Atmospheric Environment 88 (2014) 247–260

Contents lists available at ScienceDirect

Atmospheric Environment

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

Gas- and particle-phase primary emissions from in-use, on-road gasoline and diesel vehicles

Andrew A. May [a,1], Ngoc T. Nguyen [a], Albert A. Presto [a], Timothy D. Gordon [a,2], Eric M. Lipsky [a,b], Mrunmayi Karve [a], Alváro Gutierrez [c], William H. Robertson [d], Mang Zhang [c], Christopher Brandow [e], Oliver Chang [e], Shiyan Chen [c], Pablo Cicero-Fernandez [e], Lyman Dinkins [e], Mark Fuentes [c], Shiou-Mei Huang [e], Richard Ling [e], Jeff Long [f], Christine Maddox [e], John Massetti [e], Eileen McCauley [h], Antonio Miguel [e], Kwangsam Na [d], Richard Ong [e], Yanbo Pang [e], Paul Rieger [e], Todd Sax [g], Tin Truong [c], Thu Vo [c], Sulekha Chattopadhyay [f], Hector Maldonado [h], M. Matti Maricq [i], Allen L. Robinson [a,*]

[a] *Center for Atmospheric Particle Studies, Carnegie Mellon University, Pittsburgh, PA, United States*
[b] *Department of Engineering, Penn State Greater Allegheny, McKeesport, PA, United States*
[c] *Mobile Source Operations Division, California Air Resources Board, El Monte, CA, United States*
[d] *Mobile Source Control Division, California Air Resources Board, El Monte, CA, United States*
[e] *Monitoring and Laboratory Division, California Air Resources Board, El Monte, CA, United States*
[f] *Planning and Technical Support Division, California Air Resources Board, El Monte, CA, United States*
[g] *Planning and Technical Support Division, California Air Resources Board, Sacramento, CA, United States*
[h] *Research Division, California Air Resources Board, Sacramento, CA, United States*
[i] *Research and Advanced Engineering, Ford Motor Company, Dearborn, MI, United States*

HIGHLIGHTS

- Gas- and particle-phase pollutants from gasoline and diesel vehicles were quantified and compared to emissions from small off road engines.
- Measurements were extended to gasoline vehicles from more recent model years and diesel vehicles with a range of aftertreatments and fuels.
- More stringent emission standards and aftertreatment devices substantially reduce the majority of primary pollutant emissions.
- Speciated emissions data indicate that secondary organic aerosol produced from gasoline vehicle exhaust may exceed primary PM emissions.

ARTICLE INFO

Article history:
Received 5 August 2013
Received in revised form
23 December 2013
Accepted 27 January 2014
Available online 28 January 2014

Keywords:
Primary emissions
Light-duty gasoline vehicles
Heavy-duty diesel vehicles
Speciated VOCs

ABSTRACT

Tailpipe emissions from sixty-four unique light-duty gasoline vehicles (LDGVs) spanning model years 1987–2012, two medium-duty diesel vehicles and three heavy-duty diesel vehicles with varying levels of aftertreatment were characterized at the California Air Resources Board Haagen-Smit and Heavy-Duty Engine Testing Laboratories. Each vehicle was tested on a chassis dynamometer using a constant volume sampler, commercial fuels and standard duty cycles. Measurements included regulated pollutants such as carbon monoxide (CO), total hydrocarbons (THC), nitrogen oxides (NO_x), and particulate matter (PM). Off-line analyses were performed to speciate gas- and particle-phase emissions. The data were used to investigate trends in emissions with vehicle age and to quantify the effects of different aftertreatment technologies on diesel vehicle emissions (e.g., with and without a diesel particulate filter). On average, newer LDGVs that met the most recent emissions standards had substantially lower emissions of regulated gaseous pollutants (CO, THC and NO_x) than older vehicles. For example, THC emissions from the median LDGV that met the LEV2 standard was roughly a factor of 10 lower than the median pre-LEV vehicle; there were also substantial reductions in NO_x (factor of ~100) and CO (factor of ~10) emissions

* Corresponding author. Department of Mechanical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, United States.
E-mail address: alr@andrew.cmu.edu (A.L. Robinson).
[1] Now with Department of Atmospheric Science, Colorado State University, Fort Collins, CO, United States.
[2] Now with Chemical Sciences Division, National Oceanographic and Atmospheric Administration Earth System Research Laboratory, Boulder, CO, United States and Cooperative Institute for Research in Environmental Sciences, University of Colorado–Boulder, Boulder, CO, United States.

1352-2310/$ – see front matter
http://dx.doi.org/10.1016/j.atmosenv.2014.01.046

from pre-LEV to LEV2 vehicles. However, reductions in LDGV PM mass emissions were much more modest. For example, PM emission from the median LEV2 vehicle was only a factor of three lower than the median pre-LEV vehicle, mainly due to the reductions in organic carbon emissions. In addition, LEV1 and LEV2 LDGVs had similar PM emissions. Catalyzed diesel particulate filters reduced CO, THC and PM emissions from HDDVs by one to two orders of magnitude. Comprehensive organic speciation was performed to quantify priority air toxic emissions and to estimate the secondary organic aerosol (SOA) formation potential. The data suggest that the SOA production from cold-start LDGVs exhaust will likely exceed primary PM emissions from LDGVs and could potentially exceed SOA formation from on-road diesel vehicles.

1. Introduction

Mobile sources can be an important source of air pollution in urban environments. Starting in the 1970s for light-duty gasoline vehicles (LDGVs) and in 2007 for heavy-duty diesel vehicles (HDDVs), the implementation of strict emissions standards has led to the development and implementation of engine control and aftertreatment technologies that have substantially reduced the emissions of regulated pollutants such as carbon monoxide (CO), total hydrocarbons (THC), nitrogen oxides (NO_x = nitric oxide NO + nitrogen dioxide NO_2), and particulate matter (PM).

Although there is considerable literature on mobile source emissions (Rogge et al., 1993; Schauer et al., 1999, 2002; Zielinska et al., 2004; Kittelson et al., 2006; Fujita et al., 2007; Maricq, 2007; Robert et al., 2007a, 2007b; Volckens et al., 2007, 2008; Kishan et al., 2008; Biswas et al., 2009; Chirico et al., 2010; Bishop et al., 2013; McDonald et al., 2013), engine and emissions aftertreatment technology continues to be developed, requiring updated emissions data. For example, the very large Kansas City PM Characterization Study occurred nearly a decade ago (Kishan et al., 2008), when relatively few vehicles that met the latest federal emissions standard (Tier 2) were on the road. Changes in speciated THC emissions must be investigated to understand the evolution of secondary organic aerosol (SOA) production and mobile source air toxics (MSATs). Finally, few recent studies have compared emissions from different mobile source classes (e.g., on-road gasoline vehicles versus on-road diesel vehicles versus off-road gasoline equipment).

In addition to direct or primary PM emissions, it is important to understand the potential for mobile source emissions to contribute to SOA formation. SOA is organic PM that forms in the atmosphere from the oxidation of gas-phase precursors; it is a major contributor to fine PM mass, even in urban environments with substantial primary emissions (Zhang et al., 2007). Motor vehicles are a source of SOA precursors, including aromatics and larger alkanes. Although recent studies have investigated SOA formation from vehicular exhaust (Chirico et al., 2010; Bahreini et al., 2012; Ensberg et al., 2013; Nordin et al., 2013; Platt et al., 2013), there has been relatively little effort to directly relate exhaust composition with SOA formation. Improved knowledge of SOA precursor emissions should improve model predictions.

This paper describes measurements of tailpipe emissions from gasoline and diesel vehicles recruited from the in-use California vehicle fleet. Emissions data included regulated gases (e.g., CO, NO_x, THC), speciated total organic gases (e.g., single-ring aromatics, carbonyls), PM mass, and speciated PM (e.g., organic and elemental carbon). These datasets were used to investigate trends in LDGVs emissions with vehicle age and to quantify the effects of different aftertreatment technologies on HDDV emissions (e.g., with and without a diesel particulate filter). The comprehensive organic speciated data were used to estimate SOA formation potential and emissions of air toxics. The research was conducted as part of a large project that investigated linking tailpipe emissions from mobile sources to ambient PM. Companion papers describe gas-particle partitioning of primary POA emissions (May et al., 2013a, 2013b), and SOA formation from on-road gasoline vehicles (Gordon et al., 2013a), on-road diesel vehicles (Gordon et al., 2013b), and small off-road engines (Gordon et al., 2013c).

2. Methods

2.1. Fleet overview

Experiments were performed to characterize the tailpipe emissions from sixty-four unique LDGVs spanning model years 1987–2012. Fifty-seven LDGVs were recruited from southern California residents or car rental agencies; the remainder (nine) was selected from the California Air Resources Board (ARB) vehicle pool. The LDGV test fleet was not designed to represent the distribution of vehicles in the current, in-use California fleet. Instead vehicles were selected to span a wide range of model years, vehicle types, engine technologies, and emission control technologies. Vehicle recruitment was a quasi-random process. ARB solicited vehicles from southern CA drivers, and vehicles for testing were selected from the pool of responses to provide a distribution of model years, vehicle types, and manufacturer. Rental vehicles were used to fill out the LDGV fleet. Table S1 (online Supporting Information) lists the LDGV test fleet including information on model year, engine size, body type, mileage and emissions certification.

For discussion, the LDGVs were grouped based on model year: "pre-LEV" were vehicles manufactured prior to 1994; "LEV1" vehicles were manufactured between 1994 and 2003; and "LEV2" vehicles were vehicles manufactured in 2004 or later. In this work, the LEV designation is used to refer to a range of model years; it does not necessarily refer to the low emissions vehicle certification standard. For example, some of the "LEV1" vehicles were certified as Tier 1 (not LEV1) vehicles (Table S1). However, these labels still provide general information related to increased stringency of emission standards. The LDGV test fleet was comprised of 15 pre-LEV, 24 LEV1, and 25 LEV2 vehicles. Many of the pre-LEV and LEV2 vehicles were rentals, since it was difficult to recruit from these two classes. Except for three LEV2 vehicles that had a gasoline-direct-injection engine (Table S1), all of the vehicles were port-fuel-injected. Further, one vehicle was a compressed natural gas (CNG) vehicle; its emissions data are included in the Supporting Information. The vehicle naming convention: LEV designation (e.g., LEV1) followed by the vehicle number in the test sequence (e.g., LEV1-6).

The study also tested five diesel vehicles, which are summarized in Table S2, including information on model year, engine size, mileage and emissions control technology. These vehicles were selected to span a range of emission control technologies. The three HDDVs were equipped with typical 6-cylinder, in-line, direct-