrument was able to produce reliable real-time PM measurements, including those in transient conditions, in good agreement with federal filter-based reference method and with better accuracy (Khalek, 2005, 2008; Lehmann et al., 2004; Mamakos et al., 2006). Specification characteristics for the mass monitor are presented in Appendix B (Dekati, 2010).

Test Configuration and Set-Up

The study was conducted using several configurations. Test designs differed in sampling probe location and sample dilution. For the majority of tests, the sampling probe was connected to the vehicle before the DPF with exception of three tests when the exhaust was sampled after the DPF during the filter efficiency evaluation portion of the study.

Two types of sample dilution configuration were used throughout the study. The testing designs are shown in Figure 7 and 8. In the first configuration, the particle sensor, as well as the reference instrument, was measuring PM concentrations from the diluted exhaust sample. In this case, the exhaust flow from each test vehicle was first directed to a dilution system via heated line with outside diameter of 1.18 in. (3 cm) and then to both PM measurement instruments using transfer lines. Nozzles of the sampling probes were equipped with flow-regulating valves supplied by dilution system manufacturer. Typical flow rates for dilution system and its other specifications are presented in Appendix C (Dekati, 2008). Sampling transfer lines delivering the diluted exhaust flow from diluter to both instruments were of similar diameter of 0.2 in. (5 mm) and length below 39 in. (1 m).

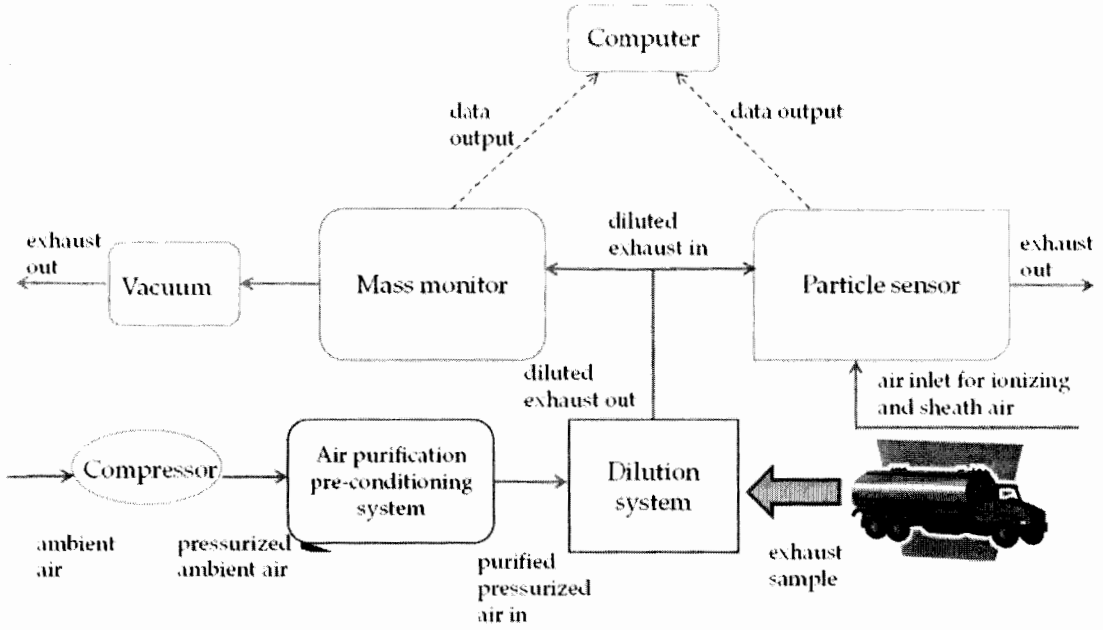


Figure 7. Test configuration diagram with the particle sensor sampling diluted exhaust.

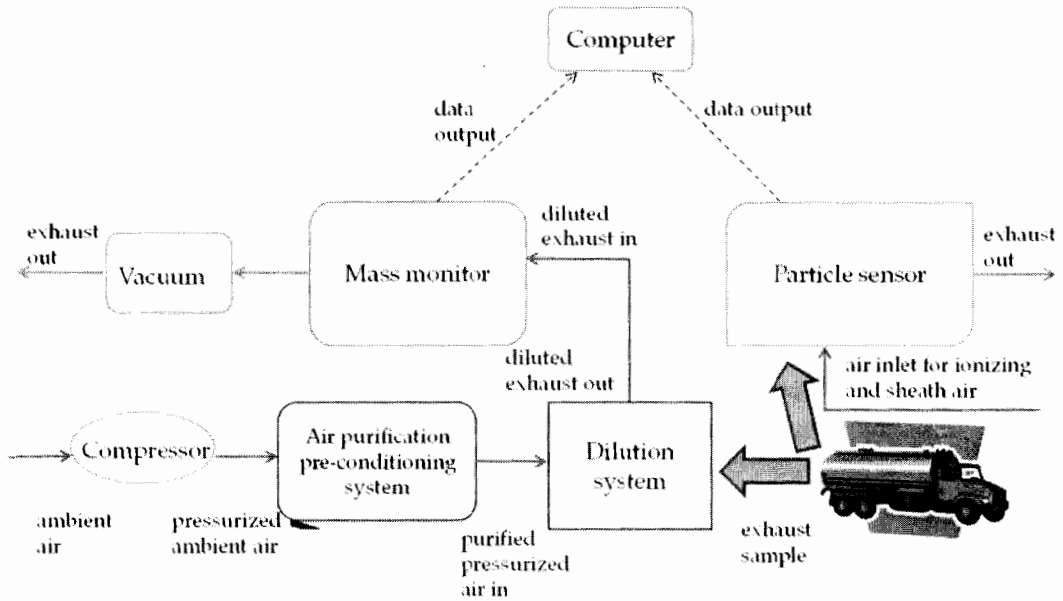


Figure 8. Test configuration diagram with the particle sensor sampling raw exhaust.

and were similarly attached to the diluter's outlet pipe parallel to each other to ensure the equivalent supply of sample to both instruments.

In the second test configuration, the particle sensor was employed to conduct measurements from the raw sample without dilution, while the reference instrument was sampling the diluted exhaust air. During raw exhaust sampling, the particle sensor was using the exhaust sample delivered using a separate sampling probe. In this case, two heated lines with outside diameter of 1.18 in. (3 cm), with one line connected to particle sensor and another line going to diluter/mass monitor, were similarly connected to the exhaust pipe using identical nozzles.

Dilution of exhaust gas was performed using ambient air. Prior to being delivered to the dilution system, the ambient air got purified and pressurized by going through a compressor and air purification pre-conditioning system. During all PM measurements the dilution system was operated at low (103:1) dilution ratio setting without pre-dilution. Operational characteristics of dilution system are summarized in Table 3. As the exhaust sample was flowing through a dilution system, it entered the first dilution stage with heated diluter. At this stage, the sample temperature was raised to 150 °C (302 °F) while sample gas got diluted about ten times. Subsequently, the exhaust sample moved to evaporation tube with temperatures at 350 °C (662 °F) where volatile particles got evaporated from the sample. Afterwards the sample got passed to the secondary dilution stage where it got diluted about ten times and cooled down to ambient temperature.

After being discharged from the dilution system, the diluted sample was fed to the PM measurement equipment. For example, when using the first test configuration, both

Table 3. Operational characteristics of dilution system (Dekati, 2008).

	Dilution system stages	Operation temperatures	Dilution ratio
1	Pre-diluter	—	0 (for low dilution setting)
2	Heated diluter	150 °C (302° F)	10
3	Evaporation tube	350 °C (662 °F)	0
4	Secondary diluter	less than 35 °C (95 °F) ambient sample temperature	10

instruments were connected to dilution system parallel to each other and performed simultaneous measurements of the diluted exhaust sample.

Additionally, the purified, dry, and pressurized air supply was connected to the particle sensor's inlet for ionizing and sheath air. This air flow was designed to shield internal parts from particle contamination, provide charged pump flow into the sensor, and assist in cooling. After the measurement, the sample gas exited the units. The reference PM measurement device also required an external vacuum pump to provide out-flow of exhaust sample.

The particle sensor and the reference instrument were connected to a computer via integrated universal serial bus (USB) interface. Prior to measurement testing, the instruments were warmed-up according to manufacturer's requirements to ensure all system parameters have reached optimum operation conditions. Zero calibration of instruments was done immediately before beginning the measurements in order to reduce zero offset and improve the measurement accuracy. During zeroing, the instruments

were drawing sampling air through a zero high efficiency particulate air (HEPA) filter. According to the USDE standards, HEPA filters remove aerosol particles greater than 0.3 micrometers in diameter with 99.97% efficiency (USDE, 1997). Throughout each test, the monitoring of PM instruments status and data collection was done using a graphical interface software program installed on computer. Upon completion of the measurement, the data from all instruments were uploaded for further post-processing and analysis.

Test Vehicle

Tests were conducted using a medium-duty diesel truck equipped with the DPF. This vehicle was powered by a 2009 model year engine. Specifications for this vehicle are listed in Table 4.

Table 4. Test vehicle specifications.

	Specifications	Test vehicle
1	Engine year	2009
2	Engine displacement	6.4 liters
3	Induction	Turbocharged
4	Injection type	Direct electronic control
5	Engine power rating	325 hp at 3000 rpm
6	DPF-equipped	Yes

Test Fuel

The test fuel used for the testing was a federal diesel No. 2 blend. It represents an ultra low sulfur fuel with sulfur content of 15 ppm. According to federal requirements,

this fuel grade has a maximum aromatic content of 35% or a minimum cetane index of 40 (DieselNet, 2009b).

Test Cycles

In order to investigate the particle sensor suitability for PM measurement during in-field environments, it was necessary to examine its performance during a variety of conditions commonly encountered in real life, including fast changing transient events as well as steady-state operating conditions. Selection of test cycles was done according to these requirements. Snap acceleration cycle, as well as typical on-road driving, was chosen for the particle sensor evaluation during transient and semi-transient operations. On-road driving cycle included a typical freeway driving segment following the traffic with speed limit of 65 miles per hour. Each driving cycle incorporated a short portion of non-congested urban street driving at the beginning and at the end. The test cycles are summarized in Table 5.

Table 5. Description of test cycles.

	Test Cycle	Description	Sampling location	Dilution
1	Snap-idle acceleration	Series of transient acceleration events	Engine-out	Diluted
2	Snap-idle acceleration	Series of transient acceleration events	Engine-out	Raw
3	Snap-idle acceleration	Series of transient acceleration events	DPF-out	Diluted
4	Urban driving	Freeway with non-congested urban portions	Engine-out	Diluted

Snap-idle acceleration cycle was used during investigation of the particle sensor performance when measuring diluted and raw exhaust (without dilution). The cycle was performed according to a J1667 diesel snap acceleration procedure recommended by the Society of Automotive Engineers (SAE) for assessing smoke emissions from in-use vehicles (SAE, 1996). During each test cycle, the test vehicles running at low idle received six snap-accelerations. While at low idle, the engine was running without any load, except the engine accessories, which were shut off during testing. While accelerating, the vehicle operator moved the throttle to the fully open position, as rapidly as possible, and hold it until the engine reached its maximum governed speed, plus an additional one to four seconds. Subsequently, the throttle was released to allow the engine to return to the idle speed. After completion of six accelerations, the test was repeated to ensure repeatability.

When conducting snap-idle accelerations, the test vehicle was in stationary condition and PM instruments were placed alongside of vehicle. In this case, the measurement devices were connected to plug-in wall mounted power supply via extension cords. The instruments were mounted and tightly secured on the test vehicle in order to conduct testing during urban driving conditions. This required the use of a portable power generator as a major power source for PM devices.

Data Processing

The first step in data processing was to retrieve PM measurement data recorded during testing. Subsequently, the data were converted from a text file to Excel format. Further processing involved conversion of PM concentrations to the same units (mg/m^3)

and correction for dilution. In order to calculate the concentration of the particles adjusted for dilution, the measurement results were multiplied by the particle concentration reduction factor of 103, corresponding to the low dilution ratio setting according to the diluter's specific data sheet. The particle sensor measurements have also been corrected for internal dilution resulting from differences between sample inlet and sample outlet flow rates due to additional sheath air supply provided through sensor's sheath air inlet. Therefore, the particle sensor measurements have been additionally multiplied by internal dilution ratio calculated by dividing a sample inlet flow by outlet flow.

Afterwards, dilution adjusted data from both instruments were aligned to match each other. Resulting output was then used to create temporal distribution graphs of PM mass concentrations. Separate line graphs were generated for each specific test in order to perform evaluative comparison of PM distribution characteristics, tendencies, and emission values measured by the particle sensor and the reference instrument.

Exploratory Data Analysis and Statistical Techniques

To undertake the analysis of emission data, the following elements were employed:

1. Exploratory data analysis (e.g., distribution evaluation and descriptive statistics).
2. Statistical techniques (e.g., correlation and regression analyses).

Exploratory Data Analysis

Exploratory analysis, using basic statistics and distribution line graphs of dilution-adjusted PM mass concentrations from both instruments, was conducted to explore, describe, and summarize the emission data, as well as to detect relationships and uncover underlying structure, and generate hypotheses regarding the data (Lind, Marchal, & Wathen, 2005). Employed basic statistics include the mean, median, and standard deviation (Westerholm & Egeback, 1994; J. S. Wang et al., 2006).

Measure of central tendency, arithmetic mean, for each data set was calculated according to Equation 1 (Lind et al., 2005):

$$\bar{x} = \frac{1}{n} \sum x \quad (1)$$

where \bar{x} is the statistical average (sample mean), mg/m³;

x is the variable, mg/m³; and

n is number of variables in the sample.

Measure of dispersion of data, standard deviation (σ), was calculated according to Equation 2 (Lind et al., 2005):

$$\sigma = \sqrt{\frac{\sum (x - \bar{x})^2}{(n-1)}} \quad (2)$$

where σ is the standard deviation of the sample; mg/m³;

\bar{x} is the sample mean, mg/m³;

x is the variable, mg/m³; and

n is the number of variables in the sample.

The temporal distribution line graphs, generated for each specific test, were used to portray distribution trends and peak concentrations of the PM emissions measured by the particle sensor and the reference instrument (Moosmüller et al., 2001a; Wallace, 2005; X. Wang, Hase, Olson, Sreenath, & Agarwal, 2009). Moreover, frequency histograms were generated for each set of data to explore the data for normality. All data sets were log transformed due to lack of normality, and consequent analysis was performed on transformed data (Yanowitz, McCormick, & Graboski, 2000).

Statistical Techniques

Correlation and linear regression analyses, using Microsoft Excel (Microsoft Inc., Redmond, Washington) software, were employed to draw conclusions regarding the performance of the particle sensor during exhaust emission measurement (Johnson et al., 2011; Moosmüller et al., 2001a; Rice, 2004; Tasić et al., 2012).

Specifically, correlation analysis was used to study the relationship between PM mass concentration measurements from the particulate sensor and the reference instrument (mass monitor). The scatter diagrams, used to explore whether there is a relationship, were developed for each test using the PM mass concentration measurement from the particle sensor as predictor X (a variable predicted from), an analog of independent variable (scaled along horizontal axis) and the PM mass concentration

measurement from the mass monitor as criterion Y (a variable predicted to), an analog of dependent variable [scaled along vertical axis] (Lind et al., 2005). Both variables satisfy the requirements of interval level of measurement of data.

Further, numerical measures of relationship—coefficient of correlation [r] (Pearson's r) and coefficient of determination (r^2)—were developed to determine the strength and direction of relationship. Coefficient of determination shows the proportion of the total variation in the dependant variable Y that is explained, or accounted for, by the variation in the independent variable X (Johnson et al., 2011; Lind et al., 2005). Significance of correlation coefficient was tested using hypothesis testing at .05 significance level, were the following hypotheses were generated:

1. Null hypothesis H_0 : there is no significant association between PM measurements produced by the particulate sensor and the mass monitor.
2. Research hypothesis H_1 : there is a significant association between PM measurements produced by the particulate sensor and the mass monitor.

Null hypothesis of not significant relationship was rejected when calculated significance coefficient was less than significance level of .05. In these instances, the research hypothesis was accepted as being true (Lind et al., 2005).

Linear regression analysis technique was used to develop the equation of the line (regression line) that best fits the data and to provide the estimates of PM mass concentration (Lind et al., 2005; Yanowitz et al., 2000):

$$Y_{\text{predicted}} = \beta_0 + \beta_1 X \quad (3)$$

where X is the PM mass concentration measurement from the particle sensor, mg/m^3 ; and

$Y_{\text{predicted}}$ is the PM mass concentration measurement from the mass monitor, mg/m^3 ;

β_0 is intercept of the regression line; and

β_1 is slope of the regression line.

To test the error in estimate, the standard error of estimate (the measure of the dispersion of observed values around the line of regression) and 95% confidence interval for the estimate have been calculated (Rice, 2004; Yanowitz et al., 2000).

The following assumptions underlying linear regression have been tested for each set of data (Lind et al., 2005):

1. For each value of X , there is a group of Y values. These Y values follow normal distribution.
2. The means of these normal distributions lie on the regression line.
3. The standard deviations of these normal distributions are all the same. The best estimate we have of this common standard deviation is the standard error of estimate.
4. The Y values are statistically independent. This means that in selecting a sample a particular X does not depend on any other value of X . This assumption is particularly important when data is collected over a period of time.

DPF Efficiency Evaluation Study

Assessment of the particle sensor's ability to be used during evaluation of DPF efficiency was also included in current study. It involved comparison of the DPF

efficiencies calculated using the PM sensor data against the DPF efficiencies computed using output from the reference instrument. To estimate the mass based filtration efficiency of the DPF, the PM mass concentrations were measured upstream and downstream of the filter using the particle sensor and the reference instrument. The setup for the DPF filtration efficiency measurement is shown below. Test vehicle with specifications summarized in Table 4 was used during the DPF efficiency study. First, the PM mass measurements were conducted after the DPF using the setup shown in Figure 9. Three successive tests were performed using this configuration. Afterwards, the measurements were conducted three consecutive times to measure PM mass concentration upstream of the DPF according to the setup depicted in Figure 10.

The DPF efficiency was determined based on comparison of the test total PM mass downstream of the filter to the total upstream PM levels in the exhaust. Equation 4 was used to calculate the DPF efficiency and is presented below (Van Asch, Verbeek, Lighterink, & Kadijk, 2009):

$$DPF_{eff} = \left(1 - \frac{PM_A}{PM_B}\right) \times 100 \quad (4)$$

where DPF_{eff} is the DPF efficiency, %;

PM_B is a sample total PM concentration before the DPF, mg/m^3 ; and

PM_A is a sample total PM concentration after the DPF, mg/m^3 .



Figure 9. After the DPF measurement of PM mass concentration setup for the DPF filtration efficiency study.



Figure 10. Before the DPF measurement of PM mass concentration setup for the DPF filtration efficiency study.

In summary, selection of procedure for data analysis was based on the ability of each procedure/method to provide information needed to address the research questions. Statistical techniques were used to generate hypothesis, confirm significance of uncovered patterns, interpret data, and draw conclusions based on research findings.

CHAPTER 4

RESULTS

Exploratory Data Analysis Results

The study investigated total number of 20 tests of PM mass concentration from exhaust gas, taken over different days with both the particle sensor and the mass monitor sensor, ranging from as low as three and as high as 10 tests per day for each instrument. The majority of tests (17 tests out of 20) were performed with the probe location before the DPF under various test cycles, including snap-idle and urban driving. The remaining three tests were conducted with the probe location connected to the vehicle exhaust system after the DPF. Out of 17 tests with sampling probe before the DPF, three tests were performed without particle sensor sample dilution according to the snap-idle test cycle; and four tests were carried out under urban driving test cycle with sample dilution.

Temporal Distribution Analysis Results

Temporal PM distribution graphs were generated using second-by-second PM measurements obtained from the particle sensor and the reference mass monitor during snap-idle acceleration and urban driving test cycles. Snap-idle acceleration cycle was used during investigation of the particle sensor performance when measuring diluted and raw exhaust (without dilution). An analysis of the results is presented below.

Snap Acceleration with Diluted Sampling

Snap-idle acceleration data obtained from vehicle engine-out diluted sampling during five testing days are displayed in Figures 11, 12, 13, 14, and 15, respectively. The continuous PM measurements revealed remarkable similarities in trends and PM

distribution patterns. Particle sensor readings were of the same order of magnitude as the values from the reference mass monitor. Temporal distribution graphs from the first, fourth, and fifth testing days show that particle sensor measurements were generally slightly lower than those from mass monitor with average percent differences of 20, 25, and 37%, respectively. However, the particle sensor read somewhat higher PM levels on the second and third testing days compared to the mass monitor with average percent differences of 22 and 29%, respectively. More studies are needed to examine the observed variation in PM measurement results between instruments.

Discrepancy between engine-out measured values was more visible at higher PM concentrations corresponding to snap-acceleration events, during which the engine throttle was moved to the fully open position. According to data from three DPF-out PM measurement tests of the diluted exhaust conducted on the fifth day, the temporal PM distributions revealed disagreement in values from both instruments. Six engine acceleration events were clearly visible on the particle sensor temporal distribution from the first of the three DPF-out tests with corresponding to only minor PM variations from mass monitor. During the second DPF-out test, the particle sensor was able to display five out of total six PM peaks; whereas mass monitor clearly displayed one major peak accompanied by four medium PM peaks. The particle sensor PM values from this test were lower compared to those from the mass monitor and to the particle sensor itself during other tests. The third test revealed three elevated PM peaks and three medium PM peaks from the particle sensor. In this case, the mass monitor detected only minor peaks. Temporal distributions for three DPF-out tests are presented in Figure 16. The particle

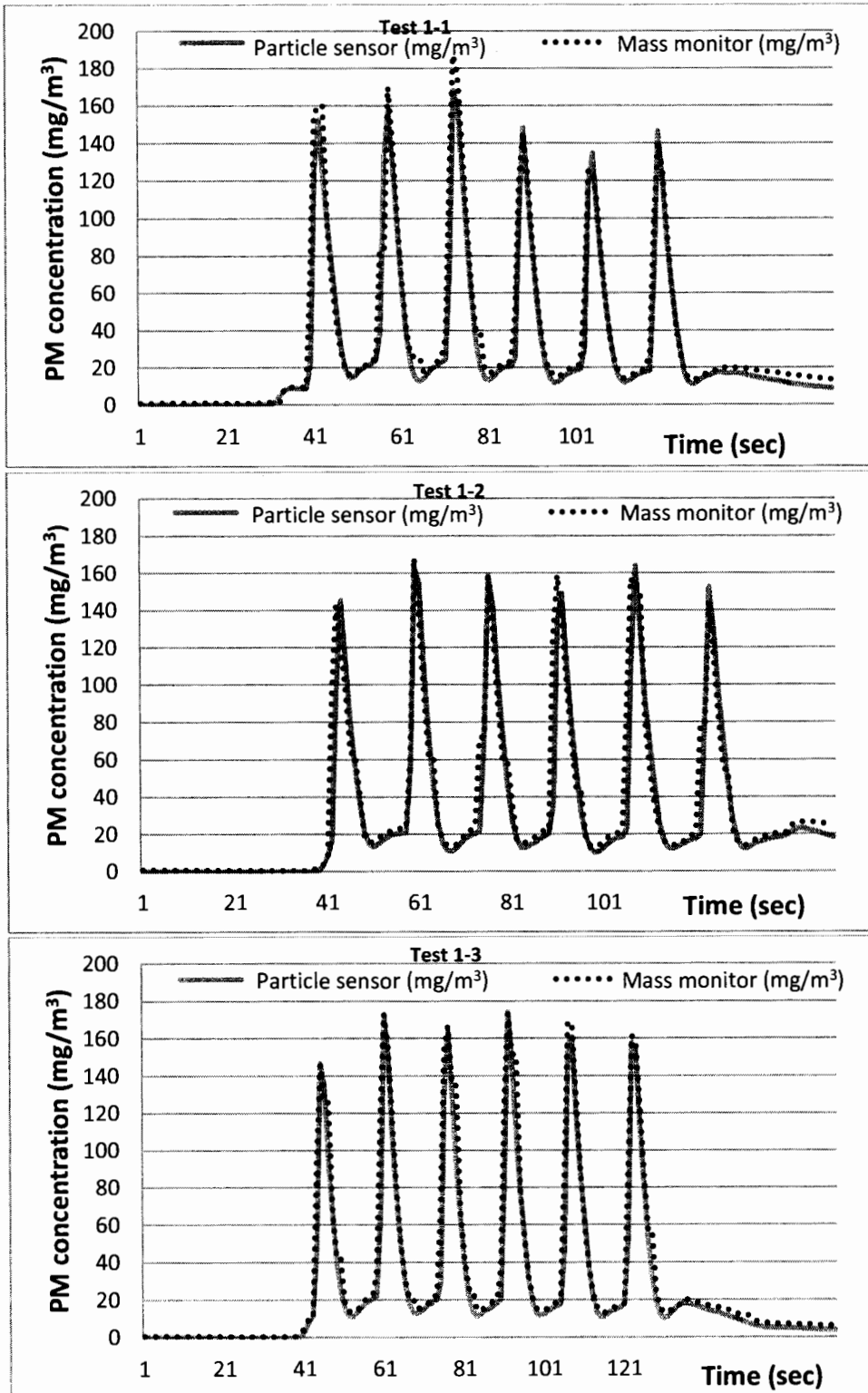


Figure 11. Snap-idle acceleration with the particle sensor measuring engine-out diluted exhaust, day 1.

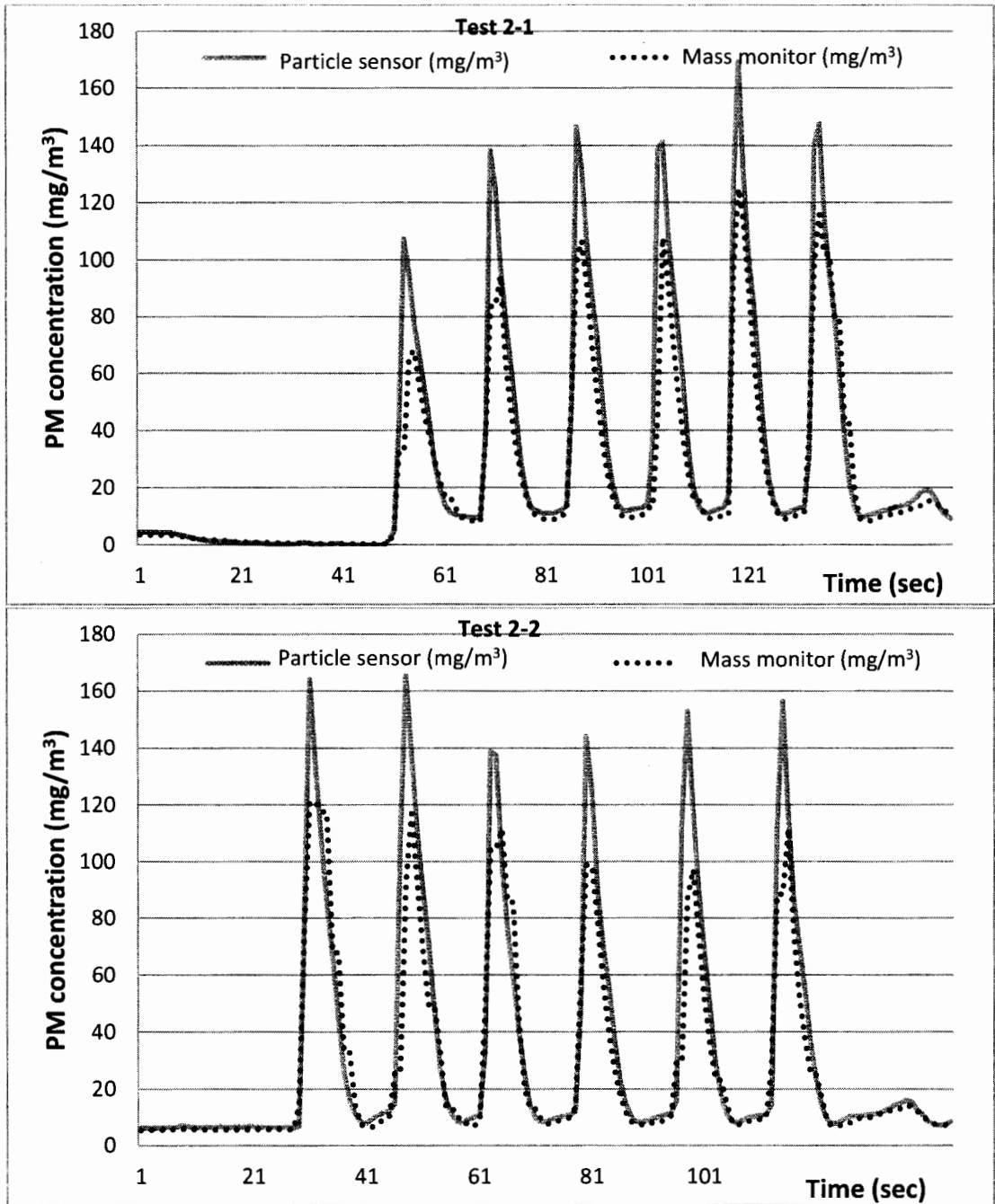


Figure 12. Snap-idle acceleration with the particle sensor measuring engine-out diluted exhaust, day 2, tests 1 and 2.

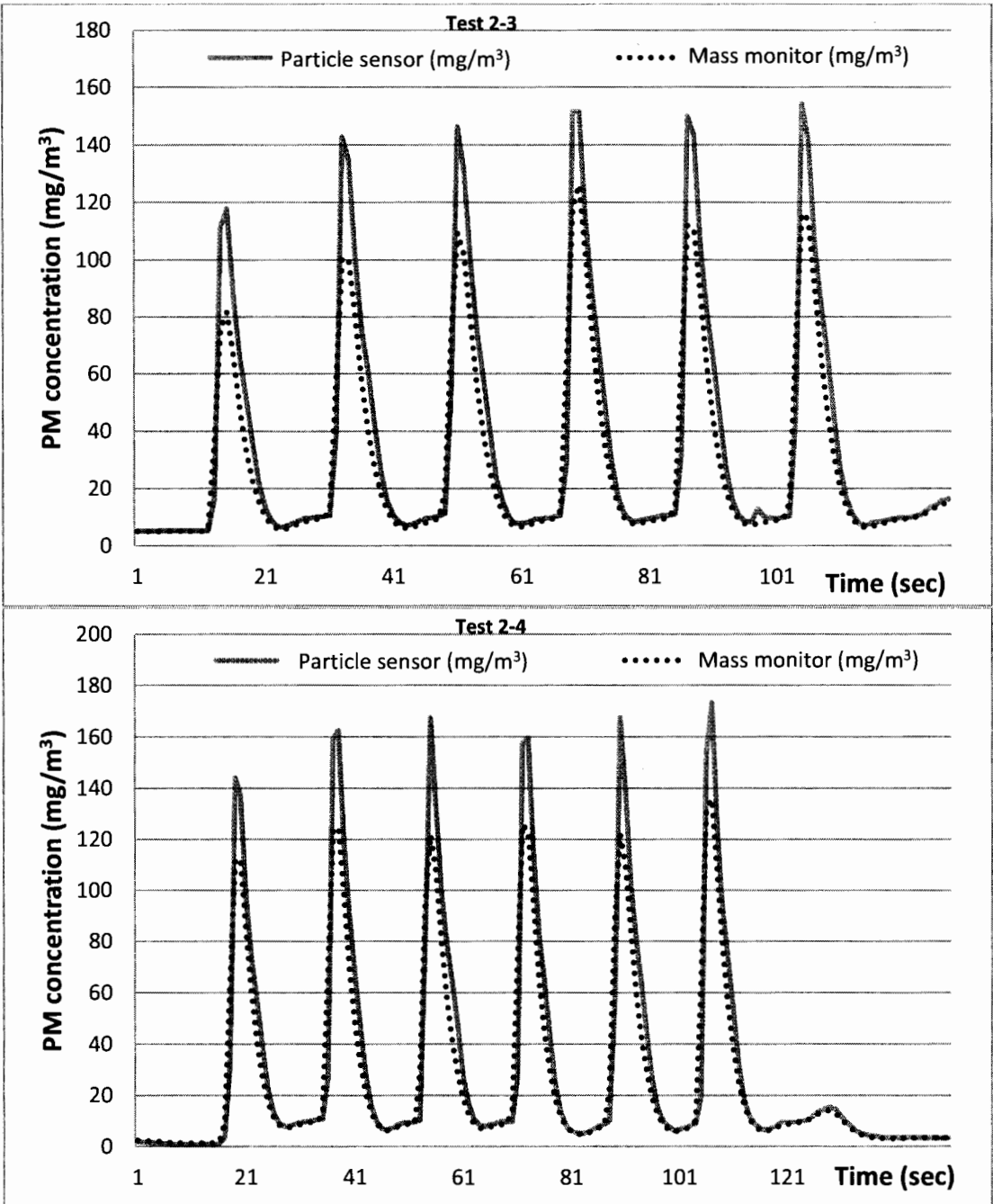


Figure 13. Snap-idle acceleration with the particle sensor measuring engine-out diluted exhaust, day 2, tests 3 and 4.

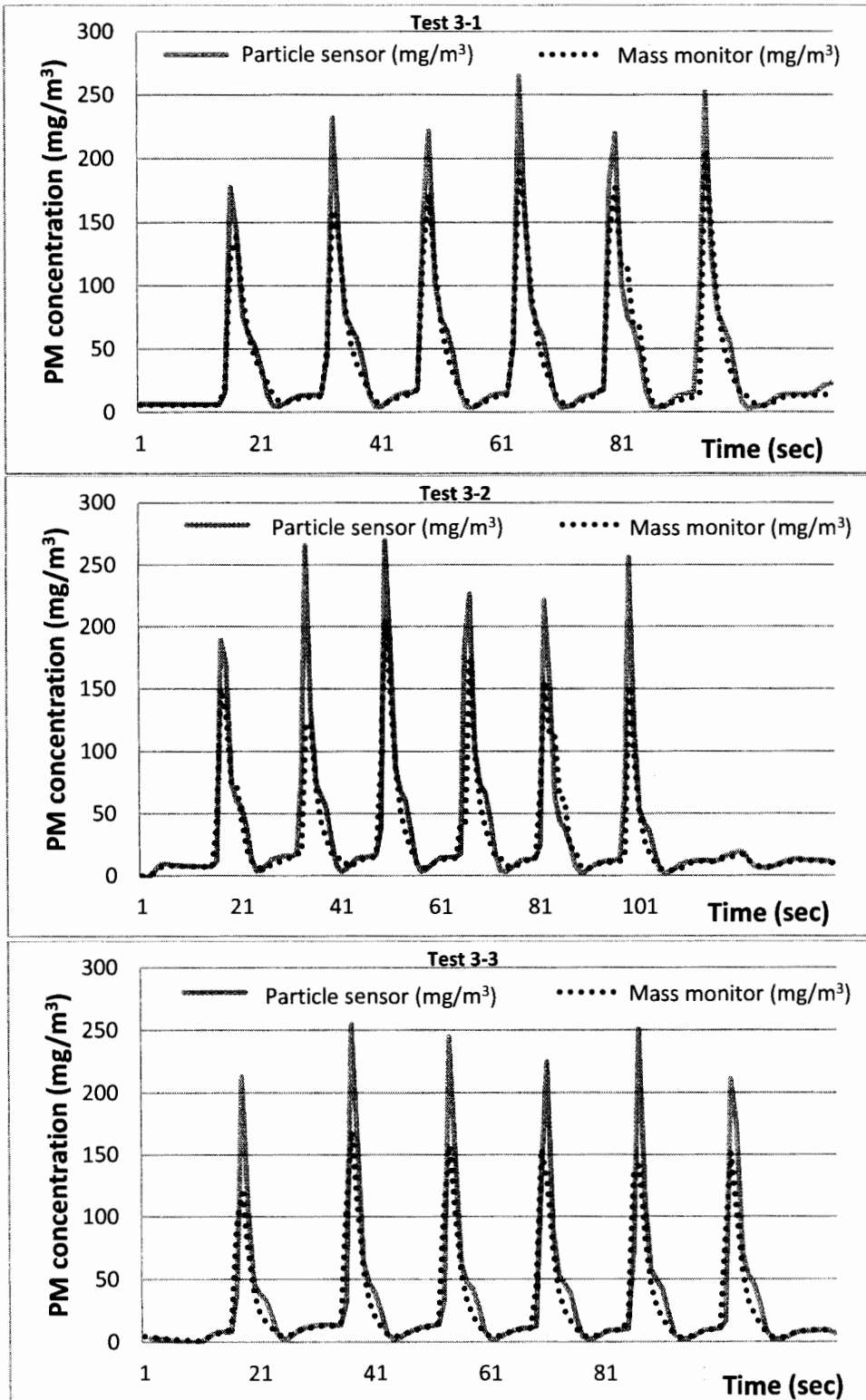


Figure 14. Snap-idle acceleration with the particle sensor measuring engine-out diluted exhaust, day 3.

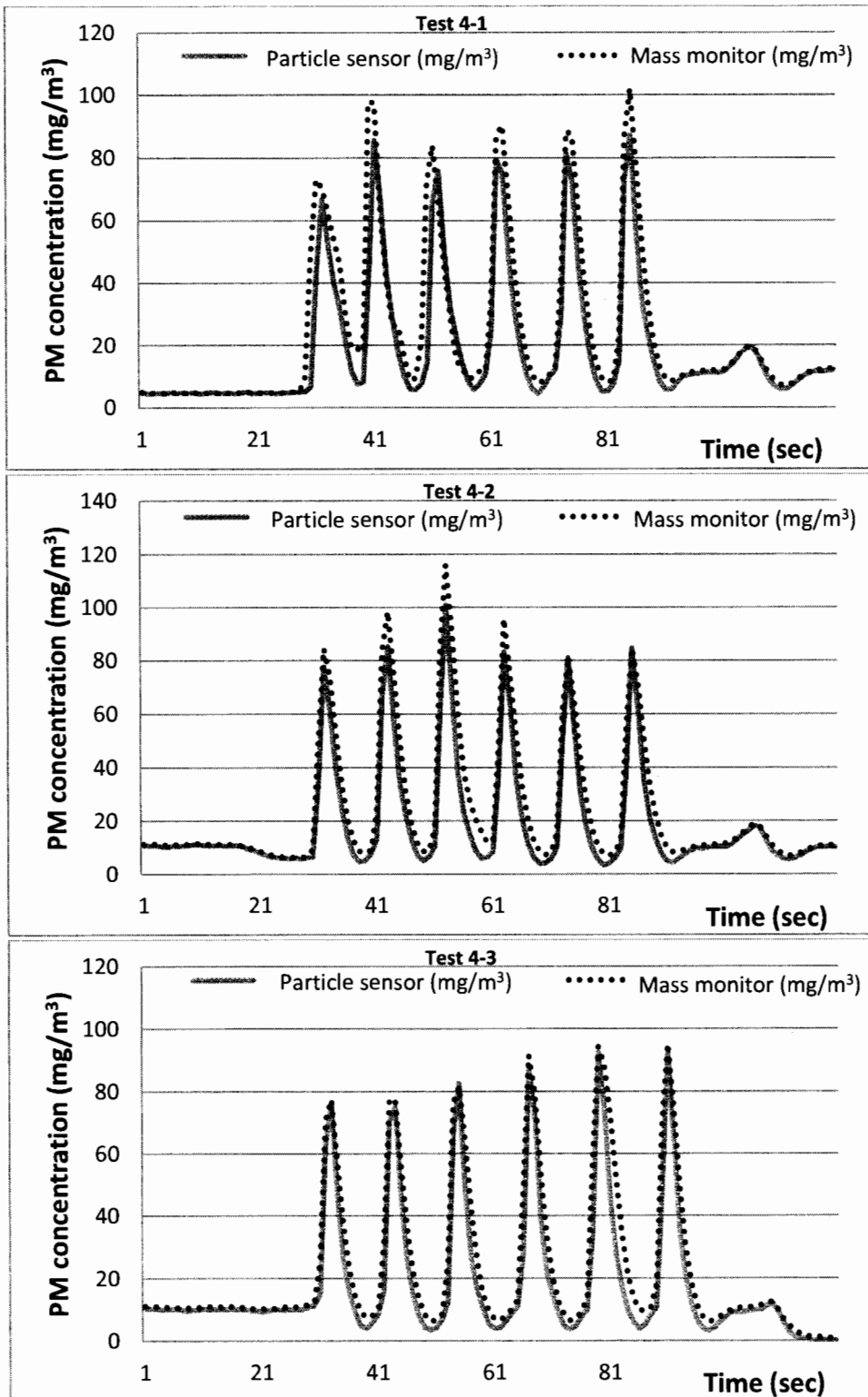


Figure 15. Snap-idle acceleration with the particle sensor measuring engine-out diluted exhaust, day 4.

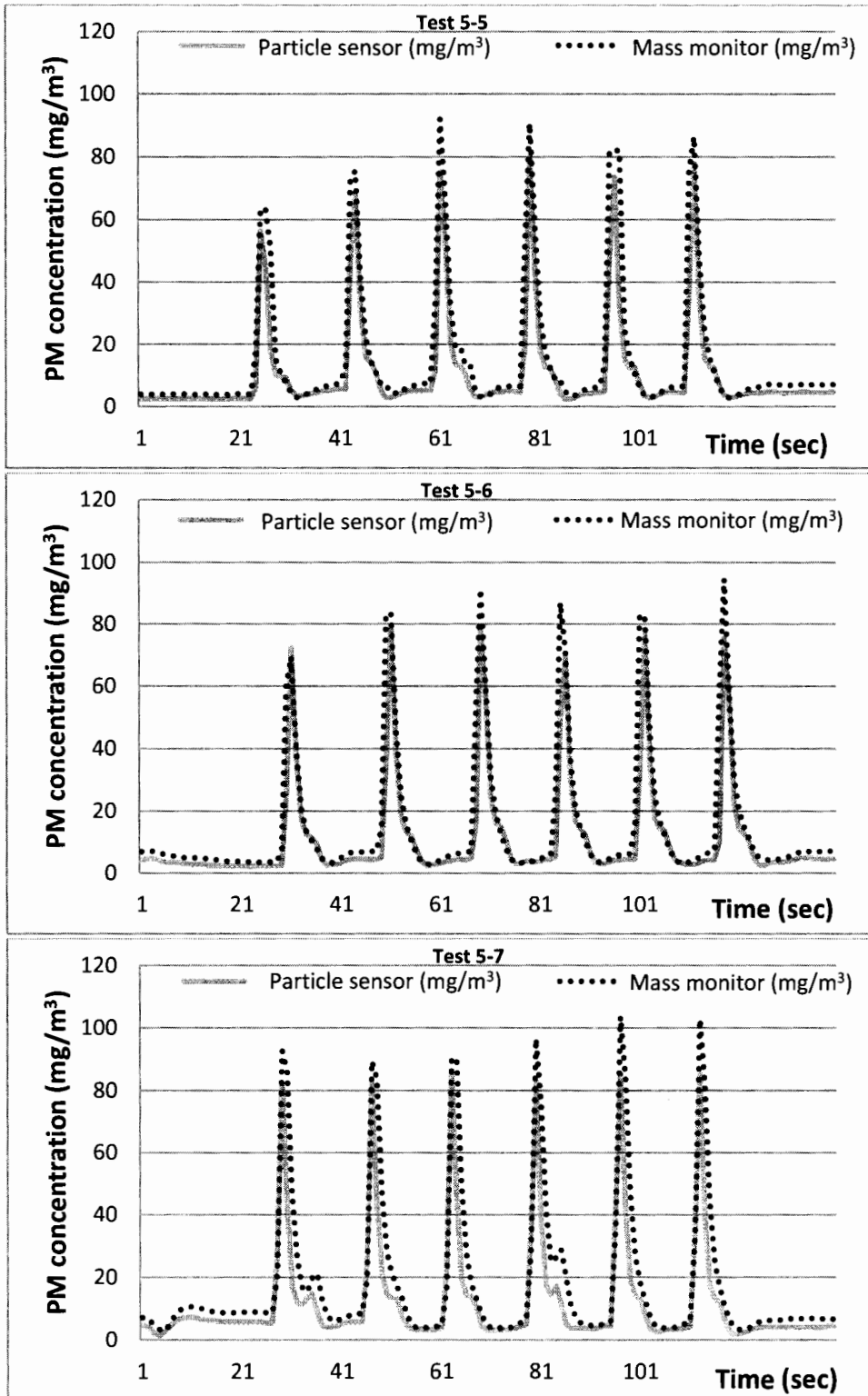


Figure 16. Snap-Idle acceleration with the particle sensor measuring engine-out diluted exhaust, day 5.

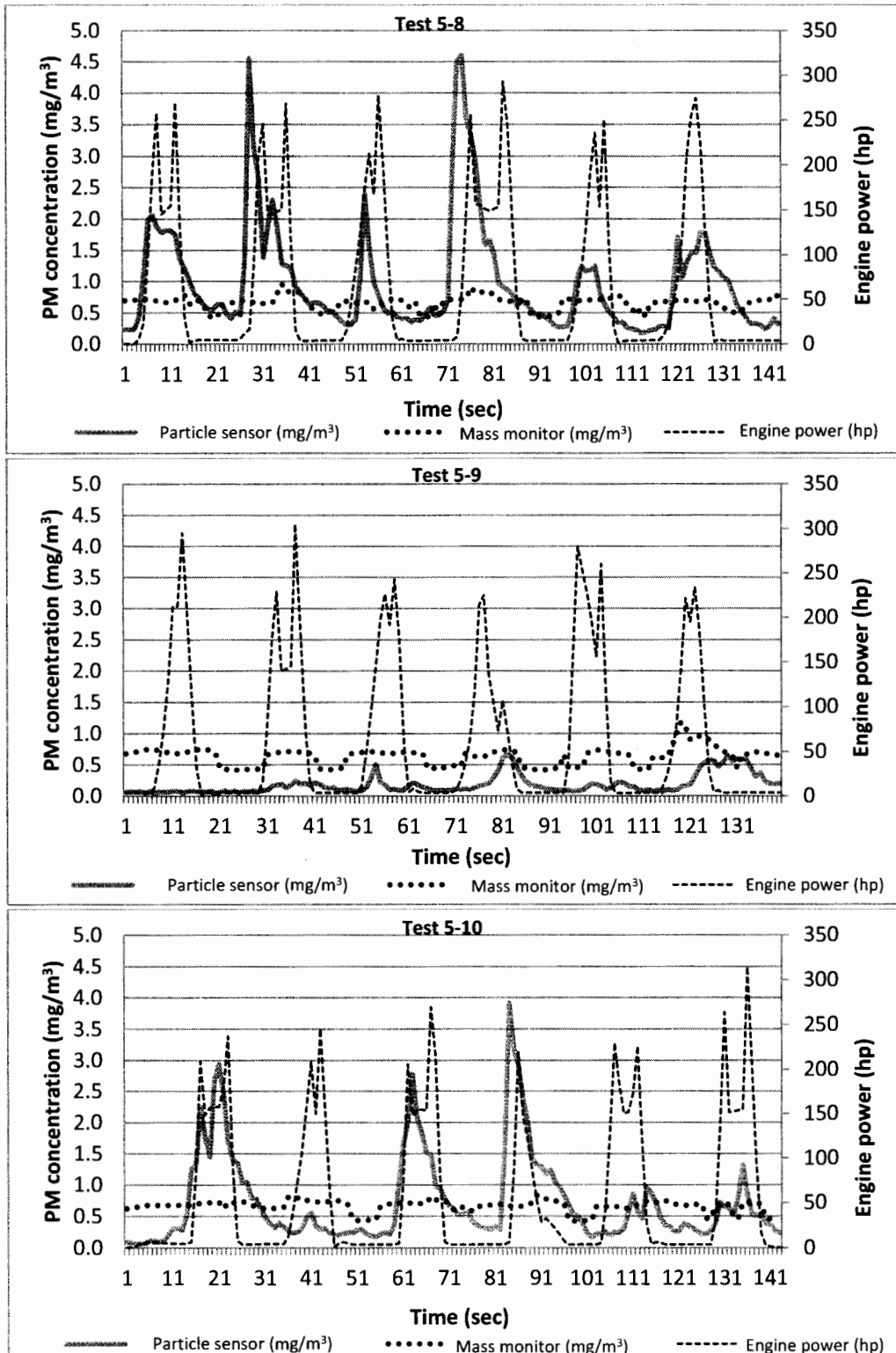


Figure 17. Snap-idle acceleration with the particle sensor measuring DPF-out diluted exhaust, day 5.

sensor measurements also displayed association with engine power during engine acceleration events. At the same time, graphs showed weak association between engine power and the mass monitor PM measurements. Overall, the particle sensor response was more consistent with increase in PM emissions during engine acceleration events compared to that from mass monitor. This is consistent with 40% increase in percent difference between PM measurements from both instruments.

Snap Acceleration with Raw Sampling

Temporal distribution graphs of snap-idle acceleration tests with raw exhaust sampling are presented in Figure 17. These tests were conducted during the third testing day after the three tests with sample dilution (shown in Figure 13) discussed earlier. Similarly to tests with sample dilution from the same day, the particle sensor measurements during raw emission testing were somewhat higher than those from the mass monitor. Average percent difference between instruments was 23% higher than results from the same day with sample dilution. The most noticeable difference in PM distribution was visible during vehicle decelerations. This could be due to the presence of condensing volatile and semi-volatile compounds in the exhaust flow sampled by the particle sensor. These compounds were removed from the mass monitor incoming exhaust sample flow during the dilution process.

Urban Driving with Diluted Sampling

Four tests with diluted exhaust sampling were conducted to evaluate the particle sensor's performance during urban driving conditions. Line graphs of PM temporal distributions from these tests are presented in Figures 18 and 19. As can be seen from

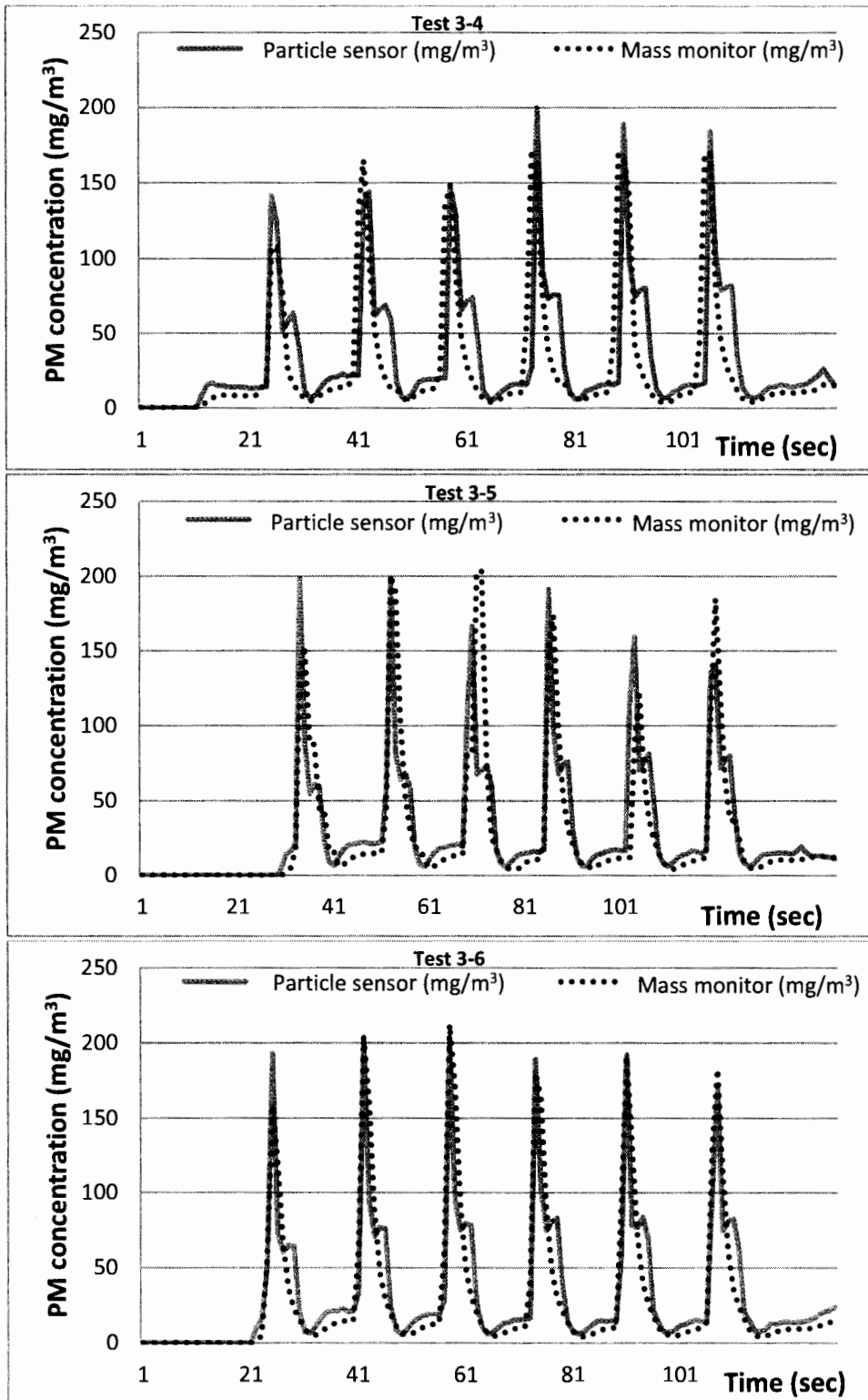


Figure 18. Snap-idle acceleration with the particle sensor measuring raw exhaust, day 3.

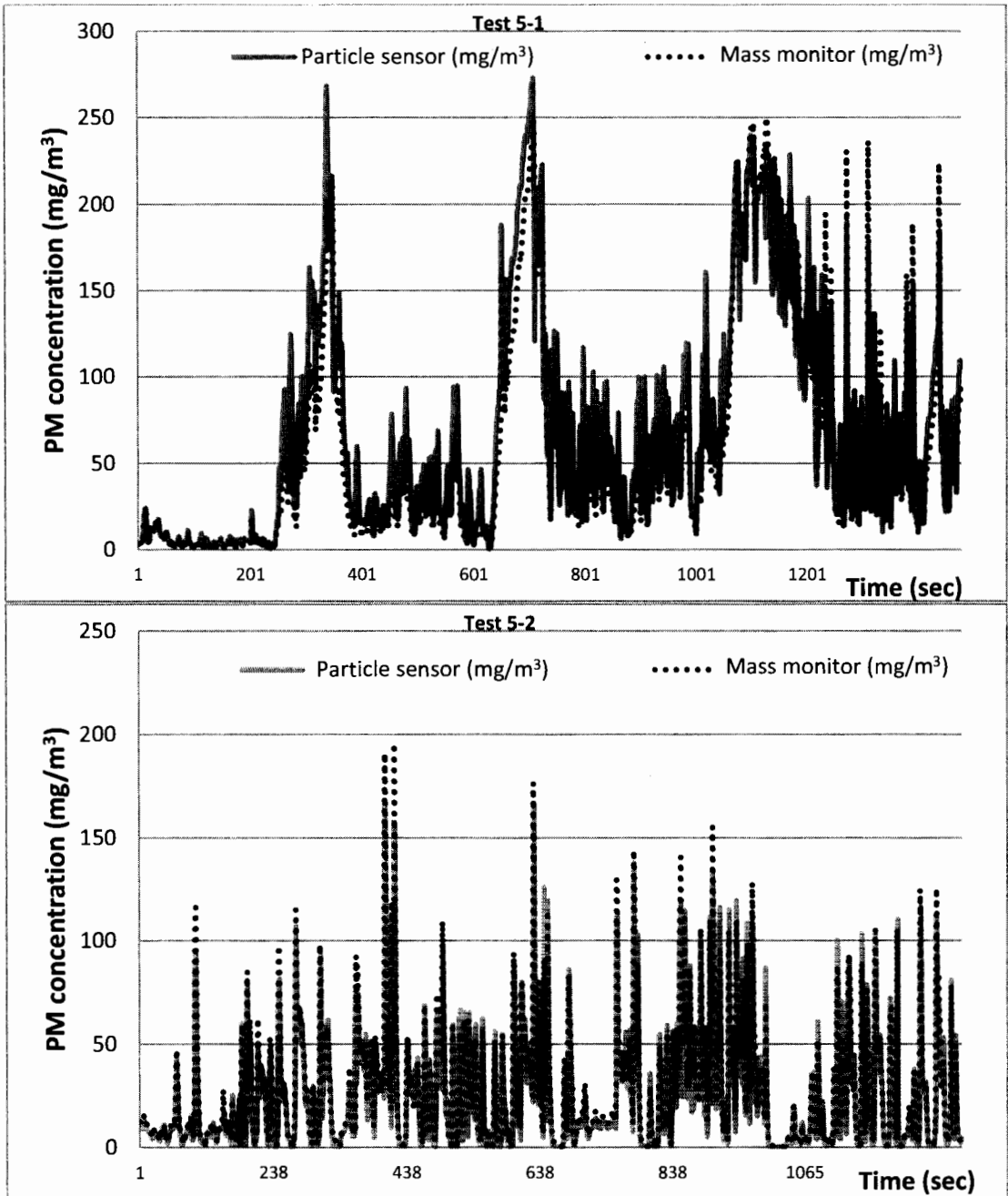


Figure 19. Urban driving tests cycle with the particle sensor measuring diluted exhaust, day 5, tests 1 and 2.

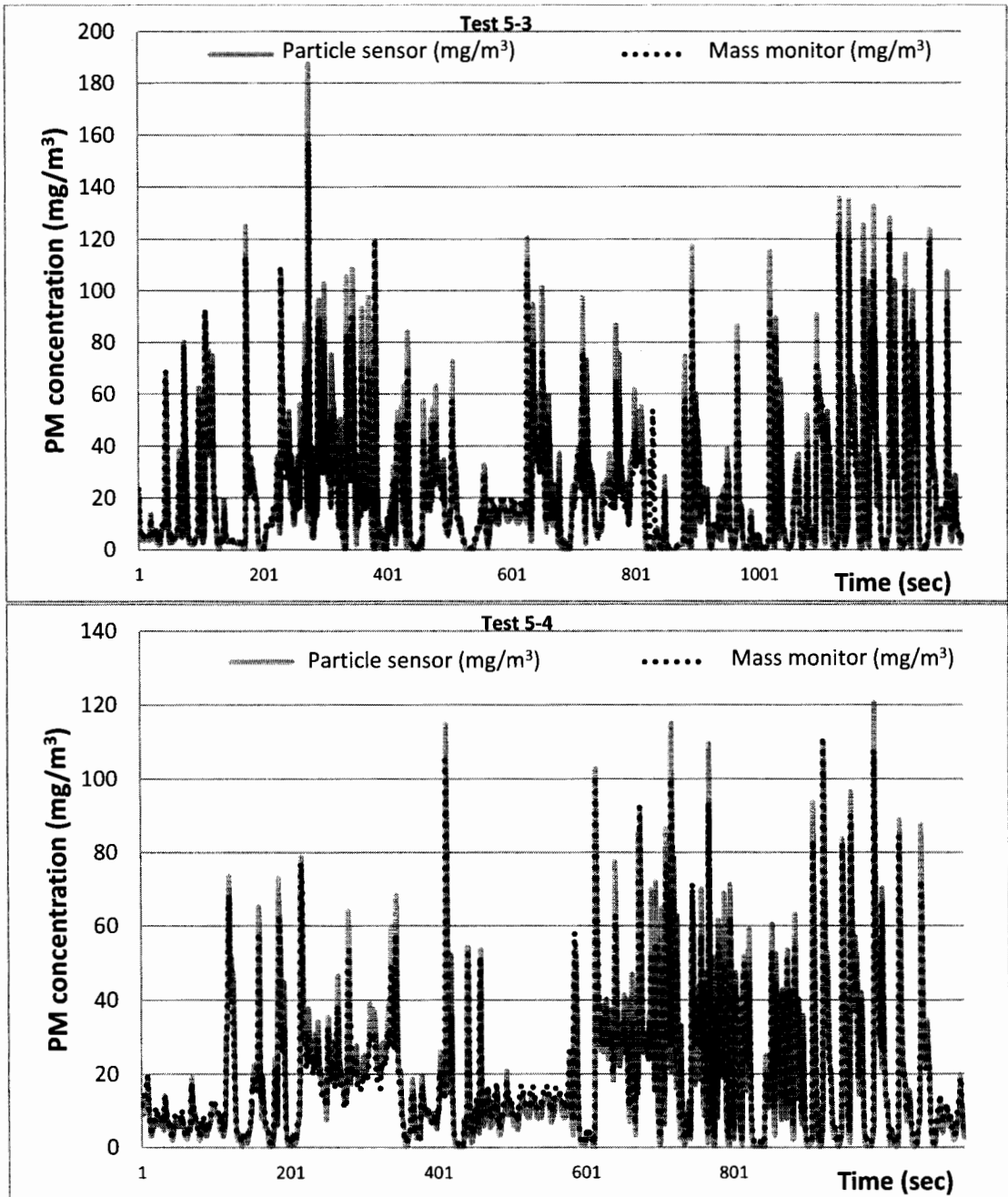


Figure 20. Urban driving tests cycle with the particle sensor measuring diluted exhaust, day 5, tests 3 and 4.

graphs, several peak values from the particle sensor appear to be slightly higher than those from the mass monitor during the first, third, and the fourth test; whereas a number of mass monitor PM peaks from the second test are slightly higher than corresponding particle sensor readings. Overall, the PM measurements from both instruments exhibit similar patterns during all four tests with average percent difference of 38%.

Descriptive Statistics

Summary statistics and frequency histograms of PM mass concentrations measured with each instrument (the particle sensor and the mass monitor) are presented in Appendices D and E. According to calculated descriptive statistics, the majority of the data sets were characterized by a little difference between the mean PM mass concentration and standard deviation suggesting a slight dispersion in emission data. However, as shown in Appendix F, the data was skewed to the right (mean was greater than median with supporting frequency histograms that did not show normal distribution). Histograms depicted frequencies of PM measurement observations occurring in certain ranges of values (i.e., bins). Accordingly, all data sets were log transformed due to lack of normality; further analysis was performed on transformed normalized data. Calculated confidence intervals for PM emission data were relatively narrow, confirming overall accuracy of generated statistics.

Correlation and Regression Analyses

Summary results of regression analysis are presented in Tables 6 and 7. According to these results, positive correlation coefficients of similar strength were found for all of the tests. However, some tests performed better than the others. Specifically,

Table 6. Summary results of regression analysis.

Test	Probe location	Test cycle	Particle sensor sample dilution	Regression statistics summary				Coefficients	
				r	r ²	r ² _{adj}	Standard error	β_1	β_0
1-1	BDPF	snap-idle	Diluted	.997	.995	.987	.107	1.043	0
1-2	BDPF	snap-idle	Diluted	.993	.987	.978	.180	1.028	0
1-3	BDPF	snap-idle	Diluted	.997	.995	.987	.106	1.054	0
Average				.996	.992	.984	.131		
2-1	BDPF	snap-idle	Diluted	.997	.995	.986	.100	0.945	0
2-2	BDPF	snap-idle	Diluted	.996	.992	.985	.117	0.961	0
2-3	BDPF	snap-idle	Diluted	.997	.995	.987	.092	0.952	0
2-4	BDPF	snap-idle	Diluted	.994	.987	.981	.138	0.963	0
Average				.996	.992	.985	.112		
3-1	BDPF	snap-idle	Diluted	.993	.986	.977	.162	0.973	0
3-2	BDPF	snap-idle	Diluted	.989	.978	.971	.191	0.958	0
3-3	BDPF	snap-idle	Diluted	.988	.976	.967	.199	0.954	0
Average				.990	.980	.971	.184		
3-4	BDPF	snap-idle	Raw	.988	.976	.967	.199	0.954	0
3-5	BDPF	snap-idle	Raw	.986	.971	.963	.229	0.927	0
3-6	BDPF	snap-idle	Raw	.974	.948	.942	.269	0.826	0
Average				.982	.965	.957	.232		
4-1	BDPF	snap-idle	Diluted	.994	.987	.979	.141	1.074	0
4-2	BDPF	snap-idle	Diluted	.996	.992	.984	.112	1.087	0
4-3	BDPF	snap-idle	Diluted	.956	.915	.906	.361	1.010	0
Average				.982	.965	.956	.205		
5-1	BDPF	driving	Diluted	.996	.992	.992	.139	0.940	0
5-2	BDPF	driving	Diluted	.971	.942	.941	.311	1.014	0
5-3	BDPF	driving	Diluted	.945	.893	.892	.394	0.917	0
5-4	BDPF	driving	Diluted	.976	.952	.951	.262	0.989	0
Average				.972	.945	.944	.277		
5-5	BDPF	snap-idle	Diluted	.991	.982	.975	.136	1.142	0
5-6	BDPF	snap-idle	Diluted	.987	.975	.967	.162	1.129	0
5-7	BDPF	snap-idle	Diluted	.991	.982	.975	.150	1.169	0
Average				.990	.980	.972	.149		
5-8	ADPF	snap-idle	Diluted	.468	.219	.212	.181	0.272	0
5-9	ADPF	snap-idle	Diluted	.889	.790	.783	.108	0.231	0
5-10	ADPF	snap-idle	Diluted	.651	.423	.416	.151	0.260	0
Average				.669	.478	.471	.147		

Table 7. Results of hypothesis testing analysis.

Test	Probe location	Test cycle	Particle sensor dilution	r	Test statistics			Conclusion
					df	F	Sig. F	
1-1	BDPF	snap-idle	Diluted	.997	129	25328.19	.000	< .05 Reject Ho (significant result)
1-2	BDPF	snap-idle	Diluted	.993	113	8285.58	.000	
1-3	BDPF	snap-idle	Diluted	.997	135	25273.36	.000	
2-1	BDPF	snap-idle	Diluted	.997	111	21838.46	.000	< .05 Reject Ho (significant result)
2-2	BDPF	snap-idle	Diluted	.996	145	17431.72	.000	
2-3	BDPF	snap-idle	Diluted	.997	128	25163.79	.000	
2-4	BDPF	snap-idle	Diluted	.994	151	11587.97	.000	
3-1	BDPF	snap-idle	Diluted	.993	116	7998.99	.000	< .05 Reject Ho (significant result)
3-2	BDPF	snap-idle	Diluted	.989	138	6096.47	.000	
3-3	BDPF	snap-idle	Diluted	.988	110	4379.84	.000	
3-4	BDPF	snap-idle	Raw	.988	110	4379.84	.000	< .05 Reject Ho (significant result)
3-5	BDPF	snap-idle	Raw	.986	114	3834.55	.000	
3-6	BDPF	snap-idle	Raw	.974	170	3094.76	.000	
4-1	BDPF	snap-idle	Diluted	.994	120	9196.35	.000	< .05 Reject Ho (significant result)
4-2	BDPF	snap-idle	Diluted	.996	120	15320.37	.000	
4-3	BDPF	snap-idle	Diluted	.956	120	1279.06	.000	
5-1	BDPF	driving	Diluted	.996	1479	190684.16	.000	< .05 Reject Ho (significant result)
5-2	BDPF	driving	Diluted	.971	1234	20069.14	.000	
5-3	BDPF	driving	Diluted	.945	1307	10899.71	.000	
5-4	BDPF	driving	Diluted	.976	1112	21906.51	.000	
5-5	BDPF	snap-idle	Diluted	.991	140	7651.50	.000	< .05 Reject Ho (significant result)
5-6	BDPF	snap-idle	Diluted	.987	140	5338.94	.000	
5-7	BDPF	snap-idle	Diluted	.991	140	7470.62	.000	
5-8	ADPF	snap-idle	Diluted	.468	140	39.09	.000	< .05 Reject Ho (significant result)
5-9	ADPF	snap-idle	Diluted	.889	140	523.53	.000	
5-10	ADPF	snap-idle	Diluted	.651	140	102.09	.000	

the highest average value of correlation coefficient ($r = .996$) was found for tests performed with the probe location before the DPF under the snap-idle acceleration test cycle (days 1 and 2). This suggests strong positive association between measurements taken with the particle sensor and those taken with the mass monitor for snap-idle tests with PM measurements sampled before the DPF.

The lowest average value of correlation coefficient ($r = .669$) was found for tests performed with the probe location after the DPF during the snap-idle test cycle (day 5). This suggests weaker association between measurements from both instruments for DPF-out snap-idle acceleration tests. Since emissions downstream of the DPF are generally low on properly maintained vehicles, a weaker association between PM measurements from both instruments was anticipated for data of such magnitude.

Somewhat weak association between PM measurements from the mass monitor and the particle sensor for tests conducted after the DPF suggests a notable disparity in measurement ability of the particle sensor compared to the mass monitor when measuring low emission levels. This is consistent with findings from Figure 16 that illustrates a steady increase of PM mass concentrations measured by the particle sensor in response to increase of engine power during engine acceleration events; while emission measurements received from the mass monitor do not portray noticeable increase corresponding to increase of engine power. Consequently, it can be suggested that the particle sensor provides advantage when measuring low emissions since it demonstrated a better ability to detect variations in lower PM levels compared to the mass monitor.

Results of comparison between measurements before the DPF performed by both instruments showed higher correlation with an average value of .991 when both the particle sensor and the mass monitor measured diluted sample, compared to average correlation ($r = .982$) when the particle sensor was measuring raw exhaust while the mass monitor was measuring diluted sample (since raw sampling is not supported by the instrument). This could be due to the presence of volatile and other materials in raw sampling flow that are removed from diluted exhaust during multi-stage sample pre-conditioning. Correlation results of raw sampling tests are consistent with corresponding PM distribution line graphs indicating fairly close transient response from both instruments. Slight increase in the particle sensor's PM levels on the downward slope of PM peaks could be explained by differences in PM composition between raw and diluted exhaust. Specifically, gas-to-particle conversion in raw exhaust is possible as volatile hydrocarbons and sulfuric acid adsorb onto existing particles and nucleate to form new particles.

Tests performed under urban driving test cycle while both instruments were sampling diluted exhaust with probe location before DPF demonstrated slightly lower coefficients of correlation with average value of .972 compared to snap-idle tests with same probe location setup and dilution setting. The highest correlation coefficient ($r = .996$) out of total four driving tests corresponds to the first one under this cycle. This test was performed right after starting the engine (i.e., during the "cold-start"), therefore PM emissions reached highest levels compared to consequent tests. The remaining "hot-start" tests with lower PM levels yielded slightly smaller correlations of up to .976.

Comparison across all tests grouped into three major categories (i.e., urban driving test cycle, before the DPF under snap-idle test cycle, and after the DPF under snap-idle test cycle) yielded correlation coefficients of .976, .986, and .712, respectively. Calculated descriptive statistics across the grouped tests is presented in Appendix G, while results of regression and hypothesis testing analyses are summarized in Appendix H. They confirmed strong and moderate positive association between PM measurements from the particle sensor and the mass monitor for tests sampled before the DPF and after the DPF, respectively.

All calculated correlation coefficients were found statistically significant with calculated significance less than selected significance level of .05. The results of linear regression analysis that showed highest correlation coefficient ($r = .997$), highest coefficient of determination ($r^2 = .995$) and lowest standard error of estimate (SEE = .107) [test 1, day 1] were used to develop the equation of the line that best fits the data and provides an estimate of PM mass concentration. It is presented in Equation 5.

$$Y_{\text{predicted}} = 0.954 \times X \quad (5)$$

where X is the normalized PM mass concentration measurement from the particle sensor, mg/m^3 ; and

$Y_{\text{predicted}}$ is the normalized PM mass concentration measurement from the mass monitor, mg/m^3 .

In this example, 99.5% of the variation in the PM mass concentration measurement from the mass monitor ($Y_{\text{predicted}}$) is explained or accounted for by the

variation in the PM mass concentration measurement from the particle sensor (X). The 95% confidence interval for the estimate is $0.954 \pm (1.96 \times \text{Standard error of estimate})$ which corresponds to a range from 1.168 to 0.740 mg/m³ for a PM measurement of 1 mg/m³. Relatively small standard errors of estimate calculated for all tests confirmed small dispersion of observed values around the regression line.

Underlying assumptions of regression analysis have been tested and found true: predicted values follow normal distribution, the means of these normal distributions lie on the regression line, the standard deviations of these normal distributions are all the same, and predicted values are statistically independent. Overall, found correlation coefficients imply statistically significant relatively strong positive association between PM mass measurements taken with the particle sensor and measurements taken with the mass monitor.

DPF Efficiency Evaluation Study

The DPF efficiency was evaluated based on PM measurements obtained during the same testing day using identical test-setup. Filtration efficiency for each instrument was estimated using three data sets, each consisting of PM values measured upstream and downstream of the filter. Calculation results are summarized in Table 8.

On average, the DPF efficiencies calculated from the particle sensor PM data were slightly lower than those from the mass monitor. Generally, the particle sensor estimated slightly lower DPF efficiency compared to the mass monitor. The absolute difference between results from both instruments ranged from 1.72 to 4.96% with average value of 1.37%.

Table 8. Comparison of DPF efficiencies.

Test	Total PM mass concentration, mg/m ³				DPF efficiency, %		Absolute difference, %
	Mass monitor		Particle sensor		Mass monitor	Particle sensor	
	After DPF	Before DPF	After DPF	Before DPF			
5-8	0.66	15.21	1.00	10.82	95.69	90.73	4.96
5-9	0.64	15.07	0.18	11.10	95.78	98.35	2.57
5-10	0.66	18.15	0.65	12.23	96.37	94.65	1.72
Average					95.95	94.58	1.37

Ultimately, the particle sensor and the mass monitor results were in good agreement with each other with a minor absolute difference. Therefore, the particle sensor can be used to provide a reasonable estimate of the DPF filtration efficiency in order to diagnose the filter operational condition and identify possible malfunctions or need in DPF maintenance. According to the performed calculations, the DPF filtration efficiency on tested vehicle was about 95 and 94%, as measured by mass monitor and particle sensor, respectively. This satisfied the minimum DPF efficiency requirements of 85% established by the EPA and the CARB (MECA, 2009; National Clean Diesel Campaign, 2010a). As a result, it can be concluded that the DPF is in good operational condition.

CHAPTER 5

DISCUSSION, CONCLUSION, AND RECOMMENDATION

Discussion

This study investigated the ability of a newly developed particle sensor to adequately measure PM mass concentration in exhaust gases of modern diesel vehicles in various real-time field settings, including snap-idle acceleration and urban driving conditions. The major findings of this limited study are as follows:

1. Temporal distributions of continuous PM mass concentration measurements from the particle sensor and the reference mass monitor revealed remarkable similarities. The particle sensor readings were found to be of the same order of magnitude as the values from the reference mass monitor.
2. As illustrated by temporal PM graphs and supported by statistical data, tests performed during different test cycles consistently demonstrated ability of the particle sensor to produce sharp transient response to fast-changing PM emission levels.
3. Correlation and regression analyses found a significant strong positive association between measurements taken with particle sensor and those taken with reference mass monitor for tests performed with the probe location before the DPF under various test cycles.
4. Correlation and regression analyses established a significant moderate positive association between measurements from the particle sensor and those from the

mass monitor for tests conducted with diluted sampling after the DPF under the snap-idle test cycle.

5. The study showed that when used during limited emission evaluation on DPF-equipped vehicles, the particle sensor provided advantage in measuring low emission levels since it was able to better respond to variation in lower PM concentrations compared to the mass monitor.
6. According to results of raw emission sampling using the particle sensor, it is capable of operating during raw emission measurements with fast transient response and good agreement with results from the mass monitor.
7. The selected particle sensor is robust enough and suitable to evaluate real-time emissions from modern diesel engines in various real-time field settings, including snap-idle acceleration and urban driving conditions.
8. The study suggests that the particle sensor can be used as a diagnostic tool for evaluation of the DPF operating condition and identify possible malfunctions or need in DPF maintenance.

The study revealed capability of the particle sensor to produce good results during fast-changing transient events. Data showed that transient peaks from both instruments consistently matched each other. Moreover, during sharp increases in PM levels, the observed discrepancy between measurements from both instruments was minimal. The ability to produce suitable second-by-second measurements is important for real-time emission evaluation. As opposed to semi-real-time instruments not capable of providing second-by-second measurements or gravimetric method that can only yield total mass,

the particle sensor not only can measure PM emissions with high second-by-second resolution, but it can also provide statistically similar results to a well-established reference instrument.

Furthermore, results of field-testing showed that the particle sensor provides several advantages over traditional instruments designed for operation only in well-controlled environments, such as laboratory conditions. Specifically, when mounted directly on a vehicle and tested under urban driving conditions, the unit was able to provide satisfactory performance. Results of tests performed under urban driving test cycle were not significantly different from tests performed under more stable conditions. Therefore, the analysis of results suggests that the particle sensor unit is robust enough to be used during field testing and able to withstand reasonable vibrations and temperatures.

As expected, better correlation between measurement results of the particle sensor and the mass monitor was observed for emissions of higher concentration levels. These emissions were measured in the exhaust upstream of the DPF and consisted of particles with bigger diameter compared to particles downstream of the filter. This is because the exhaust gas contains mostly fine particles after passing through a well-functioning DPF, whereas coarse particulates get trapped by the filter. Thus, a stronger correlation established for PM measurements with probe location BDPF, confirms that higher concentrations of larger agglomerates were detected by both PM analyzers more effectively than lower concentrations of finer particles.

Moreover, the particle sensor was found to be more sensitive when measuring low emissions during a limited number of tests after the DPF compared to the mass monitor.

As evidenced by line graphs (Figure 16), the particle sensor detected the majority of emission events confirmed by spikes in engine power; whereas the mass monitor measurements were less defined. Therefore, it can be suggested that the particle sensor is more responsive to fine particles than the mass monitor. This characteristic is especially valuable for emission measurement applications on vehicles equipped with DPFs.

In addition, during raw exhaust sampling, the particle sensor operation was able to handle non-conditioned engine-out high temperature exhaust. Moreover, PM data produced during these tests were in agreement with data from the mass monitor with only slight discrepancies that could be attributed to differences in raw and diluted exhaust composition and measurement principle assumptions used by two instruments. The particle sensor's capability to conduct raw exhaust sampling represents a major advantage to that of the mass monitor since it requires sample pre-conditioning of exhaust in order to remove moisture, lower its temperature and, in case of high emitting vehicles, protect unit from contamination. Besides, raw exhaust sampling can be considered the most favorable method for lower PM emissions measurement due to increased PM levels with absence of dilution. Moreover, equipment setup used during raw emission measurement can be considerably simplified due to elimination of extra equipment used for dilution of exhaust sample flow.

Overall, the findings support the study hypothesis and infer that the particle sensor is suitable for adequate emission measurement due to revealed correlation between PM measurements from the particle sensor compared to the reference instrument.

Detailed examination of observed differences between measured PM values from particle sensor and mass monitor needs to be conducted in the future.

Conclusion and Recommendation

This study provided further evidence that selected PM sensor is robust and suitable to evaluate real-time emissions from modern diesel engines in various real-time field settings, including snap-idle acceleration and urban driving conditions.

Results of the study revealed remarkable similarities of temporal distributions of continuous PM mass concentration measurements from the particle sensor and the reference mass monitor.

The study also identified the ability of the particle sensor to produce sharp transient response to fast-changing PM emission levels under different test cycles.

Statistically significant strong positive association has been found between measurements taken with the particle sensor and those taken with the reference mass monitor for tests performed with the probe location before the DPF under various test cycles.

There are four implications from this study. First, the study found that the particle sensor represents an attractive and practical alternative for field PM measurements. The unit was found robust enough to be used during real-world testing (as opposed to the laboratory conditions) and able to withstand reasonable vibrations and temperatures.

Second, the particle sensor demonstrated the potential of being effective in evaluating raw emissions which offers the advantage of measuring low PM levels due to eliminating the need of further sample dilution and, thus, increasing the instruments'

detection limits. Moreover, this would also eliminate bulky dilution equipment, reduce the costs, and simplify testing setup and operation.

Third, the particle sensor was found to be more sensitive when measuring low emissions (e.g., after the DPF) compared to the mass monitor during a limited number of tests. Therefore, it can be recommended for emission evaluation on modern vehicles. This is especially valuable since these vehicles are required to comply with the increasingly tighter EPA emission standards.

Finally, based on the particle sensor performance during the DPF efficiency evaluation portion of the study, the results suggest that PM sensor can effectively identify DPF filtration efficiencies. As a result, it can help determine the DPF operating condition and identify its possible malfunctions or need in DPF maintenance.

The findings support the study hypothesis that the newly developed particle sensor is able to adequately measure PM mass concentration in exhaust gases of modern diesel vehicles based on identified strong positive correlation between measurements from the particle sensor and the reference instrument.

Still, several limitations of this study should be noted. First, the study was conducted on one engine. Since emission levels can vary considerably between different engines, additional testing needs to examine whether measurements from the particle sensor will vary depending on engine characteristics (e.g., model and year).

Second, the comparison was performed to only one reference instrument. It is recommended to further investigate the particle sensor performance in comparison to other real-time instruments. Also a direct comparison to a standard filter-based

gravimetric method would benefit the results. Third, additional tests under different cycles, such as transient and constant speed tests on vehicle dynamometer would provide further information on instrument performance during tests with pre-determined and controlled speed and load. Fourth, further study of raw exhaust sampling using the particle sensor is needed to compare measurements before and after the DPF without sample dilution.

Ultimately, since the current study is based on a limited number of tests, it is recommended to conduct additional investigation to confirm the study results. Detailed examination of the observed difference between PM measurement values from both instruments across different testing days is also recommended.

In summary, additional study is needed to confirm identified trends and further investigate the particle sensor's performance in comparison to other real-time instruments, to make direct comparison to a standard filter-based gravimetric method, and to test its performance using different engines under additional testing cycles.

The results of this study could be useful to researchers and governmental agencies involved in regulation, control, and monitoring of diesel engine emissions. Specifically, findings of this research provide an insight on usability of the novel sensor to adequately measure PM emissions of mobile diesel engines in real-time field conditions. This addresses the need in reliable real-time sensors to monitor lower PM levels in order to comply with new stringent diesel emission regulations. Additionally, adequate measurements of PM emissions on vehicles equipped with exhaust after-treatment devices, such as DPFs, is useful to confirm proper operation of the engine and the DPF,

as well as diagnose any operational problems. The results of the study may serve as the basis for establishment of future recommendations for engine emission measurement.

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APPENDIX A
MEASURED AMBIENT ENVIRONMENTAL PARAMETERS

Test day	Temperature (F)	Humidity (%)	Pressure (inHg)
1	75	56	29.8
2	75	56	29.9
3	69	55	29.9
4	72	37	30.1
5	70	31	30.2

APPENDIX B
MASS MONITOR SPECIFICATION CHARACTERISTICS

Parameter	Specification
Particle size range	0-1.5 μm
Number of stages	6 impactor stages + one mobility channel
Volumetric flow rate	10 lpm (nominal)
Lowest stage pressure	100 mbars absolute pressure
Operation temperature	15-30 $^{\circ}\text{C}$
Operation humidity	0-70 % R.H., non-condensing
Response time	< 5 seconds with 400, 000fA measurement range ~ 15 seconds with 10, 000fA measurement range
Concentration	From 1 to 1,000 $\mu\text{g}/\text{m}^3$ (momentarily up to 5000 $\mu\text{g}/\text{m}^3$)
Unit dimensions	Main unit (installable to a 19 in. rack, 6 U high): W x L x H 490 x 400 x 270 mm Outer cabinet (accessory): W x L x H 550 x 400 x 300 mm
Unit weight	40 kg
Inlet	G 3/8 in. thread, 12 mm male pipe connector
Outlet	G 3/8 in. thread, NW-16-connector
Required accessories	Vacuum pump Computer Sampling system
Computer requirements	Pentium III processor, 128 MB RAM, MS-WINDOWS2000 or XP
Pump requirements	7 m^3/h at 100 mbar absolute pressure

APPENDIX C
DILUTION SYSTEM SPECIFICATION CHARACTERISTICS

Parameter	Specification
Dilution ratio	~ 100 with low dilution ratio ~ 1000 with high dilution ratio
Dilution ratio accuracy	± 5% typical, ± 10% maximum
Evaporation efficiency	> 99% for > 30 nm tetracontane particles, with inlet concentration above 10,000 #/cm ³
Sample flow rate	~ 7 slpm
Outlet flow rate	0-60 slpm (depending on the measurement instruments)
Exhaust flow rate	60-120 slpm (depending on the measurement instruments)
Operation conditions	5-40 °C
Ambient humidity	0-90%, non-condensing
Dilution air specifications:	
- Particle concentration	< 0.1 #/cm ³
- Relative humidity	Non-condensing at -20 °C
- Minimum intake pressure	5 bar absolute
- Maximum intake pressure	9 bar absolute
- Flow rate in normal configuration	120 slpm
- Flow rate with pre-DPF sampler	160 slpm

APPENDIX D
 DESCRIPTIVE STATISTICAL SUMMARY OF PM MASS MEASUREMENTS
 FROM PARTICLE SENSOR AND MASS MONITOR

Table D1. Descriptive statistics, day 1.

Descriptive statistics	Snap acceleration test cycle before the DPF					
	Sample dilution: mass monitor and particle sensor					
	Test 1-1		Test 1-2		Test 1-3	
	Particle sensor	Mass monitor	Particle sensor	Mass monitor	Particle sensor	Mass monitor
Mean	31.18	35.46	42.52	46.18	36.70	42.60
Standard error	3.24	3.47	4.11	4.06	3.91	4.23
Median	15.81	18.31	19.90	23.26	15.88	17.82
Mode	—	1.55	—	22.10	—	15.67
Standard deviation	40.89	43.69	43.70	43.14	45.48	49.11
Sample variance	1672.11	1909.07	1909.62	1861.01	2068.54	2411.68
Kurtosis	2.36	2.30	1.06	0.88	1.86	0.89
Skewness	1.84	1.79	1.52	1.43	1.75	1.52
Range	170.56	186.44	163.72	164.76	173.21	171.27
Minimum	-2.65	0.72	0.23	3.54	0.62	1.97
Maximum	167.91	187.15	163.95	168.30	173.83	173.25
Sum	4958.23	5638.60	4805.14	5217.90	4954.14	5751.43
Count	159.00	159.00	113.00	113.00	135.00	135.00
Confidence level (95%)	6.41	6.84	8.15	8.04	7.74	8.36

Table D2. Descriptive statistics, day 2.

Descriptive statistics	Snap acceleration test cycle before the DPF							
	Snap acceleration test cycle before the DPF							
	Test 2-1		Test 2-2		Test 2-3		Test 2-4	
	Particle sensor	Mass monitor	Particle sensor	Mass monitor	Particle sensor	Mass monitor	Particle sensor	Mass monitor
Mean	29.92	34.29	33.55	28.57	35.33	29.14	31.76	27.30
Standard error	3.21	3.12	3.53	2.79	3.78	2.94	3.70	2.97
Median	12.25	16.27	10.88	10.07	10.64	9.95	9.51	8.74
Mode	—	32.59	—	5.39	—	65.93	—	13.97
Standard deviation	40.77	32.85	42.51	33.61	42.73	33.31	45.47	36.53
Sample variance	1662.39	1079.18	1806.99	1129.80	1826.02	1109.57	2067.41	1334.14
Kurtosis	1.77	0.15	1.55	0.92	1.25	0.88	2.17	1.41
Skewness	1.68	1.21	1.66	1.50	1.57	1.47	1.82	1.64
Range	169.42	120.41	159.33	115.75	149.17	121.01	172.85	134.54
Minimum	-0.18	4.53	159.33	5.27	5.09	4.96	0.50	1.22
Maximum	169.24	124.94	165.41	121.03	154.25	125.97	173.35	135.75
Sum	4817.70	3806.37	4865.10	4142.21	4521.84	3729.32	4795.56	4122.60
Count	161.00	111.00	145.00	145.00	128.00	128.00	151.00	151.00
Confidence level (95%)	6.35	6.18	6.98	5.52	7.47	5.83	7.31	5.87

Table D3. Descriptive statistics, day 3, tests from 1 to 3.

Descriptive statistics	Snap acceleration test cycle					
	Sample dilution: mass monitor and particle sensor					
	Test 3-1		Test 3-2		Test 3-3	
	Particle sensor	Mass monitor	Particle sensor	Mass monitor	Particle sensor	Mass monitor
Mean	40.99	36.53	35.59	28.98	36.94	30.02
Standard error	5.38	4.51	4.77	3.39	5.54	3.94
Median	13.95	12.56	12.71	11.99	11.11	10.73
Mode	—	114.74	—	11.00	—	—
Standard deviation	57.91	48.58	55.99	39.83	58.12	41.30
Sample variance	3353.70	2360.00	3134.99	1586.78	3377.86	1705.67
Kurtosis	4.53	2.46	7.00	5.25	5.62	2.69
Skewness	2.21	1.86	2.71	2.38	2.49	1.94
Range	261.62	201.73	268.45	203.80	252.70	165.49
Minimum	2.87	4.27	0.78	1.59	1.75	3.33
Maximum	264.49	206.00	269.23	205.38	254.45	168.82
Sum	4754.59	4237.25	4912.03	3998.82	4063.54	3301.69
Count	116.00	116.00	138.00	138.00	110.00	110.00
Confidence level (95%)	10.65	8.93	9.42	6.71	10.98	7.80

Table D4. Descriptive statistics, day 3, tests from 4 to 6.

Descriptive statistics	Snap acceleration test cycle					
	Dilution: mass monitor only (particle sensor without dilution)					
	Test 3-4		Test 3-5		Test 3-6	
	Particle sensor	Mass monitor	Particle sensor	Mass monitor	Particle sensor	Mass monitor
Mean	34.11	28.74	37.99	36.00	25.67	23.66
Standard error	3.67	3.88	4.05	4.68	3.18	3.46
Median	15.52	10.33	16.97	12.60	12.81	7.14
Mode	—	0.06	—	87.39	—	14.64
Standard deviation	41.72	44.11	43.23	49.97	41.52	45.15
Sample variance	1740.91	1945.68	1869.00	2497.49	1724.10	2038.95
Kurtosis	4.11	3.83	4.13	3.51	7.16	6.44
Skewness	2.08	2.23	2.07	2.09	2.58	2.66
Range	199.65	172.17	192.76	199.14	203.47	212.21
Minimum	0.01	0.04	5.59	4.07	0.01	0.07
Maximum	199.66	172.22	198.34	203.22	203.48	212.28
Sum	4400.14	3707.15	4330.68	4103.99	4364.68	4022.99
Count	129.00	129.00	114.00	114.00	170.00	170.00
Confidence level (95%)	7.27	7.68	8.02	9.27	6.29	6.84

Table D5. Descriptive statistics, day 4.

Descriptive statistics	Snap acceleration test cycle before the DPF					
	Sample dilution: mass monitor and particle sensor					
	Test 4-1		Test 4-2		Test 4-3	
	Particle sensor	Mass monitor	Particle sensor	Mass monitor	Particle sensor	Mass monitor
	Mean	19.25	24.31	19.29	24.49	18.33
Standard error	1.96	2.40	1.96	2.33	2.02	2.27
Median	10.47	11.91	10.40	11.32	9.92	10.90
Mode	—	4.69	—	11.13	—	10.83
Standard deviation	21.51	26.29	21.52	25.54	22.17	24.83
Sample variance	462.82	691.01	462.95	652.21	491.37	616.32
Kurtosis	2.20	1.22	3.28	1.98	3.10	1.26
Skewness	1.80	1.55	2.04	1.74	2.02	1.60
Range	82.69	97.27	97.48	109.61	93.00	94.56
Minimum	4.30	4.52	3.27	6.26	0.00	0.63
Maximum	86.99	101.79	100.76	115.88	93.00	95.19
Sum	2309.41	2917.33	2314.58	2939.06	2199.51	2773.61
Count	120.00	120.00	120.00	120.00	120.00	120.00
Confidence level (95%)	3.89	4.75	3.89	4.62	4.01	4.49

Table D6. Descriptive statistics, day 5, tests from 1 to 4.

Descriptive statistics	Urban driving test cycle before the DPF							
	Sample dilution: mass monitor and particle sensor							
	Test 5-1		Test 5-2		Test 5-3		Test 5-4	
	Particle sensor	Mass monitor	Particle sensor	Mass monitor	Particle sensor	Mass monitor	Particle sensor	Mass monitor
Mean	69.16	58.39	24.14	27.77	22.44	21.87	19.54	19.88
Standard error	1.68	1.61	0.76	0.81	0.71	0.63	0.60	0.56
Median	49.40	37.39	12.83	17.09	13.02	14.62	12.32	14.21
Mode	76.41	107.33	0.00	37.33	0.00	6.88	0.32	13.49
Standard deviation	64.57	61.81	26.98	28.59	25.76	22.75	20.08	18.52
Sample variance	4169.67	3820.94	727.91	817.25	663.81	517.56	403.31	342.99
Kurtosis	0.33	1.03	3.55	3.84	4.08	3.58	3.20	3.68
Skewness	1.10	1.41	1.70	1.71	1.84	1.75	1.68	1.75
Range	272.44	249.36	167.24	195.41	187.39	156.78	120.52	110.93
Minimum	0.42	0.52	0.00	0.29	0.00	0.61	0.00	0.51
Maximum	272.86	249.88	167.24	195.70	187.38	157.38	120.52	111.45
Sum	102287.86	86354.79	30223.23	34773.65	29536.95	28777.99	21744.34	22121.85
Count	1479.00	1479.00	1252.00	1252.00	1316.00	1316.00	1113.00	1113.00
Confidence level (95%)	3.29	3.15	1.50	1.59	1.39	1.23	1.18	1.09

Table D7. Descriptive statistics, day 5, tests from 5 to 7.

Descriptive statistics	Snap acceleration test cycle before the DPF					
	Sample dilution: mass monitor and particle sensor					
	Test 5-5		Test 5-6		Test 5-7	
	Particle sensor	Mass monitor	Particle sensor	Mass monitor	Particle sensor	Mass monitor
Mean	10.82	15.21	11.10	15.07	12.29	18.24
Standard error	1.38	1.83	1.43	1.82	1.53	2.05
Median	4.56	6.45	4.34	6.23	5.17	8.21
Mode	—	6.45	2.17	3.61	—	6.30
Standard deviation	16.33	21.71	16.96	21.57	18.10	24.21
Sample variance	266.53	471.36	287.52	465.07	327.56	585.93
Kurtosis	7.70	4.27	7.39	4.71	8.62	4.44
Skewness	2.85	2.33	2.80	2.39	2.95	2.32
Range	77.69	89.30	77.27	91.62	85.38	100.95
Minimum	2.24	2.80	2.16	2.82	1.19	2.67
Maximum	79.93	92.09	79.43	94.44	86.58	103.62
Sum	1514.23	2129.68	1554.58	2109.67	1720.91	2553.50
Count	140.00	140.00	140.00	140.00	140.00	140.00
Confidence level (95%)	2.73	3.63	2.83	3.60	3.02	4.04

Table D8. Descriptive statistics, day 5, tests from 8 to 10.

Descriptive statistics	Snap acceleration test cycle after the DPF					
	Sample dilution: mass monitor and particle sensor					
	Test 5-8		Test 5-9		Test 5-10	
	Particle sensor	Mass monitor	Particle sensor	Mass monitor	Particle sensor	Mass monitor
Mean	1.00	0.66	0.18	0.64	0.74	0.66
Standard error	0.07	0.01	0.01	0.01	0.06	0.01
Median	0.65	0.68	0.12	0.68	0.46	0.68
Mode	—	0.66	0.07	0.44	1.41	0.72
Standard deviation	0.88	0.11	0.16	0.15	0.73	0.09
Sample variance	0.77	0.01	0.02	0.02	0.53	0.01
Kurtosis	4.99	0.33	2.09	0.73	3.66	1.42
Skewness	2.07	-0.48	1.77	0.32	1.90	-1.37
Range	4.44	0.53	0.62	0.78	3.85	0.41
Minimum	0.18	0.42	0.05	0.41	0.06	0.42
Maximum	4.62	0.96	0.66	1.18	3.91	0.83
Sum	140.36	91.77	25.62	88.98	103.59	92.30
Count	140.00	140.00	140.00	140.00	140.00	140.00
Confidence level (95%)	0.15	0.02	0.03	0.03	0.12	0.02

APPENDIX E
 FREQUENCY HISTOGRAMS
 FOR PARTICLE SENSOR AND MASS MONITOR
 PM MEASUREMENT DATA SETS

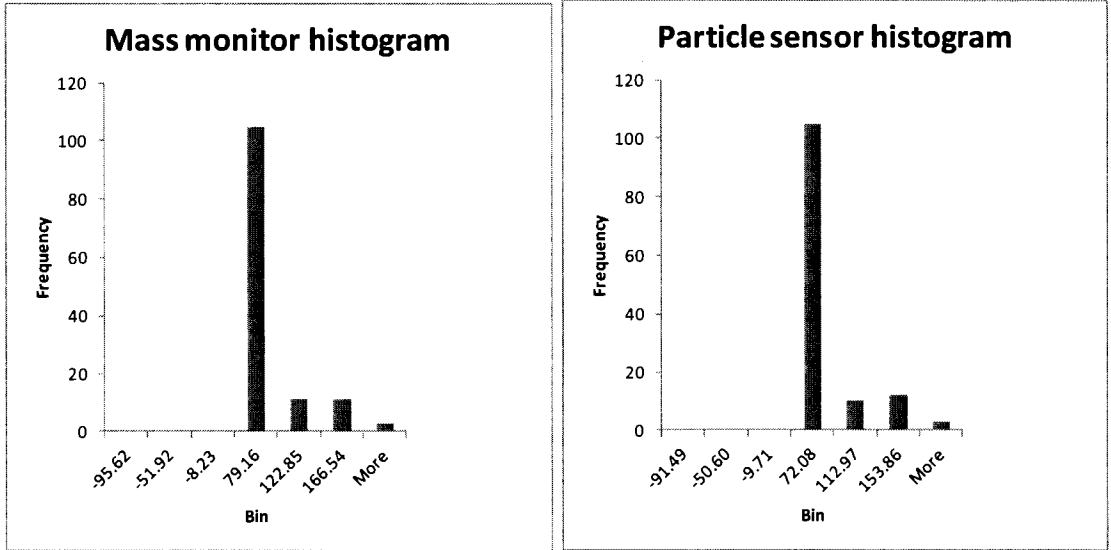


Figure E1. Frequency histograms of measured PM concentrations, day 1, test 1.

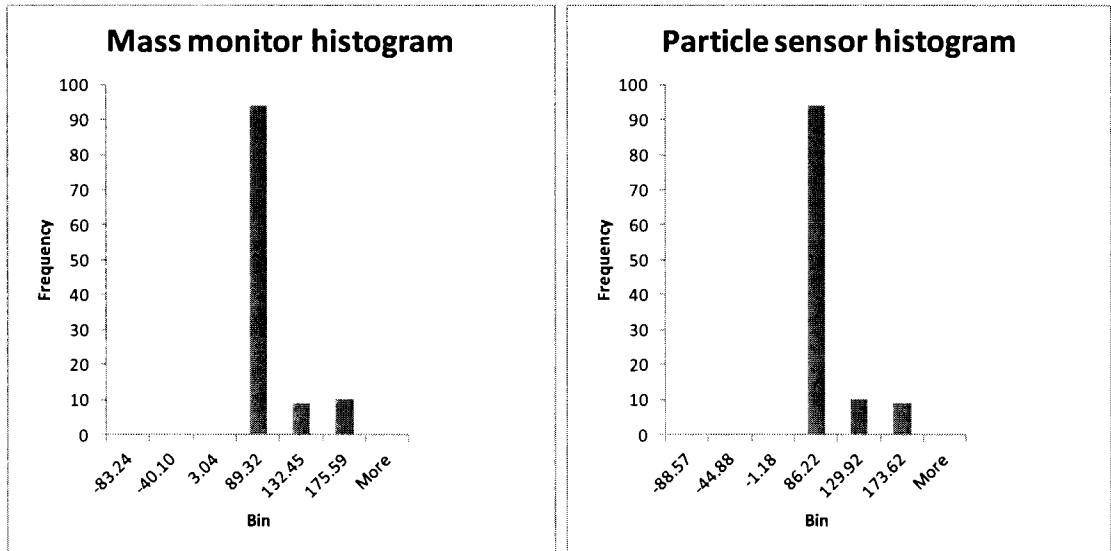


Figure E2. Frequency histograms of measured PM concentrations, day 1, test 2.

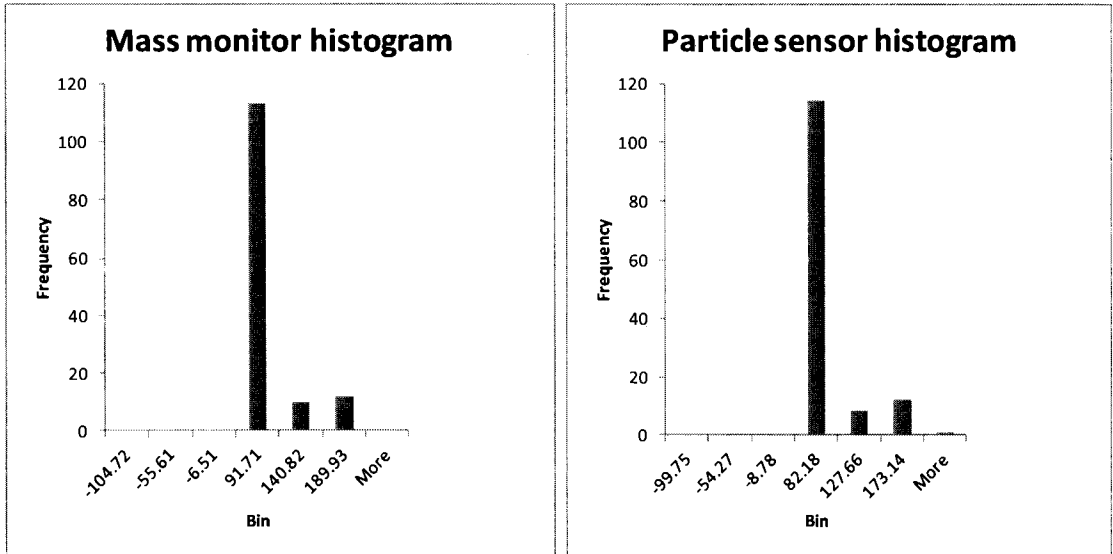


Figure E3. Frequency histograms of measured PM concentrations, day 1, test 3.

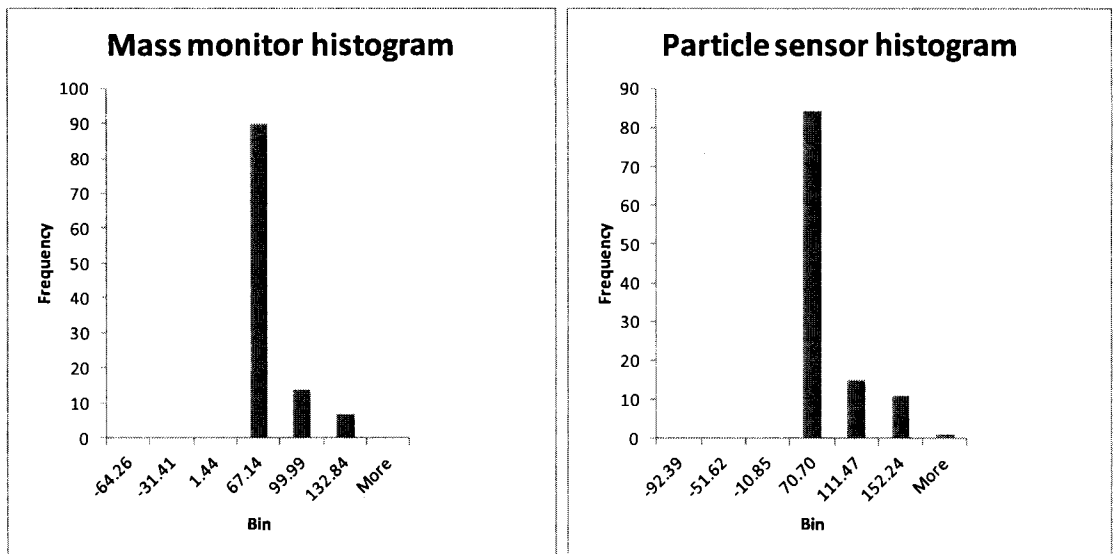


Figure E4. Frequency histograms of measured PM concentrations, day 2, test 1.

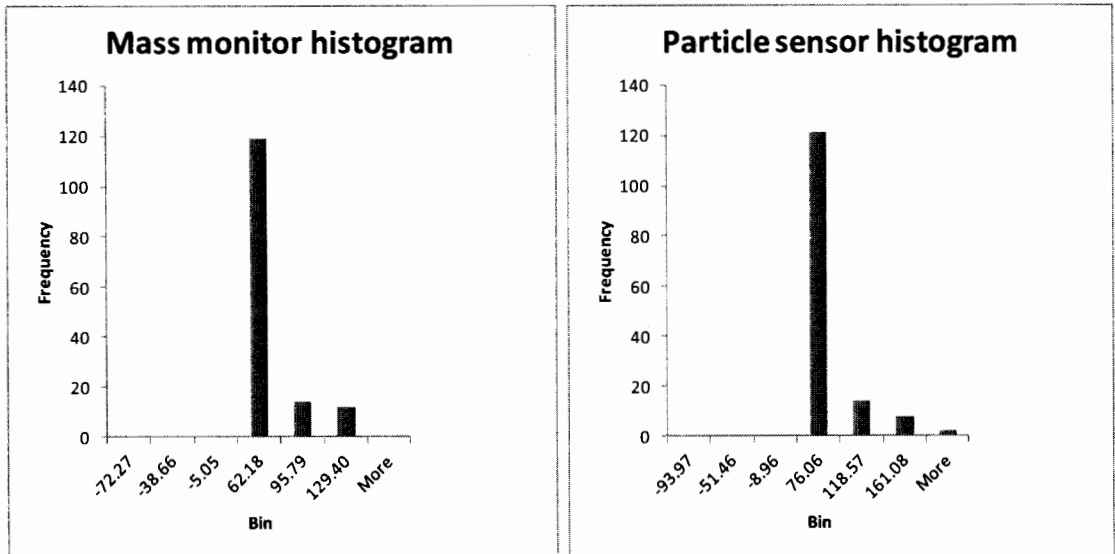


Figure E5. Frequency histograms of measured PM concentrations, day 2, test 2.

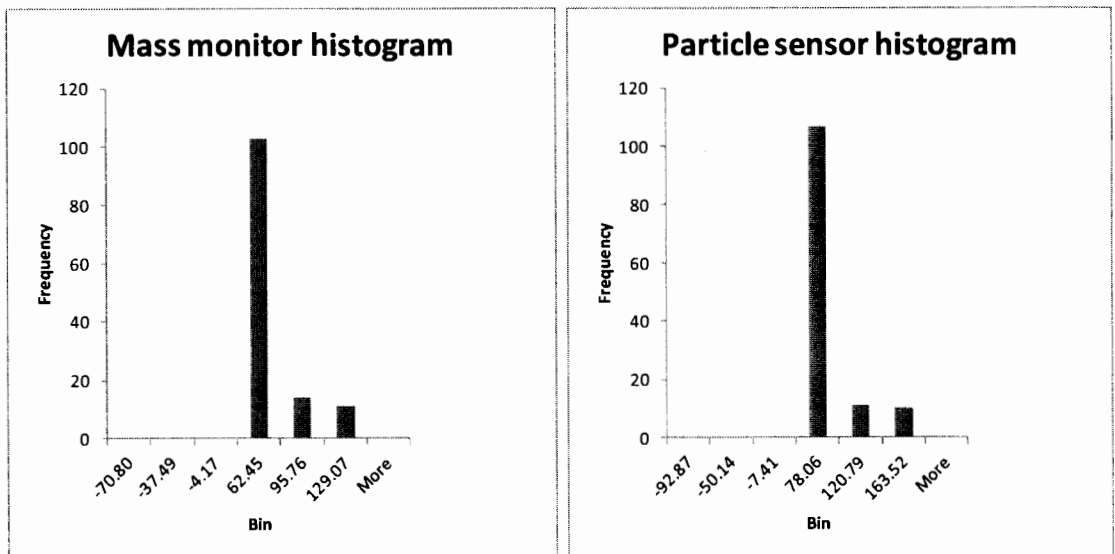


Figure E6. Frequency histograms of measured PM concentrations, day 2, test 3.

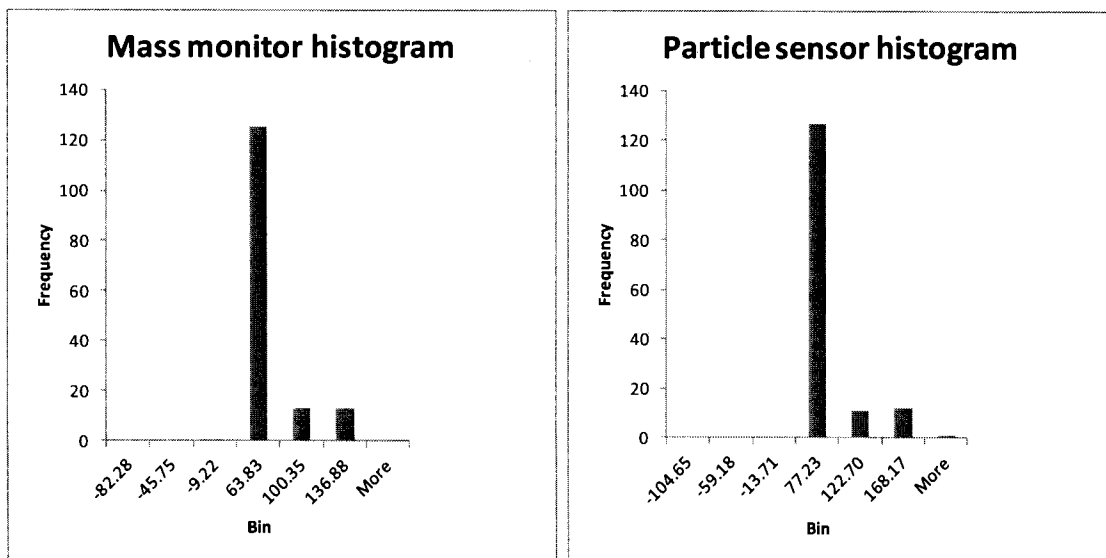


Figure E7. Frequency histograms of measured PM concentrations, day 2, test 4.

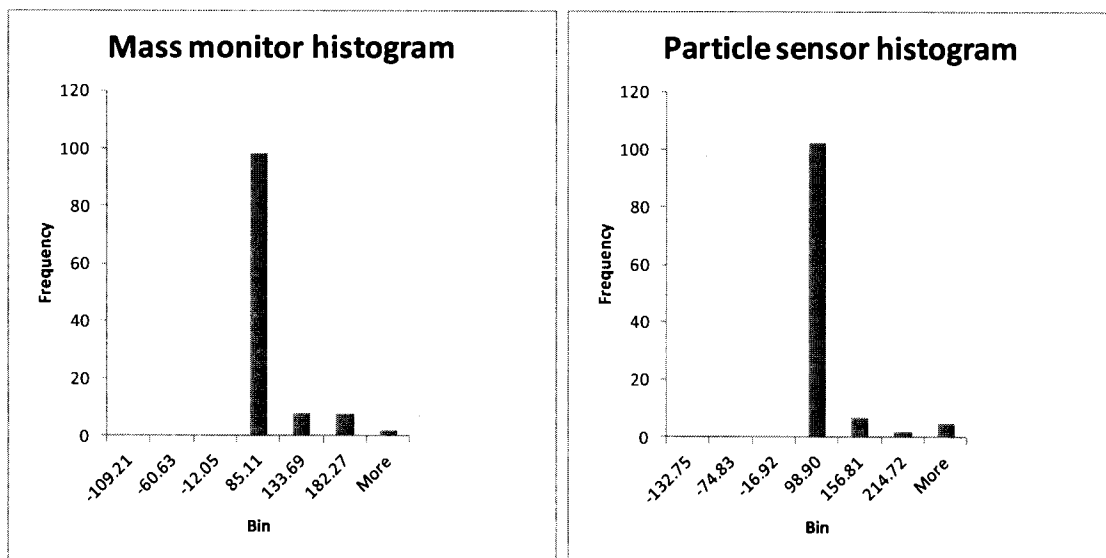


Figure E8. Frequency histograms of measured PM concentrations, day 3, test 1.

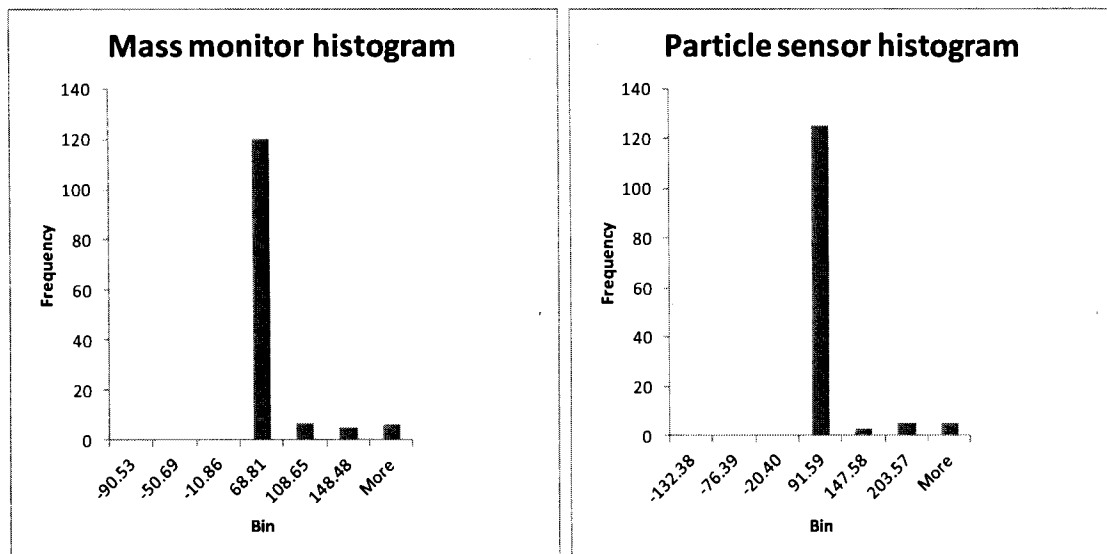


Figure E9. Frequency histograms of measured PM concentrations, day 3, test 2.

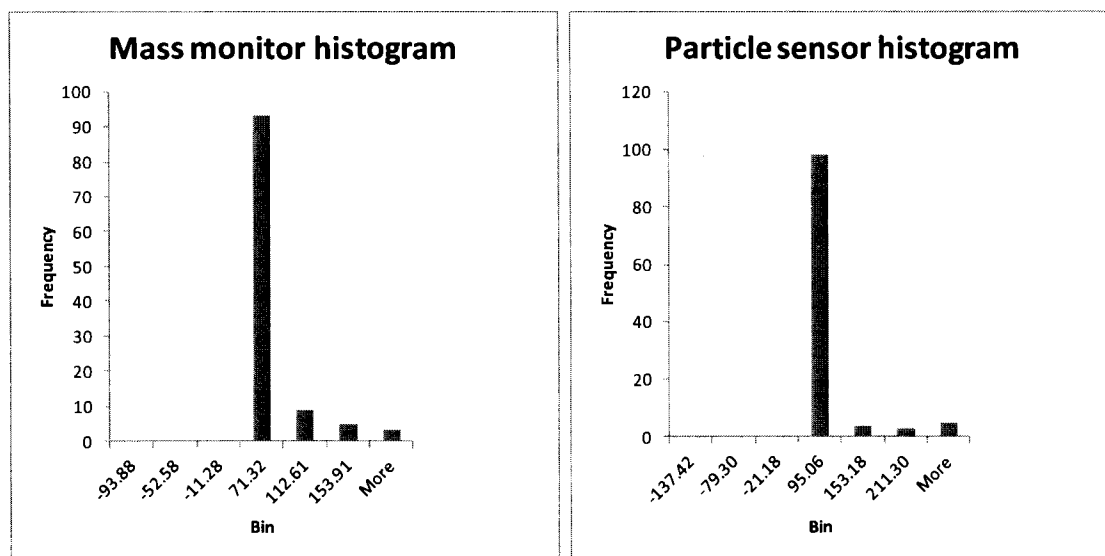


Figure E10. Frequency histograms of measured PM concentrations, day 3, test 3.

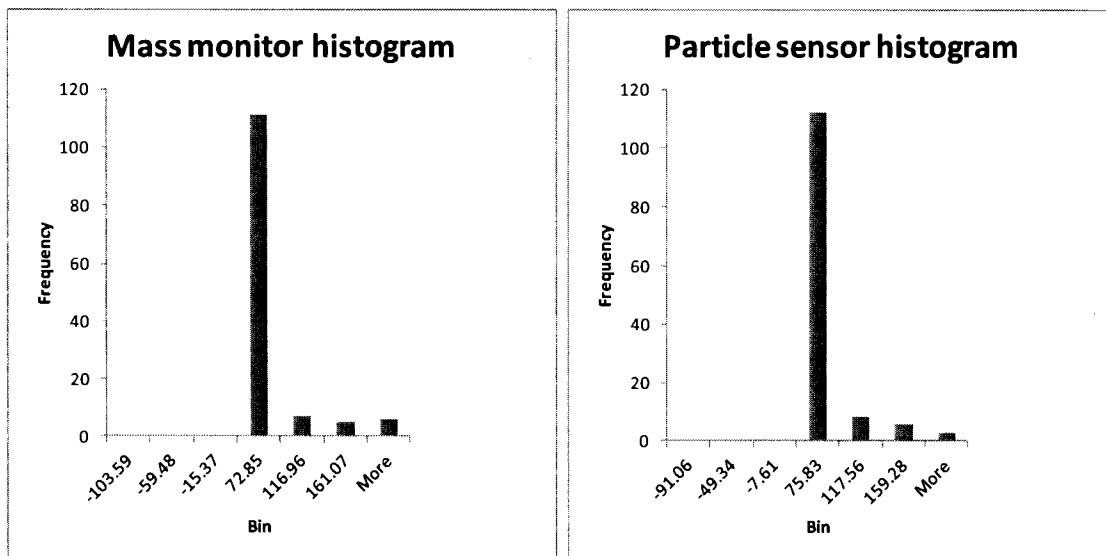


Figure E11. Frequency histograms of measured PM concentrations, day 3, test 4.

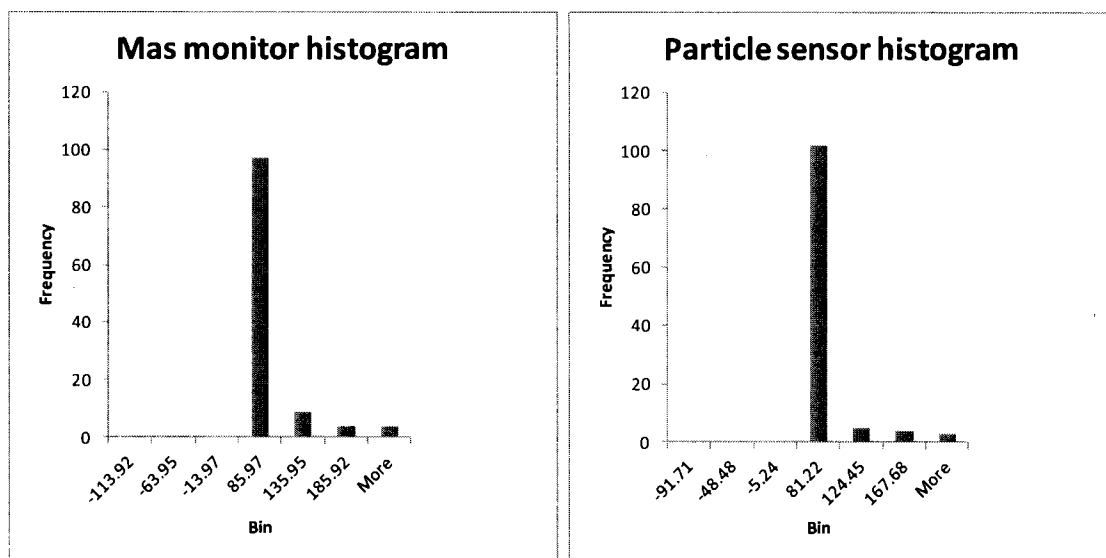


Figure E12. Frequency histograms of measured PM concentrations, day 3, test 5.

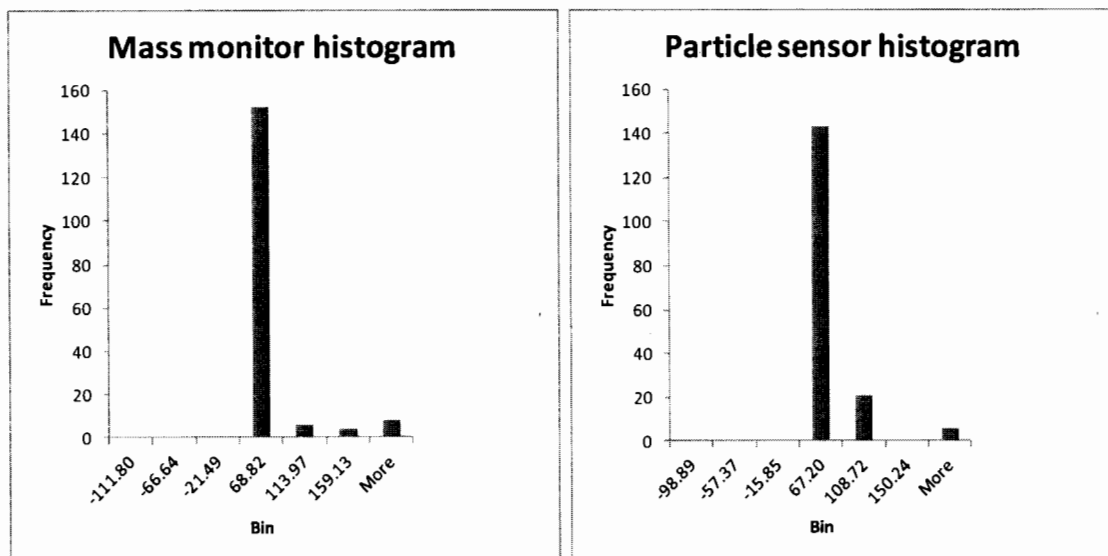


Figure E13. Frequency histograms of measured PM concentrations, day 3, test 6.

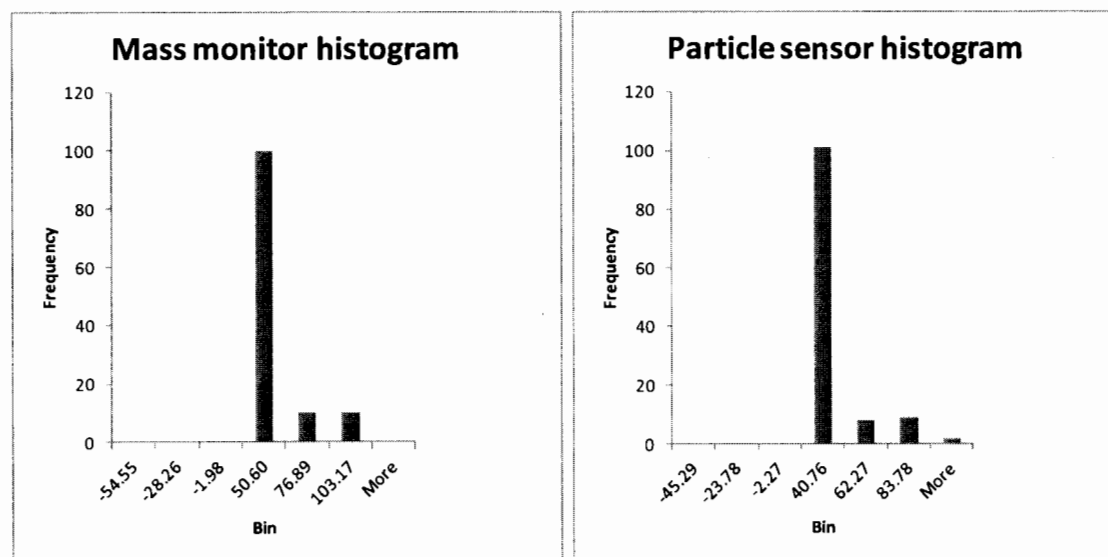


Figure E14. Frequency histograms of measured PM concentrations, day 4, test 1.

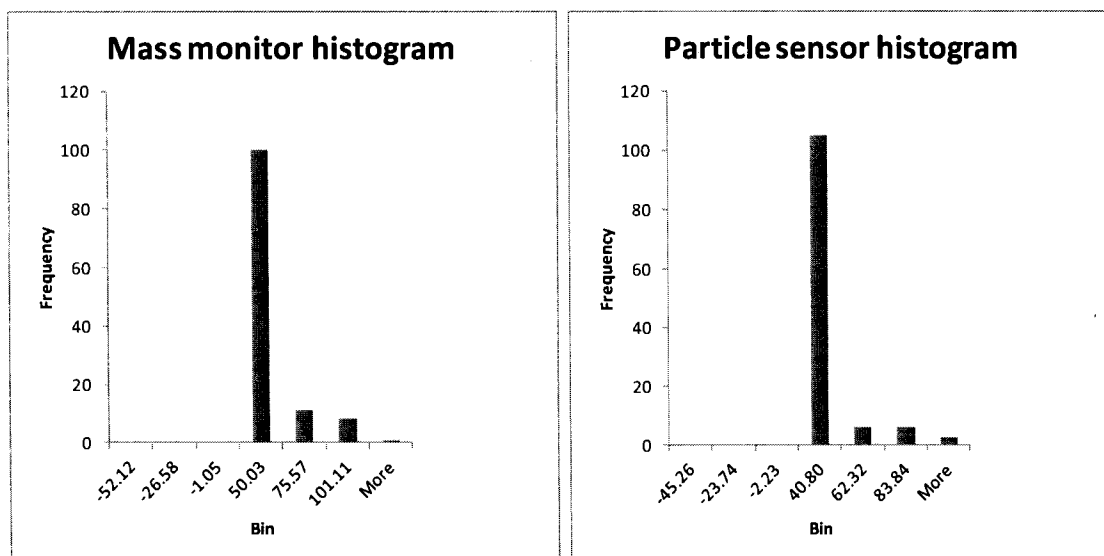


Figure E15. Frequency histograms of measured PM concentrations, day 4, test 2.

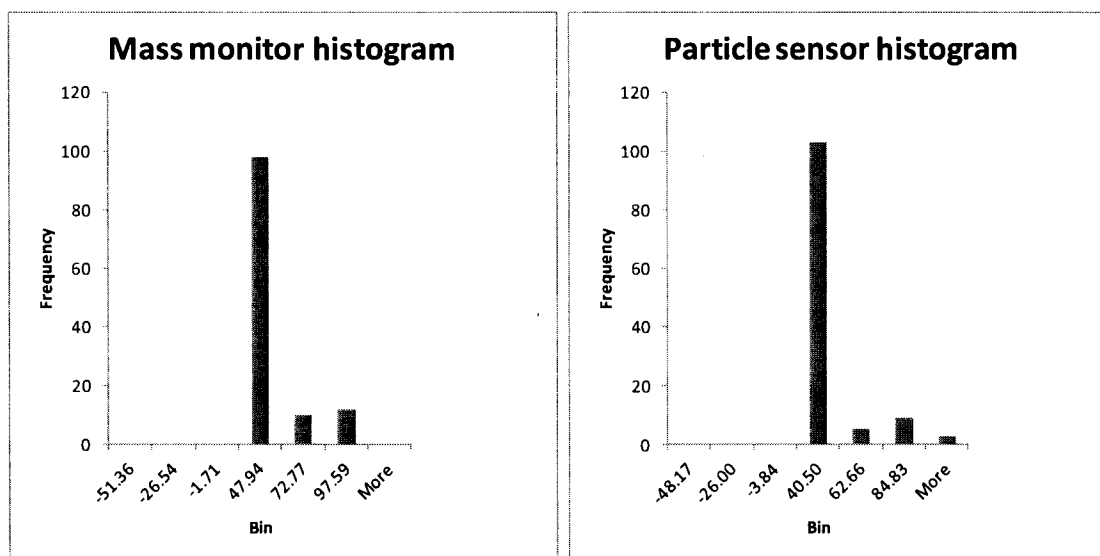


Figure E16. Frequency histograms of measured PM concentrations, day 4, test 3.

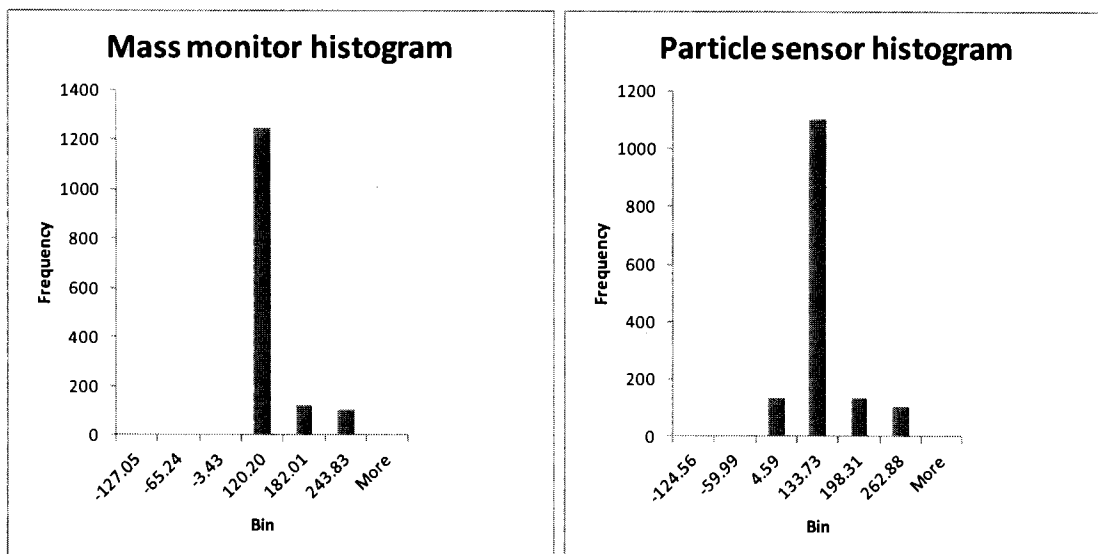


Figure E17. Frequency histograms of measured PM concentrations, day 5, test 1.

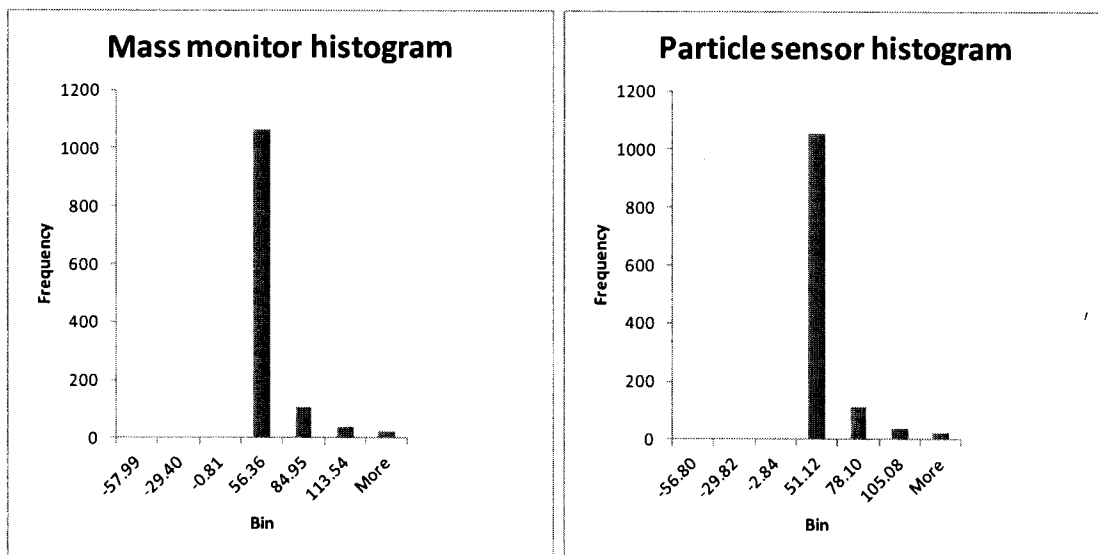


Figure E18. Frequency histograms of measured PM concentrations, day 5, test 2.

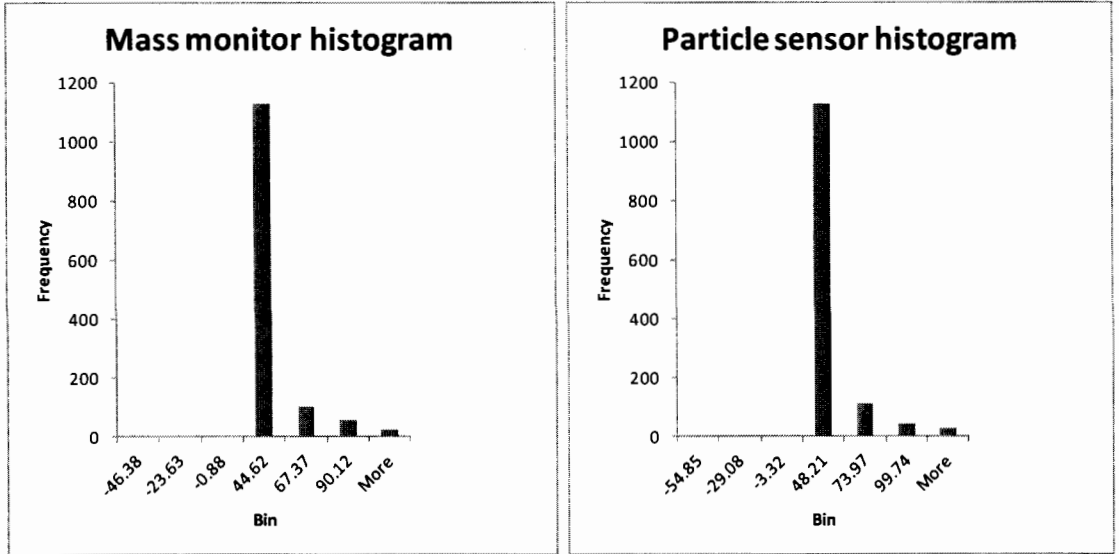


Figure E19. Frequency histograms of measured PM concentrations, day 5, test 3.

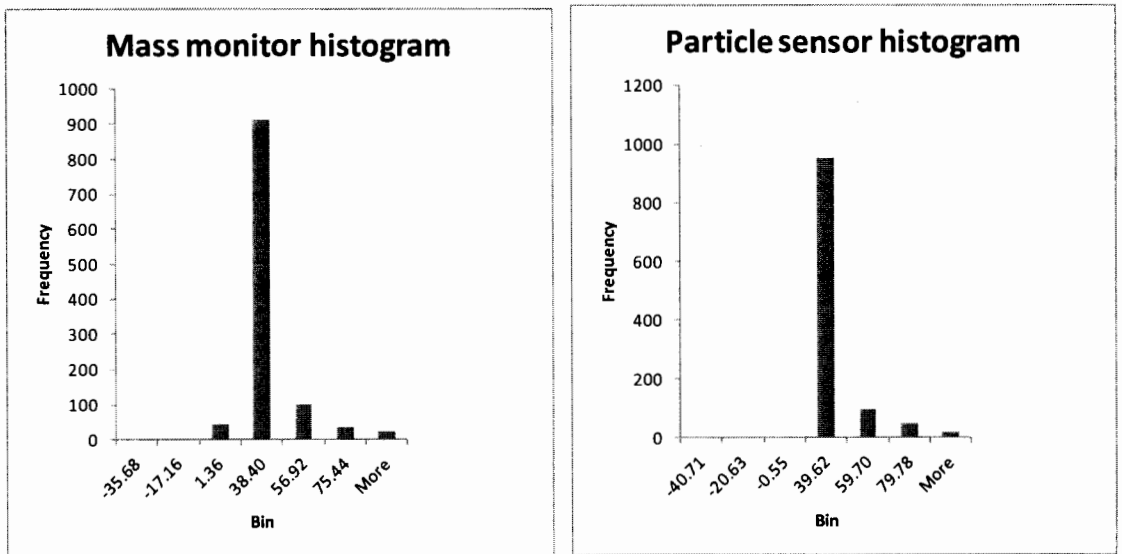


Figure E20. Frequency histograms of measured PM concentrations, day 5, test 4.

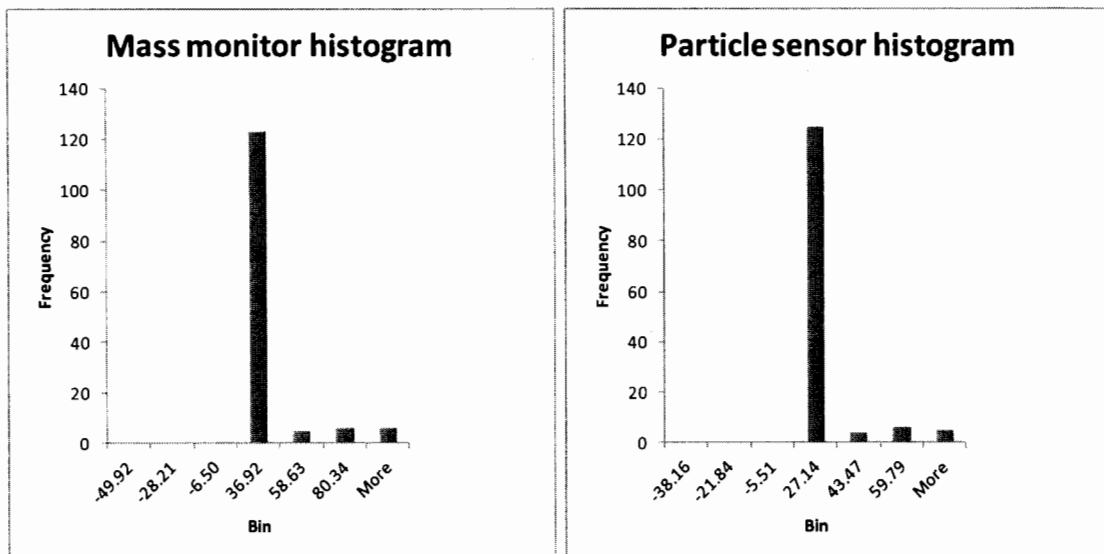


Figure E21. Frequency histograms of measured PM concentrations, day 5, test 5.

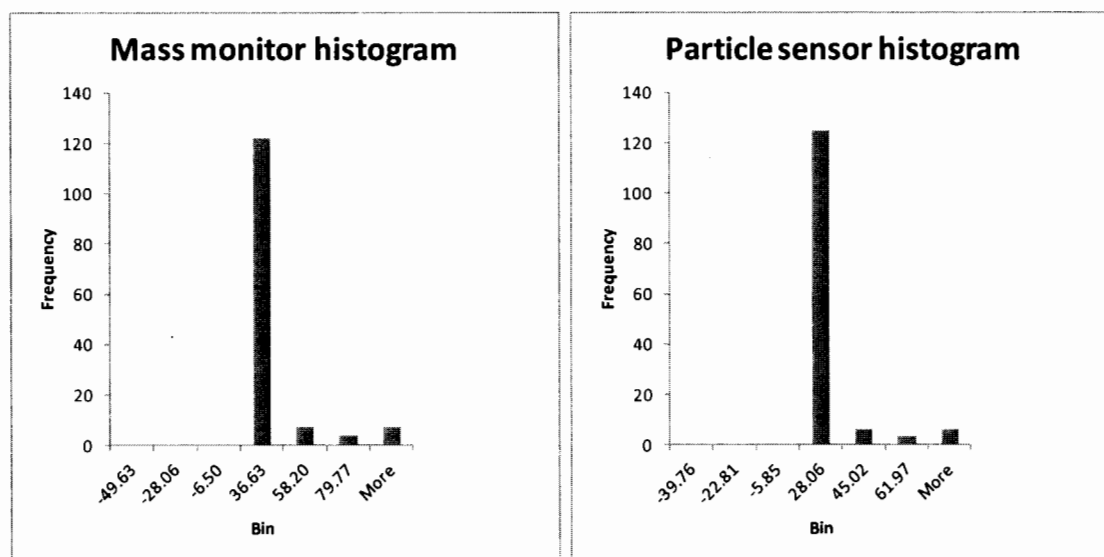


Figure E22. Frequency histograms of measured PM concentrations, day 5, test 6.

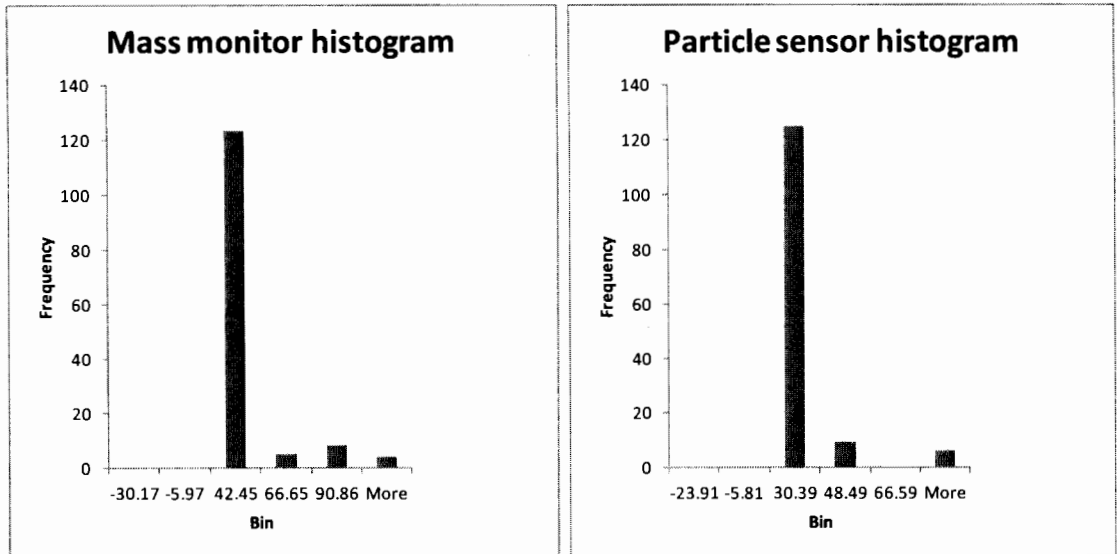


Figure E23. Frequency histograms of measured PM concentrations, day 5, test 7.

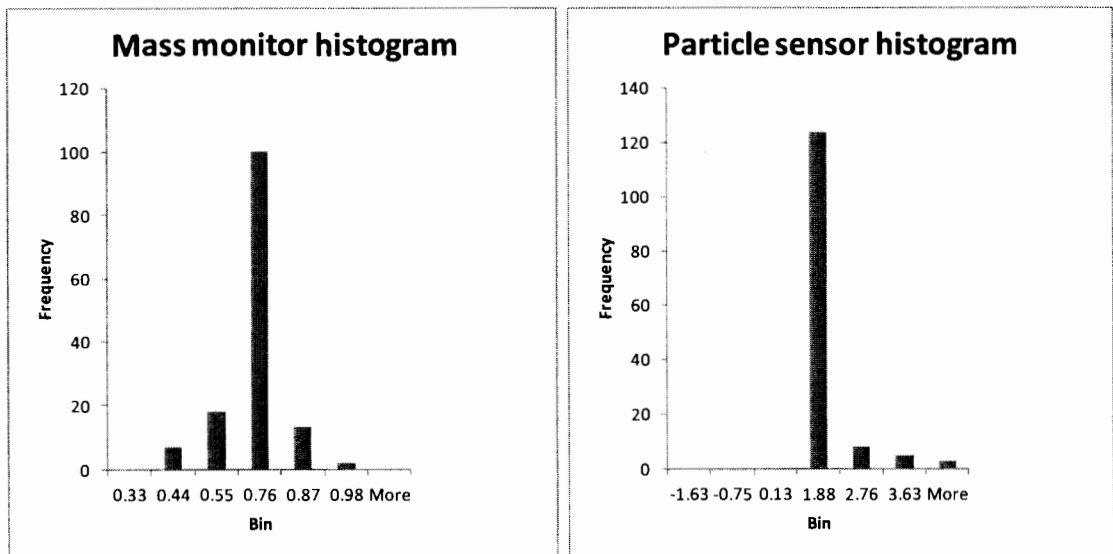


Figure E24. Frequency histograms of measured PM concentrations, day 5, test 8.

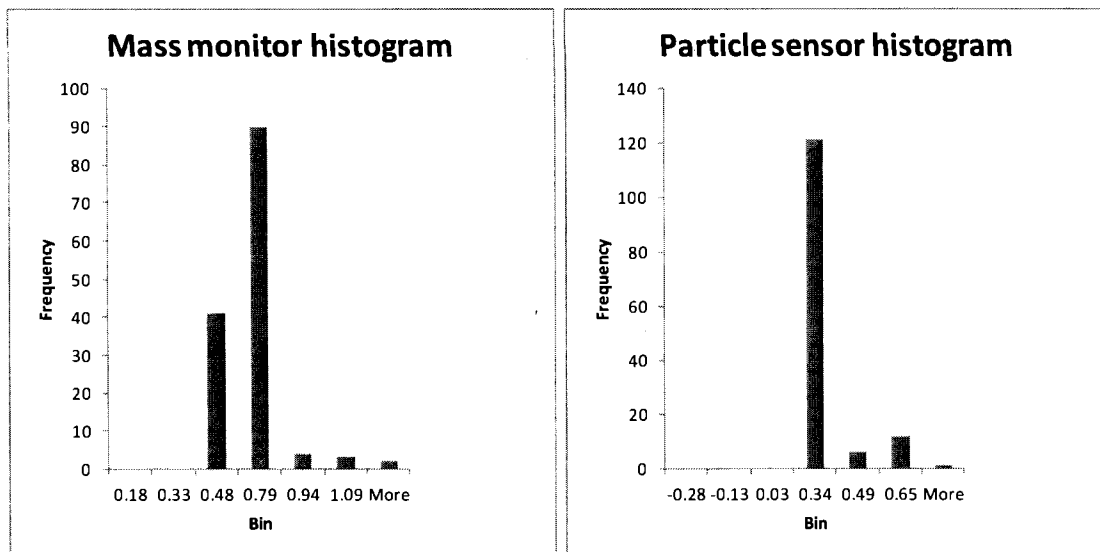


Figure E25. Frequency histograms of measured PM concentrations, day 5, test 9.

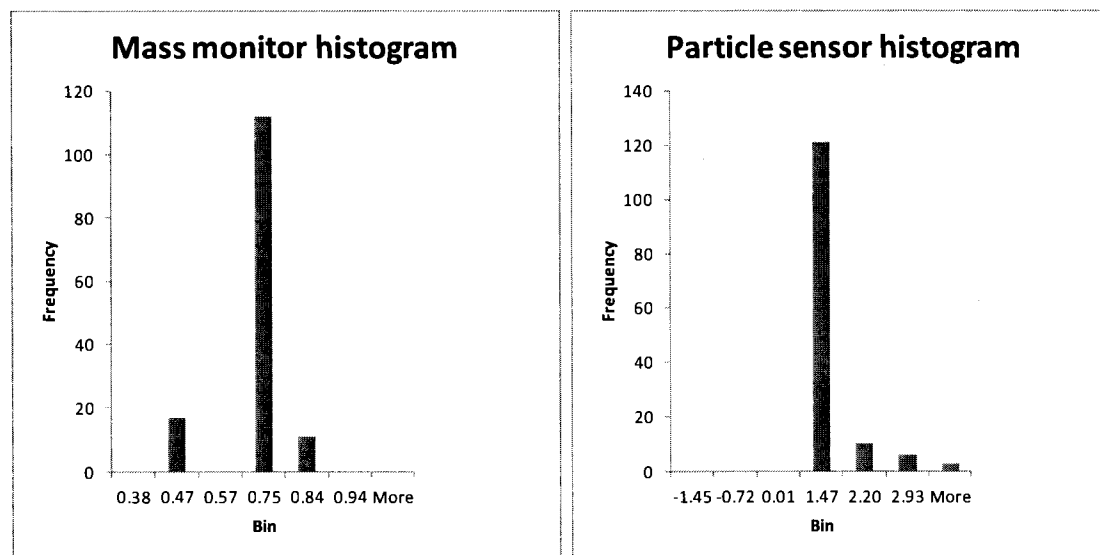


Figure E26. Frequency histograms of measured PM concentrations, day 5, test 10.

APPENDIX F
SCATTER PLOTS OF PARTICLE SENSOR AND MASS MONITOR
PM MEASUREMENT DATA SETS

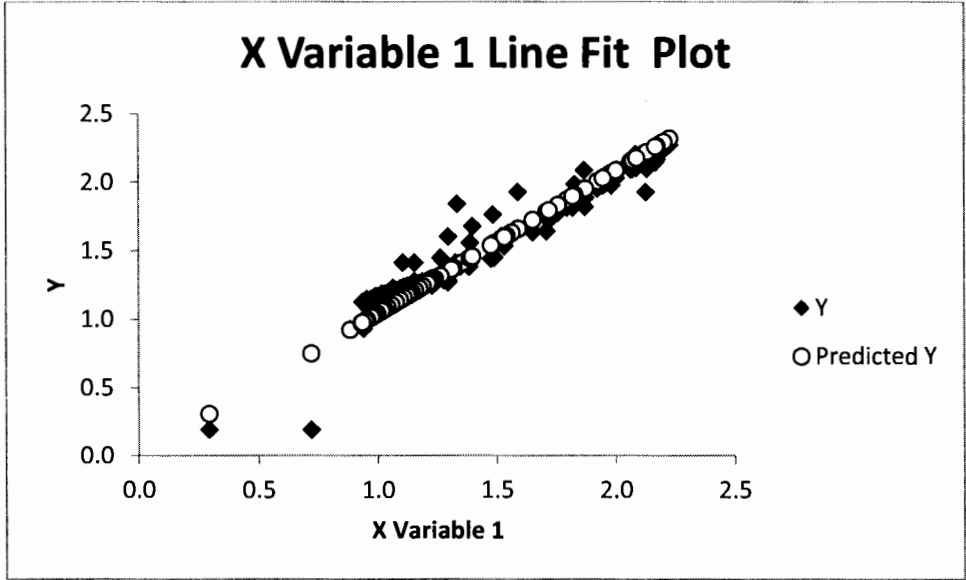


Figure F1. Scatter plot of particle sensor and mass monitor data sets, day 1, test 1.

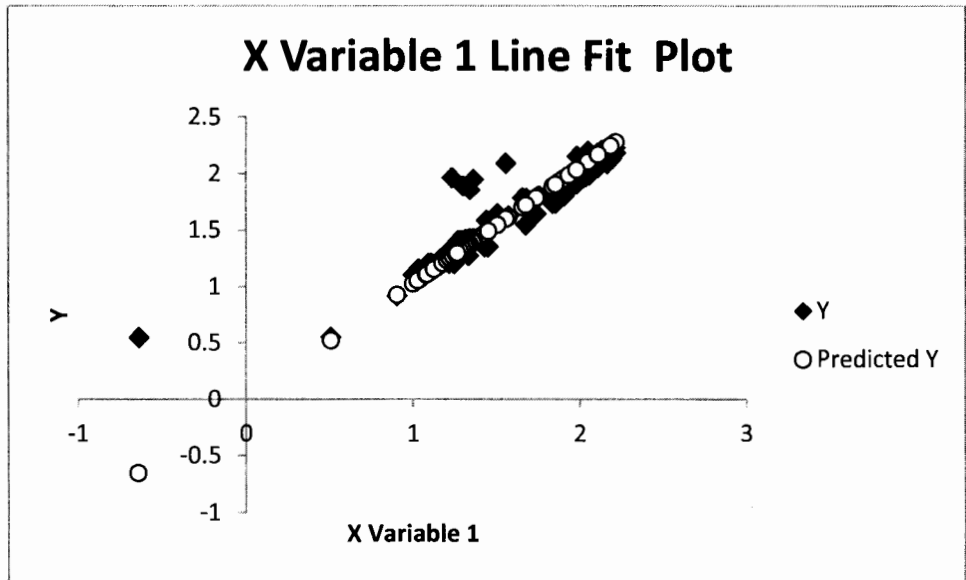


Figure F2. Scatter plot of particle sensor and mass monitor data sets, day 1, test 2.

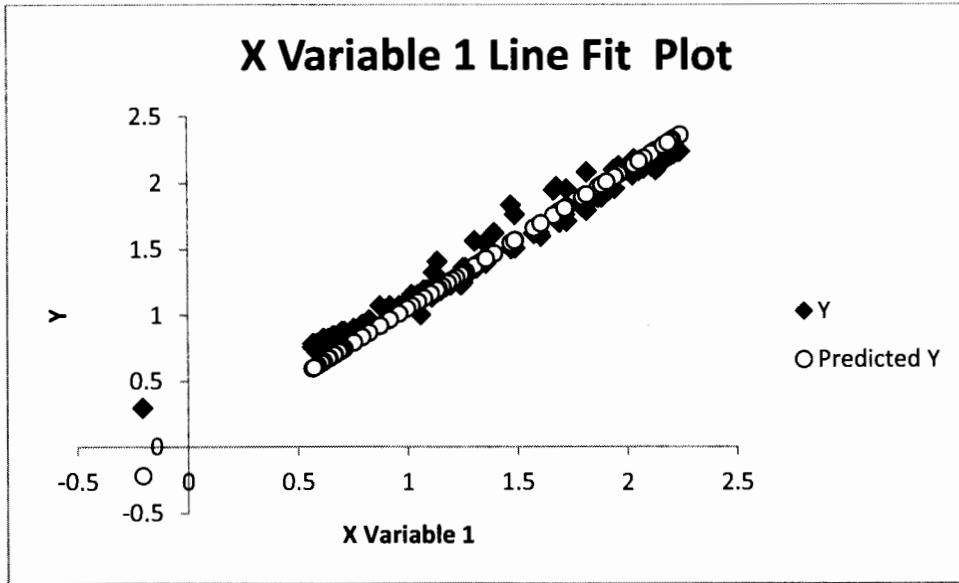


Figure F3. Scatter plot of particle sensor and mass monitor data sets, day 1, test 3.

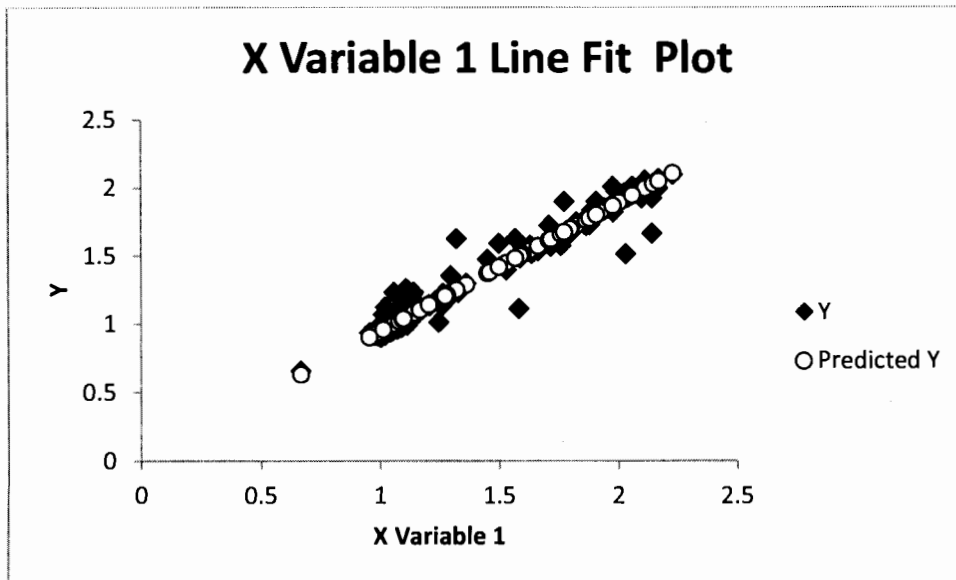


Figure F4. Scatter plot of particle sensor and mass monitor data sets, day 2, test 1.

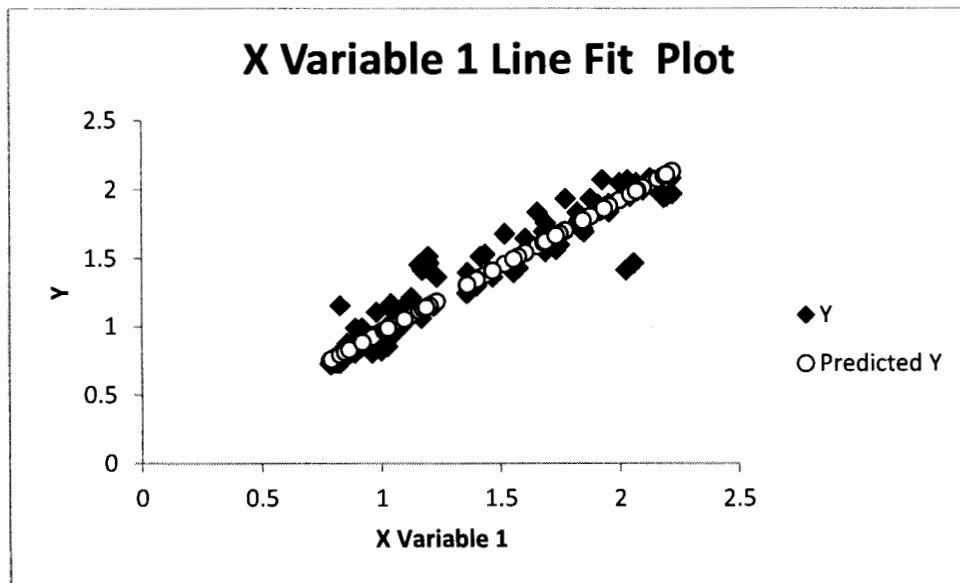


Figure F5. Scatter plot of particle sensor and mass monitor data sets, day 2, test 2.

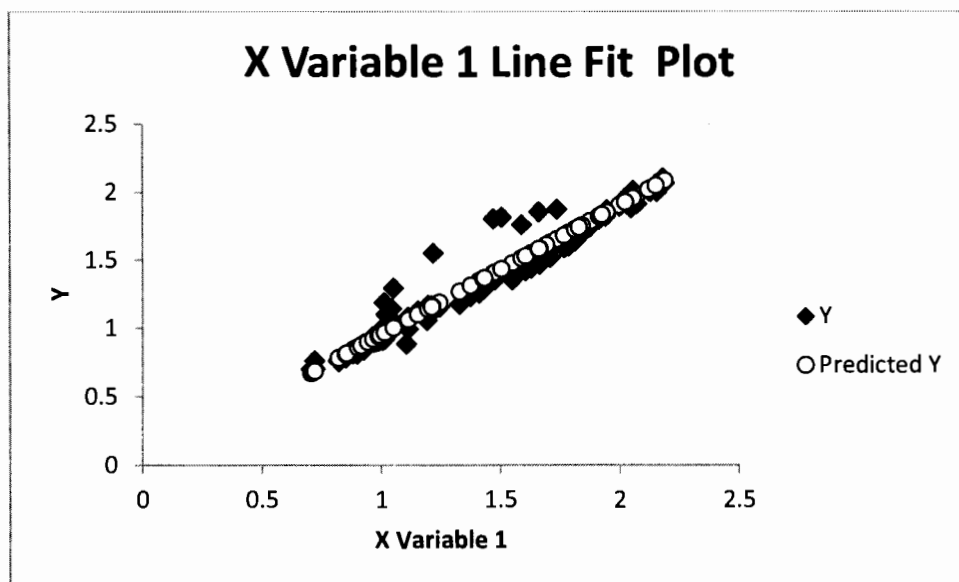


Figure F6. Scatter plot of particle sensor and mass monitor data sets, day 2, test 3.

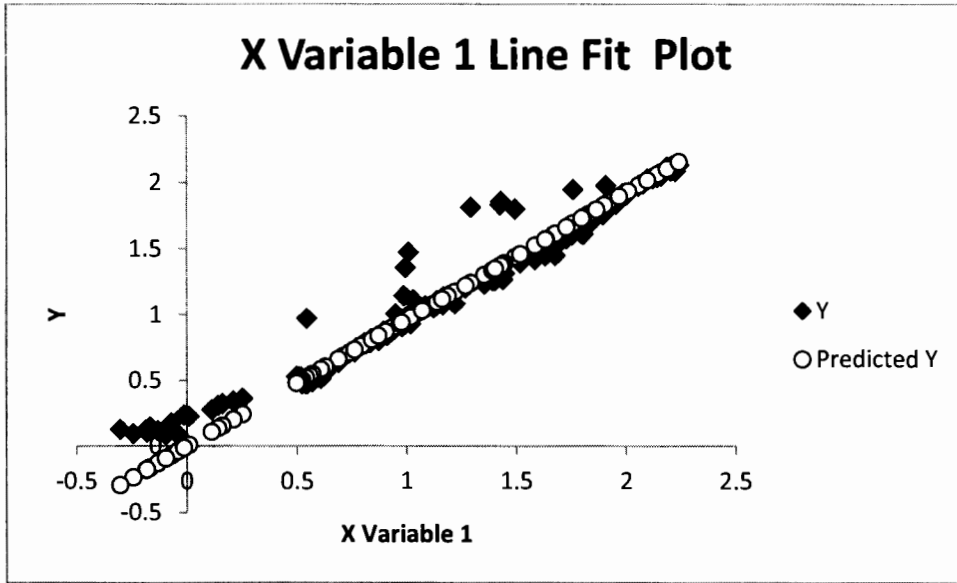


Figure F7. Scatter plot of particle sensor and mass monitor data sets, day 2, test 4.

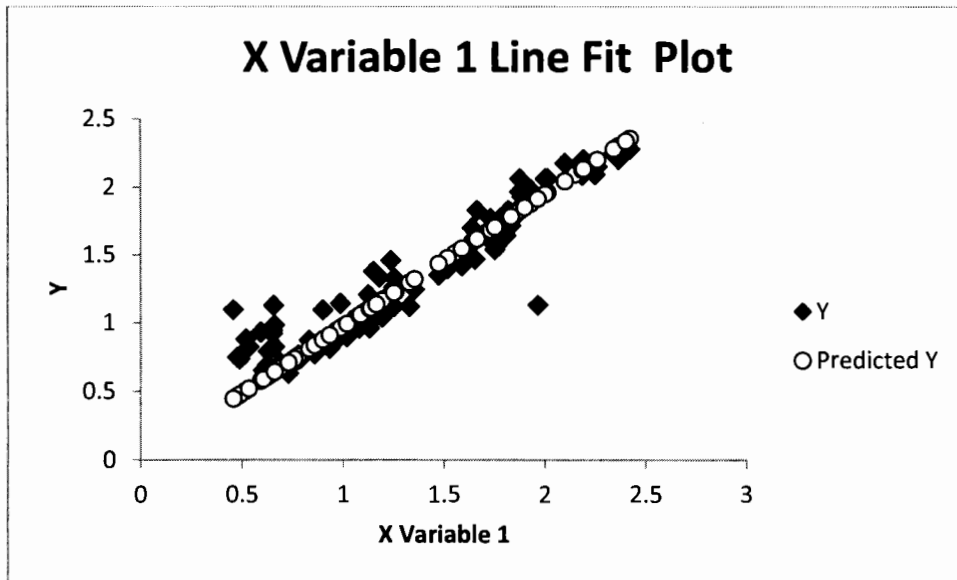


Figure F8. Scatter plot of particle sensor and mass monitor data sets, day 3, test 1.

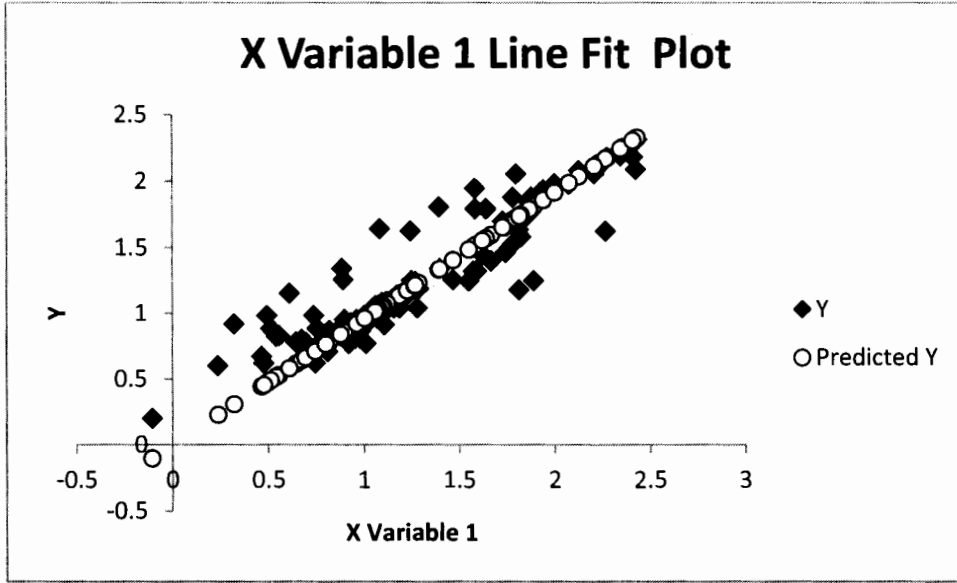


Figure F9. Scatter plot of particle sensor and mass monitor data sets, day 3, test 2.

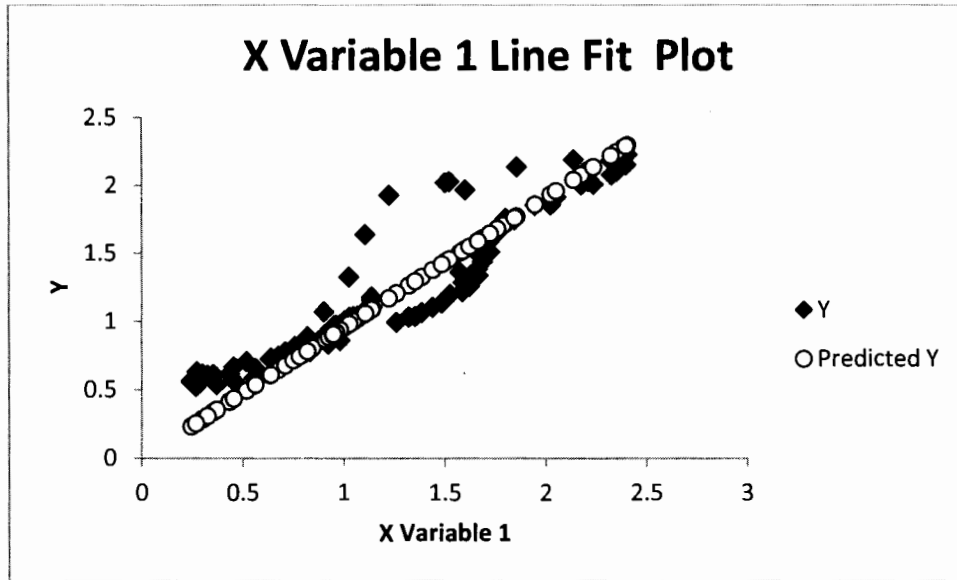


Figure F10. Scatter plot of particle sensor and mass monitor data sets, day 3, test 3.

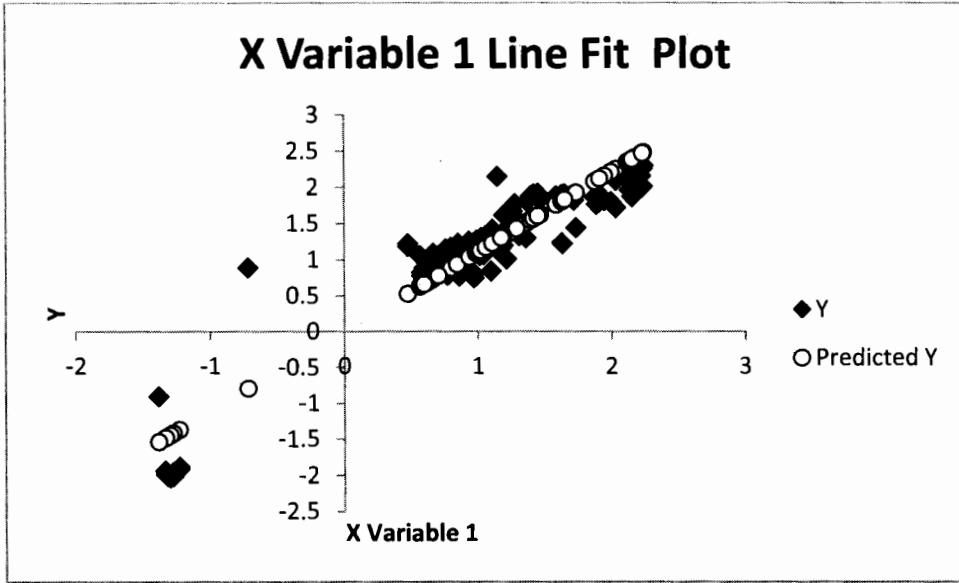


Figure F11. Scatter plot of particle sensor and mass monitor data sets, day 3, test 4.

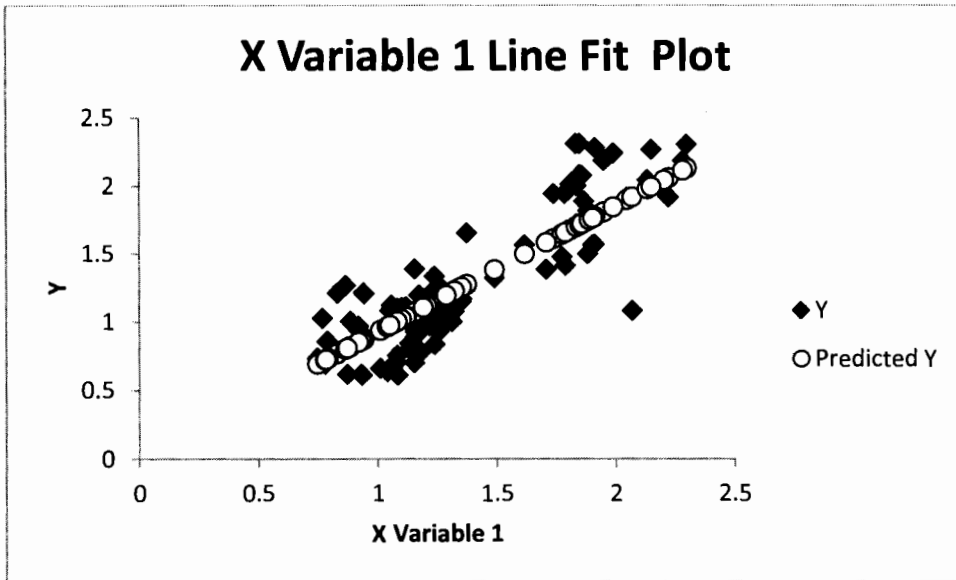


Figure F12. Scatter plot of particle sensor and mass monitor data sets, day 3, test 5.

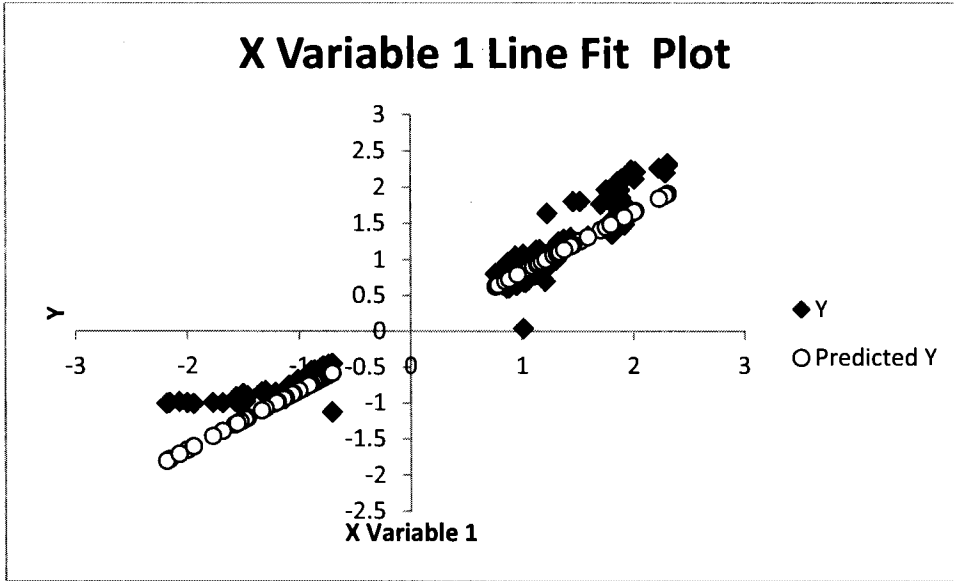


Figure F13. Scatter plot of particle sensor and mass monitor data sets, day 3, test 6.

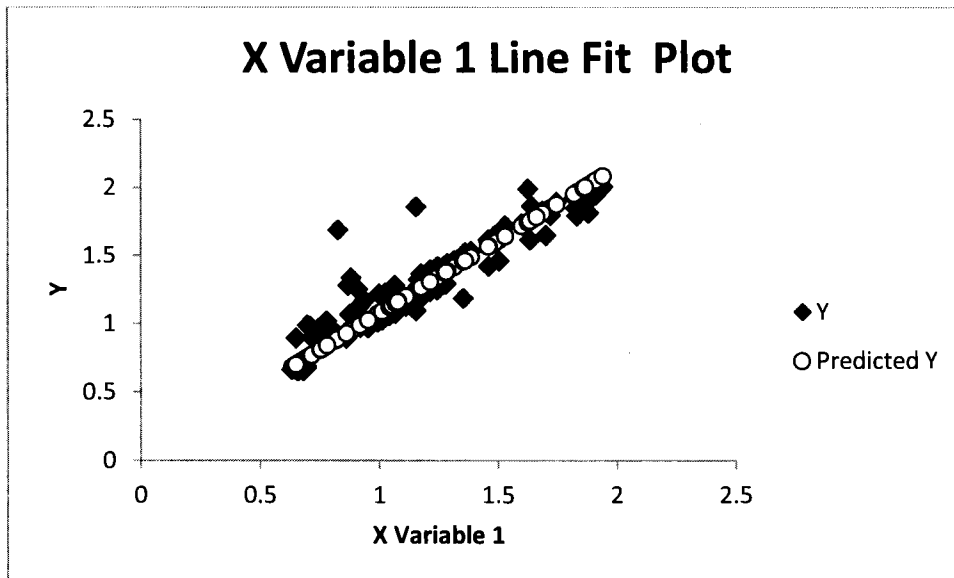


Figure F14. Scatter plot of particle sensor and mass monitor data sets, day 4, test 1.

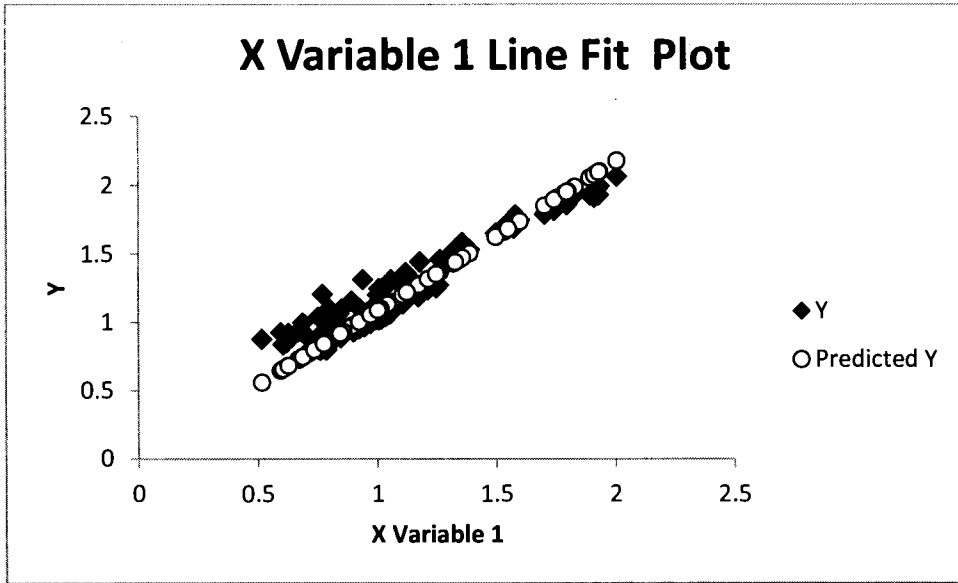


Figure F15. Scatter plot of particle sensor and mass monitor data sets, day 4, test 2.

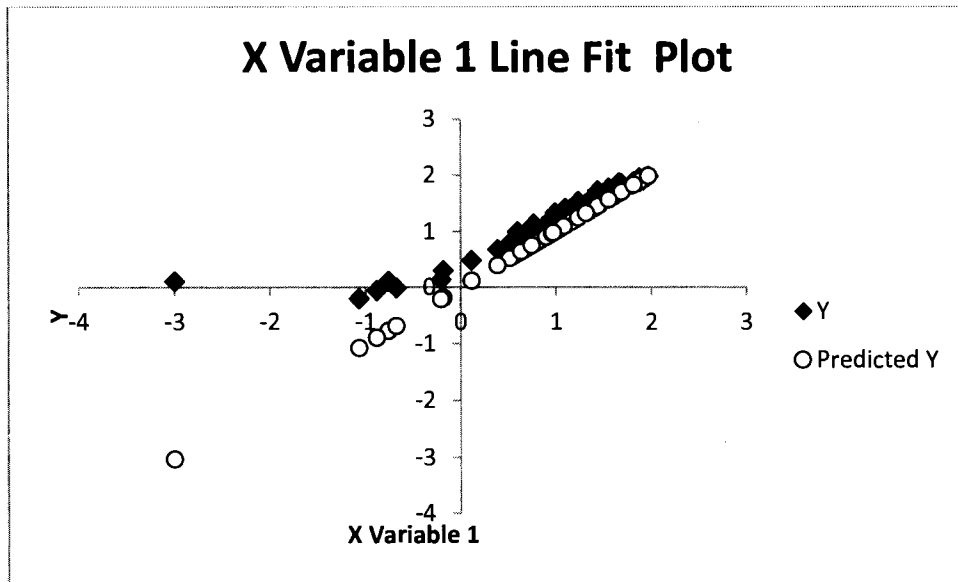


Figure F16. Scatter plot of particle sensor and mass monitor data sets, day 4, test 3.

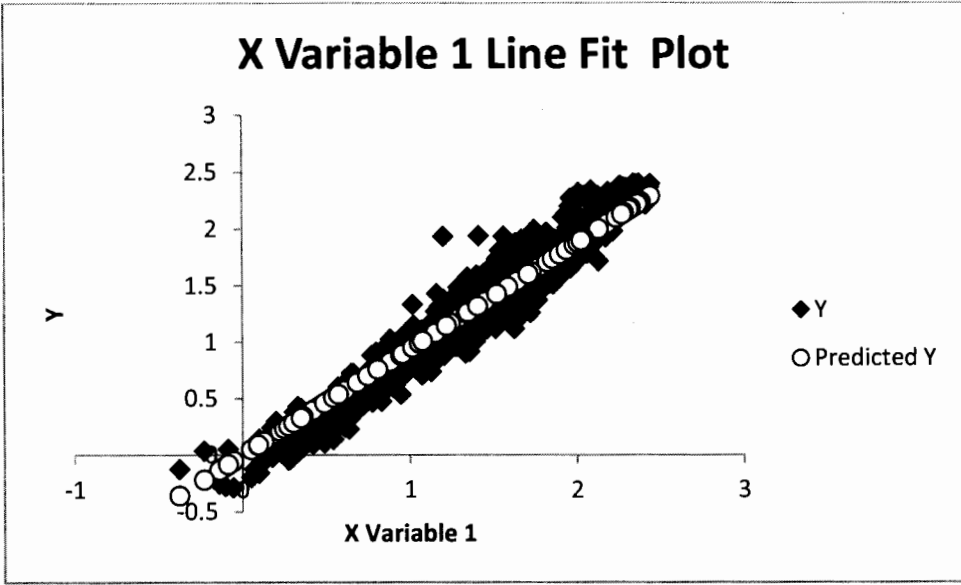


Figure F17. Scatter plot of particle sensor and mass monitor data sets, day 5, test 1.

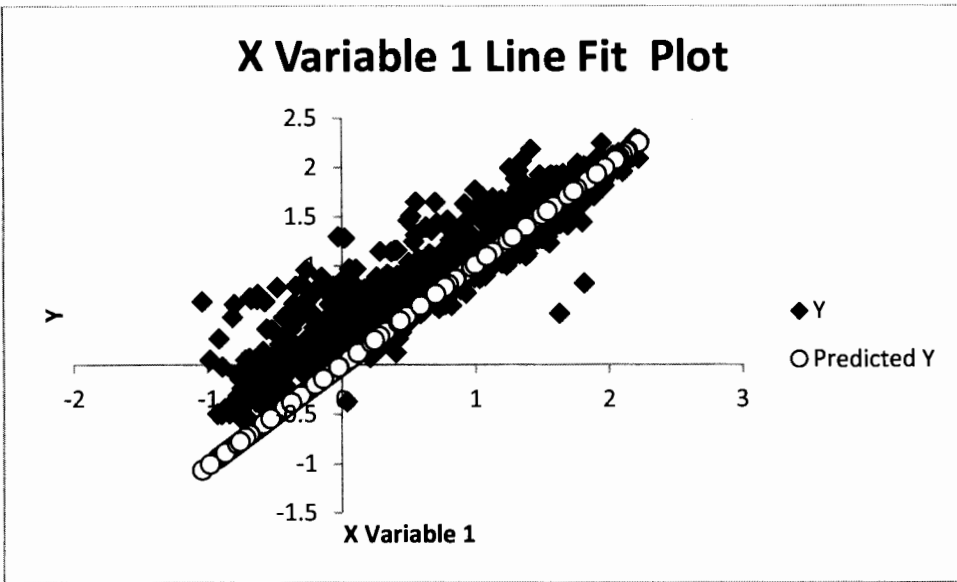


Figure F18. Scatter plot of particle sensor and mass monitor data sets, day 5, test 2.

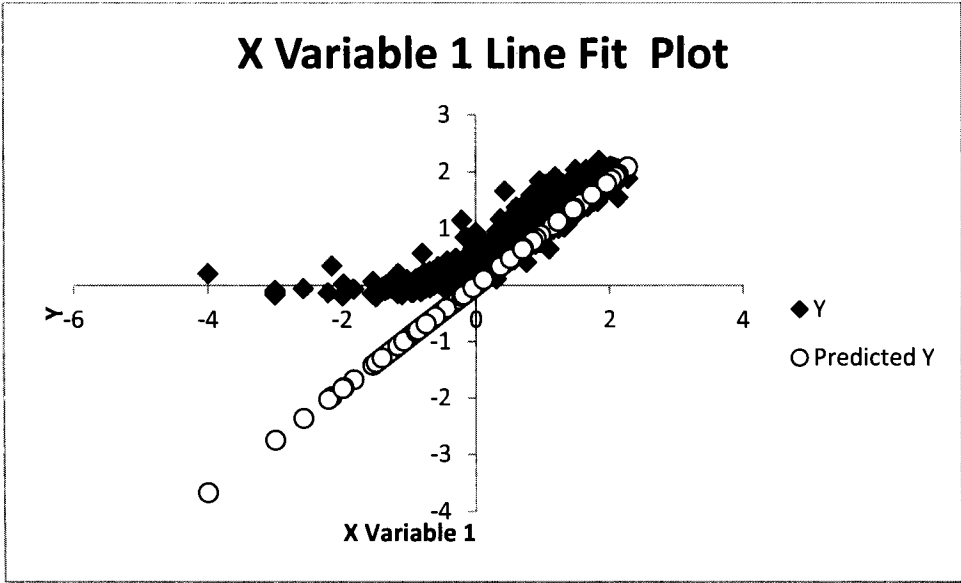


Figure F19. Scatter plot of particle sensor and mass monitor data sets, day 5, test 3.

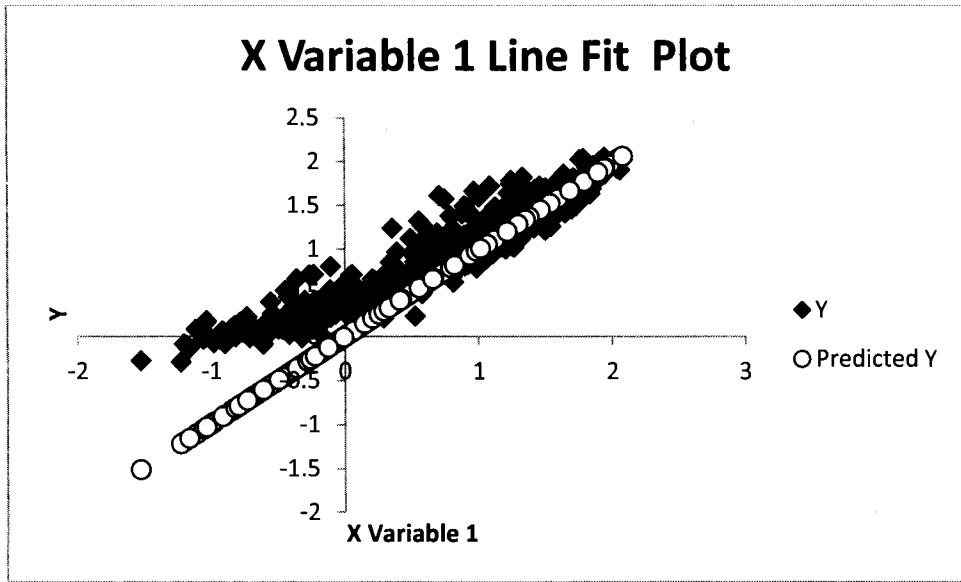


Figure F20. Scatter plot of particle sensor and mass monitor data sets, day 5, test 4.

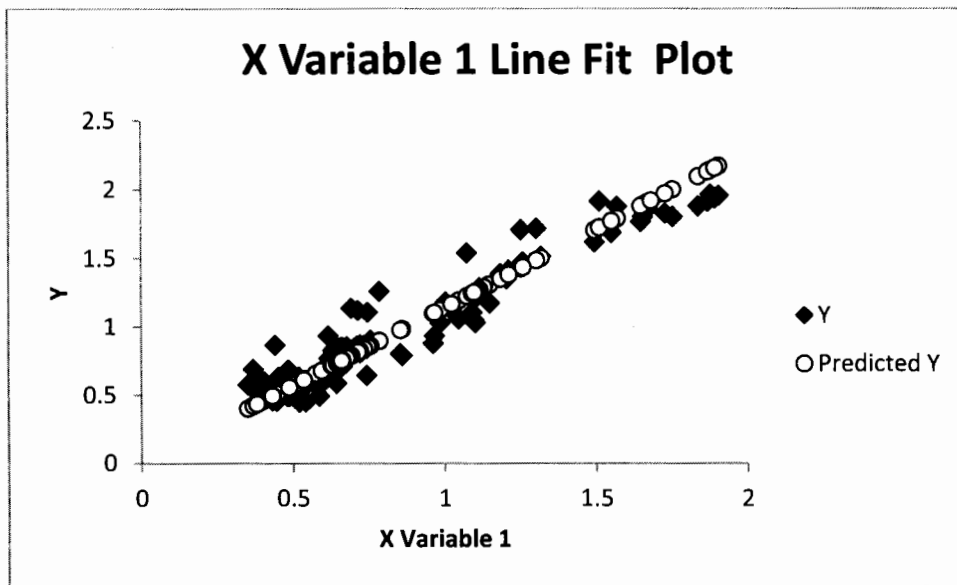


Figure F21. Scatter plot of particle sensor and mass monitor data sets, day 5, test 5.

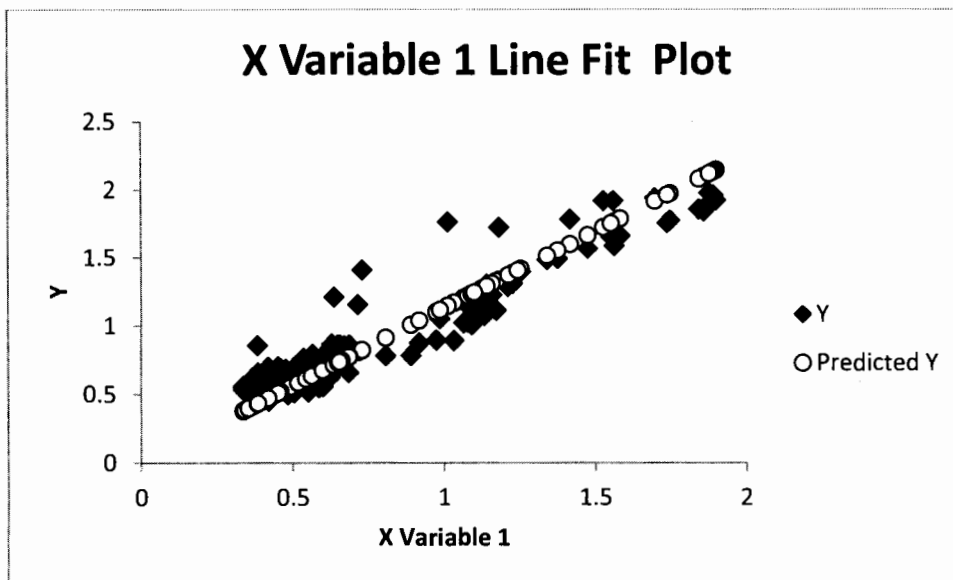


Figure F22. Scatter plot of particle sensor and mass monitor data sets, day 5, test 6.

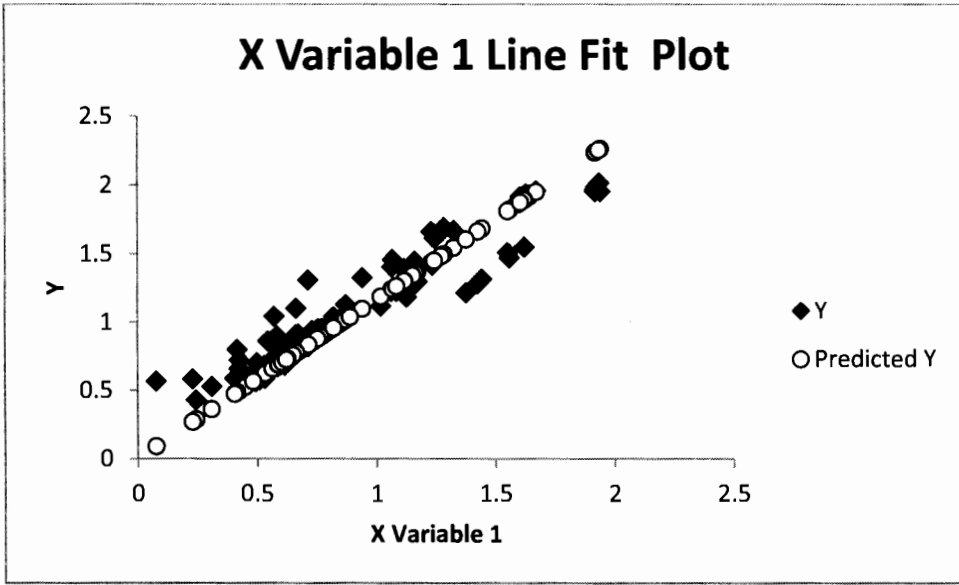


Figure F23. Scatter plot of particle sensor and mass monitor data sets, day 5, test 7.

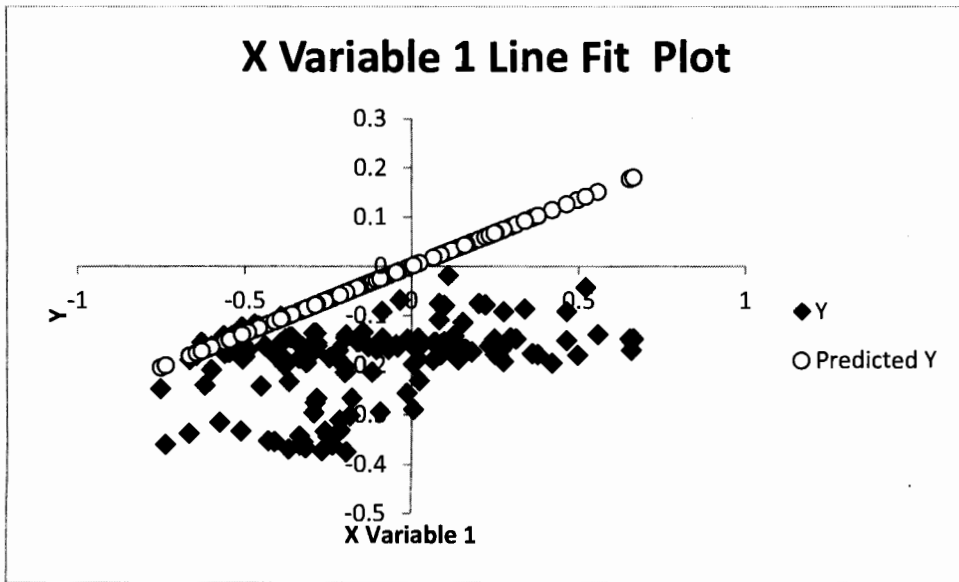


Figure F24. Scatter plot of particle sensor and mass monitor data sets, day 5, test 8.

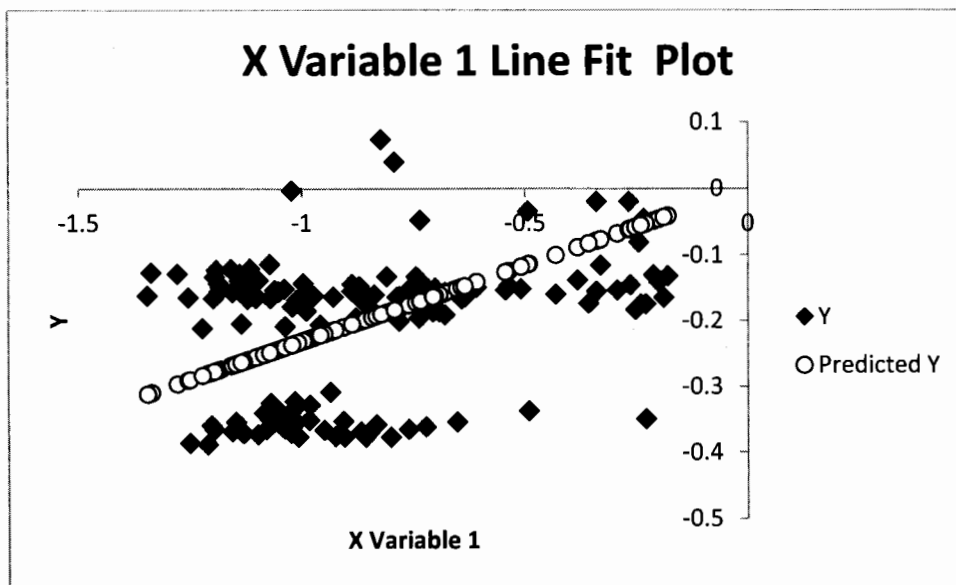


Figure F25. Scatter plot of particle sensor and mass monitor data sets, day 5, test 9.

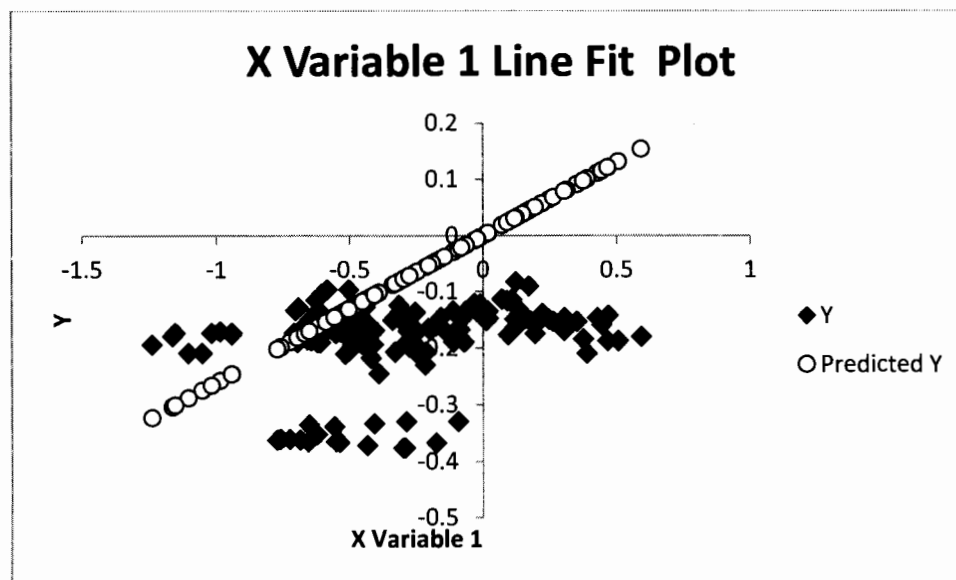


Figure F26. Scatter plot of particle sensor and mass monitor data sets, day 5, test 10.

APPENDIX G
 DESCRIPTIVE STATISTICAL SUMMARY OF PM MASS MEASUREMENTS
 FROM PARTICLE SENSOR AND MASS MONITOR
 GROUPED ACROSS MAJOR TEST CONFIGURATIONS

Descriptive statistics	Snap acceleration test cycle				Urban driving test cycle	
	Tests BDPF		Tests ADPF		BDPF	
	Particle sensor	Mass monitor	Particle sensor	Mass monitor	Particle sensor	Mass monitor
Mean	1.10	1.14	-0.43	-0.20	1.15	1.21
Standard error	0.01	0.01	0.02	0.004	0.01	0.01
Median	1.10	1.07	-0.43	-0.17	1.30	1.27
Mode	0.65	1.07	-1.16	-0.15	-3.00	1.03
Standard deviation	0.66	0.58	0.46	0.09	0.74	0.58
Sample variance	0.43	0.33	0.21	0.01	0.55	0.33
Kurtosis	5.15	2.85	-0.74	0.25	1.90	-0.28
Skewness	-1.39	-0.82	0.12	-0.83	-1.08	-0.40
Range	5.43	3.71	2.01	0.46	6.44	2.94
Minimum	-3.00	-1.38	-1.35	-0.39	-4.00	-0.54
Maximum	2.43	2.33	0.66	0.07	2.44	2.40
Sum	2722.42	2816.91	-180.00	-81.91	5889.61	6219.30
Count	2470.00	2470.00	420.00	420.00	5132.00	5132.00
Confidence level (95%)	0.03	0.02	0.04	0.01	0.02	0.02

APPENDIX H
 SUMMARY RESULTS OF REGRESSION AND
 HYPOTHESIS TESTING ANALYSES OF PM MEASUREMENT TESTS
 GROUPED ACROSS MAJOR TEST CONFIGURATIONS

Regression test statistics	Snap-acceleration test cycle		Urban driving test cycle
	BDPF	ADPF	BDPF
r	.986	.712	.976
r ²	.971	.507	.952
r ² adj	.971	.504	.952
Standard error	.216	.150	.293
β_1	0.982	0.242	0.958
β_0	0	0	0
df	2470	420	5132
F	84083.6	430.6	102715.8
Significance F	.000	.000	.000
Conclusion	Significance F < .05; reject Ho (significant result)		