Atmospheric Environment 191 (2018) 9–18

Contents lists available at ScienceDirect

Atmospheric Environment

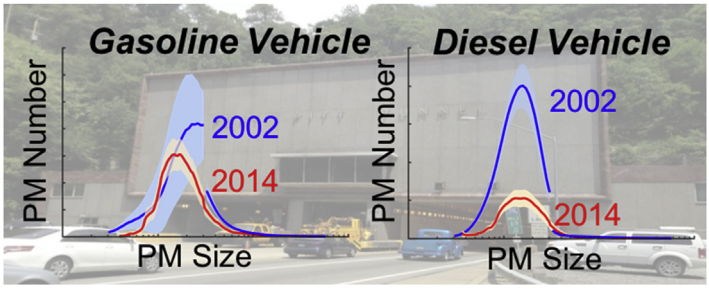
journal homepage: www.elsevier.com/locate/atmosenv

Size distribution of vehicle emitted primary particles measured in a traffic tunnel

Xiang Li[a,b,1], Timothy R. Dallmann[a,b,2], Andrew A. May[c], Charles O. Stanier[d], Andrew P. Grieshop[e], Eric M. Lipsky[a,b,f], Allen L. Robinson[a,b,g], Albert A. Presto[a,b,*]

[a] Center for Atmospheric Particle Studies, Carnegie Mellon University, Pittsburgh, PA, 15213, United States
[b] Department of Mechanical Engineering, Carnegie Mellon University, Pittsburgh, PA, 15213, United States
[c] Department of Civil, Environmental and Geodetic Engineering, The Ohio State University, Columbus, OH, 43210, United States
[d] Department of Chemical and Biochemical Engineering, The University of Iowa, Iowa City, IA, 52242, United States
[e] Department of Civil, Construction and Environmental Engineering, North Carolina State University, Raleigh, NC, 27695, United States
[f] Penn State University Greater Allegheny Campus, McKeesport, PA, 15132, United States
[g] Department of Engineering and Public Policy, Carnegie Mellon University, Pittsburgh, PA, 15213, United States

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Emissions
Diesel
Size distribution
Aerosol

ABSTRACT

Total and size-resolved concentrations and emission factors are used to compare fleet-averaged vehicle emissions in Pittsburgh, PA in 2002 and 2014. Winter-time traffic tunnel measurements acquired using dual scanning mobility particle sizers (SMPS) over the size range 3–500 nm form the key input for the analysis. Size-resolved mass emission factors were calculated assuming a nanoparticle aggregate model. The ultrafine particle (< 30 nm) emissions of diesel vehicles significantly dropped from 2002 to 2014. In the 2014 study, a thermodenuder (TD) was deployed together with the SMPS to measure emissions of non-volatile particles. After evaporation at 250 °C inside the TD, the median diameter of the number-size distribution shifted from 16 nm to ~7 nm. The total particle number decreased significantly (69%) after evaporating inside the TD, indicating that vehicle emitted particles may be largely externally mixed and that a large fraction of these particles may be purely composed of volatile components. Based on the SMPS-TD measurements, we report a size-resolved volatile-to-non-volatile-component-ratio for vehicle emitted particles. It shows that smaller particles (10–60 nm) emitted by vehicles are dominantly (over 75%) composed of volatile components. We also apportioned the size-resolved particles and non-volatile particle mass and number emission factors for both gasoline and diesel

* Corresponding author. Center for Atmospheric Particle Studies, Carnegie Mellon University, Pittsburgh, PA, 15213, United States.
E-mail address: apresto@andrew.cmu.edu (A.A. Presto).
[1] Current address: Department of Soil, Water, and Climate, University of Minnesota, Saint Paul, MN, 55108, United States.
[2] Current address: International Council for Clean Transportation, Washington, DC, United States.

https://doi.org/10.1016/j.atmosenv.2018.07.052
Received 7 February 2018; Received in revised form 5 June 2018; Accepted 28 July 2018
Available online 31 July 2018
1352-2310/

vehicles. Diesel vehicles emitted significantly more ultrafine particles and non-volatile particles than gasoline vehicles.

1. Introduction

On-road gasoline and diesel vehicles are a major source of particulate matter (PM) in the urban environment (Zhang et al., 2015; Dallmann and Harley, 2010; U.S. EPA, 2011), and emissions from these sources can have a significant impact on both climate and human health. Vehicle PM emitted to the atmosphere could potentially become cloud condensation nuclei (CCN) and impact climate through the aerosol indirect effect (Lohmann and Feichter, 2004). The size distribution of primary emissions is a major source of uncertainty in quantifying global CCN concentrations (Lee et al., 2013; Pierce and Adams, 2008). Specifically, for reduction in uncertainty on CCN quantification, constraints on the size distribution of primary particle emissions are as important or more important than improved estimates of mass emissions (Lee et al., 2013).

Combustion sources, especially on-road gasoline and diesel vehicles, are a major source of ultrafine particles (Hinds, 1999), defined as particles with diameter smaller than 100 nm. Ultrafine particles are able to penetrate and deposit in the human lung and cause respiratory diseases and other health concerns (Brown et al., 2002; Oberdörster, 2000; Peters et al., 1997; HEI, 2010). Exposure to ultrafine particles may have health effects distinct from exposure to fine particle mass alone due to differences in lung deposition and toxicity (HEI, 2013). Therefore, to better estimate the effect of PM on climate change and public health, it is necessary to better quantify the size-distribution of vehicle emitted particles.

Vehicle emitted particles are mostly fractal aggregates of small spherical primary particles, as reported by both vehicle engine tests (Lu et al., 2012; Kocbach et al., 2005; Park et al., 2003; Bérubé et al., 1999) and near-road and tunnel studies (Ancelet et al., 2011; Paoletti et al., 2002; Dye et al., 2000). Particle size distributions are typically measured with electrical mobility (e.g., with a scanning mobility particle sizer, SMPS), and particle number counts are converted to volume or mass by assuming spherical particles. Since a fractal particle is less massive than a sphere with the same mobility diameter (Sorensen, 2011), using the standard SMPS spherical assumptions can overestimate mass emissions (Lall and Friedlander, 2006; Park et al., 2003). Among all tunnel studies that measured the size-distribution of vehicle emitted particles, very few studies reported the volume or the mass size distributions. Ban-Weiss et al. (2010) conducted measurements in the Caldecott tunnel and reported size-resolved particle volume emission factors of both gasoline and diesel vehicles. However, the particle volume calculated by Ban-Weiss et al. (2010) assumes spherical particles, and thus volume emissions could be potentially overestimated. No tunnel studies have compared the particle mass calculated from the size-distribution measurement with direct particle mass measurements.

A large fraction of vehicle PM is organic and will therefore undergo evolving gas-particle partitioning in the atmosphere (May et al., 2013b, 2013a; Robinson et al., 2007). The evaporated organic vapors can react to form secondary organic aerosol (SOA) (Gentner et al., 2017; Tkacik et al., 2014). Therefore, in order to better quantify the contribution of vehicle emissions to the total particle burden in the atmosphere, it is necessary to have a better quantification of both the particle size and volatility.

Numerous studies measured the chemical composition (e.g., contributions of organic and black carbon to PM mass) and the volatility of vehicle emitted organic aerosol (Li et al., 2016; Worton et al., 2014; May et al., 2013b, 2013a; Pant and Harrison, 2013; Lu et al., 2012; Maricq, 2007; Kleeman et al., 2000), but very few studies reported size-dependent chemical composition or volatility. Kleeman et al. (2000) reported size-dependent contributions of organic carbon, elemental carbon, and inorganic ions to particles larger than ~50 nm for both gasoline and diesel vehicles, and Lu et al. (2012) measured the size-dependent volatile and non-volatile components of particles emitted by diesel vehicles. Those studies were lab engine tests on a limited number of vehicles and may not represent emissions from the much larger real-world fleet.

Recent near-roadway studies investigated the size-dependent volatility of particles emitted by a mixed real-world vehicle fleet. Saha et al. (2018b) and Wang et al. (2017) both showed that emissions of particle number are higher in winter than summer. A significant fraction of these particles, especially at smaller sizes (< 50 nm) evaporate downwind from the roadway (Saha et al., 2018b), suggesting that organics rapidly repartition in the near-road environment. Saha et al. (2018a) and Wang et al. (2017) also show that emissions of the less-volatile particle fraction (e.g., particles that do not evaporate in a thermodenuder at T > 180 °C) are correlated with black carbon, and that these emissions are less seasonally variable than total particle number emissions.

Previous studies also show that particle mass emission factors have decreased over past decades in the United States (McDonald et al., 2015; May et al., 2014). These emissions reductions are attributable to stricter emissions limits and introduction of control technologies such as diesel particulate filters. However, these studies only report the total particle emission factors. To our knowledge, no studies show how the emitted particle size distribution, or emission factors as a function of particle size, have changed over the same period.

In this work, we measured the size distribution of vehicle emitted primary particles in traffic tunnels in Pittsburgh, PA in winter 2014 and winter 2002. We report both number-size distribution and mass-size distribution of vehicle emitted particles. A thermodenuder (TD) was used to determine the size-dependent volatility. Finally, we apportioned the size-resolved emission factors to both gasoline and diesel vehicles, and compared the size-resolved emission factors measured in winter 2014 and winter 2002.

2. Methods

2.1. Size distribution measurement

The winter 2014 measurement was conducted in the Fort Pitt Tunnel, a tunnel on Interstate-376 in Pittsburgh, PA, from Jan 31 to Feb 4, 2014. Information about the Fort Pitt Tunnel, the traffic conditions in the tunnel, and the measurement station can be found in Li et al. (2016). During this study, approximately 140,000 light duty vehicles (LDV) and 3600 heavy duty diesel vehicles (HDDV) passed through the tunnel. The presence of HDDV is quantified with the diesel fuel fraction ($\%fuel_D$) derived by Li et al. (2016) from traffic volume data. Short sampling lines (< 1 m) were used to minimize particle sampling losses. The mean temperature during the sampling period was 0.4 ± 4.7 °C.

Particle size distributions were concurrently measured with a pair of scanning mobility particle sizers (SMPS, TSI Inc.). Particles with a diameter of 4–120 nm were measured by a nano-SMPS consisting of a nano Differential Mobility Analyzer (DMA, model 3085) and an ultrafine Condensation Particle Counter (CPC, model 3025A). In order to minimize the diffusion loss of smaller particles, the ultrafine CPC was run under high-flow mode. Particles with a diameter of 12–550 nm were measured by a long-SMPS (model 3081 DMA and model 3772 CPC). The particle number concentrations measured by the nano-SMPS and the long-SMPS are consistent in the size range of 20–120 nm (Figure S1).

For particles with a diameter less than 15 nm, particle number