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To link to this article: http://dx.doi.org/10.1080/10473289.2004.10470905

Published online: 21 Feb 2012.

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A Quantitative Description of Vehicle Exhaust Particle Size Distributions in a Highway Tunnel

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ABSTRACT
During the period May 18–May 22, 1999, a comprehensive study was conducted in the Tuscarora Mountain Tunnel on the Pennsylvania Turnpike to measure real-world motor-vehicle emissions. As part of this study, size distributions of particle emissions were determined using a scanning mobility particle sizer. Each measured size distribution consisted of two modes: a nucleation mode with midpoint diameter less than 20 nm and an accumulation mode with midpoint diameter less than 100 nm. The nucleation and accumulation components in some distributions also exhibited second maxima, which implies that such particle size distributions are superpositions of two particle size distributions. This hypothesis was utilized in fitting the particle size distributions that exhibited second maxima with four lognormal distributions, two for the nucleation mode and two for the accumulation mode. The fitting assumed that the observed particle size distribution was a combination of two bimodal lognormal distributions, one attributed to the heavy-duty diesel (HDD) vehicles and another attributed either to a different class of HDD vehicles or to the light-duty spark ignition vehicles. Based on this method, estimated particle production rates were $1.8 \times 10^{13}$ and $2.8 \times 10^{14}$ particles/vehicle-km for light-duty spark ignition and HDD vehicles, respectively, which agreed with independently obtained estimates.

INTRODUCTION
Monitoring and characterizing airborne particulate matter (PM) has been an important research area because of the impact of PM on human health, visibility reduction, and material damage. As major contributors to ambient airborne PM levels, both light-duty spark ignition (LDSI) and heavy-duty diesel (HDD) vehicles are subject to numerous control measures. To comply with PM standards, low PM emission diesel engines also have been developed. Despite reduced PM emissions, motor vehicles may emit significant numbers of nanoparticles (particles with aerodynamic diameter less than 50 nm). Because of their low inertia, these particles can penetrate deeply into the lungs, increasing negative health effects.

Previous studies have found that HDD vehicle exhaust fine particle size distributions tend to be bimodal and lognormal in form, with a nuclei mode in the 7–20 nm diameter range and an accumulation mode in the 30–70 nm range. Researchers have hypothesized that nanoparticles are formed during exhaust dilution and cooling from sulfuric acid ($H_2SO_4$), the soluble organic fraction, and polycyclic aromatic hydrocarbons (PAHs).

Available information on vehicle exhaust fine particles is based, mainly, on dynamometer studies, where the dilution ratio is on the order of 1:10. In many cases, high inlet temperature evaporates volatile hydrocarbons from the surface of the sampling system, producing artifact particles in the nucleation mode. An on-road experiment conducted in the Tuscarora Mountain Tunnel located on the Pennsylvania Turnpike observed nanoparticle formation and preservation under dilution ratios exceeding 1:1000.

One interesting feature of the Tuscarora experiment data is that in some of the particle size distributions (e.g., Figures 3, 5, and 11 in Abu-Allaban et al.,), the nucleation and accumulation modes exhibited clear second local maxima. Two different scenarios might account for the origin of the second maxima. One is the measurement method, but the second maximum was observed in only a few cases. Had it been an instrumental issue, the second maximum should have appeared in all cases. A second
The scenario is the existence of another nucleation or accumulation mode, analogous to the presence of a shoulder on a spectroscopic peak. Based on this observation, it is hypothesized that the multiple maxima observed in some of the Tuscarora size distributions are caused by superposition of particle size distributions.

Whether the distributions are caused by different emission sources (e.g., HDD vs. LDSI vehicles) is not straightforward because distributions depend on both the age of those particles and the emission source. If particles form in the ambient air a long time after leaving the tailpipe, it is difficult to determine which particles come from which vehicle. Each vehicle contributes to the ambient concentration of the gaseous precursors until reaching supersaturation, which leads to heterogeneous particle formation. In addition, aged particles from different sources tend to coagulate with time to form larger particles. These phenomena could limit the appearance of distinct size modes from the different source emissions.

According to Kittelson and Abdul-Khalek, nucleation of most of the observed nanoparticles takes place in the exhaust just after gases exit the tailpipe and mix with ambient air. This finding resolves the issue of where particle formation takes place. The particle age issue could be resolved by sampling fresh particles close to their sources. Because the average air residence time within the Tuscarora Tunnel was 5 ± 1 min, it is thought that fresh nanoparticles emitted from different engines are different and their identities are preserved for short time periods.

Another issue is whether the measured particle size distribution can be decomposed into different components and each component attributed to its source. To be able to do this, it is necessary to characterize the particle size distribution for each particle source. This may include distribution shape, peak position, and peak width. If the characteristics of the particle size distribution depend on the source and each source has a unique characteristic, it is possible to apportion any given particle size distribution into distributions that can be attributed to specific local sources.

Traditional source attribution techniques, such as the chemical mass balance (CMB) receptor model, depend on the chemical profile of species present in the collected PM samples. These techniques require extensive sampling and laboratory analysis. In addition, in some cases there is chemical colinearity between different sources (e.g., gasoline/diesel, diesel/meat cooking, motor vehicles/vegetative burning). Source apportionment based on the particle size distribution may complement the CMB to a significant degree by resolving the issue of chemical colinearity. In this paper, steps are outlined for applying this technique to apportion particle emissions based on the observed size distributions and to estimate particle production rates for LDSI and HDD vehicles measured in a highway tunnel study.

**EXPERIMENTAL DESCRIPTION**

Measurements were made of mobile source particle emission size distributions in the south bore of the Tuscarora Mountain Tunnel. The Tuscarora Mountain Tunnel is a two-bore tunnel, two lanes each bore, 1623.2 m long that carries the Pennsylvania Turnpike (Interstate 76) through Tuscarora Mountain in south-central Pennsylvania at an altitude of ~305 m. The road is flat (an upward grade of 0.30% toward the middle from either end) and straight. Average vehicle speed in the tunnel was 92 ± 5 km/hr determined using a radar gun. Vehicle emissions were measured in the eastbound bore. The nearest interchange (10 km west of the tunnel entrance) is lightly used. Other accesses are the Sideling Hill service plaza (22 km to the west), the interchange with Interstate 70 (40 km to the west, heavily used), and other interchanges and service plazas farther west. Effectively, the minimum trip length before reaching the tunnel is 15 min (much of it after a hot start). It is estimated that 75% of all vehicles are on the road for more than 50 min before reaching the tunnel. Accordingly, cold-start and hot-start operations are inconsequential in the eastbound traffic.

The tunnel is ventilated entirely by the traffic piston effect and the prevailing winds. The mechanical ventilation system was not operated during the experiment (as is typical during normal operation of this tunnel). This situation is ideal for emission rate calculations and is well suited to the study of PM: no fresh air injections occur within the tunnel and none of the particulates are removed, as is the case in many transverse-vented U.S. tunnels. Mean residence time of air within the tunnel was 5 ± 1 min, calculated from the tunnel length and measured wind speeds. Wind speed was measured using two propeller anemometers situated on the walkways adjacent to the road inside the tunnel.

Experimental runs were performed between May 18 and May 23, 1999. All experimental periods lasted 1 hr. Within each period, 5–10 scans were performed. Periods were chosen to maximize the range in the fraction of HDD vehicles. For example, late night runs tended to be dominated by HDD traffic, while midday and weekend runs had high fractions of LDSI vehicles. The LDSI vehicles traveling through the tunnel tended to be quite new, with a mean age of 5 yr (the mean model year ranged from 1991.1 to 1995.0). Very few pre-1980 vehicles were observed.

Particle number distribution measurements were made using a scanning mobility particle sizer (SMPS). The SMPS utilizes an electrostatic classifier (Model 3071; TSI, Inc.) to bin the particles according to electrical
mobility. The unit effectively operates on the size axis of 0.01–0.5 μm mobility-equivalent diameter. Once the particles are size-binned by the classifier, they are counted by the second component of the SMPS, which is a condensation particle counter (CPC Model 3022A; TSI). The CPC counts particles by optically detecting butanol droplets that have grown on the particles under high-supersaturation conditions. Size distribution measurements were conducted at either 60- or 90-sec scan times. SMPS measurement uncertainties are discussed in Rogers et al.14

The SMPS is conventional in all respects except for the interfacing aerosol connections. Because of the age of the 85Kr neutralizer, an external neutralizer was connected to the SMPS inlet. This unit contained 3.0 mCi 210Po in commercial antistatic strips manufactured in April 1999. The sample inlet of the SMPS was reconfigured to a total length of 25 cm to minimize the time delay. The aerosol sample line connecting the electrostatic classifier and the CPC is not a standard TSI product but rather is 29% longer than standard. A longer tubing connection implies that an increased delay time is needed (measure of the travel time of the aerosol sample between the classifier and the CPC). Therefore, the default delay time settings were increased by 29% in post-processing of the data. The delay time can be varied in post-processing these data. For the Tuscarora Tunnel data, the effect of the delay time correction was a decrease of 2–4 nm in the count median diameter of the size distributions. The peak diameters of the most prominent modes of each distribution decreased 2–3 nm. Therefore, the decreases caused by the delay time correction were not much above uncertainty levels.14 The daily measurement routine included checking the zero of the instrument. The indicated particle count typically was less than 0.01 particle/cm³ before each scan.

RESULTS AND DISCUSSION

Table 1 shows the number of vehicles and average particle concentrations within each of four sampling intervals. It is clear from Table 1 that the intervals with highest fraction of HDD vehicles exhibited the highest particle concentrations. This implies that HDD vehicles are a potential source of observed particles, which agrees with previous studies.4,5

The particle size distributions consisted of two modes (Figure 1), a dominant nucleation mode in the size range 8–40 nm and a shallow accumulation mode in the size range 40–300 nm. The presence of the dominant nucleation mode supports laboratory findings that diesel vehicles emit a high number of ultrafine particles in the nucleation mode.4,5 Furthermore, results indicate that nanoparticles are preserved under real-world conditions of dilution and mixing.

| Table 1. Sampling intervals, traffic features, and average particle concentrations. |
|---------------------------------|-----------------|-----------------|-----------------|-----------------|
| Date                            | Interval 1      | Interval 2      | Interval 3      | Interval 4      |
| Start time                      | May 18, 1999    | May 20, 1999    | May 20, 1999    | May 22, 1999    |
| End time                        | 10:00 p.m.      | 1:00 a.m.       | 5:00 p.m.       | 1:00 p.m.       |
| End time                        | 11:00 p.m.      | 2:00 p.m.       | 6:00 p.m.       | 1:00 p.m.       |
| Relative humidity (%)           | 69              | 62              | 28              | 37              |
| LDSI vehicles                   | 104             | 43              | 505             | 467             |
| HDD vehicles                    | 189             | 158             | 225             | 80              |
| Total                           | 293             | 201             | 730             | 547             |
| HDD (%)                         | 64.5            | 78.6            | 30.8            | 15.2            |
| Average particle concentration  | 104,136         | 108,502         | 74,740          | 61,715          |

To provide a quantitative description of size distributions, averages of particle size distributions were fitted within each measurement interval with bimodal logarithmic distributions using Origin-6 software (OriginLab Corp.). Figure 1 presents the result for the 64.5% HDD run. The fitting parameters (mean diameter and standard deviation) are summarized in Table 2. In all cases, the nucleation mode constituted more than 86% of the total area, supporting the observation that the emitted particle counts are dominated by the nucleation mode.

Second maxima are observed in the nucleation or accumulation peaks of some size distributions (Figures 2–5). As mentioned previously, if the particle size distribution of each vehicle class (HDD and LDSI) is lognormal by itself, then we could be seeing a superposition of size distributions from LDSI and HDD vehicles analogous to the spectroscopic case. Alternatively, the measured particle size distributions could be a superposition of size distributions from HDD vehicles operating under different conditions including speeds, loads, years of manufacture, engine manufacture, and maintenance. Based on a multiple sources assumption, nucleation mode peaks of the particle size distribution in Figures 2–5 were fitted with two lognormal distributions.

For the case where the HDD fraction was 15.2%, two peaks in the nucleation mode were located at 9.2 and 15.9 nm and 9.3 and 15.0 nm for the size distributions in Figures 2 and 3, respectively. Following the argument that HDD vehicles emit more particles than LDSI vehicles, it was hypothesized the smaller peaks in the nucleation mode (with midpoint diameters of 9.2 and 9.3 nm) to be caused by LDSI vehicle emissions, while the larger nucleation peaks (with midpoint diameters of 15.0 and 15.9 nm) were caused by HDD vehicle emissions.

Alternatively, splitting of the nucleation peak in Figures 4 and 5 may not be caused by LDSI vehicles because HDD vehicles dominated these measurement periods. The observed second maximum could be caused by class
4–6 vehicles (medium diesel and spark ignition trucks), model year distribution of trucks, or speed variability during the measurement period. Providing an exact explanation requires further investigation.

**Estimate of Particle Production Rates**

To further investigate the hypothesis that double maxima in the particle size distributions indicate two or more sources, particle production rates were examined for different vehicle classes. Particle production rates were calculated using the following equation:15

\[ P = \frac{C \times V \times A}{L \times N} \]  

(1)

where \( P \) is the particle production rate per vehicle-km; \( C \) is the particle concentration measured at the outlet and corrected for the inlet contribution; \( L \) is the length of the tunnel; \( N \) is the number of vehicles passing through the tunnel in 1 hr; \( V \) is the measured wind speed in the tunnel; and \( A \) is the cross-sectional area of the tunnel.

To calculate particle production rates for different vehicle classes, it is necessary to know the particle concentration (\( C \)) related to each vehicle class. In the Tuscarrora Mountain Tunnel experiment, this was difficult because any measured concentration was a result of a mixture of different vehicle classes. The particle size distribution decomposing technique, however, provides fractional LDSI and HDD contributions to the measured concentration. Figures 2 and 3 show a superposition of two distributions, each a bimodal lognormal distribution. The first distribution consists of the first and third components and was attributed to LDSI vehicles. The second distribution consists of the second and the fourth components and was attributed to HDD vehicles. According to the fitting results, LDSI and HDD fractional contributions to the total measured particle size distribution were 0.27 and 0.73, respectively. This implies that for the 61,715 cm\(^3\) concentration measured in case 4 (see Table 1), 16,725 cm\(^3\) were caused by LDSI vehicles and 44,990 cm\(^3\) were caused by HDD vehicles. Based on these concentrations, emission rates were \(1.8 \times 10^{13}\) and \(2.8 \times 10^{14}\) particles/vehicle-km for LDSI and HDD vehicles, respectively.

The particle emission rate of \(2.8 \times 10^{14}\) for HDD vehicles calculated in this study is consistent with the results of Abu-Allaban et al.\(^6\) for the 78.6% HDD case (i.e., \(3.1 \times 10^{14}\) particles/vehicle-km). The slight increment in the 78.6% case may be because the ambient temperature was lower than in the 15.2% case, which can enhance the particle formation rate.\(^{16}\)

Rickeard et al.\(^{17}\) reported that at 120 km/hr, the gasoline car production rate was \(\sim 1 \times 10^{14}\) particles/vehicle-km.
vehicle-km, while at 50 km/hr, the production rate was reduced by a factor of $\frac{1}{2}$ from this value. Graskow et al.\textsuperscript{18} found that at low-load, “cruising” conditions of 120 km/hr, emission rates were as low as $3 \times 10^{11}$ particles/vehicle-km. The particle production rate was found to increase by more than 2 orders of magnitude as the rpm and manifold pressure were increased from 1500 to 2500 rpm, and from 50 to 90 kPa, respectively.

The finding for LDSI vehicles ($1.8 \times 10^{13}$ particles/vehicle-km) is not too dissimilar with the Rickeard et al.\textsuperscript{17} and Graskow et al.\textsuperscript{18} results when it is considered that the LDSI vehicles were operating at 80–100 km/hr under minimal load and hot stabilized conditions, which minimizes particle production rates.

**CONCLUSIONS**

Vehicle particle size distributions were determined using an SMPS. In some of the size distributions, the nucleation and accumulation modes exhibited a clear second maximum. The existence of the maximum in the nucleation or accumulation mode was hypothesized to be because of the superposition of two bimodal lognormal particle size distributions.
distributions that were attributed to LDSI and HDD vehicle emissions. Fitting the observed size distributions with two bimodal lognormal size distributions enabled the calculation of the fractional contribution of each vehicle class to the total size distribution. The LDSI and HDD fractional contributions then were utilized to estimate particle production rates. Using this methodology, estimated particle production rates were $1.8 \times 10^{13}$ and $2.8 \times 10^{14}$ particles/vehicle-km for LDSI and HDD vehicles, respectively. These results were consistent with LDSI and HDD particle production rate estimates obtained by other methods and investigators, which provide confidence that the particle-size distribution decomposing technique is effective and could be used to apportion ambient nanoparticles to specific local sources.

**ACKNOWLEDGMENTS**
The Tuscarora Mountain Tunnel study was funded by the Health Effects Institute under Research Agreement #98-26.
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