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

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**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 $3.0 \pm 0.2$ 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 $0.07 \pm 0.02$ and $1.4 \pm 0.3\,g\,kg^{-1}$. The ratio of particulate black carbon to organic mass (BC/OM) for LD vehicles was $0.71 \pm 0.15$. For diesel trucks, BC/OM was $2 \pm 1$, 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 $6 \pm 1$ to $13 \pm 1$ 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 $0.31 \pm 0.06$ and $0.20 \pm 0.05$ 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

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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.

## 2. Experimental methods

### 2.1. Field site

Motor vehicle emissions were measured in the Caldecott tunnel during July and August of 2006. The 1.0 km long tunnel is located on highway 24 in the San Francisco Bay area, and has three separate two-lane traffic bores, with a grade of 4% uphill in the eastbound direction. Each bore consists of a traffic tube, through which the vehicles travel, and ventilation air ducts located above the traffic tubes through which fresh outside air and polluted tunnel air flow into and out of the tunnel, respectively. Air flow through the ventilation ducts is facilitated by large fans; these fans were turned off during intensive observation periods to simplify mass balance calculations, which are further discussed in Section 2.5. Turning off the fans meant that airflow in the tunnel was longitudinal in the direction of traffic flow. A schematic of the tunnel is available in Kirchstetter et al. (1996). Pollutant concentrations were measured in bores 1 and 2. Bore 1 carries a mix of LD passenger vehicles and medium-duty (MD) and HD trucks; HD trucks are restricted from traveling through bore 2. This special feature of the Caldecott tunnel allows direct determination of LD vehicle emission factors without having to apportion pollutant concentrations, as must be done in situations with mixed LD and HD traffic (Fraser et al., 2003; Grieshop et al., 2006; Imhof et al., 2006). Bore 1 traffic flows eastbound (uphill) at all times, whereas bore 2 traffic switches from westbound to eastbound at approximately noon on weekdays to accommodate commuter traffic. All results reported here are for uphill traffic conditions in both bores.

Measurements at the tunnel were conducted on 8 weekdays in each traffic bore, for a total of 16 days. Exact dates are listed in Table S1 in the Appendix. Although most pollutant analyzers ran 24 h a day, intensive observations were made 12:00–14:00 h in bore 1 and 16:00–18:00 h in bore 2. These times were chosen to maximize the diesel truck fraction of traffic in bore 1, and LD vehicle traffic volume in bore 2.

### 2.2. Traffic characterization

Manual traffic counts of LD vehicles, MD trucks, and HD trucks were performed each day. Vehicles were categorized by number of axles and tires: LD = 2-axle/4-tire, MD = 2-axle/6-tire, and HD = 3 or more axles. Vehicles passing through the tunnel were also recorded using video cameras at the tunnel entrance and exit. Camera clocks were synchronized and used to calculate vehicle transit times, which were combined with known tunnel length to compute average vehicle speeds inside the tunnel. This was done for every individual HD truck entering bore 1 between 12:00 and 14:00 h on 7 out of 8 sampling days. In bore 2, speeds were calculated every 5 min using vehicles observed in both lanes. This was done between 16:00 and 18:00 h on 6 out of 8 sampling days. On the remaining sampling days, one of the video cameras was repositioned outdoors and used for license plate surveys. License plates were transcribed and matched with registration data to determine vehicle age distributions.

### 2.3. Gas-phase measurements

$CO_2$, $NO_x$, and CO concentrations were measured using analyzers set up at the traffic entrance and exit of the tunnel. At the exit (east end), air was drawn from the traffic tube using a pump through a ~40 m Teflon sample line (ID = 1.2 cm) to the pollutant analyzers. The residence time of air in the sample line was approximately 20 s. At the entrance (west end), tunnel air was drawn through a ~5 m Teflon sample line (ID = 0.48 cm) from the traffic tube to the pollutant analyzers using only the internal instrument pumps.

$CO_2$ concentrations were measured using LI-COR (Lincoln, NE) model 820 non-dispersive infrared gas analyzers at both ends of the tunnel. $NO_x$ concentrations were measured using chemiluminescent analyzers (Thermo Environmental Instruments (TEI), Franklin, MA, models 42A and 42C at the entrance and exit, respectively). CO was measured using gas filter correlation spectrometers at both ends of the tunnel (TEI model 48). Calibration of all gas-phase analyzers was checked daily prior to sampling. The Bay Area Air Quality Management District's Quality Assurance group audited the gas analyzers in the tunnel to ensure measurement accuracy.

### 2.4. Particle measurements

Two-hour average measurements of $PM_{2.5}$, BC, and OC were made at both the entrance and exit of the tunnel using matching experimental systems.

The aerosol samples were drawn from approximately 15 cm below the ceiling of the traffic bore. Filter samples were collected downstream of sharp cut cyclones (BGI, Waltham, MA, model VSCCA) at 16.7 L $min^{-1}$ to achieve a particle size cut of 2.5 μm. This flow rate was maintained during sample collection using calibrated mass flow controllers (Alicat Scientific, Tucson, AZ, model MC-50SLPM-D) with the following exceptions. On the first 3 days of the study (18–20 July, in bore 1) at the exit end of the tunnel, valves and rotameters that were calibrated with a primary air flow standard (Sensidyne/Gilian, Clearwater, FL, model Gilibrator-2) were used to maintain a steady flow rate through the filters. On the 9th day of the study (i.e., 31 July, the first sampling day in bore 2) at the exit end of the tunnel, a mass flow controller power supply malfunctioned and the tandem quartz filters sampled tunnel air at 13 L $min^{-1}$ instead of 16.7 L $min^{-1}$. For this sample, the cyclone established a particle size cut of 3.2 μm instead of 2.5 μm. Particulate carbon concentrations of BC and OC for this sample were calculated using the reduced flow rate. The $PM_{2.5}$ sample was not affected. The total carbon (TC) concentration (i.e., BC plus OC) determined for this sample was equal to the average concentration in bore 2, indicating there were no large changes in carbon particle mass collected.

Quartz (Pallflex, East Hills, NY, model 2500QAT-UP) and Teflon (Gelman Sciences, Teflon membrane, 2.0 μm pore size) filters were used to collect PM samples. To determine $PM_{2.5}$ mass, Teflon filters were weighed before and after sampling; the mass difference was divided by the volume of air sampled. Prior to use, quartz filters were baked at 800 °C for 6 h to remove carbonaceous impurities. In the tandem filter sampling method (Turpin et al., 2000) employed here, two quartz filters in series are collected in parallel with a Teflon filter and a quartz filter in series. The first filter in the tandem quartz pair collects PM that is subsequently analyzed for carbon content, and the quartz filter behind the Teflon filter is used to correct particulate carbon concentrations for the positive OC sampling artifact, as described by Kirchstetter et al. (2001) and Subramanian et al. (2004).

The carbon content of quartz filter samples was measured using thermal optical analysis (TOA). Filter samples were heated at a constant rate of 40 °C $min^{-1}$ from 50 to 700 °C in a pure oxygen atmosphere. The evolved carbon was fully oxidized over a platinum coated ceramic catalyst maintained at 800 °C, and the resultant $CO_2$ was measured with a non-dispersive infrared analyzer (LI-COR, Lincoln, NE, model 7000). The intensity of light transmitted through the sample was continuously monitored during analysis and was used to differentiate between organic and light-absorbing black carbon. Transmission was measured using a white light source and a spectrometer (Ocean Optics, Dunedin, FL, model S2000). The recovery of TC concentrations measured with this approach was determined to be $100 \pm 5\%$ by analysis of prepared samples of potassium hydrogen phthalate and glucose. This TOA protocol will be referred to as the LBNL TOA method henceforth. The LBNL TOA method differs from the more common IMPROVE thermal–optical reflectance (TOR) (Chow et al., 2001) and NIOSH thermal–optical transmission (TOT) (Birch and Cary, 1996) protocols, which expose the sample to helium followed by a helium/oxygen mixture (as opposed to pure oxygen), raise the sample temperature stepwise (as opposed to at a constant rate), and measure optical reflectance or transmission at a single wavelength (as opposed to over a broad spectral region). In a previous sampling campaign at the same tunnel (Kirchstetter et al., 1999a), particulate carbon was determined using the NIOSH TOT protocol. Therefore, in the current study, selected samples were analyzed according to both the NIOSH TOT and LBNL TOA protocol to ensure the two protocols yielded similar results, as shown in Fig. 1.

In addition to 2-h average particle measurements, the physical and optical aerosol properties were measured in real-time at both ends of the tunnel. BC concentrations were measured using aethalometers (Magee Scientific, Berkeley, CA, models AE-2 and AE-1 at the entrance and exit, respectively), and scattering coefficients ($b_{scat}$) were measured at $\lambda = 530$ nm using nephelometers (Radiance Research, Seattle, WA, model M902). Aerosol was sampled at both ends of the tunnel through an AIHL cyclone (John and Reischl, 1980) to a common sample manifold to achieve a size-cut of 2.5 μm at a flow rate of 25 L $min^{-1}$. The aethalometers and nephelometers drew isokinetic samples from the common manifolds.

Aerosol optical properties of $b_{scat}$ and extinction coefficient ($b_{ext}$) were measured with 1-s temporal resolution at $\lambda = 675$ nm using the cavity ring-down instrument of Strawa et al. (2003, 2006) known as Cadenza. PM concentrations and emission factors

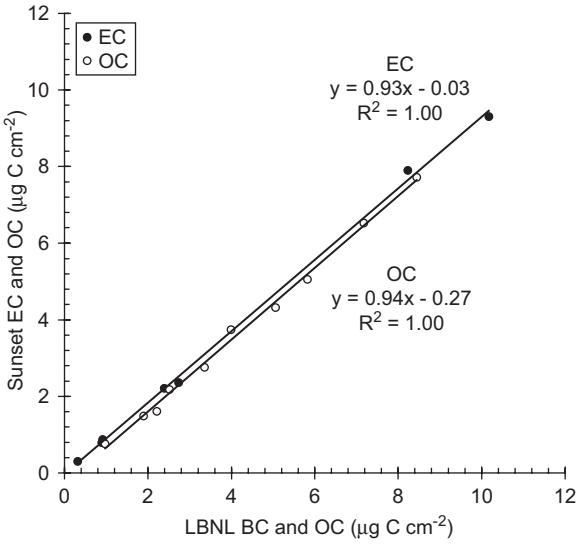


Fig. 1. Comparison of EC and OC measured by Sunset Labs (NIOSH TOA method), vs. BC and OC measured by LBNL (LBNL TOA method). Slopes near unity and $y$-intercepts near zero indicate good agreement between the two TOA methods.

are generally reported on a mass basis. However, for purposes of visibility or climate effects the relevant parameter is $b_{ext}$, which is a measure of the amount of light attenuated over a linear distance, typically reported as $10^{-6}\,m^{-1}$ or $Mm^{-1}$ for atmospheric conditions. Measurement of $b_{ext}$ is generally difficult because long path lengths are required. The cavity ring-down technique in this study employs a long effective path-length ($\sim$1 km) in a $20 \times 20\,cm^2$ optical cell. Cadenza simultaneously measures $b_{scat}$ in the same measurement cell as $b_{ext}$ using a reciprocal nephelometer technique (Mulholland and Bryner, 1994). Absorption coefficient ($b_{abs}$) can be subsequently calculated as $b_{ext}$ minus $b_{scat}$. Based on laboratory calibrations and airplane flight experience, the measurement uncertainty for $b_{ext}$ is 2%. Strawa et al. (2006) compared Cadenza measurements of $b_{ext}$ with the sum of $b_{scat}$ measured with a nephelometer and $b_{abs}$ measured with a Particle Soot Absorption Photometer (PSAP), and found agreement within 2%.

### 2.5. Emission factors

Emission factors were computed using the carbon balance method shown by Eq. (1), and expressed per unit mass of fuel burned separately for LD vehicles and MD/HD diesel trucks. Most of the carbon in gasoline and diesel fuel is emitted as $CO_2$, with smaller amounts emitted as CO. Even smaller amounts of fuel carbon emitted as PM (Kirchstetter et al., 1999a) and unburned hydrocarbons (Kirchstetter et al., 1999b) are neglected in the denominator of Eq. (1). The emission factor $E_p$ (g of pollutant $P$ $kg^{-1}$ fuel burned) can be calculated as

$$E_p = \left(\frac{\Delta[P]}{\Delta[CO_2] + \Delta[CO]}\right) w_c, \quad (1)$$

where $\Delta[P]$ is the background-subtracted (exit minus entrance) mass concentration of pollutant $P$ ($\mu g\,m^{-3}$), and $\Delta[CO_2]$ and $\Delta[CO]$ are background-subtracted concentrations in $mg\,C\,m^{-3}$. The fuel carbon mass fraction is $w_c = 0.87$ for diesel, and 0.85 for oxygenated gasoline (Kirchstetter et al., 1999a). To calculate emission factors from $b_{abs}$, $b_{scat}$, and $b_{ext}$, $P$ in Eq. (1) is replaced by $b$ ($Mm^{-1}$). The resulting emission factors are total optical cross-section of particles emitted per unit mass of fuel burned ($m^2\,kg^{-1}$). Background (tunnel entrance) values of light absorption, scattering, and extinction were not measured by Cadenza. To estimate background values for use in Eq. (1), we used the ratio of tunnel entrance to exit values of BC and $b_{scat}$ from the aethalometer and nephelometer, respectively. For bores 1 and 2, the entrance to exit ratio for BC was 0.11 and 0.19, and $b_{scat}$ was 0.23 and 0.32, respectively. Entrance values of $b_{ext}$ were estimated by summing entrance values of $b_{abs}$ and $b_{scat}$.

Emission factors for LD vehicles were calculated with Eq. (1) using measured concentrations from bore 2. To calculate emission factors for MD/HD diesel trucks, contributions to bore 1 concentrations first need to be apportioned between LD vehicles and MD/HD diesel trucks. Pollutants other than $CO_2$ in bore 1 were apportioned using estimates of $CO_2$ emissions from gasoline engines, and LD vehicle pollutant to $CO_2$ emission ratios measured in bore 2:

$$\Delta[P]_{1,D} = \Delta[P]_1 - \Delta[CO_2]_{1,G}\left(\frac{\Delta[P]_2}{\Delta[CO_2]_2}\right), \quad (2)$$

where subscripts D and G indicate diesel and gasoline, subscripts 1 and 2 outside the brackets indicate tunnel bore number, and $\Delta[CO_2]_{1,G}$ is the concentration of $CO_2$ in bore 1 attributed to LD vehicles, calculated using Eq. (3). Previously (Kirchstetter et al., 1999a), CO was used as a tracer for LD vehicle emissions assuming that diesel trucks and LD vehicles emit similar amounts of CO per vehicle-km traveled. This assumption is questionable especially in 2006 since CO emissions

have been reduced more for LD vehicles than for MD/HD diesel trucks (e.g. Bishop and Stedman, 2006; Burgard et al., 2006). Fuel economy, and thus $CO_2$ emissions per vehicle-km traveled have changed little in the last 10 years (Heavenrich, 2006).

Diesel contributions to $CO_2$ concentrations in the mixed traffic bore were calculated from observed traffic counts, estimated fuel economies, and known fuel properties using Eq. (3a). This is similar to the method used previously by Kirchstetter et al. (1999a), but is revised to include MD trucks explicitly rather than counting half of them as HD trucks:

$$\frac{\Delta[CO_2]_{1,D}}{\Delta[CO_2]_1} = \frac{\rho_D w_D (f_{hd} U_{hd} + f_{md} F U_{mdD})}{\rho_D w_D (f_{hd} U_{hd} + f_{md} F U_{mdD}) + \rho_G w_G (f_{ld} U_{ld} + f_{md}(1-F) U_{mdG})}. \quad (3a)$$

LD vehicle contributions to $CO_2$ concentrations in bore 1 were subsequently calculated as

$$\Delta[CO_2]_{1,G} = \Delta[CO_2]_1 - \Delta[CO_2]_{1,D}. \quad (3b)$$

In Eq. (3a), $f_x$ are the observed fractions of total vehicles that are LD, MD, and HD vehicles. $F$ is the fraction of MD vehicles equipped with diesel engines, as determined from truck census data for the US (TIUS, 1992; VIUS, 1997, 2002). For 1997, $F = 0.42$, and by extrapolating to 2006, $F$ was projected to have increased to 0.53. Gasoline and diesel (subscripts G and D, respectively) fuel densities $\rho$ and carbon weight fractions $w$ are from Kirchstetter et al. (1999a). Estimates of fuel consumption rates by vehicle category are presented in Table 2; these are derived from uphill traffic results in the Ft. McHenry tunnel reported previously by Pierson et al. (1996), and an extrapolation of in-use truck census data to 2006. Only the *relative* magnitudes of fuel consumption for different vehicle categories matter for the $CO_2$ apportionment in Eq. (3a); absolute amounts of $CO_2$ emitted inside the tunnel are measured directly to calculate emission factors via Eq. (1).

Table 2
Parameters used for bore 1 $CO_2$ apportionment in Eq. (3)

| Parameter | Symbol | Value (L 100 km$^{-1}$) |
|---|---|---|
| LD vehicle fuel consumption | $U_{ld}$ | 10.3 |
| MD diesel fuel consumption | $U_{mdD}$ | 27.0 |
| MD gasoline fuel consumption | $U_{mdG}$ | 28.4 |
| HD diesel fuel consumption | $U_{hd}$ | 49.5 |

## 3. Results

### 3.1. Traffic characterization

Table 3 shows traffic volumes by vehicle category observed on each day for 12:00–14:00 h in bore 1 (mixed traffic) and 16:00–18:00 h in bore 2 (LD only). Midday traffic volumes in bore 1 are approximately half of those observed in bore 2 during the afternoon peak period. The MD and HD truck fraction in bore 2 is low (<1%) in contrast to midday traffic in bore 1, which includes 7.3±0.6% MD plus HD trucks. Since bore 2 opens to eastbound (uphill) traffic around noon on weekdays,

Table 3
Traffic volumes (vehicles h$^{-1}$) in Caldecott tunnel, summer 2006

| Date | LD | MD | HD | MD + HD fraction of total (%) |
|---|---|---|---|---|
| | 2-axle/ 4-tire | 2-axle/ 6-tire | ⩾3-axle | |
| *Bore* 1 (12:00–14:00 h) | | | | |
| 18-Jul | 1877 | 88 | 58 | 7.2 |
| 19-Jul | 1897 | 106 | 49 | 7.6 |
| 20-Jul | 1949 | 111 | 50 | 7.6 |
| 21-Jul | 2011 | 105 | 64 | 7.7 |
| 24-Jul | 1853 | 105 | 57 | 8.0 |
| 25-Jul | 1835 | 89 | 54 | 7.2 |
| 26-Jul | 1933 | 99 | 48 | 7.1 |
| 27-Jul | 2308 | 90 | 52 | 5.8 |
| Average[a] | 1958±127 | 99±8 | 54±4 | 7.3±0.6 |
| *Bore* 2 (16:00–18:00 h) | | | | |
| 31-Jul | 3955 | 13 | 0 | 0.3 |
| 1-Aug | 3973 | 24 | 0 | 0.6 |
| 2-Aug | 3660 | 30 | 0 | 0.8 |
| 3-Aug | 3913 | 16 | 0 | 0.4 |
| 7-Aug | 3783 | 19 | 0 | 0.5 |
| 8-Aug | 3524 | 26 | 0 | 0.7 |
| 9-Aug | 3732 | 28 | 1 | 0.8 |
| 10-Aug | 3861 | 27 | 0 | 0.7 |
| Average[a] | 3800±131 | 23±5 | 0±0 | 0.6±0.2 |

[a]Reported as mean±95% confidence interval.

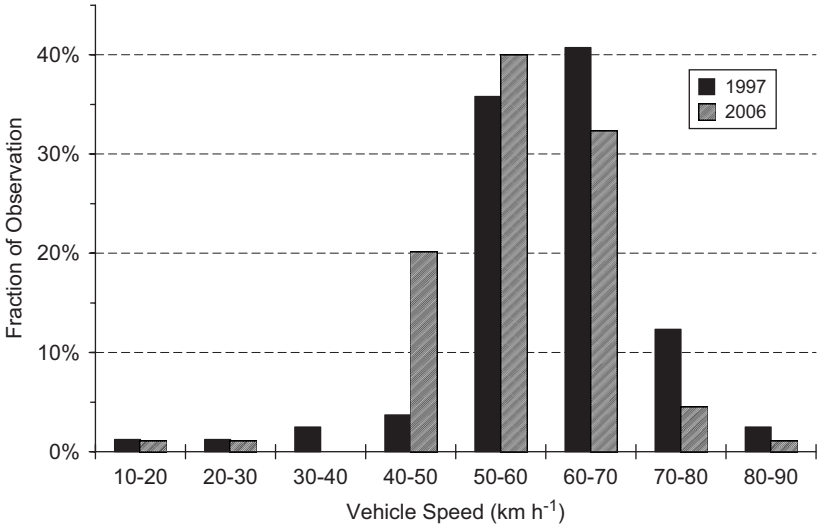


Fig. 2. Vehicle speed distribution for light-duty vehicles in bore 2. Mean vehicle speed was 57 km $h^{-1}$ ($N = 288$ vehicles) in 2006, and 59 km $h^{-1}$ ($N = 27$) in 1997.

many eastbound LD vehicles use bore 2 instead of bore 1 in the afternoon, increasing the MD/HD truck fraction in bore 1 compared to morning hours.

Fig. 2 shows a histogram of LD vehicle speeds from bore 2 including data from 2006 and 1997 for comparison. Average LD vehicle speeds were similar in both years (57 and 59 km $h^{-1}$). The mean speed of all HD trucks observed driving through bore 1 ($N = 702$) was 64 km $h^{-1}$, which is similar to the mean truck speed from the 1997 study of 65 km $h^{-1}$. The average LD vehicle model year was $2000.3 \pm 0.2$ ($N = 1711$ vehicles). The average model year of diesel trucks was $2000 \pm 1.6$ for MD trucks ($N = 22$) and $1997 \pm 2.2$ for HD trucks ($N = 24$). This matches results of an earlier study from 1997 (Kirchstetter et al., 1999a), which found average LD vehicle and HD truck ages of 6 and 9 years, respectively, corresponding to the same vehicle ages as the present study.

### 3.2. Carbon particle comparisons

BC concentrations measured in this study with the aethalometer agree well with those measured using TOA, as shown in Fig. S1 in the Appendix. This is despite reports of large discrepancies between BC measured with these two methods (Kirchstetter and Novakov, 2007). On average, the aethalometer yielded BC concentrations that were 1.09 and 1.21 times those determined by TOA in tunnel bores 1 and 2, respectively. Aethalometer-derived BC emission factors were therefore higher than those calculated from TOA.

Ten filter samples were analyzed by Sunset Laboratory (Tigard, OR) using the NIOSH TOT method to compare with parallel analysis of the same samples by the LBNL TOA method. This comparison was done because previous Caldecott tunnel data from 1997 used the NIOSH TOT method, whereas this study used the LBNL TOA method. A comparison of carbon contents of PM samples analyzed by the two methods is shown in Fig. 1. While different TOA methods can yield large differences in OC and BC contents, there is generally good agreement when the analyzed samples do not contain organic compounds that pyrolyze to form char during analysis (Watson et al., 2005; Kirchstetter and Novakov, 2007). Charring was not observed during analysis of any of the tunnel samples in this study. As a result, the OC and BC contents measured with the LBNL and NIOSH TOA methods are in good agreement.

### 3.3. Emission factors

Table 4 shows emission factors for LD vehicles and MD/HD diesel trucks, calculated using Eqs. (1)–(3). Both $NO_x$ and $PM_{2.5}$ emission factors decreased between 1997 and 2006 for all vehicle categories. The largest decrease was for $NO_x$ from LD vehicles.

Note that reported emission factors for MD/HD diesel trucks in Table 4 are less certain than

Table 4
Trends in measured LD vehicle and MD/HD diesel truck emission factors[a]

| Pollutant | Units | LD vehicles | | | MD/HD diesel trucks | | |
|---|---|---|---|---|---|---|---|
| | | 1997 | 2006 | % Change | 1997[b] | 2006 | % Change |
| $NO_x$[c] | $g\,kg^{-1}$ | $9.0\pm0.4$ | $3.0\pm0.2$ | $-67\pm3$ | $57\pm7$ | $40\pm3$ | $-30\pm9$ |
| $PM_{2.5}$ | $g\,kg^{-1}$ | $0.11\pm0.01$ | $0.07\pm0.02$ | $-36\pm17$ | $2.7\pm0.3$ | $1.4\pm0.3$ | $-48\pm12$ |
| TOA OM[d] | $g\,kg^{-1}$ | $0.05\pm0.01$ | $0.031\pm0.005$ | $-38\pm16$ | $0.59\pm0.08$ | $0.41\pm0.07$ | $-31\pm14$ |
| TOA BC[e] | $g\,kg^{-1}$ | $0.035\pm0.004$ | $0.022\pm0.004$ | $-37\pm13$ | $1.4\pm0.6$ | $0.86\pm0.07$ | $-39\pm26$ |
| Aeth BC[f] | $g\,kg^{-1}$ | | $0.026\pm0.004$ | | | $0.92\pm0.07$ | |
| Sct cross-sec[g,i] | $m^2\,kg^{-1}$ | | $0.10\pm0.02$ | | | $2.0\pm0.3$ | |
| Sct cross-sec[h,i] | $m^2\,kg^{-1}$ | | $0.090\pm0.009$ | | | $1.1\pm0.2$ | |
| Abs cross-sec[h,i] | $m^2\,kg^{-1}$ | | $0.20\pm0.05$ | | | $4.4\pm0.8$ | |
| Ext cross-sec[h,i] | $m^2\,kg^{-1}$ | | $0.29\pm0.05$ | | | $5.6\pm1.0$ | |

[a]Emission factors reported per unit mass of fuel burned. All values are mean $\pm$ 95% confidence interval.
[b]Diesel truck emission factors for 1997 differ from those reported previously due to changes in apportionment method (see text).
[c]$NO_x$ is reported as $NO_2$ equivalents (i.e., a molecular mass of $46\,g\,mol^{-1}$ was used to convert ppm to $\mu m^{-3}$).
[d]OM is obtained by multiplying measured OC by 1.4 to account for mass of hydrogen and oxygen (Gray et al., 1986). OC is corrected for the positive sampling artifact by subtracting the quartz behind Teflon filter mass (see text).
[e]BC from thermal optical analysis of filters.
[f]BC reported as 2 h average of continuous aethalometer data.
[g]Measured by nephelometer at $\lambda = 530$ nm.
[h]Measured by Cadenza at $\lambda = 675$ nm.
[i]As with all emission factors, the denominator is kg of fuel burned. These values should not be confused with mass specific cross sections in units of $m^2\,g^{-1}$ of particle mass.

corresponding results for LD vehicles due to the need to apportion pollutant concentrations in the mixed traffic bore (bore 1) between gasoline and diesel contributions. Values reported in Table 4 for 1997 differ from those in Kirchstetter et al. (1999a) due to changes in data analysis methods (Eqs. (2) and (3)). The diesel $NO_x$ emission factor for 1997 is 36% higher than reported previously, and lies at the high end of the range reported in other studies as reviewed by Jimenez et al. (2000).

Also note that the measured OC concentrations used to calculate emission factors have been corrected for the positive sampling artifact (see the Methods section), and have been multiplied by 1.4 to account for the mass of hydrogen and oxygen (Gray et al., 1986), as was done by Kirchstetter et al. (1999a). Thus, we report organic mass (OM) emission factors, an estimate of total particulate OM, not just the mass of OC. Carbon particles (BC plus OM) account for $76\pm16\%$ and $91\pm18\%$ of $PM_{2.5}$ mass for LD vehicles and diesel trucks, respectively.

Although absolute PM emission factors have decreased since 1997, ratios of BC to OM (BC/OM) and BC to TC (BC/TC) did not change significantly. The BC/OM ratio was $0.71\pm0.15$ and $2\pm1$ for LD vehicles and MD/HD diesel trucks, respectively. Likewise, the BC/TC ratio was $0.42\pm0.08$ and $0.7\pm0.3$ for LD vehicles and diesel trucks, respectively. BC dominates $PM_{2.5}$ emissions for diesel trucks.

Table 4 also shows absorption, scattering, and extinction cross-section emission factors. The optical cross-section emission factors for diesel trucks are an order of magnitude larger than for LD vehicles.

## 4. Discussion

### 4.1. $NO_x$ emission trends

Bishop and Stedman (2006) measured LD vehicle NO emission factors by remote sensing in Los Angeles in 1999, 2001, 2003, and 2005. As shown in Fig. 3a, this study indicates a 9% $year^{-1}$ reduction in the NO emission factor from LD vehicles, similar to the 7% $year^{-1}$ reduction observed at the Caldecott tunnel. Absolute NO emission factors in Los Angeles are higher than $NO_x$ emissions at the Caldecott tunnel due to an older vehicle fleet (6.5 vs. 5.7 years old). Note that NO emission factors by remote sensing are reported as $NO_2$ equivalents to allow for comparison with tunnel results.

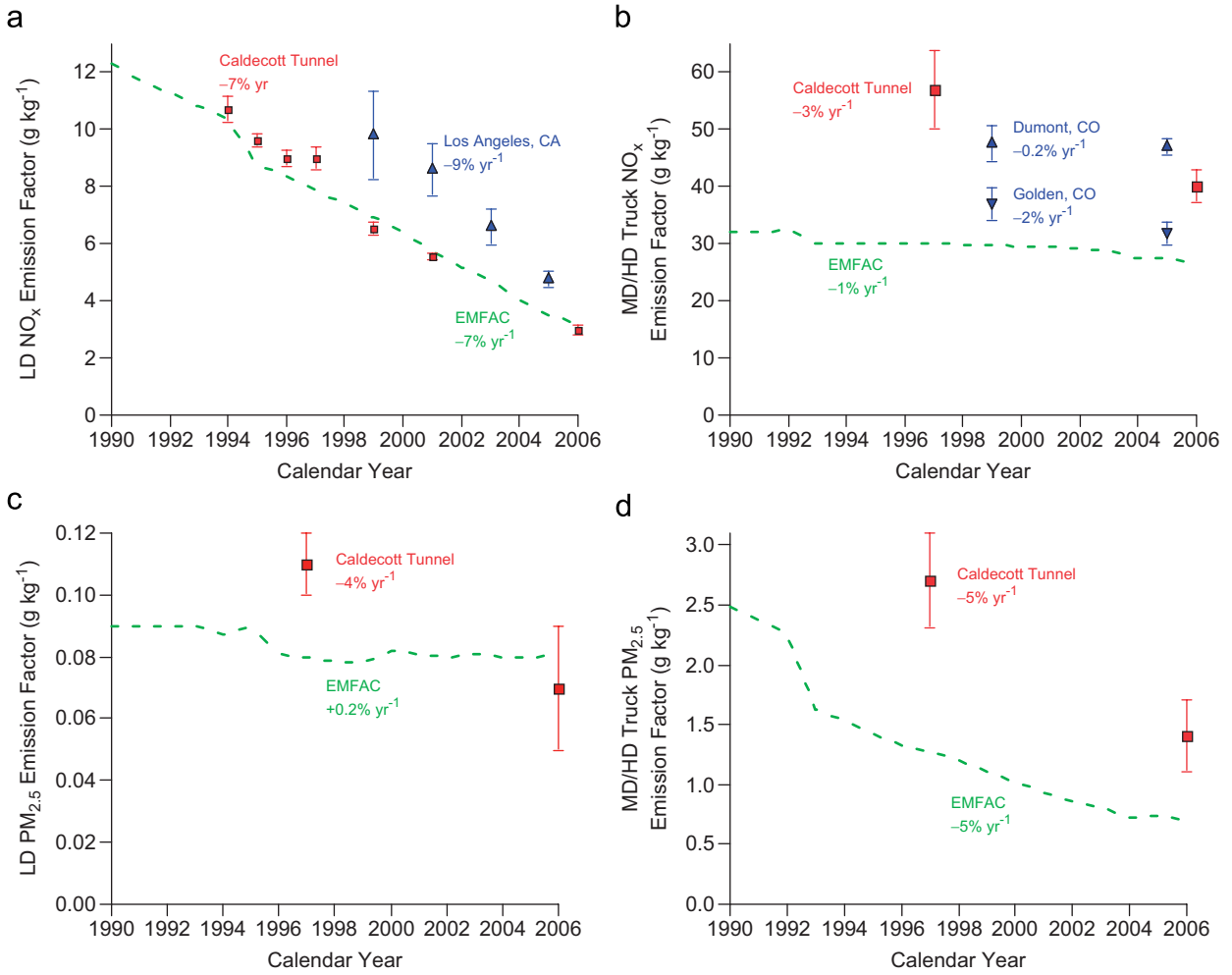


Fig. 3. $NO_x$ emission factors for (a) LD vehicles and (b) MD/HD diesel trucks. $PM_{2.5}$ emission factors for (c) LD vehicles and (d) MD/HD diesel trucks. Tunnel studies are squares, and remote sensing studies are triangles, both shown as mean ± 95% confidence interval. EMFAC predictions shown as the dashed line. Caldecott tunnel and EMFAC yearly percent reductions are for 1997–2006, even when data prior to 1997 are shown. NO emission factors are expressed as $NO_2$ equivalents for remote sensing studies.

Burgard et al. (2006) measured HD diesel truck NO and $NO_2$ emission factors by remote sensing at two Colorado locations in 2005, as shown in Fig. 3b. Dumont is at an elevation of 2530 m with a +1.8% grade, and Golden is at 1695 m with a grade of +0.2%. Burgard et al. suggest that the difference in $NO_x$ between these two sites is likely due to altitude effects. NO at Golden is 20% below the 2006 Caldecott tunnel result, and NO at Dumont is 17% higher. When compared to remote sensing data at the same locations from 1999 (Bishop et al., 2001), the annual rate of reduction in $NO_x$ emission factor at Golden was similar to the Caldecott tunnel. The lack of change in NO at Dumont is attributed to a different engine calibration that is allowed under high-altitude operating conditions (Stedman, 2007).

### 4.2. Particulate matter

Geller et al. (2005) measured PM emissions at the Caldecott tunnel in 2004. Their emission factors for LD vehicles and MD/HD diesel trucks was $0.07 \pm 0.03$ and $1.02 \pm 0.06\,g\,kg^{-1}$, respectively, matching 2006 results from the Caldecott tunnel within the stated uncertainties (95% confidence interval). However, note that Geller et al. used a longer sample period in both bores (12:00–18:00 h), whereas we measure bore 1 in the early afternoon (12:00–14:00 h) and bore 2 in the late afternoon

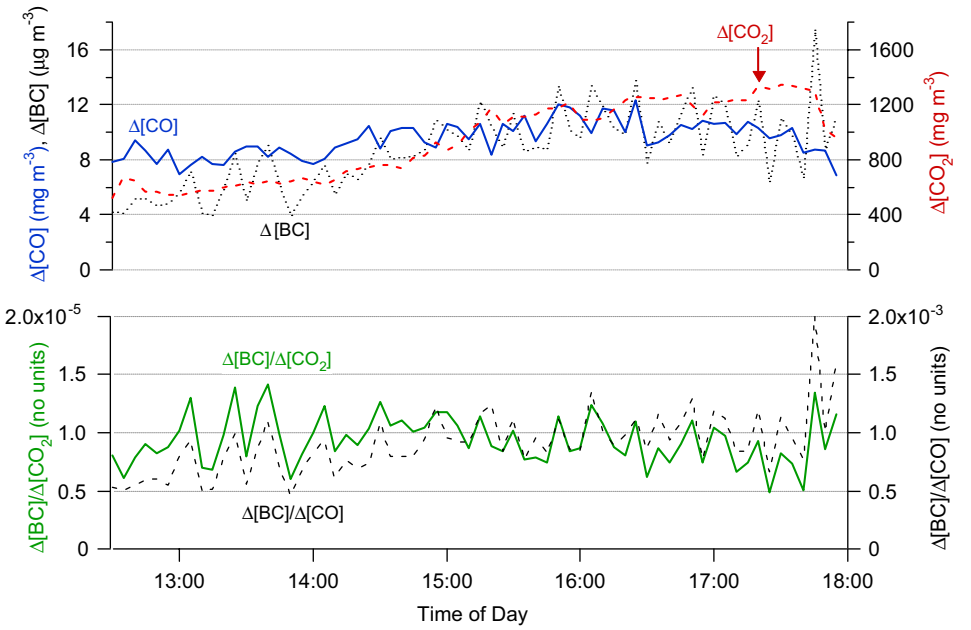


Fig. 4. Pollutant concentrations and BC ratios to CO and $CO_2$. Five minute averages from 12:30 to 18:00 h, averaged over the eight sampling days for bore 2. Top: pollutant concentrations (exit-entrance) of CO, black carbon, and $CO_2$. Bottom: ratios of Δ[BC] to Δ[CO] and Δ[$CO_2$].

(16:00–18:00 h). Thus, our average pollutant concentrations for bore 2 are more heavily weighted toward high-traffic conditions during rush hour, which lead to higher $CO_2$ concentrations. Note in Fig. 4 that Δ[$CO_2$] is higher from 16:00–18:00 h than 12:00–16:00 h. Geller et al. note that their Δ[$CO_2$] in bore 2 is lower than reported by Kirchstetter et al. (1999a) and hypothesize that the change is due to increased fuel efficiency of the on-road vehicle fleet. This $CO_2$ difference is actually due to comparing 6- vs. 2-h average concentration levels.

### 4.3. Comparison to EMFAC

EMFAC, a statistical model of on-road vehicle emissions (CARB, 2007), was used to predict LD vehicle and MD/HD diesel truck emissions. Emission factors for the San Francisco Bay area during the summers 1990–2006 are plotted in Fig. 3. Idle emissions and cold engine-starts were not included in this analysis since vehicles driving through the tunnel were already warmed up. EMFAC brake-wear emissions were not included in $PM_{2.5}$ as eastbound vehicles are generally accelerating or cruising after they enter the tunnel. Emission factors were calculated at vehicle speeds matching averages for LD vehicles and diesel trucks observed in the tunnel. EMFAC labels in Fig. 3 show yearly percent reductions from 1997 to 2006 for direct comparison with the Caldecott tunnel results. EMFAC predicts matching rates of emission reduction vs. those observed at the tunnel for LD $NO_x$ and diesel truck $PM_{2.5}$. EMFAC predicts lower rates of reduction for LD $PM_{2.5}$ and diesel $NO_x$ relative to the tunnel results.

### 4.4. Gasoline vs. diesel

$NO_x$ and particle emission factors for diesel trucks are higher than LD vehicles by an order of magnitude or more (Table 4). $NO_x$ from diesel trucks has decreased at a slower rate than for LD vehicles; the ratio of HD to LD emission factor for $NO_x$ increased from $6\pm1$ to $13\pm1$ between 1997 and 2006, which indicates an increase in the *relative* importance of diesel trucks as a source of $NO_x$ emissions. Comparing absolute values of LD vs. diesel truck PM emission factors may be misleading due to the load sensitivity of diesel engines for which BC emissions especially are expected to increase for uphill driving. In this case, comparing percent reductions is preferred. At first glance, Table 4 suggests that $PM_{2.5}$ emission factors for diesel trucks decreased more since 1997 than for LD vehicles. Given associated uncertainties, however, the changes in $PM_{2.5}$ emissions since 1997 could be

the same for both vehicle categories. Since sulfate comprised only 1.8% of $PM_{2.5}$ mass from HD trucks in 1997 (Kirchstetter et al., 1999a), the introduction of ULSD fuel in 2006 is likely to have played a small direct role in $PM_{2.5}$ reduction thus far. This fuel change is intended mainly to enable use of catalytic exhaust after-treatment devices starting in 2007. Therefore, $PM_{2.5}$ emission decreases reported here are attributed to vehicle technology improvements that occurred prior to the fuel change.

The scattering cross-section emission factors measured by the nephelometer and Cadenza are nearly the same for LD vehicles, but differ by a factor of two for diesel trucks (Table 4). This is likely due to the wavelength dependence of light scattering by diesel engine BC emissions. Single-scattering albedo $\omega$ is an important parameter in assessing the climate and visibility impacts of an aerosol, and is calculated by taking the ratio of scattering to extinction coefficient. Using emission factors measured by Cadenza shown in Table 4, $\omega$ was found to be $0.31 \pm 0.06$ and $0.20 \pm 0.05$ for LD vehicles and diesel trucks, respectively. Two caveats should be noted. First, $\omega$ is a strong function of wavelength. Values of $\omega$ measured by Cadenza at $\lambda = 675$ nm are expected to be lower than atmospheric values typically reported at $\lambda = 550$ nm. Second, the relatively fresh aerosol found in the tunnel has not significantly aged in the atmosphere. Further interpretation of these optical properties can be found in Strawa et al. (in preparation).

### 4.5. Modal effects

In order to apportion pollutant concentrations in bore 1 accurately, the emission ratio, $\Delta[P]/\Delta[CO_2]$, for LD vehicles needs to be well-represented by bore 2 measurements. Since the mean vehicle speed in bore 2 from 16:00 to 18:00 h is slightly lower than bore 1 from 12:00 to 14:00 h (Kirchstetter et al., 1999a), driving mode effects (i.e., effects due to changes in vehicle speed and engine load) on $\Delta[P]/\Delta[CO_2]$ may be an issue. Modal effects on LD vehicle CO and $NO_x$ emission factors have already been discussed for the Caldecott tunnel (Kirchstetter et al., 1999b; Kean et al., 2003). These studies found that both CO, and to a lesser extent, $NO_x$ emission factors increased with vehicle speed through the tunnel. Here we consider modal effects on BC emissions from LD vehicles.

Fig. 4 plots values of $\Delta[CO]$, $\Delta[BC]$, $\Delta[CO_2]$, $\Delta[BC]/\Delta[CO]$, and $\Delta[BC]/\Delta[CO_2]$ measured in bore 2. Generally, as the afternoon progresses vehicle speeds decrease due to increased traffic. It can be seen that $CO_2$, BC, and CO concentrations increase during the afternoon rush hour due to increased traffic volumes. Fig. 4 also shows that BC and $CO_2$ increase by similar relative amounts. Thus, $\Delta[BC]/\Delta[CO_2]$ does not show any trend over the afternoon, whereas $\Delta[BC]/\Delta[CO]$ increases over the same period. This illustrates an added advantage of using $CO_2$, rather than CO, as the basis for tracking LD vehicle emissions: $\Delta[BC]/\Delta[CO_2]$ is more stable than $\Delta[BC]/\Delta[CO]$. The lack of any trend in $\Delta[BC]/\Delta[CO_2]$ (proportional to BC emission factor) in Fig. 4 suggests that LD vehicle emissions of BC per unit fuel do not show strong dependence on driving mode over the range of uphill driving conditions observed here.

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## Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.atmosenv.2007.09.049.

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