



A Report from the University of Vermont Transportation Research Center

Vehicle Driver & Atmospheric Factors in Light-Duty Vehicle Particle Number Emissions

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Vehicle, Driver and Atmospheric Factors in Light-Duty Vehicle Particle Number Emissions

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Abstract

Made possible by the collection of on-board tailpipe emissions data, this research identifies road and driver factors that are associated with a relatively understudied tailpipe pollutant from light-duty vehicles: ultrafine particle number emissions. High emission events (HEE) of ultrafine particle number (PN) emissions occurred most frequently at locations with steep upgrades or locations that required moderate to rapid accelerations (>3 mph/s). The analysis revealed that less than 2% of the time driving was responsible for almost a third of all ultrafine particles emitted along the designated 17-mile test route for a sample of 22 drivers. Variables identified in a generalized linear model as significant to PN emissions include measures of engine speed (RPM), driver behavior (speed and acceleration rates), and road geometry (grade). These factors account for approximately 61% of the variability measured. Few modal emissions models estimate PN emissions; however, this research has revealed that the same predictor variables used to model gas pollutants are significant predictors of PN emissions. Therefore, the addition of PN emissions estimates to existing models would require little effort if these relationships were developed with larger datasets. This project also documented additional challenges related to on-board PN data collection related to temperature, humidity, and background PN concentrations. Finally, a large amount of the variation in light-duty PN emissions remains unexplained, suggesting a need for more comprehensive on-board datasets, including data on particle size distribution.

1.0 Introduction

State-of-the-art models for vehicle tailpipe emissions use a one-second vehicle operating mode as a main predictor for gas-phase pollutants (CO, HC, NO_x). Mode or operating bin is often quantified as a function or category of vehicle specific power (VSP), velocity, acceleration, and grade. Modal emissions models, such as CMEM, VT-MICRO or EPA's MOVES, can be used to estimate emissions produced by a vehicle fleet or the transportation network as a whole (Rakha et al 2004, Barth et al. 2001). Given the complexity of delineating time- and space-resolved operating mode of vehicles within the large and variable transportation system, there is a relative dearth of data, especially on-board real-world data for light-duty vehicles. Ideally, data that describes the spatial distribution of vehicle operating mode and the associated emissions are collected together. These data, especially particle number or particulate mass, are rarely collected due to the intensive personnel time and equipment needs; furthermore, only recently have portable emissions measurement systems (PEMS) become available to quantify particle number emissions during real-world vehicle travel. This study includes a large dataset of tailpipe gas and particle number emissions for a single vehicle operated by 22 drivers over a specified driving route.

Models and research efforts have typically focused on the EPA's regulated pollutants, including total particulate matter (PM) and increasingly, carbon dioxide, but have neglected to consider the number of individual particles, especially distinguished by size. The number of particles in the ultrafine (diameter < 100 nm) size range is critical when considering public health impacts in part due to toxicity, but also because a high proportion of ultrafine particles quickly reaches the bloodstream and is not removed by the respiratory system (Nemmar et al. 2002).

Thus, the distinction between PN and PM emissions is critical to public health impacts, yet remains relatively understudied. PN refers to the number of particles emitted; the majority of regulation and research quantifies PM, which only describes the total mass-based amount of particle emissions to which the ultrafine particles contribute little. A measure of PM per unit time or distance could signify either a small number of large particles or a very large number of small particles.

In order to understand the relationship between air quality and the transportation network, it is essential to consider the combination of vehicle operating mode, tailpipe emissions, environmental conditions, and spatial location data. This study is focused on total particle number (PN) in the 3 nanometers to 3 micrometers (or 3-3,000 nm) particle diameter range, using second-by-second data collected from one instrumented light-duty vehicle. We include the following data collected on a one-second basis: vehicle velocity and acceleration, PN tailpipe emissions, temperature, humidity, and vehicle location. This study's first objective is defining and quantifying high PN emission events within the transportation network and identifying spatial patterns and associated factors in PN emissions rate for a single 17-mile route driven multiple times by 22 volunteers in the same vehicle. These high emission events are not modeled in current regional emissions models despite their significant contributions to overall emissions and their potential to significantly degrade air quality. The second objective of the paper is to consider the one-second PN emission rates and their relationship to vehicle velocity,

acceleration, VSP, road grade, and engine load. Advancing our understanding of these relationships is critical to achieving more accurate estimates of the environmental impacts for alternative facility designs and traffic control strategies.

2.0 Background

This paper aims to contribute to the understanding of on-road particle number emissions from light-duty vehicles using on-board tailpipe data collection. Therefore, background studies related to particulate matter (PM) and its regulation as well as more recent efforts to consider particle number (PN) are summarized. The second subsection of background addresses data collection methods for tailpipe emissions study.

2.1 Particulate Matter (PM) and Particle Number (PN) Emissions

Particulate matter is defined as a complex mixture of solid and liquid particles that are suspended in air (Kittelson, 1998). PM can be generated from many different sources, both natural and manmade. In urban areas, local air quality is greatly degraded due to the transportation network (Weijers et al., 2004). Particulate matter and the number of particles are increasingly being recognized as an important part of this transportation system problem (Wang and Gao, 2011; Johnson and Ferreira, 2001; Harrison and Jones, 2005).

The combustion of fuel in a vehicle produces very small (chiefly $<1 \mu\text{m}$) particles that are then expelled from the combustion chamber and into the exhaust system. In the exhaust system, primary nanoparticles of $\sim 10 \text{ nm}$ diameter can link together with one another to form particle chains and in the process become interlocked with volatile hydrocarbons and sulfates (Heywood, 1988). This makes particles themselves difficult to study; the sampling condition, time and location of measurement are important study design considerations.

In the United States, National Ambient Air Quality Standards (NAAQS) have targeted only PM emissions (EPA, 2004). To achieve the NAAQS, federal regulations have called for the reduction of diesel vehicle PM emissions. For example, 2007 model-year *heavy-duty* diesel engines were required to meet PM *mass-based* emission rates of 0.01 g/hp-hr . This represented a 90 percent reduction in PM mass from the 2004 rate. The EPA placed these standards on PM tailpipe emissions for heavy-duty diesel vehicles to reduce ambient PM concentrations. However, note that these standards are based on particle mass emitted from the engines of newly manufactured heavy-duty engines. There are currently no PM tailpipe emission standards for light-duty gasoline vehicles. These mass-based emission standards focus primarily on particles emitted by diesel engines because diesel engines typically emit 10–100 times more total particulate *mass* than light-duty engines (Kittelson, 1998). Note further that current U.S. vehicle emission limits do not address the number of particles of any size, but that the 2012 European Union emission standards do include particle number.

The majority of the attention to air quality has been given to gas-phase pollutants (CO , NO_x , HC , VOCs and Ozone). However, PM has gained a great deal of attention since being listed as one of the EPA's six criteria air pollutants (EPA, 2004). Measurement of PM is challenging, and uncertainty remains about the details of the formation and transformation

processes (Heywood, 1988; Burtscher, 2001). The exact processes that produce high numbers of particles emitted from tailpipes are not well known for diesel engines and even less known for light-duty engines. Recent research suggests that PM is having a significant impact on public health and the quality of life in urban areas; especially in young children and the elderly (Riedl and Diaz-Sanchez, 2005; Harrison and Yin, 2000; HEI, 1999; HEI, 2003; Englert, 2004). When considering the impacts of PM on health, particle size is very important. Many health-related studies conclude that there is a link between fine and ultrafine particles (less than 2.5 micrometers and less than 100 nanometers, respectively) and a host of respiratory and circulatory conditions (Dockery et al., 1993; Pope et al., 1995; Seaton et al., 1995). This suggests a need to move from PM-based research to PN-based research and data collection for both heavy- and light-duty vehicles.

Kittelson, along with most other researchers, focuses on diesel particulate emissions, primarily because diesel engines produce more PM than spark ignition engines. However, these researchers have started to document important findings with implications for PN. For example, Kittelson et al. (2006) analyzed data obtained in the Northern Front Range Air Quality Study (NFRAQS) and reported that a spark ignition vehicle contributed 2.5 to 3 times the amount of PM_{2.5} (the mass of particles of less than 2.5 micrometers diameter) emissions when compared to a diesel vehicle. The particles emitted from spark ignition vehicles range in size from 10 to 80 nm. They are deemed to mostly be a result of incomplete fuel combustion in the engine and are very often coated with various organic compounds (Hildemann et al., 1991; Ristovski et al., 2005; Kleeman et al., 2000). While light-duty particulate mass emissions are less than those found in diesel exhaust on a per-vehicle basis, it does not mean that we can ignore this source of particle emissions. Furthermore, Lawson and Smith (1998) conducted dynamometer tests of in-use vehicles in Denver, CO and concluded that in real-world driving conditions, the gasoline-powered vehicle population generated three times more fine PM emissions by mass than the diesel-powered vehicle population.

When one moves to consider these smaller particles, the distinction between mass and number becomes more critical because the smaller particles are greater in number and do not contribute significantly to total particulate mass. Particle number (PN) emissions remain unregulated, and light-duty vehicles remain understudied due to their low *mass* PM emissions compared to diesel engines. Kittelson et al. (2006) reports that there is little to no correlation between mass of particle emissions and number of particle emissions. One on-board, on-road PN emissions study conducted at the University of Connecticut in cooperation with Connecticut Transit (CT Transit) (Vikara et al., 2007) investigated real-world emissions of four in-use CT Transit buses (two diesel and two diesel-electric hybrid). No significant difference in ultrafine PN emissions was found between bus type, but road grade was noted as a significant factor when considering PN emissions. Robinson and Holmén (2011) used similar methods to collect on-board on-road data for both a conventional and hybrid light-duty vehicle and illustrated that PN size distributions vary with operating conditions. The same dataset indicated that elevated PN emissions are associated with engine start events in light-duty vehicles (Robinson et al., 2010).

Given the challenges of measuring PM and PN, researchers have attempted to find relationships between particle and gas emissions with mixed results. Qu et al. (2008) used on-board data and found promising results suggesting that PN can be modeled as a function of gas

emissions that are more routinely measured. Others (Mazzoleni et al., 2004; Harrison and Jones, 2005) found weak correlations between particle and gas emissions using remotely sensed data.

In summary, we do not have adequate knowledge regarding the production of PN emissions from light-duty vehicles and the factors affecting PN emissions levels. This research focuses on measuring the number of particles in the ultrafine particle size ranges because they have been shown to have the most significant impact on human health yet still remain unregulated. Moreover, we focus on light-duty as opposed to diesel ultrafine particle number emissions because even though data remains limited, these emissions may be an important future policy consideration related to public health.

2.2 Data Collection Methods

Conventional vehicle emissions data collection has typically been conducted in the laboratory using chassis dynamometers (Frey et al., 2003; Joumard et al., 1995). A significant amount of laboratory dynamometer emissions data has been collected thus far over limited ranges of operating conditions (determined by a programmed speed profile known as a driving cycle). Although, the full range of real-world driving conditions are difficult to replicate in a lab setting, these data have formed a solid base for the first operating mode emissions models, including models for PM (North et al., 2006).

Over the years, researchers have turned to other methods, including remote roadside pollutant measurement, to collect real-world emissions data (Johnson and Ferreira, 2001; McGregor et al., 2003). On-road remote sensing devices measure the concentration of pollutants in the exhaust plume from the tailpipe of vehicles as they pass through an infrared or UV light beam emitted by the emissions monitoring device located on the side of a roadway (Singer and Harley, 2000). However, remote sensing collects only a finite snapshot of a vehicle's tailpipe emissions and is limited in its ability to collect data specific to the vehicle, driver and second-by-second sustained operation of the vehicle (Singer and Harley, 2000).

On-road, on-board emissions monitoring is the most desirable method of collecting real-world tailpipe emissions data due to its ability to collect comprehensive data about the driver, vehicle, roadway and engine operating conditions simultaneously (Frey et al., 2001; Frey et al., 2003). Most studies using on-road on-board techniques to date have focused on heavy-duty diesel vehicles including modal emissions models for PN (Jackson and Holmén, 2009; Sonntag and Gao, 2009; and Kamarianakis et al., 2011). Domínguez-Sáez et al. (2012) focused on a light-duty diesel and notably included particle size distribution.

Even though measuring on-board vehicle emissions allows for real-world second-by-second collection of vehicle operation and emissions output, there are many challenges in developing an accurate method for data collection. These include: time synchronization, adequate power supply, instrument sensitivity to motion and vibration, instrument size, instrumentation drift, and instrument errors. Furthermore, challenges in on-board emissions testing are caused by the use of adapted technology and instruments originally intended for laboratory use. The instrumentation used to collect the data for this analysis was developed and tested for this experiment and many of the challenges identified above were overcome. In the

results section of this paper, we identify atmospheric conditions as a remaining challenge that requires more consideration. The data collection procedures for this project are discussed below, but are also described in greater detail elsewhere (Jackson et al., 2006).

3.0 Data and Methodology

3.1 Instrumentation

The objectives of this study were to identify spatial patterns of high PN emission events and to consider PN emissions as a function of second-by-second vehicle operations and road characteristics. Therefore, a diverse set of volunteers was recruited to repeatedly drive an identical test route in order to include a range of driver behaviors. A single vehicle was used to control engine factors and ease the challenges of in-vehicle equipment installation. The data used in this analysis were collected between October 11th and October 27th, 2006 using an instrumented light-duty minivan¹ driven by 22 different volunteer drivers over a 17-mile predefined test route in northeast Connecticut (Figure 1). The route chosen for this research was selected to contain multiple road types (freeways, rural two-lane highways, and local stop-controlled roads) that would require drivers to use different driving patterns while traveling.

A 1999 Toyota Sienna minivan (7 years old at the time of experiment, odometer ~ 120,000 miles) was instrumented to simultaneously collect spatial location, vehicle/engine operating parameters, and tailpipe emissions data (Table 1). Spatial data and vehicle speed data were collected using a Garmin 16 HVS Global Positioning System (GPS) antenna on Fugawi navigation software. An AutoEnginuity ST01 ScanTool collected vehicle velocity, engine load, and engine RPM from the vehicle's engine computer. Vehicle acceleration data were collected using a Crossbow CXLO2LF3 accelerometer mounted on the roof. This accelerometer is a three-axis accelerometer with a measurable range of $-2 g^2$ to $+2 g$ and a 50 Hz response rate. The accelerometer selected for this research has a low noise density and a reported accuracy of ± 0.01 volts (0.2 mph/s). The GPS receiver and ScanTool were powered by the vehicle, and the accelerometer was powered through the data collection port of a desktop computer.

¹ 1999 Toyota Sienna, mileage \approx 120K, 3.0 liter V6, automatic, Unloaded weight: 3,890 lbs

² g = acceleration due to gravity 32.2 ft/s^2 (9.81 m/s^2)

Table 1: Equipment and Recorded Variables

Instrument	Parameters Measured	Recording Frequency
Accelerometer	Instantaneous acceleration (g)	10 Hz
ScanTool	Velocity (mph) and Engine RPM	4 Hz
Garmin 16 HVS GPS	Vehicle position (latitude, longitude) Velocity (mph)	1 Hz
Canon Digital Video Camera	Forward windshield view	30 Hz
Condensation Particle Counter	Ultrafine particle number concentrations ($\#/cm^3$) TSI Model 3025A, 3 – 3000 nm aerodynamic particle diameter	1 Hz
Thermocouples	Exhaust temperature at tailpipe and in dilution system ($^{\circ}C$)	10 Hz
Differential Pressure Transducer 1	Exhaust flow rate: Range: 0-10 volts corresponds to 0-30 inch water (linear). Calibration curve: (V=volts) $\text{Log}_{10}(Q \text{ (L/m)}) = 0.74141 * \text{log}_{10}(\text{volts}) + 3.2165$	10 Hz
Differential Pressure Transducer 2	Exhaust flow rate: Range: 0-10 volts corresponds to 0-5 inch water (linear). Calibration curve: (V=volts) $\text{Log}_{10}(Q \text{ (L/m)}) = 0.5842 * \text{log}_{10}(\text{volts}) + 2.814$	10 Hz
Differential Pressure Transducer 3	Exhaust flow rate: Range: 0-10 volts corresponds to 0-1 inch water (linear). Calibration curve: (V=volts) $\text{Log}_{10}(Q \text{ (L/m)}) = 0.5925 * \text{log}_{10}(\text{volts}) + 2.449$	10 Hz
Differential Pressure Transducer 4	Exhaust flow rate: Range: 0-10 volts corresponds to -0.125 to 0.125 inch water (linear). Calibration curve: (V=volts) $\text{Log}_{10}(Q \text{ (L/m)}) = 2.0337 * \text{log}_{10}(\text{volts}) + 0.5917$	10 Hz

In order to collect accurate particle number data, a data collection system had to be constructed (Qu et al., 2008). This system consisted of a TSI, Inc. model 3025A ultrafine Condensation Particle Counter (CPC) (3-3,000 nm size range) and a Matter Engineering MD19-2E rotating disk (six 0.04 cm^3 cavity, 0.15 Hz rotation) exhaust mini-diluter. A heated exhaust line was also used to transport the sample from the tailpipe into the dilution system. To monitor exhaust temperature entering the dilution system, an Omega model G type-K thermocouple was located at the inlet of the mini-diluter prior to dilution with HEPA-filtered, heated ($80^{\circ}C$) ambient air from the mini-diluter. The range of particle concentration detection extends from less than $0.01 \text{ particles/cm}^3$ to $9.99 \times 10^4 \text{ particles/cm}^3$. The CPC reports particle concentrations at 10 Hz. However, concentrations were recorded using Aerosol Management Instrument software (version 5.2.0) at 1 Hz intervals using a running average.

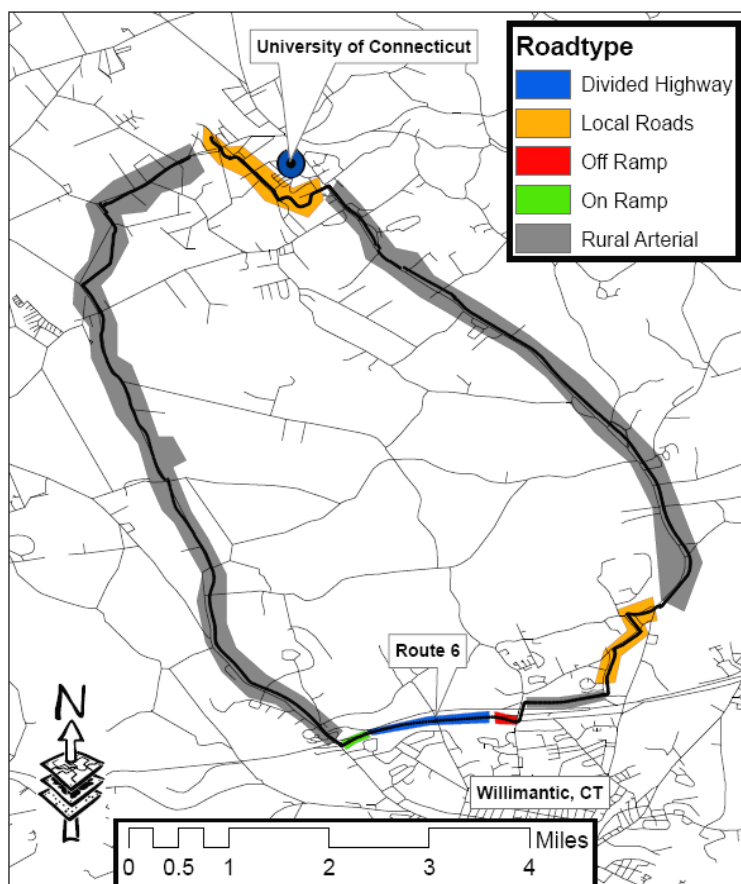


Figure 1: Connecticut Data Collection Test Route

The emissions monitoring devices were connected to the vehicle's tailpipe via an adapter. The tailpipe adapter (Qu et al., 2008) contained five ports with the following functions: 1) an Omega type-K model-E thermocouple to measure tailpipe exhaust temperature; 2) a stainless steel Swagelok fitting to transport the exhaust to the mini-diluter; 3) a stainless steel Swagelok fitting to transport the exhaust to an Autologic 5-Gas analyzer; 4) an Omega model PX181 pressure transducer to measure total exhaust pressure; and 5) United Sensor Corporation pitot tube to measure the exhaust flow rate by recording differential pressure. The pitot tube was connected in parallel to four different differential pressure transducers. The last four rows in Table 1 describe the pressure transducers and include the calibration equations used to convert a recorded voltage to exhaust flow rate in Liters/minute. The tailpipe exhaust temperature and pressure readings were recorded at 10 Hz using National Instruments Labview 7.0 software. In order to power all the instruments in the minivan, two marine batteries and two large deep-cycle RV batteries were connected to power inverters.

3.2 On-Board On-Road Data Collection

Twenty-two drivers were recruited from the University of Connecticut community via email and personal communication. All drivers were asked to drive the route at least twice in a row, resulting in a minimum of 3,200 one-second data records per driver. However, there were some drivers that volunteered to drive the test route more than twice. One volunteer drove the

test route 10 times, 3 volunteers drove 4 replicates, and the other 18 drivers drove 2 test runs consecutively. These drivers included undergraduates, graduate students, and faculty members with between 1 and 40 years of driving experience.

To ensure data consistency, a strict setup and warm-up procedure was created. Each run began with the synchronization of the laptop clock with the GPS. The drivers were then allowed to practice driving the minivan in a parking lot until they were comfortable with the vehicle. Then a research assistant seated in the passenger seat directed the driver along the test route. The drivers were advised to drive the test route as they would normally drive and that they were free to stop driving at any time if they felt uncomfortable driving the vehicle. Comparisons between runs discussed below and elsewhere (Jackson and Aultman-Hall, 2010) indicate differences in driving style between drivers, but consistency for each individual driver.

In addition to the emissions and vehicle operating data that were collected by the research team, road grade data were also collected. The Connecticut Department of Transportation (ConnDOT) collected these data by using an ARAN photologging van (Roadware, 2007). Grade and curvature data were collected every 10 meters along the entire length of the test route. Using the GPS location information in the emissions dataset and the GPS data from the ARAN van, the geometry data were added to the emissions dataset using ArcGIS and a spatial join. Therefore, every record in the emissions dataset was assigned a grade (in percent) and horizontal curvature (radius in meters) based on the ARAN data point that was closest to it. Despite its known importance, road grade is not frequently available in on-road studies of emissions.

3.3 Data Tabulation

The data from each instrument were merged into one master database. Other research studies have identified surrogate variables that can be calculated to describe how a vehicle is operating. One such variable is vehicle specific power (VSP). VSP is a measure of engine power demand that is calculated from the measured velocity, acceleration, and road grade. The joining of road grade to the emissions dataset allows the calculation of VSP when investigating causal factors in vehicle emissions. VSP is highly correlated to increased concentrations of gas phase vehicle emissions (Huai et al., 2005; Jimenez, 1999; Kuhns, 2004; Pokharel, 2001). VSP for each second of data was calculated using Equation 1 (Jimenez, 1999):

$$\mathbf{VSP = 1.1(v * a) + 9.81(grade * v) + 0.213(v) + 0.000305(v^3)} \quad \mathbf{[Equation 1]}$$

VSP = vehicle specific power (kW/ton)

v = vehicle velocity (m/s)

a = acceleration (m/s²)

grade = road grade (%)

The calculation of VSP is useful in emissions research. However, the calculation of a variable from three different sources propagates errors. Specifically, when calculating VSP, potential sources of measurement error (velocity (at ± 1 mph), acceleration (at ± 0.2 mph/s), and

grade (no reported error) are multiplied, added, and cubed. While the propagation of error has been limited by instrumentation selection, it cannot be eliminated.

With the collection of vehicle emissions data, there are time lags that are inherent in the system due to the travel time of the exhaust sample from the engine through the tailpipe and muffler, sampling hoses, and then into the sensors. To account for these lags in the exhaust and instruments, a cross correlation analysis was conducted to determine an appropriate time lag to be applied to a particular data series. By comparing the covariance between two time series at multiple time lags, an estimate of an appropriate time lag was determined. The results indicated a time lag of two seconds. This lag was applied to the PN count data to ensure that the vehicle engine operations data were temporally aligned with the emissions data.

The final step in creating the dataset for use in the analysis was calculating the PN emissions rate. Using the calculated exhaust flow rate, the dilution ratio of exhaust to high efficiency particulate air (HEPA)-filtered ambient air of 1:64 and the temporally aligned PN concentration, a PN number emissions rate in particles per second (#/s) was calculated using Equation 2:

$$\text{PN_Rate} = \text{PN} \times [\text{EX_rate} * (1000\text{cm}^3/\text{L}) * (1 \text{ min}/60 \text{ sec})] * \text{Dil} \quad \text{[Equation 2]}$$

PN = Particle Number Concentration (#/cm³)

EX_Rate = Exhaust Flow Rate (L/Min)

Dil = Dilution Ratio of raw exhaust with HEPA-filtered air = 64

Once again, with the multiplication of data from separate instruments, errors may propagate. Using four differential pressure sensors at various measurement ranges limited the errors associated with measuring exhaust flow rate at the upper and lower limits of the sensors.

The final dataset contains more than 105,000 complete 1-Hz observations from 22 different drivers. This dataset contains a rare combination of field data, including vehicle operation (speed and acceleration), engine operation (load and RPM), driving conditions (road grade and curvature), and emissions data (PN emissions rate as well as gases).

4.0 Results

The analysis and results of this experiment are described in four parts: 1) the repeatability and consistency of individual driving behavior and the associated PN emissions; 2) consistency of PN emissions over the course of the experiment (16 days); 3) the spatial pattern of High Emission Events; and 4) analysis and modeling of normalized PN emission rates as a function of vehicle operating parameters.

4.1 Consistency of driving behavior and the associated PN emissions

The data used in this research are unique because multiple drivers drove a prescribed real-world test route multiple times. Having a volunteer drive the test route more than once allows for an analysis of the consistency and repeatability of both the driving and emissions

patterns. The investigation into differences between drivers is important in understanding how individual driving habits affect vehicle emissions (Jackson et al., 2006).

To investigate the relationship between PN emissions and driver, box plots were generated (Figure 2). The median is represented by the horizontal line within the box, the mean is represented by a black cross, and the upper and lower quartiles are represented by the extent of the boxed region. The whiskers in Figure 2 extend 1.5 times the inter-quartile range beyond the upper and lower quartiles. Figure 3 displays the natural log transformed PN number emission rate for the same drivers.

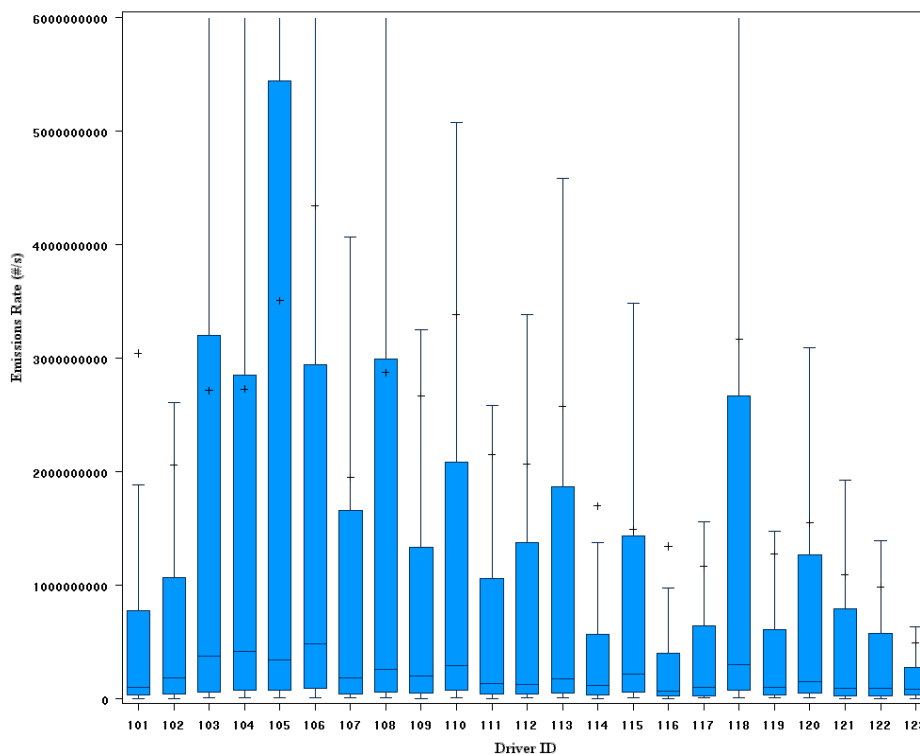


Figure 2: PN Emissions Rate by Driver

In addition to the wide variation between drivers, note first that the mean is notably larger than the median in all cases. This suggests that there are a small number of large spikes in PN emission rates that contribute significantly to the mean but have minimal impact on the median. This motivates the investigation of high emission events in the section that follows. These large spikes in PN emissions are not unexpected as emissions vary with respect to vehicle and engine operation (Clean Air Technologies, 2001; Frey et al., 2003). When evaluating the differences between drivers for raw PN there is a wide range in means and upper quartiles, but the medians are relatively consistent. For the natural log of PN emissions rate, the quartiles are more consistent across drivers. The log transform aids in the reduction of the variability from driver to driver and produces a dataset that is more normally distributed and might be more suitable to satisfy modeling assumptions associated with linear modeling techniques.

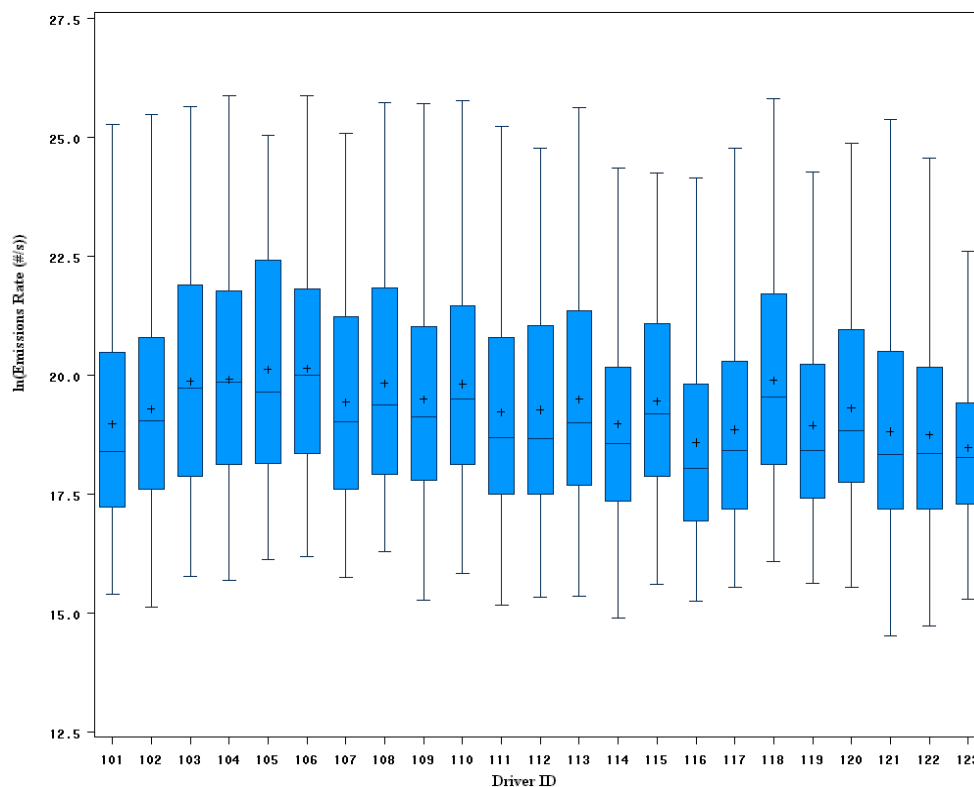


Figure 3: Log Transformed PN Emissions Rate by Driver

While many drivers drove only two consecutive runs, there were four drivers that drove more than two runs of the test route and these test runs were not run consecutively. Investigation of driving consistency and repeatability is of interest for two reasons. First, volunteers may have been uncomfortable driving an unfamiliar vehicle or route. If comfort and familiarity with the vehicle or test route were the issue, we would expect the vehicle operations data to be consistently more aggressive on the second test run. In particular, we hypothesize that drivers might on average drive faster on the second run and have more rapid accelerations that are considered to be more aggressive.

Second, emissions are presumed to vary (we do not know by how much) due to the driver behavior. It is important in this research to determine to what extent driving behavior is consistent even for a single individual. This affects future experimental design and also contributes to our understanding of emissions variation. Considering the consistency of the driver behavior and consistency of emissions during repeated runs on this single route is a first step in establishing any source of relative variability.

The hypothesis related to consistency in driving style is that multiple runs of the test route by the same driver should have the same or at the least similar speed and acceleration profile. Then by extension, if the same driver drove the vehicle in a similar manner for each run, one would predict the same emissions. Given that the environmental conditions are also unlikely to change dramatically from one consecutive run to the next (30 minutes), variation in emissions

between adjacent runs by the same driver given constant behavior would suggest that other uncontrolled factors play a significant role in PN emission rate. Initial investigations using time series plots of speed and acceleration suggest vehicle operating trends are consistent between test runs (see Figure 4). The plots in Figure 4 only represent a single driver on a small segment (approximately 3.5 minutes) of the test route. The acceleration plots have more variation from second to second, but the overall trend is consistent between the two test runs.

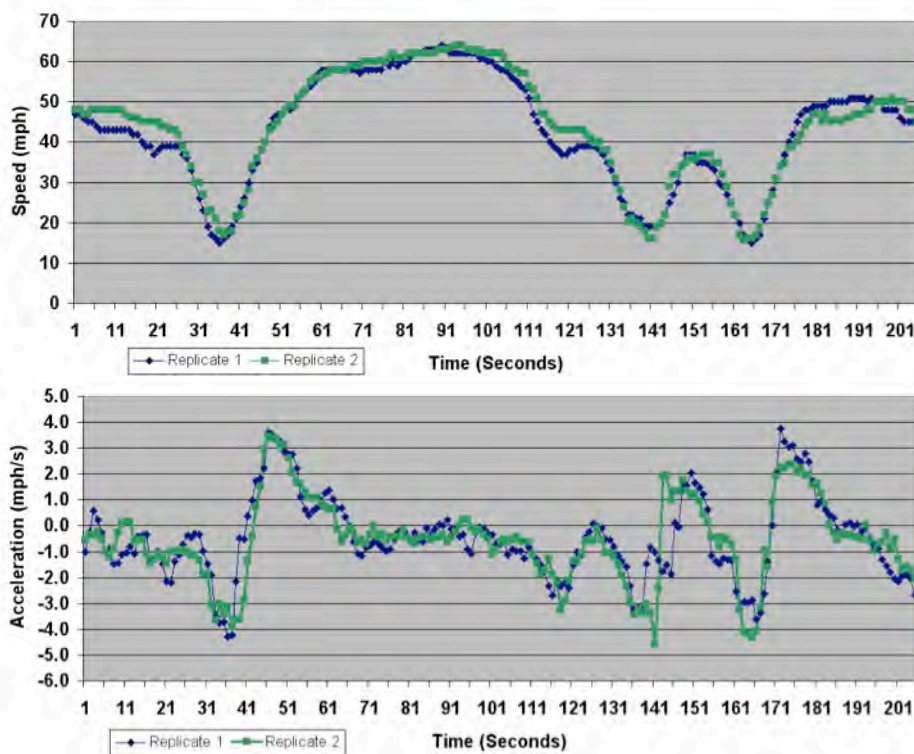


Figure 4: Temporal Patterns in Speed and Acceleration for same driver, successive runs

To compare repeatability between replicate runs driven by the same driver, the individual replicated runs for each driver were overlaid in ArcGIS. Then, by using a spatial join, the data from the two test runs were linked. The joining process uses every point in the first run file and then selects a point in the second run file that is spatially closest to that individual point. Once that point has been located, the data from the second run is linked to the data point of the first run. Note that multiple runs for each driver may not be exactly the same because traffic conditions change over time and route familiarity is likely to have an impact on an individual's driving style.

To test the repeatability between replicate runs by the same driver aggregate statistics were compared using a paired student's t-test for each driver. To limit the impact of traffic conditions on this analysis, only sections of the test route where both the first and second runs were classified as unconstrained by a vehicle ahead or by traffic control were used (determined by video). Furthermore, only sections that occurred on Route 32 and away from major

intersections were used in the replicate analysis. The speed limit on this section of the route was 40 mph. The number of drivers that had overlapping sections of unconstrained driving was limited, so not every driver was eligible for this analysis. Table 2 shows the mean speeds and acceleration for each driver with the associated standard deviation in parentheses. No pairs were different based on a 95% significance level. Based on this, we conclude that drivers were consistent from run to run and assume that PN emissions between runs for a single driver would likewise be consistent.

Table 2: Mean and Standard Deviation Between Runs for Unconstrained Driving

Driver	Speed (MPH)		Acceleration (MPH/S)	
	Replicate 1	Replicate 2	Replicate 1	Replicate 2
101	47 (5)	46 (6)	-0.5 (0.9)	-0.4 (1.0)
103	42 (9)	43 (8)	0.2 (1.3)	0.1 (1.2)
104	40 (8)	41 (7)	0.1 (1.1)	0.1 (1.0)
105	41 (11)	40 (11)	0.4 (1.7)	0.2 (1.5)
107	40 (6)	40 (6)	-0.2 (1.0)	-0.1 (1.2)
109	43 (7)	43 (9)	0.2 (1.1)	0.3 (1.1)
111	40 (7)	42 (6)	0.0 (0.9)	0.0 (0.9)
112	41 (6)	40 (5)	0.2 (1.0)	0.1 (0.9)
113	41 (7)	43 (9)	0.3 (1.0)	0.2 (1.0)
114	41 (8)	42 (8)	0.1 (1.1)	0.0 (1.3)
116	38 (7)	39 (7)	-0.2 (0.9)	-0.1 (0.8)
117	40 (9)	39 (9)	0.1 (1.2)	0.0 (1.3)
118	49 (3)	50 (3)	1.0 (1.4)	1.0 (1.6)
119	42 (9)	42 (10)	0.4 (1.3)	0.3 (1.2)
123	40 (7)	40 (8)	0.0 (1.2)	-0.1 (1.3)

4.2 Consistency of PN Emissions over time

Assuming consistent driver behavior we also compared PN emissions rates by run. Figure 5 illustrates PN emissions by run number in the sequence the data was collected. Ten boxes are colored red and correspond to the driver that drove 10 replicate runs. Two key observations can be made from this figure. The mean (+ symbol) is much larger than the median (line in center of box) for all drivers. This suggests that there are outlier events of large magnitude and of short duration for all drivers (the high emission events discussed in section 4.3).

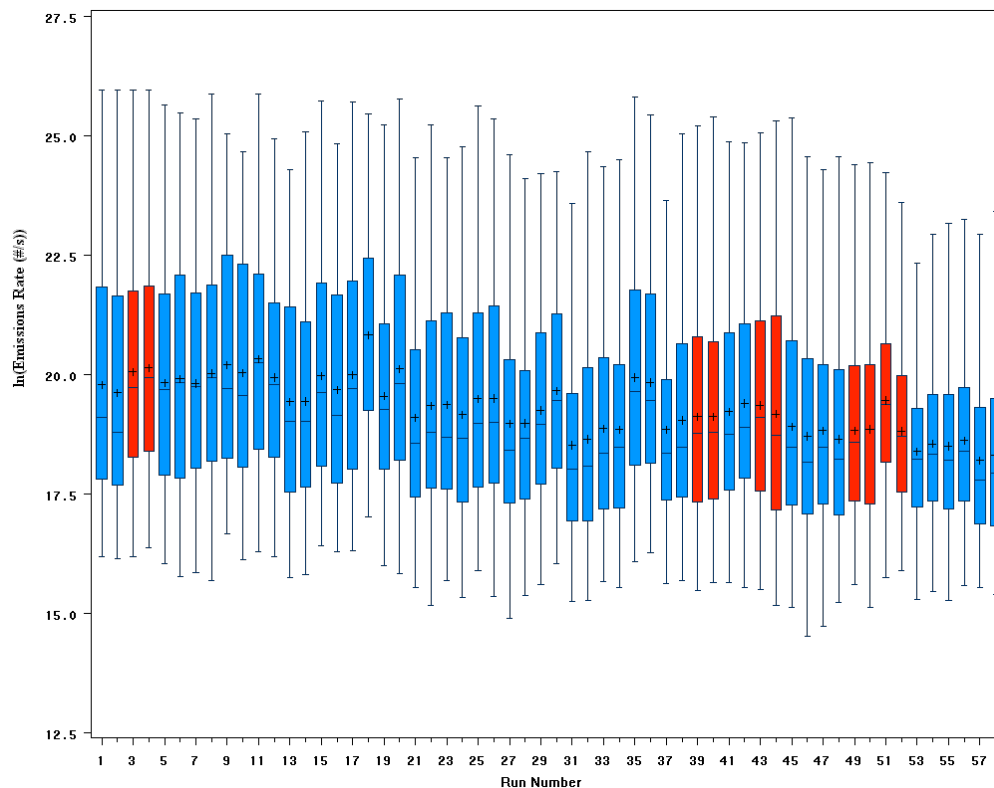


Figure 5: PN Emissions Rate By Run Number (Outliers Not Shown)

The second observation drawn from Figure 5 is that there is a decreasing trend in PN emissions rates as the run number increases (i.e., decreasing over the duration of the experiment). This trend is troubling because specific drivers that drove at the beginning and end of the experiment have dramatically different mean emissions rate. For example, the first and last drivers in Figure 5 were the same person. The first and second runs by this driver had a mean PN emission rate of 5.5×10^9 particles-per-second. When this driver drove two additional runs at the end of the experiment, the mean emission rate was 5.5×10^8 particles-per-second, which represents an order of magnitude decrease over the duration of the experiment.

In order to rule out randomness, a regression analysis was conducted to determine if the decreasing mean was statistically different than zero. The resulting model ($\alpha=0.05$) has an R^2 of 0.63 and the probability that the slope was zero was less than 0.0001. Therefore, we are able to reject the null hypothesis and conclude that there is a difference in PN emission rate means over time. This test does not indicate what factors are responsible, but just that there is a statistically significant trend in the means.

This difference could be due to any number of factors, some of which have implications for future data collection. There are several hypotheses that could explain this drop in mean PN emissions rates over time. The first hypothesis is instrument problems or error, but inspection of the instruments gave no evidence for this suggestion.

The second hypothesis involves the effects of a sitting vehicle followed by constant use of the vehicle over the test period. Prior to data collection, the van sat for several weeks while instruments were installed and tested. Once data collection began, the van was run almost daily for at least five hours of continuous operation. The hypothesis is that particles lodged in the engine or exhaust system were dislodged at a higher rate in the beginning of the experiment than towards the end. This is a very difficult hypothesis to test without letting the van sit for an extended period of time and then repeating the experiment. Time and cost prevent the testing of this hypothesis and current PN literature offers no clear answers to this question.

The third hypothesis for the downward mean PN trend involves the effects of ambient conditions on particle formation. As part of this experiment, the relative humidity and temperature conditions outside the vehicle were recorded every 10 seconds. Figure 6 shows the trends in ambient temperature and ambient relative humidity throughout the course of the experiment. Runs 5-14 were driven on days with low humidity and an increasing temperature. Furthermore the temperature and humidity data collected after run number 18 show an overall downward trend similar to the trend in the mean PN emissions rate. These observations suggest that temperature and humidity may have a large impact on mean particle emission rate. Figure 7 shows the mean PN emission rate plotted against relative humidity. This figure shows that there are a group of runs (5-14) that are distinct from the rest of the dataset with run 11 being an outlier. These data runs were collected during a period of low humidity and average temperature. These runs have an upward trend in mean particle counts as relative humidity increased. When the remaining data are examined (the upper portion of the Figure) there is also an upward trend in mean particles as relative humidity increased. These results indicate a need to measure both relative humidity and temperature in future experiments and to include these variables in models.

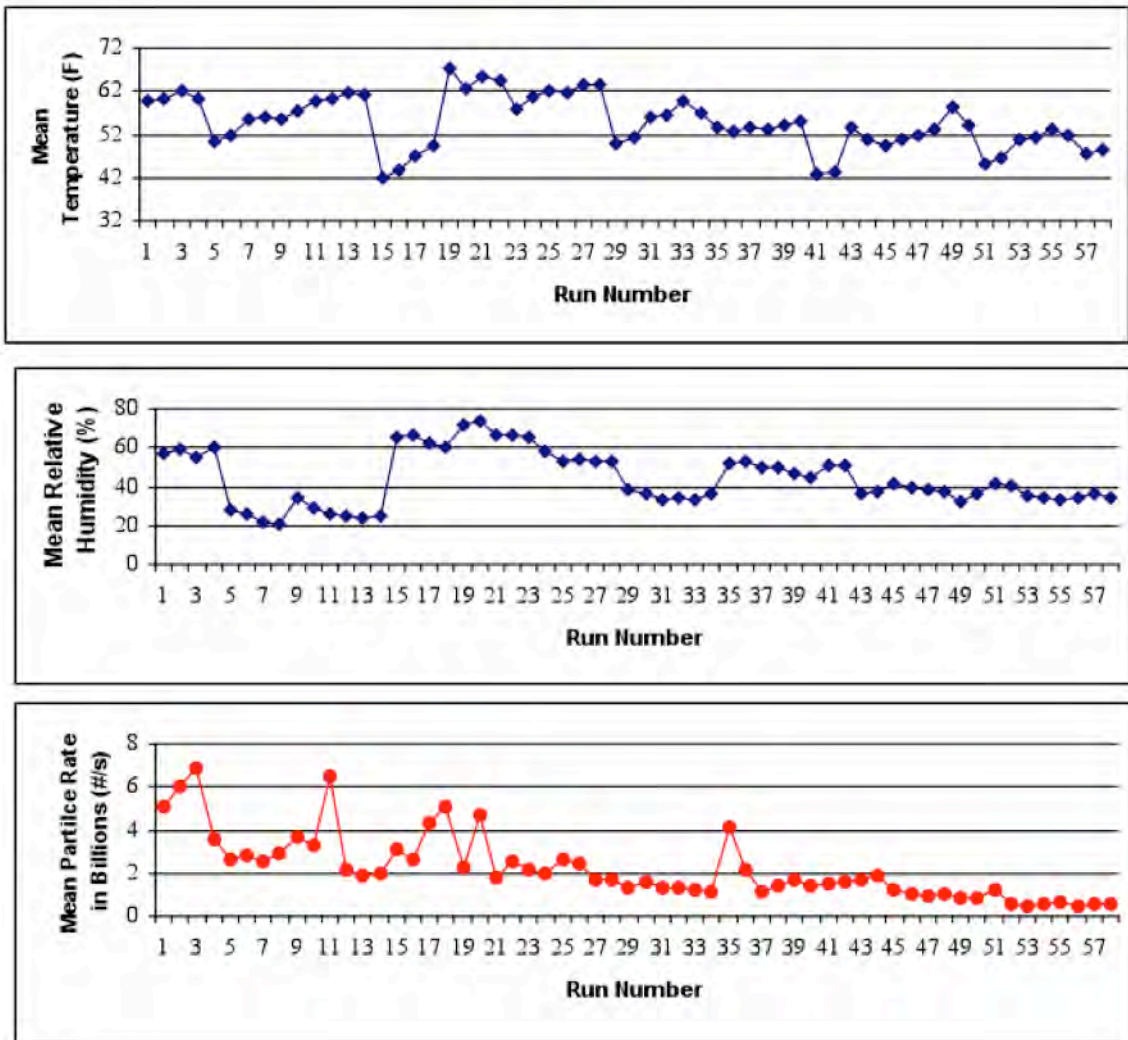


Figure 6: Temperature, Humidity, and PN Trends over Time

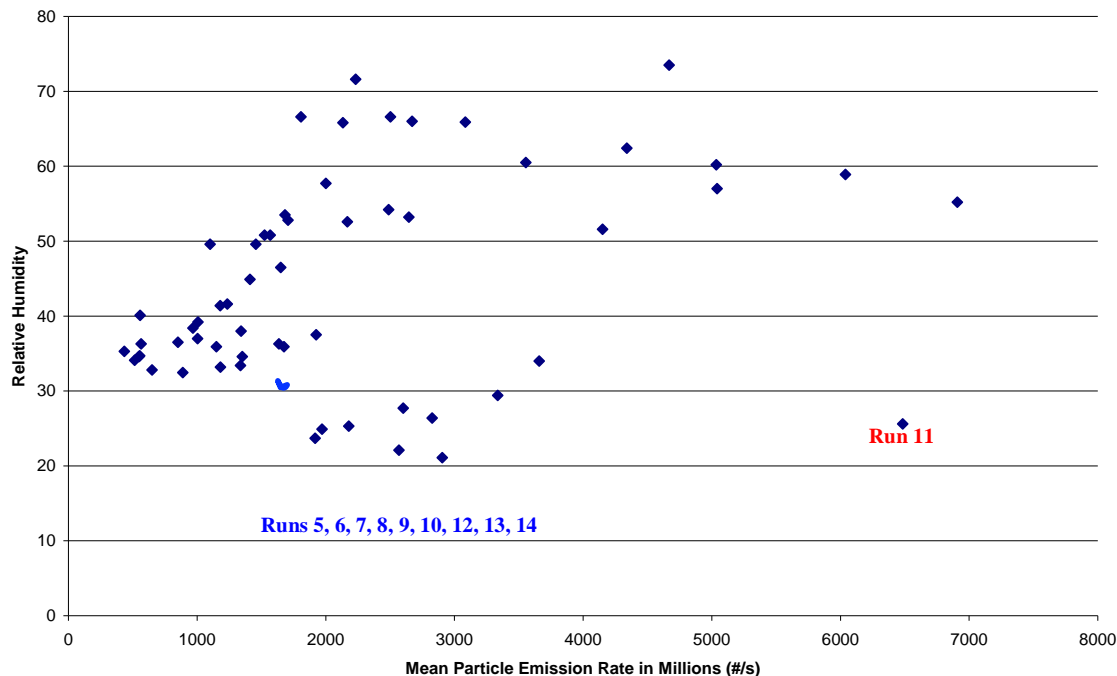


Figure 7: Mean Particle Emissions Rate by Relative Humidity

The exact cause of the decreasing trend seen in the mean PN emissions rate over the extent of the experiment is not known and without further testing, it is difficult to determine if the vehicle, instruments, or ambient environmental conditions were responsible. The video data from the forward facing camera was scanned for periods of high PN emissions during the experiment. This revealed one further indication that ambient environmental conditions have a significant impact on the number of particles measured in the exhaust. One day, while driving on the test route, the test vehicle approached a large dump truck that was hauling dry dead corn stalks recently cut out from a neighboring field. The large truck was covered in dust and the load was not secured or covered, allowing debris to fly out of the truck and onto the road and trailing vehicles. As the van approached the back of the dump truck, the particulate counts shown on the CPC screen began to rise and held fairly constant at the midpoint of the PN count scale (approximately 45,000 #/cm³). Nowhere else in the entire data collection effort did PN counts reach a sustained level this high for an extended period of time. Furthermore, once the dump truck pulled off the main road, the PN counts returned to levels consistent with other test runs. This event suggests that the number of particles measured from a vehicle's tailpipe vary along with the number of ambient particles in the background. This circumstantial evidence suggests that ambient conditions or atmospheric conditions are responsible for at least some variation in the PN emissions rate and potentially the downward trend. It is possible that the air intake of the vehicle is pulling in particles (from the dump truck in this case) and then through the combustion chamber, thus changing exhaust emissions. While vehicles have air filters on their intake system, they are not high-efficiency particulate air filters (HEPA), which could remove these ultrafine particles. There was no background monitor in the vicinity that could be used to consider ambient PN or PM through this time period.

The data in this experiment suggest that atmospheric conditions, including temperature, RH, and ambient particle concentrations have a large impact on tailpipe PN emission rates.

4.3 Identifying High Emissions Events

To gain a better understanding of high ultrafine PN emission events, one-second time periods with particularly elevated PN emissions were termed a High Emissions Event (HEE). Evaluation of the histogram in Figure 8 supports observations from the prior section and the need to consider HEEs. Observations for the full x-axis in the histogram were measured. The tail sections of the histogram at different emissions rate scales (x-axis) are inset in Figure 8 in order to observe how the tail behaves. The increase in frequency at the end of the histogram is due to the limited range of the CPC. All number counts that should have been above this value were assigned the maximum PN count by the CPC. There appears to be no natural breakpoint at which one might define an HEE.

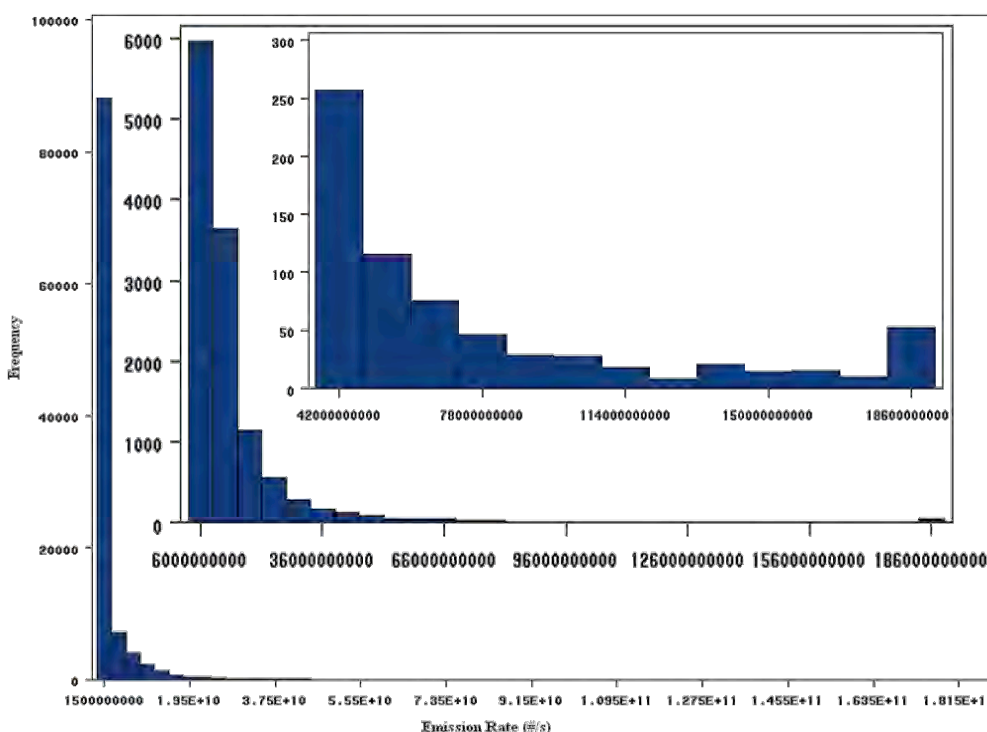


Figure 8: Histogram of PN Emission Rate

Given the lack of an obvious threshold in Figure 8, multiple criteria for an HEE were established:

- 1) HEEs should be defined such that each driver and each run contain HEEs. This requirement allows for quick visual analysis of sections along the test route where each driver's PN emissions were significantly larger than the driver's baseline emissions rate.

- 2) An HEE should be a function of a PN emissions baseline. This will aid in accounting for differences in driving style and ambient weather conditions.
- 3) Less than 10% of the data for each driver should be classified as an HEE. As stated earlier, an HEE should be an event that is of low frequency and large magnitude.

Therefore, a record was classified as an HEE if the PN emissions rate was three standard deviations above the mean PN emissions rate for a given run. This definition accounts for differences in data distributions by using the standard deviation of the emissions rate for each run.

The percent of data classified as HEEs varied from as low as 0.9 % of the run to 2.9 % of the run. For the dataset as a whole, 1704 of the 105,943 (1.6 %) of the data were classified as HEEs. **However, this small percentage of the dataset accounted for over 30% of the total number of particles emitted during data collection.**

To investigate whether these high emissions events occur randomly along the transportation route or at finite spatial locations, the HEE data were plotted spatially in ArcGIS. Figure 9 represents the frequency of HEEs along the test route for all 22 drivers. The plot in Figure 9 shows that there are specific locations along the test route that have a high density of HEEs. Plots of the 22 individual driver HEEs (not shown) also support that there are two main locations along the test route where HEEs occur. The first clustering of HEEs is located in the bottom center of the figure where a freeway on-ramp is located. At this location on the test route, there is a relatively steep upgrade along the ramp and most drivers required acceleration from a stop due to a traffic signal or making a left turn across opposing traffic. The second location where HEEs are clustered is on the right of the figure. At this location, there is a significant upgrade, (>9 % for 925 ft), and just south of this section there is another steep grade of short length (>10 % for 250 ft). Note that even though some sections of the predominantly rural route have no HEEs, most sections do have some and the sections with no HEEs are not particularly unique.

The plots for each individual driver indicate that high emission PN events are typically located at intersections where the vehicle makes a turn at an intersection causing the driver to slow down or stop and then accelerate from the intersection. However, there are other HEEs that occur for no apparent spatial reason and are totally unique to an individual test run. These events are hypothesized to be caused by instrument error, abnormal transient engine operation (such as rich fuel, downshifting gears, high RPM, etc.), or driver reaction to the roadway or other drivers.

Overall, in terms of high emissions events, this spatial analysis reveals PN HEEs are associated with grade and acceleration and the dataset clearly conveys that HEEs are not completely spatially random. However, it is clear that other factors are often at play in creating PN HEEs.

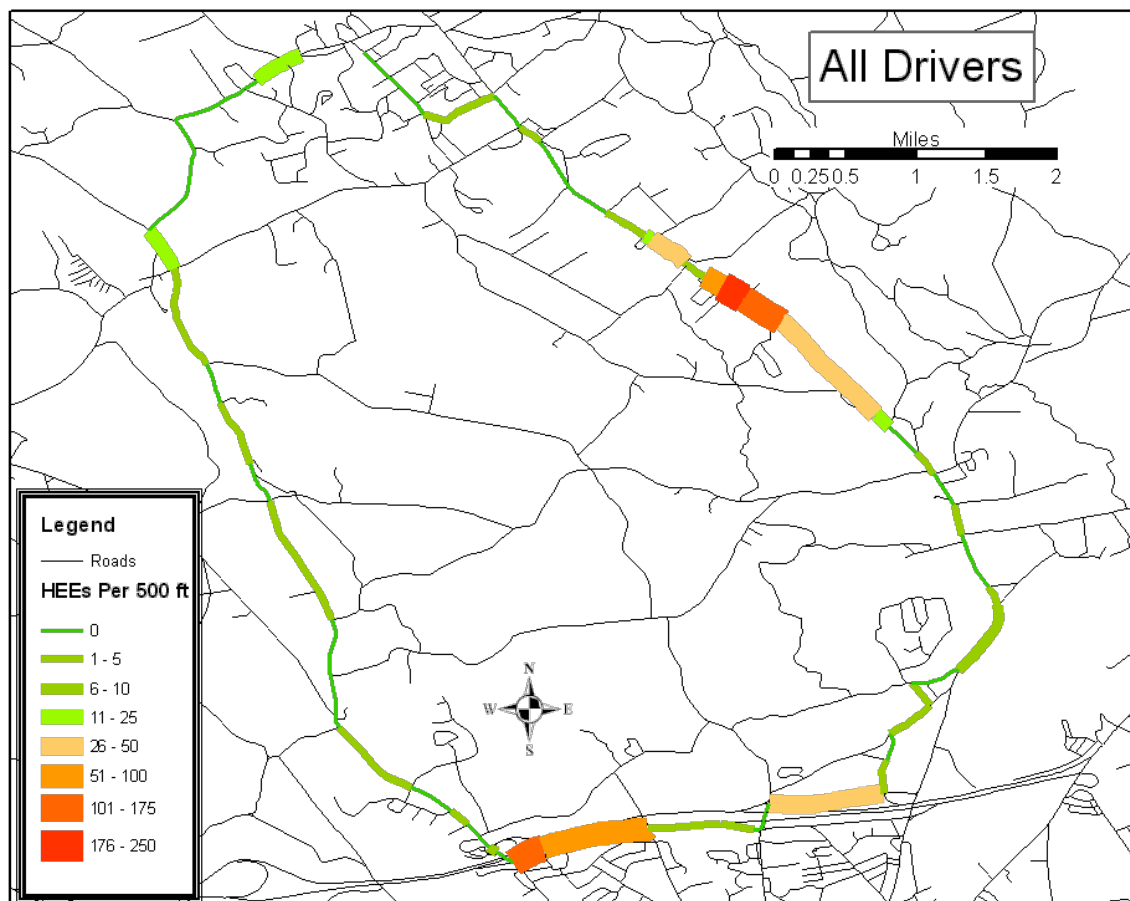


Figure 9: Spatial Location of High Emission Events

4.4 Particle Number Emissions and Vehicle Operating Parameters

Considering the downward trend in mean PN counts over the course of the experiment, the PN data was normalized by the mean PN emissions rate for each driver. The emission rate observations within the run were divided by the associated PN emissions rate mean for that run. This method preserves the time series pattern in PN emissions.

To explore PN emissions rates, box plots were generated to investigate the relationship between vehicle operation and emissions rate. In Figures 10, 11, and 12, the y-axis represents PN emissions rate expressed as the factor above the mean (FAM). In these figures, the median is again represented by the line within the box, the mean is represented by a black cross, and the upper and lower quartiles are represented by the extent of the boxed region. Note that the number of observations on the right side of the plots can be low. For example, accelerations over 5 mph/s are infrequent (< 0.5 % of the data) and of very short duration (most are less than 2 seconds in duration). This low frequency and duration may contribute to this unexpected drop in PN counts from the bins larger than 3 mph/s. Overall, the average PN emissions rate convincingly increases with vehicle speed, vehicle acceleration, and road grade.

Recent research suggests that VSP has the most significant relationship with increased tailpipe emissions. VSP is a combination of velocity, acceleration, and road grade and is a measure of power demand.

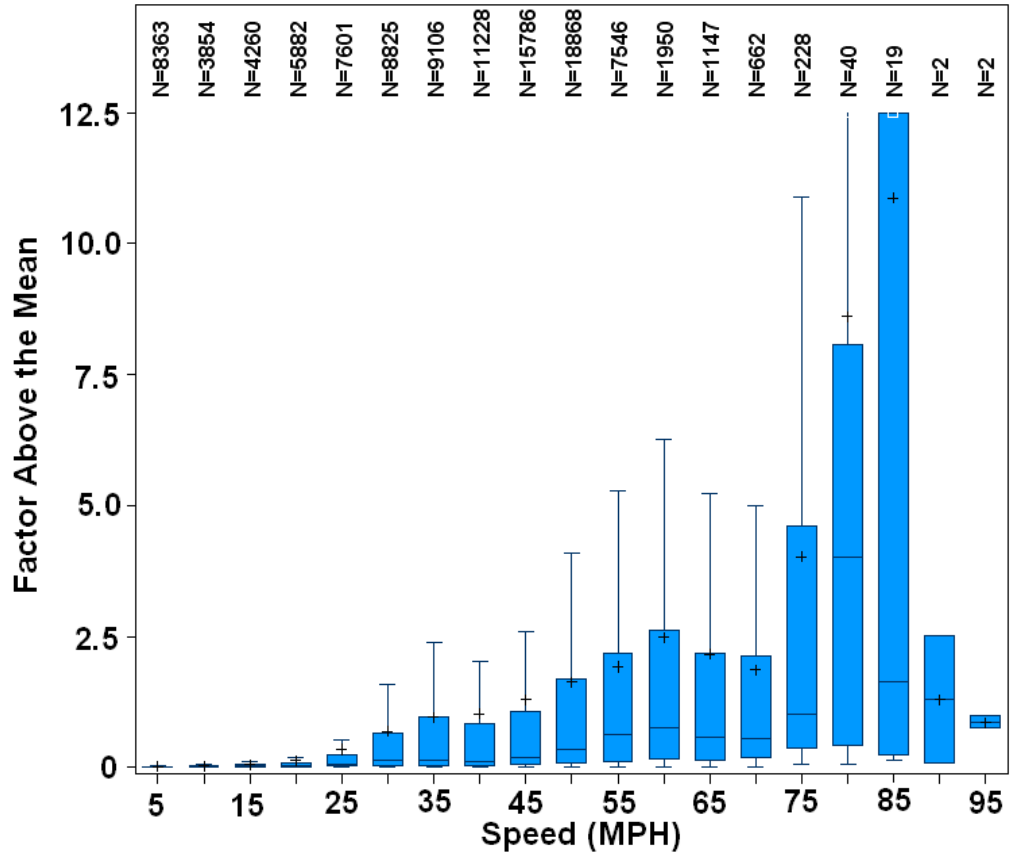


Figure 10: PN Emissions FAM Versus Speed Bin (MPH)

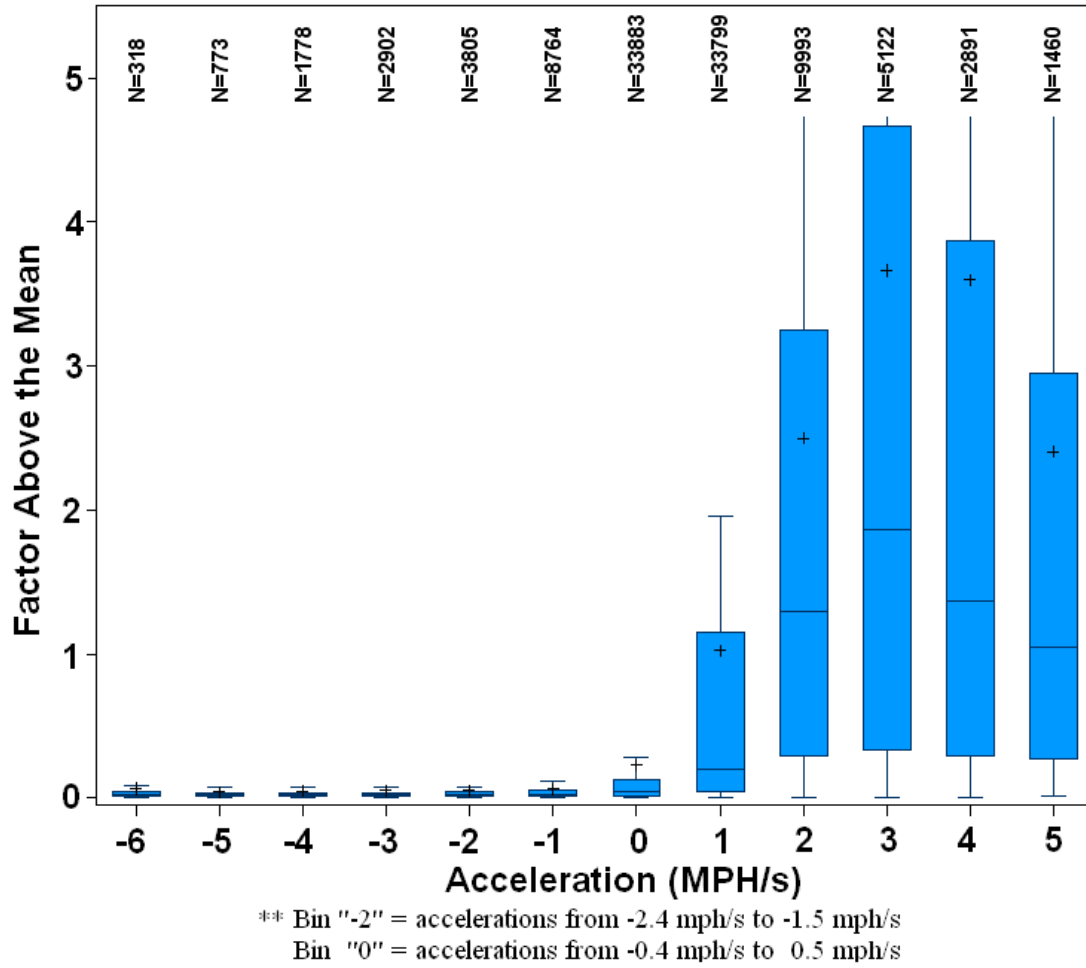


Figure 11: PN Emissions FAM Versus Acceleration Bin (MPH/s)

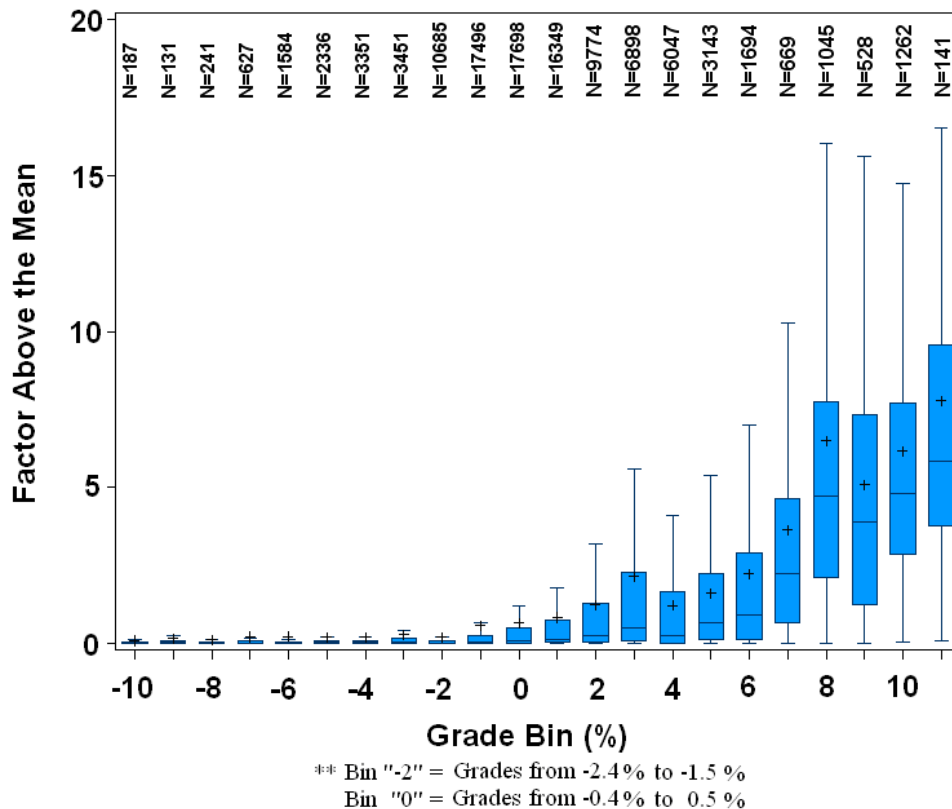


Figure 12: PN Emissions FAM Versus Grade Bin (%)

A correlation analysis was conducted to determine the extent to which each of the operating parameters was correlated with ultrafine PN number emission FAM (Table 3). The Pearson correlation analysis assumes that there is a linear relationship between the dependent and independent variables. Therefore, the log transform of the PN emission rate data had the highest correlation coefficients. RPM and VSP are highly correlated with ln(PN rate). Furthermore, speed, acceleration, and grade are also moderately correlated with ln(PN rate). The correlation between engine load and ln(PN rate) is relatively weak.

Not surprisingly, several predictor variables are correlated with each other. This correlation analysis suggests that there is a potential problem for development of a model due to multicollinearity between these predictor variables. While this multicollinearity will not affect the model outcome, it will cause the estimated parameter coefficients to be invalid, thus not allowing an accurate interpretation of how individual predictors influence PN emissions. For this reason and due to the undetermined influence of environmental factors described above, a model is developed here only for the purpose of assessing the extent to which vehicle and road parameters explain overall variance in PN emissions.

Table 3: Pearson Correlation Coefficients (All Are Significant $p < 0.001$)

Variable	Speed	Acceleration	Grade	VSP	RPM	Engine Load
Speed	1	-	-	-	-	-
Acceleration	0.25	1	-	-	-	-
Grade	0.09	0.32	1	-	-	-
VSP	0.54	0.79	0.46	1	-	-
RPM	0.62	0.68	0.32	0.77	1	-
Engine Load	0.16	0.33	0.21	0.37	0.38	1
Ln(PN Rate)	0.52	0.53	0.42	0.69	0.75	0.34
PN Rate	0.2	0.24	0.24	0.38	0.37	0.18

The model shown in Table 4 was estimated using a generalized linear model (GLM) least squares method. The extent of multicollinearity in the modeling process is assessed using variance inflation factor (VIF). There is no set cutoff for acceptable maximum values for VIF; however, Kleinbaum et al. (1998) state that a VIF above 10 suggests that there are strong multicollinearity issues between predictors and the parameter estimates may be inaccurate. No serious multicollinearity problems are suggested in this case. Table 4 contains the model parameters and statistics of significant predictors of the log-transformed particulate number emission rate.

Table 4: Model Parameters for Preliminary Linear PN Emissions Model

Parameter	Data Range	DF	Estimate	Standard Error	t value	Pr > t	VIF
Intercept	---	1	13.080	1.8E-02	16.1	<.0001	0.0
Engine RPM	750 to 4600	1	0.002	1.2E-05	149.5	<.0001	2.8
Speed	0 to 95	1	0.001	7.3E-05	8.2	<.0001	1.1
Acceleration	-10 to 9	1	0.052	3.9E-03	13.1	<.0001	2.1
Grade	-11% to 12%	1	0.097	1.9E-03	51.5	<.0001	1.6
Engine Load	0% to 100%	1	0.001	1.3E-04	7.2	<.0001	1.2
VSP	-300 to 300	1	0.008	1.4E-04	58.5	<.0001	2.9

The primary purpose of this preliminary modeling exercise was to identify relative importance of vehicle and road factors in ultrafine PN emissions. This model had a resulting R^2 of 0.61. Therefore, the operating and road factors can explain a majority of the variance in PN emissions and might be higher with a more sophisticated model.

5.0 Summary and Conclusions

Although state-of-the-art tailpipe emissions models are now based on vehicle operating mode, existing models and data collection have focused on either regulated gas emissions or mass-based particle emissions (PM). Yet public health research findings and policy questions now clearly suggest the importance of focusing on the number of fine and ultrafine particles emitted

from tailpipes (PN). While few databases can be found that contain time-resolved on-road vehicle emissions data, even fewer databases exist that focus on light-duty ultrafine particles. Therefore, the data in this research fill an important data gap. While a single vehicle was used, allowing driver and transportation network conditions to vary allowed focus on high particle number emission events and consideration of the relationship between PN emission rate and driver and road attributes. These unique time- and space-resolved data allow for conclusions of three types: 1) new knowledge regarding real-world light-duty ultrafine particle number emissions; 2) direction for future data collection; and 3) positive reinforcement of the possibility to extend current modeling frameworks for inclusion of ultrafine particle number emissions.

In terms of new knowledge, these data have demonstrated that high ultrafine PN emissions events, each of one second in duration, are very important to overall PN emissions levels. Only 1.6% of the driving time monitored in the experiment resulted in 30% of ultrafine particles emitted. Road grade and vehicle acceleration were key factors in the occurrence of high emissions events. Engine operating variables and road conditions account for approximately 60% of the variation in PN emissions. This is high enough to encourage the use of existing modeling frameworks that employ operating model for prediction, but it is low enough to recommend further data collection. In particular, the importance of humidity, temperature, and background atmospheric conditions require focus in future work.

Important directions for future data collection are gleaned from this work. First, on-road time-resolved emissions data are critical to allow for spatially disaggregate models that will allow evaluation of alternative designs for physical infrastructure as well as traffic control. Lab dynamometers can be used for emissions rate development, but on-road experiments such as the one described here are essential to understanding which operating modes occur where in the transportation system and also to consider the impact of real-world environmental conditions on emissions. This experiment clearly demonstrated that driving style is consistent. This will allow future work to include more individual drivers and fewer repeats by each individual. The next field experiments might use the same drivers in different vehicles. This experiment also revealed the need for more robust methods to capture atmospheric conditions such as background PN concentrations, temperature, and humidity.

In terms of modeling, it is important to be reassured based on real-world data that existing operating mode-based models are viable frameworks for expansion to include PN emissions. Moreover, within existing models the grade component is often not included in vehicle-specific power calculation because grade is not known for the real-world network. This study clearly demonstrated that grade must be used in regional and project-level modeling.

In summary, this study has established that vehicle operation, driver style, and atmospheric factors are important predictors of light-duty PN emissions. Given public health concerns, existing models could be extended to include these emissions if more time- and space-resolved data were collected. Dynamometer studies will play an important role in gathering the large, comprehensive data needed for defensible emissions rates. But future on-road data collection will be needed to advance our understanding of the role of atmospheric conditions as well as to better refine driving style by location to enable models to be used for project-level analysis,

including identification of pollution “hot-spots”. Future traffic control and roadway design considerations may then be evaluated to reduce the occurrence of hot-spots.

References

Barth, M., An, F., Younglove, T., Scora, G., Levine C., 2001. Development of a Comprehensive Modal Emissions Model. NCHRP Web-Only Document 122: National Cooperative Highway Research Program Project 25-11.

Burtscher, H., 2001. Literature Study on Tailpipe Particulate Emission Measurement for Diesel Engines. Report for the Particle Measurement Program (PMP) for BUWAL/GPRE, Windisch Switzerland.

Clean Air Technologies, 2001. Erie County Portable Emissions Monitoring Project. Project Report by Clean Air Technologies International. <http://www.cleanairt.com/eriecoMAIN.pdf>.

Dockery, D., Pope III, A., Xu, X., Spengler, J., Ware, J., Fay, M., Ferris Jr., B., Speizer, F., 1993. An Association Between Air Pollution and Mortality in Six U.S. Cities. *Journal of Medicine* 329, 1753–1759.

Domínguez-Sáez, A., Viana, M., Barrios, C., Rubio, J., Amato, F., Pujadas, M., Querol, X., 2012. Size-Resolved Particle Number Emission Patterns under Real-World Driving Conditions Using Positive Matrix Factorization. *Environ. Sci. Technol.*, 2012, 46 (20), 11187–11194.

Englert, N., 2004. Fine Particles and Human Health – A Review of Epidemiological Studies. *Toxicology Letters* 149, 235-242.

EPA, 2004. Air Quality Criteria for Particulate Matter (October 2004). U.S. Environmental Protection Agency, Washington, DC, EPA 600/P-99/002aF-bF.

Frey C., Unal, A., Roupail, N., Coylar, J., 2003. On-Road Measurement of Vehicle Tailpipe Emissions Using a Portable Instrument. *Journal of the Air and Waste Management Association* 53, 992-1002.

Frey C., Roupail, N., Unal, A., Colyar, J., 2001. Emissions Reduction Through Better Traffic Management: An Empirical Evaluation Based Upon On-Road Measurements, North Carolina State University Department of Civil Engineering, Raleigh, NC.

Harrison, R., Jones A., 2005. Multisite Study of Particle Number Concentrations in Urban Air. *Environ. Sci. Technol.*, 39 (16), 6063–6070.

Harrison, R., Yin. J., 2000. Particulate Matter in the Atmosphere: Which Particle Properties are Important for its Effects on Health? *The Science of the Total Environment* 249 85-101.

Health Effects Institute (HEI), 1999. Program Summary: Research on Diesel Exhaust. Available at <http://pubs.healtheffects.org/>.

Health Effects Institute (HEI), 2003. Program Summary: Research on Diesel Exhaust and Other Particles. Available at <http://pubs.healtheffects.org/>.

Heywood, J.B. 1988. Internal Combustion Engine Fundamentals, McGraw-Hill, New York.

Hildemann, J., Markowski, G., Cass, G., 1991. Chemical Composition of Emissions from Urban Sources of Fine Organic Aerosol. *Environmental Science and Technology* 25, 744–759.

Huai, T., Durbin, T., Yunglove, T., Scora, G., Barth, M., Norbeck, J., 2005. Vehicle Specific Power Approach to Estimating On-Road NH₃ Emissions from Light-Duty Vehicles. *Environmental Science and Technology*, 39(24), 9595-9600.

Jackson, E., Aultman-Hall, L., 2010. Analysis of Real-World Lead Vehicle Operation for Integration of Modal Emissions and Traffic Simulation. *Transportation Research Record*, 2128.

Jackson, E., Holmén, B., 2009. Modal Analysis of Vehicle Operation and Particulate Emissions from Connecticut Transit Buses. *Transportation Research Record* 2123, 76-87.

Jackson, E., Qu, Y., Holmén, B., Aultman-Hall, L., 2006. Driver and Road Type Effects on Light-duty Gas and Particulate Emissions. *Transportation Research Record* 1987, 118-127.

Jimenez, J., 1999. Understanding and Quantifying Motor Vehicle Emissions with Vehicle Specific Power and TILDAS Remote Sensing. Ph.D. Thesis, Massachusetts Institute of Technology.

Johnson, L., Ferreira, L., 2011. Modeling particle emissions from traffic flows at a freeway in Brisbane, Australia. *Transportation Research Part D: Transport and Environment*, 6(5), 357-369.

Joumard, R., Jost, P., Hickman, J., Hassel, D., 1995. Hot passenger car emissions modeling as a function of instantaneous speed and acceleration. *Science of The Total Environment*, 169(1-3); *Transport and Air Pollution*, 167-174.

Kamarianakis, Y., Gao, O., Holmén, B., Sonntag, D., 2011. Robust modeling and forecasting of diesel particle number emissions rates. *Transportation Research Part D: Transport and Environment*, 16(6) 435-443.

Kittelson, D., Watts, W., Johnson, J., Schauer, J., Lawson, D., 2006. On-Road and Laboratory Evaluation of Combustion Aerosols—Part 2: Summary of Spark Ignition Engine Results. *Journal of Aerosol Science*, 37(8), 931-949.

Kittelson, D., 1998. Engines and Nanoparticles: A Review. *Journal of Aerosol Science*, 29(5/6), 575-588.

Kleeman, M., Schauer, J., Cass, G., 2000. Size and Composition Distribution of Fine Particulate Matter Emitted from Motor Vehicles. *Environmental Science and Technology*, 34, 1132–42.

Kleinbaum, D., Kupper, L., Muller, K., Nizam, A., 1998. Applied Regression Analysis and Other Multivariable Methods, 3rd ed. Pacific Grove, CA: Brooks/Cole Publishing Company.

Kuhns, H., Mazzoleni, C., Moosmuller, H., Nikolic, D., Keislar, R., Barber, P., Li, Z., Etyemezian, V., Watson, J., 2004. Remote sensing of PM, NO, CO, and HC emission factors for on-road gasoline and diesel engine vehicles in Las Vegas, NV. *Science of the Total Environment*, 322(1-3), 123-137.

Lawson, D., and Smith, R., 1998. The Northern Front Range Air Quality Study A Report to the Governor and General Assembly. Colorado State University, December.

Mazzoleni, C., Moosmüller, H., Kuhns, H., Keislar, R., Barber, P., Nikolic, D., Nussbaum, N., Watson, J., 2004. Correlation between automotive CO, HC, NO, and PM emission factors from on-road remote sensing: implications for inspection and maintenance programs. *Transportation Research Part D: Transport and Environment*, 9(6), 477-496.

McGregor, F., Ferreira, L., Morawska, L., 2003. Modelling of sub-micrometer particle concentrations in free-flowing freeway traffic, Brisbane, Australia: some empirical results. *Transportation Research Part D: Transport and Environment*, 8(3), 229-241.

Morawska, L., Jamriska, M., Thomas, S., Ferreira, L., Mengersen, K., Wraith, D., McGregor, F., 2005. Quantification of Particle Number Emission Factors for Motor Vehicles from On-Road Measurements. *Environ. Sci. Technol.*, 39 (23), 9130–9139.

Nemmar, A., Hoet, P., Vanquickenborne, B., Dinsdale, D., Thomeer, M., Hoylaerts, M., (2002). Passage of inhaled particles into the blood circulation in humans. *Circulation* 105:411–414.

North, R., Noland, R., Ochieng, W., Polak, J., 2006. Modelling of particulate matter mass emissions from a light-duty diesel vehicle. *Transportation Research Part D: Transport and Environment*, 11:5, 344-357.

Pokharel, S., Bishop, G., Stedman, D., 2001. On-Road Remote Sensing of Automobile Emissions in the Los Angeles Area: Year 2. Univ. of Denver final report to the Coordinating Research Council, Inc., available at <http://www.crcao.com>.

Qu, Y., Ravishanker, N., Holmén, B., 2008. Predicting Light-Duty Gasoline Vehicle On-Road Particle Number Emissions From Gas Emissions Using A Time-Series Cross-Section Regression Approach. *Transportation Research Record* 2058, 97-105.

Pope III, C., Thun, M., Namboodiri, M., Dockery, D., Evans, J., Speizer F., Heath Jr. C., 1995. Particulate Air Pollution as a Predictor of Mortality in a Prospective Study of U.S. Adults. *American Journal of Respiratory Critical Care Medicine*, 151, 669–674.

Rakha H., Ahn K., and Trani A., 2004. Development of VT-Micro Framework for Estimating Hot Stabilized Light Duty Vehicle and Truck Emissions. *Transportation Research, Part D: Transport & Environment*, Vol. 9(1), January, 49-74.

Riedl, M., Diaz-Sanchez, D., 2005. Biology of Diesel Exhaust Effects on Respiratory Function. *Molecular Mechanisms in Allergy and Clinical Immunology. Journal of Allergy and Clinical Immunology*, 115(2) 221-228.

Ristovski, Z., Jayaratne, E., Morawska, L., Ayoko G., Lim, M., 2005. Particle and carbon dioxide emissions from passenger vehicles operating on unleaded petrol and LPG fuel. *Science of the Total Environment*, 345, 93-98.

Roadware Inc., 2007. Roadware ARAN brochure
http://www.roadware.com/_lib/pdf/datasheet.icon_sheet.pdf.

Robinson, M., Holmén, B., 2011. Onboard, Real-World Second-by-Second Particle Number Emissions from 2010 Hybrid and Comparable Conventional Vehicle, *Journal of the Transportation Research Board*, 2233, 63-71.

Robinson, M., Sentoff, K., Holmén, B., 2010. Particle Number and Size Distribution Emissions During Light-Duty Vehicle Cold Start Using the Total On-Board Tailpipe Emissions Measurement System. *Transportation Research Record*, 2158, 86-94.

Singer, B., Harley, R., 2000. A Fuel-Based Inventory of Motor Vehicle Exhaust Emissions in the Los Angeles Area During Summer 1997. *Atmospheric Environment*, 34, 1783-1795.

Sonntag, D., Gao, O., 2009. Developing link-based particle number emission models for diesel transit buses using engine and vehicle parameters. *Transportation Research Part D: Transport and Environment*, 14(4), 240-248.

Vikara, D., Holmén, B., 2007. Ultrafine Particle Number Concentrations from Hybrid Urban Transit Buses Using Onboard Single-Diameter Scanning Mobility Particle Size Measurements. *Transportation Research Record*, 1987, 54-61.

Wang, X., Gao, O., 2011. Exposure to fine particle mass and number concentrations in urban transportation environments of New York City. *Transportation Research Part D: Transport and Environment*, 16(5), 384-391.

Weijers, E., Khlystov, A., Kos, G., Erisman, J., 2004. Variability of Particulate Matter Concentrations Along Roads and Motorways Determined by a Moving Instrument Unit. *Atmospheric Environment*, 38, 2993-3002.