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Size-resolved particle number and volume emission factors for on-road gasoline and diesel motor vehicles

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ABSTRACT

Average particle number concentrations and size distributions from ~61,000 light-duty (LD) vehicles and ~2500 medium-duty (MD) and heavy-duty (HD) trucks were measured during the summer of 2006 in a San Francisco Bay area traffic tunnel. One of the traffic bores contained only LD vehicles, and the other contained mixed traffic, allowing pollutants to be apportioned between LD vehicles and diesel trucks. Particle number emission factors (particle diameter $D_p > 3$ nm) were found to be $(3.9 \pm 1.4)\times10^{14}$ and $(3.3 \pm 1.3)\times10^{15}$ # kg^{-1} fuel burned for LD vehicles and diesel trucks, respectively. Size distribution measurements showed that diesel trucks emitted at least an order of magnitude more particles for all measured sizes ($10 < D_p < 290$ nm) per unit mass of fuel burned. The *relative* importance of LD vehicles as a source of particles increased as D_p decreased. Comparing the results from this study to previous measurements at the same site showed that particle number emission factors have decreased for both LD vehicles and diesel trucks since 1997. Integrating size distributions with a volume weighting showed that diesel trucks emitted 28 ± 11 times more particles by volume (per unit fuel) than LD vehicles, consistent with the diesel/gasoline emission factor ratio for $PM_{2.5}$ mass measured using gravimetric analysis of Teflon filters, reported in a companion paper.

1. Introduction

Particulate matter (PM) emissions from motor vehicles are regulated on a mass basis, whereas particle number emissions are unregulated. Particulate mass and number concentrations show little correlation since most particles are emitted in the nuclei mode (particle diameter $D_p < 50$ nm), which have negligible mass. There are fewer particles emitted in the accumulation mode ($100 < D_p < 2000$ nm), but these are responsible for the majority of exhaust particulate mass (Kittelson, 1998).

Particles of various sizes and compositions originate from different phases of the combustion process (Kittelson, 1998; Seigneur, 2009). Accumulation mode particles in diesel engines are carbonaceous soot agglomerates, formed early in the combustion process within fuel-rich pockets inside the engine cylinder. Nuclei mode particles are formed in diesel and gasoline engines when hydrocarbons and sulfates, stemming from vaporized lubricating oil and fuel, nucleate as exhaust dilutes and cools. The nuclei mode is highly dependent on the degree of supersaturation of the nucleating species. High concentrations of accumulation mode particles in engine exhaust suppress particle formation by scavenging or sorbing precursors needed for nucleation to occur (Ban-Weiss, Lunden, Kirchstetter, & Harley, 2009; Kittelson, Watts, & Johnson, 2006).

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Particle size is important in determining health and environmental impacts of PM. Ultrafine particles ($D_p < 100$ nm) have been identified as a particular concern for human health (Pope & Dockery, 2006; Pope et al., 1995). Laboratory studies have shown that particles that are non-toxic with $D_p \sim 1$ μm can be toxic when $D_p \sim 10$ nm (Donaldson, Beswick, & Gilmour, 1996; Seaton, Macnee, Donaldson, & Godden, 1995). Nuclei mode particles can penetrate deeply into the lung and enter the circulatory system, whereby they may deposit in other vital organs such as the brain or heart (Kennedy, 2007). It has been suggested that adding particle number-based air quality standards and/or engine emissions limits to the current mass-based limits could help in identifying and reducing adverse health impacts (Kennedy, 2007). Since nuclei mode particles in the atmosphere coagulate with accumulation mode particles in minutes to hours, and can evaporate at even faster time scales (Seigneur, 2009), health effects are especially detrimental to those in close proximity of fresh emissions. The residence time of accumulation mode particles in the atmosphere is ~1 week, orders of magnitude longer than that of ultrafine particles. Light absorption is stronger than scattering for soot particles in the accumulation mode with important implications on regional visibility degradation and global warming (Ramanathan & Carmichael, 2008).

There is an ongoing debate about the importance of gasoline vs. diesel engines as sources of fine PM (Johnson, Kittelson, & Watts, 2005). Results from a companion paper (Ban-Weiss et al., 2008) indicate that on average, diesel engines emit an order of magnitude more PM mass than gasoline engines, per unit of fuel burned. But the *relative* importance of gasoline vehicles as a source of PM is higher when considering number rather than mass emissions; the *fraction* of particles that are emitted in the nuclei mode is higher in gasoline vs. diesel engines (Graskow, Kittelson, Abdul-Khalek, Ahmadi, & Morris, 1998; Johnson et al., 2005; Kayes, Hochgreb, Maricq, Podsiadlik, & Chase, 2000; Maricq, Podsiadlik, & Chase, 1999a, 1999b). However, variations in sampling protocols, both in terms of driving cycles and PM measurement methods (Ayala, Olson, Cantrell, Drayton, & Barsic, 2003), and a lack of definitive "real-world" studies, have made direct comparison of gasoline vs. diesel particle number emissions difficult. Further complications arise due to dilution and atmospheric aging of aerosols; semi-volatile organic compounds that are emitted in the particle-phase from engines can evaporate upon dilution in the atmosphere. These gas-phase species may undergo subsequent photochemical reactions to form secondary organic aerosol (Robinson et al., 2007).

The results presented in this paper are unique in that they directly compare fresh particle emissions from a very large sample of light-duty (LD) vehicles and diesel trucks under similar driving conditions, using identical particle analyzers and sampling protocols. Size-segregated particle number emissions ($10 < D_p < 290$ nm) and number concentrations ($D_p > 3$ nm) were measured in a highway tunnel where the aerosol had undergone real-world "tailpipe-to-roadway" dilution (Zhang & Wexler, 2004), but had not further aged in the atmosphere. We report particle number and volume size distributions, size segregated number and volume emission factors (per unit fuel), and total particle number and volume emission factors, separately for LD vehicles and for medium- (MD) and heavy-duty (HD) diesel trucks (henceforth referred to as "diesel trucks"). We also compare results reported here to previous measurements from 1997 (Kirchstetter, Harley, Kreisberg, Stolzenburg, & Hering, 1999) at the same site to quantify long-term temporal trends for total particle number emission factors.

2. Experimental methods

2.1. Field measurements

Emissions from motor vehicles were measured at the Caldecott tunnel on California Highway 24, located in the San Francisco Bay area. The 1 km long tunnel has three separate traffic bores, each with two lanes. Measurements were made in two of the three traffic bores. Bore 1, the southernmost bore, carried a mix of LD vehicles and MD and HD trucks. Vehicles were defined as LD = 2-axle/4-tire, MD = 2-axle/6-tire, and HD = 3 or more axles (Ban-Weiss et al., 2008). Average traffic flow rates in bore 1 were 1958 ± 127 LD vehicles h^{-1}, and 153 ± 9 MD/HD trucks h^{-1} from 1200–1400 h. Bore 2 carried mostly LD vehicles (3800 ± 131 h^{-1}) with a small fraction of MD trucks (< 1%) from 1600 to 1800 h. Pollutant concentrations were measured from 1200 to 1400 h in the mixed traffic bore to maximize the truck fraction, and from 1600 to 1800 h in the LD-only bore to maximize traffic volumes. Traffic was always traveling eastbound and uphill on a 4% grade. Measurements were made on eight weekdays in each traffic bore during July and August of 2006 (see Table 1 for exact dates).

Pollutant concentrations were measured simultaneously at the traffic entrance (west end) and exit (east end) of the tunnel. Particle analyzers were located in the exhaust duct directly above the traffic. Sample air was carried from the sample inlet, located ~15 cm below the ceiling of the traffic bore, through approximately 1 m of conductive silicone tubing to the particle analyzers. An upper size cutoff of 2.5 μm was achieved using sharp cut cyclones (BGI, Waltham, MA, model VSCCA). Particle size distributions were measured at each end of the tunnel using TSI (Shoreview, MN) model 3080L scanning mobility particle sizers (SMPS) paired with TSI model 3025A ultrafine condensation particle counters (CPC). This system was configured to measure number concentration as a function of particle size for $10 < D_p < 290$ nm. Separate standalone CPCs were used to determine total particle number concentrations. An ultrafine water CPC (TSI model 3786) was used to measure particle number concentrations ($D_p > 3$ nm) at the traffic exit, whereas a butanol-based TSI 3022A CPC ($D_p > 7$ nm) was used at the traffic entrance. Due to the high particle concentrations inside the tunnel, the CPC sample air at the tunnel exit was diluted prior to being analyzed. The sample line was split into two parallel lines. One line passed through an orifice and the other through a HEPA filter; the lines were recombined prior to passing through the CPC. The pressure drop across the orifice caused a large and stable fraction of the sample flow to pass through the lower pressure drop line where the HEPA filter removed all of the particles. Because two different orifices were used throughout the measurement campaign, dilution ratios varied as follows. All measurements in the