Available online at www.sciencedirect.com
ScienceDirect
Atmospheric Environment 42 (2008) 220–232
ATMOSPHERIC ENVIRONMENT
www.elsevier.com/locate/atmosenv

Long-term changes in emissions of nitrogen oxides and particulate matter from on-road gasoline and diesel vehicles

George A. Ban-Weiss[a], John P. McLaughlin[b], Robert A. Harley[b,*], Melissa M. Lunden[c], Thomas W. Kirchstetter[c], Andrew J. Kean[d], Anthony W. Strawa[e], Eric D. Stevenson[f], Gary R. Kendall[f]

[a]*Department of Mechanical Engineering, University of California, Berkeley, CA 94720-1740, USA*
[b]*Department of Civil and Environmental Engineering, University of California, Berkeley, CA 94720-1710, USA*
[c]*Lawrence Berkeley National Laboratory, Department of Atmospheric Science, Berkeley, CA 94720, USA*
[d]*Department of Mechanical Engineering, California Polytechnic State University, San Luis Obispo, CA 93407, USA*
[e]*NASA-AMES Research Center, Moffett Field, CA 94035, USA*
[f]*Bay Area Air Quality Management District, Technical Services Division, 939 Ellis Street, San Francisco, CA 94109, USA*

Received 11 July 2007; received in revised form 15 September 2007; accepted 20 September 2007

Abstract

Gas- and particle-phase pollutants were measured separately for (a) light-duty (LD) vehicles and (b) medium-duty (MD) and heavy-duty (HD) diesel trucks. Measurements were made during summer 2006 at the Caldecott Tunnel in the San Francisco Bay area as part of a continuing campaign to track changes in vehicle emissions over time. When normalized to fuel consumption, NO_x emission factors were found to be 3.0 ± 0.2 and $40 \pm 3\,g\,kg^{-1}$ for LD vehicles and MD/HD diesel trucks, respectively. Corresponding particulate matter ($PM_{2.5}$) emission factors were 0.07 ± 0.02 and $1.4 \pm 0.3\,g\,kg^{-1}$. The ratio of particulate black carbon to organic mass (BC/OM) for LD vehicles was 0.71 ± 0.15. For diesel trucks, BC/OM was 2 ± 1, indicating that $PM_{2.5}$ was dominated by BC. Results from 2006 are compared to similar measurements made at the same site in 1997. For LD vehicles, NO_x and $PM_{2.5}$ emission factors decreased by $67 \pm 3\%$ and $36 \pm 17\%$, respectively. Corresponding decreases for diesel trucks were $30 \pm 9\%$ for NO_x and $48 \pm 12\%$ for $PM_{2.5}$. The ratio of HD to LD emission factor for NO_x increased from 6 ± 1 to 13 ± 1 between 1997 and 2006, which indicates an increase in the *relative* importance of diesel trucks as a source of NO_x emissions. The absorption, scattering, and extinction cross-section emission factors parameters relevant to climate change and atmospheric visibility, were an order of magnitude higher for diesel trucks than LD vehicles. Single-scattering albedo, measured at $\lambda = 675$ nm, was 0.31 ± 0.06 and 0.20 ± 0.05 for LD vehicle and diesel truck PM emissions, respectively.

Keywords: Motor vehicles; Tunnel study; Aerosol optical properties; Nitrogen oxides; Exhaust particulate matter

1. Introduction

Motor vehicles emit gaseous air pollutants including nitrogen oxides (NO_x), volatile organic compounds (VOC), carbon monoxide (CO), and

*Corresponding author. Tel.: +1 510 643 9168; fax: +1 510 642 7483.
E-mail address: harley@ce.berkeley.edu (R.A. Harley).

1352-2310/$ - see front matter © 2007 Elsevier Ltd. All rights reserved.
doi:10.1016/j.atmosenv.2007.09.049

carbon dioxide (CO_2). They also emit particulate matter (PM), mostly in the form of black carbon (BC) and organic carbon (OC) (Sawyer et al., 2000). These gaseous and particulate air pollutants raise public health concerns (Lloyd and Cackette, 2001). In 2006, on-road motor vehicles were responsible for 51% of total NO_x, 34% of VOC, 60% of CO, and 9% of $PM_{2.5}$ in California (CARB, 2006). In addition to contributing to local and regional air pollution problems, vehicle exhaust contributes to climate change. Motor vehicles are responsible for 35% of California CO_2 emissions (CEC, 2006), the greenhouse gas responsible for the greatest amount of global warming. NO_x is a precursor to tropospheric ozone, which also contributes to global warming. PM has direct and indirect effects on radiative forcing, leading to both global warming and cooling; the direct effect of BC emissions is positive forcing (IPCC, 2007).

Motor vehicles span a wide range of sizes from light-duty (LD) vehicles, which in the US are mostly fueled by gasoline, to heavy-duty (HD) trucks, mostly diesel-powered. The *relative* importance of HD diesel truck exhaust as a source of NO_x emissions has increased in the last 15 years because control of LD gasoline vehicle emissions has progressed more than HD diesel truck emissions, and on-road use of diesel fuel has grown faster than gasoline since 1990 (Harley et al., 2005). As shown in Table 1, NO_x emission standards for HD diesel engines were gradually reduced during the 1990s. However, most 1990s engines met these standards only during emission certification tests but not while being used on-road (Yanowitz et al., 2000). As a result of increased regulatory pressure, most new engines met the 2004 NO_x standard 2 years early using exhaust gas recirculation (e.g. Volvo, 2007). To meet increasingly stringent HD diesel NO_x standards in the future, urea-based selective catalytic reduction systems will likely be used in new engines starting in 2010 (Johnson, 2004). Such systems are already being used in HD diesel trucks to meet European emission standards (e.g. Mack, 2006). Trends in on-road LD and HD vehicle emissions in Europe have been reported by Schmid et al. (2001) and Colberg et al. (2005).

Table 1
US heavy-duty diesel truck emission standards[a]

Model year	NO_x	PM
1979–1984	13.4[b]	
1985–1987	14.4	
1988–1989	14.4	0.80
1990	8.1	0.80
1991–1993	6.7	0.34
1994–1997	6.7	0.13
1998–2003	5.4	0.13
2004–2006	3.2	0.13
2007–2009	1.7[c]	0.013
>2010	0.27	0.013

[a]Units of grams per kilowatt-hour ($g\,kW^{-1}h^{-1}$): mass emitted per unit of engine brake work output.

[b]Total hydrocarbon (THC) + NO_x.

[c]The NO_x standard for 2007 is $0.27\,g\,kW^{-1}h^{-1}$, but is being phased in over 3 years. 50% of total sales for each engine manufacturer must meet the $0.27\,g\,kW^{-1}h^{-1}$ standard from 2007 to 2010, thus the effective standard is $1.7\,g\,kW^{-1}h^{-1}$.

PM emission standards for HD diesel engines underwent larger reductions during the 1990s than NO_x (Table 1). Yanowitz et al. (2000) have shown that unlike NO_x, exhaust PM mass emissions from HD trucks decreased during this time. New HD diesel trucks started using diesel particle filters in 2007 to meet new emission standards, which required PM emissions to be reduced by an order of magnitude. To enable catalytic diesel exhaust emission controls, the sulfur content of diesel fuel was reduced in the US to <15 ppm by weight starting in 2006. Less is known about PM emission trends for LD vehicles relative to HD trucks; there is continuing controversy about the relative importance of gasoline vs. diesel vehicles as sources of exhaust PM emissions (Gertler, 2005), indicating the need for additional measurements.

This paper reports measured NO_x and exhaust PM emissions from large numbers of on-road vehicles during 2006 in a San Francisco Bay area highway tunnel, and compares with data from the same site from previous years to quantify trends over time. Measurements were made immediately after the switch to ultra-low sulfur diesel (ULSD) fuel in California, and immediately prior to the deployment of PM and NO_x control technologies on new HD diesel trucks, so this study can also serve as a baseline to quantify on-road emission trends after the 2007–2010 emission standards take effect. Light-absorbing and scattering properties of exhaust PM emissions that are relevant to understanding visibility and climate-forcing effects of vehicle emissions are also quantified here.