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Measurements and predictors of on-road ultrafine particle concentrations and associated pollutants in Los Angeles

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Abstract

Motor vehicles are the dominant source of oxides of nitrogen (NO_x), particulate matter (PM), and certain air toxics (e.g., benzene, 1,3-butadiene) in urban areas. On roadways, motor vehicle-related pollutant concentrations are typically many times higher than ambient concentrations. Due to high air exchange rates typical of moving vehicles, this makes time spent in vehicles on roadways a major source of exposure. This paper presents on-road measurements for Los Angeles freeways and arterial roads taken from a zero-emission electric vehicle outfitted with real-time instruments. The objective was to characterize air pollutant concentrations on roadways and identify the factors associated with the highest concentrations. Our analysis demonstrated that on freeways, concentrations of ultrafine particles (UFPs), black carbon, nitric oxide, and PM-bound polycyclic aromatic hydrocarbons (PM-PAH) are generated primarily by diesel-powered vehicles, despite the relatively low fraction (~6%) of diesel-powered vehicles on Los Angeles freeways. However, UFP concentrations on arterial roads appeared to be driven primarily by proximity to gasoline-powered vehicles undergoing hard accelerations. Concentrations were roughly one-third of those on freeways. By using a multiple regression model for the freeway measurements, we were able to explain 60–70% of the variability in concentrations of UFP, black carbon, nitric oxide, and PM-PAH using measures of diesel truck density and hour of day (as an indicator of wind speed). Freeway concentrations of these pollutants were also well correlated with readily available annual average daily truck counts, potentially allowing improved population exposure estimates for epidemiology studies. Based on these roadway measurements and average driving time, it appears that 33–45% of total UFP exposure for Los Angeles residents occurs due to time spent traveling in vehicles.
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1. Introduction

Motor vehicles are a dominant source of air pollution in urban areas. In California, motor vehicles are the primary source of oxides of nitrogen

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(NO_x), particulate matter o 2.5 mm diameter (PM2.5), carbon monoxide (CO), benzene, polycyclic aromatic hydrocarbons (PAH), and 1,3-butadiene (CARB, 2006). Exposure to PM is associated with significantly increased rates of respiratory and cardiovascular illness, hospitalization, and pre-mature mortality (e.g., Dockery, 2001; Pope et al., 1995, 2002; Samet et al., 2000; Schwartz et al., 2002; Zanobetti et al., 2003). As recent studies have indicated, the size of airborne particulate is an important factor in generating health effects; ultrafine particulate (PMo 0.1 mm) is associated with greater toxicity on a per-mass basis than larger particles (Donaldson et al., 1998; Li et al., 2003; Nel et al., 2006; Obersdörster, 2001; Xia et al., 2004).

Most studies linking illness or death to particles use ambient concentrations to estimate exposures, but actual exposures to motor vehicle emissions are generally higher than ambient concentrations indicate. One important source of exposure to vehicle-related pollutants is time spent traveling in vehicles. Due to the typically high air exchange rates of moving vehicles, in-vehicle concentrations are typically close to roadway concentrations. For example, Rodes et al. (1998) found 15–40 air changes per hour at 55 mph with windows closed and fans set on "recirculation mode," the lowest air exchange rate condition in three vehicles, two of which were new and one was 6 years old. As a result, time spent in vehicles contributes a large fraction of total exposure, such as 30–55% of Californian's total exposure to diesel PM (Fruin et al., 2004).

Recent studies have used real-time mobile monitoring technologies to assess on-road concentrations with spatial and temporal resolution (Bukowiecki et al., 2002a, b; Canagaratna et al., 2004; Kittelson et al., 2004; Pirjola et al., 2004, 2006; Weijers et al., 2004; Westerdahl et al., 2005). Real-time instruments are necessary to characterize the highly variable and rapidly changing concentrations generated by mobile sources. When combined with video analysis and other measures of traffic and driving conditions, real-time instruments can be used to unambiguously identify associations between high roadway concentrations and particular vehicles types and traffic conditions.

We recently used real-time mobile measurements to characterize ultrafine particle (UFP) and related vehicle air pollutant concentrations on roadways in the Los Angeles region (Westerdahl et al., 2005). Our freeway route covered the widest range of traffic and truck traffic densities of any mobile monitoring study to date, with widely varying contributions from gasoline- and diesel-powered vehicles during each sampling run. This paper demonstrates strong associations between heavy-duty diesel truck counts and high concentrations of (UFP), nitric oxide (NO), black carbon (BC), and particle-bound PAHs (PM-PAH) that are relatively independent of total vehicle traffic volumes.

2. Methods

2.1. Data collection

A zero-emitting 1998 electric Toyota RAV4 sport utility vehicle served as a mobile monitoring platform (MMP). Using a vehicle having no emissions eliminated the possibility of sampling the MMP's own exhaust plume when stationary. Table 1 provides a summary of the instruments used. Sampling intervals were 10 s or less, except for the Aethalometer, which sampled BC for 1-min intervals. BC, PM-PAH, and NO were measured as indicators of diesel exhaust, along with NO_2, UFP number, particle size distributions, and particle length (via the EAD 3070A from TSI). CO was measured as an indicator of gasoline-powered vehicle exhaust, particularly high emitters and/or hard accelerations, and CO_2 was measured to indicate total traffic volumes, as it is produced in large quantities by all vehicles. To remove the effect of study car occupants' own CO_2 contributions, outside air was pulled into a 6-in diameter galvanized steel duct, located 1.5 m above the roadway in the rear passenger space of the vehicle, and exhausted out of the rear, driver-side window. Each instrument sampled from ports in the duct, connected by short lengths (0.5–2 m) of conductive tubing. This arrangement also allowed a faster instrument response to changing conditions outside the vehicle when the vehicle was not moving. A complete description of instrumentation, quality assurance, calibration, and platform assembly is provided in Westerdahl et al. (2005).

The MMP was driven on a fixed, 33-mile arterial road route and a 75-mile freeway route as shown in Fig. 1. The freeway route covered nearly an order of magnitude difference in freeway traffic volumes, from 46 000 vehicles per day on the northern portion of CA-110 near Pasadena to 330 000 vehicles per day on I-10. The distance-weighted route average was 200 000 vehicles per day (CalTrans, 2004). The volume of diesel truck traffic

Table 1
Monitoring instruments employed in the Mobile Monitoring Platform (reproduced from Westerdahl et al., 2005)

Instrument	Parameter measured	Time resolution
TSI Portable CPC, Model 3007	Particle count, 10 nm–1 mm	10 s
TSI CPC, Model 3022A	Particle count, 7 nm–1 mm	10 s
TSI Electrical Aerosol Detector, Model 3070A	Particle length mm cm^{-3}	2 s
Magee Scientific Portable Aethalometer	Black carbon	1 min
TSI Scanning Mobility Particle System, Model 3080 classifier:	Particle counts/size	1 min scan rate
Nano DMA, w/3025 CPC	5–153 nm	
(long) DMA, w/3025 CPC	16–600 nm	
EcoChem PAH Analyzer, Model PAS 2000	PM-PAH ($ng\,m^{-3}$)	2 s
TSI DustTrak	PM2.5	10 s
Teledyne-API NO_x Analyzer, Model 200e	NO, NO_x, NO_2	20 s
TSI Q-Trak Plus Monitor, Model 8554	CO, CO_2, temp., humidity	10 s

(1.5 ton with dual rear tires or larger) varied by nearly two orders of magnitude, from less than 300 trucks per day near Pasadena on northern CA-110, to 28 000 trucks per day on I-710, the major truck route from the Ports of Los Angeles and Long Beach. Truck fractions ranged from less than 2% on the CA-110 to 25% at times of peak midday truck traffic on I-710 (CalTrans, 2004). The route average truck count was about 18 000 trucks per day.

The arterial route was driven on Jefferson Boulevard west to Western Avenue, north to Wilshire Boulevard, west to La Cienega Boulevard, north to Santa Monica Boulevard, east to Western Boulevard, south to El Segundo Boulevard, east to Avalon Boulevard, and north to USC. These streets are all major arterial roads, typically two lanes wide in each direction, with stop lights at major intersections and consistently heavy traffic during most of the day.

Measurements were conducted from February through April 2003. Of the freeway route runs, two weekdays in February and two in April were randomly selected for detailed analysis from days with typical meteorology (e.g., no recent rains). For these days, measurements were conducted during 2- to 4-h periods, from 9 to 11 a.m. on 7 April, 11 to 2 p.m. on 20 February and 16 April, and 2 to 6 p.m. on 14 February. These times included waxing and waning portions of rush hour traffic, which allowed a full mix of conditions and speeds to all be included in a single run, from high speed, free-flowing conditions to congested, stop-and-go traffic. The arterial route was analyzed for two days, 14 April, from 11:45 a.m. to 12:45 p.m. and 15 April, 2:30 to 4:00 p.m.

Meteorological data were collected from four stations nearest the route (Pasadena, downtown Los Angeles, Long Beach, and Lynwood). Wind speeds and directions on each of these days were typical of Los Angeles during springtime, except for 16 April where wind speeds were about 50% higher than normal. The morning on-shore and afternoon off-shore winds typical of Los Angeles during most of the year occurred on all days. Meteorological conditions are described more fully in Westerdahl et al. (2005).

A digital video camera was mounted above the RAV4 dashboard to record traffic conditions during all runs. Digital video tapes were archived to DVD for subsequent review and analysis. The video camera recorded time, driver observations, the lane driven, surrounding traffic, the type of vehicle followed, and general driving conditions such as relative speed and congestion. Location was recorded with a global positioning system at 1-min intervals.

2.2. Digital video disc (DVD) analysis

A key component to this study was the analysis of the correlation between measured pollutant concentrations and traffic conditions such as speed and

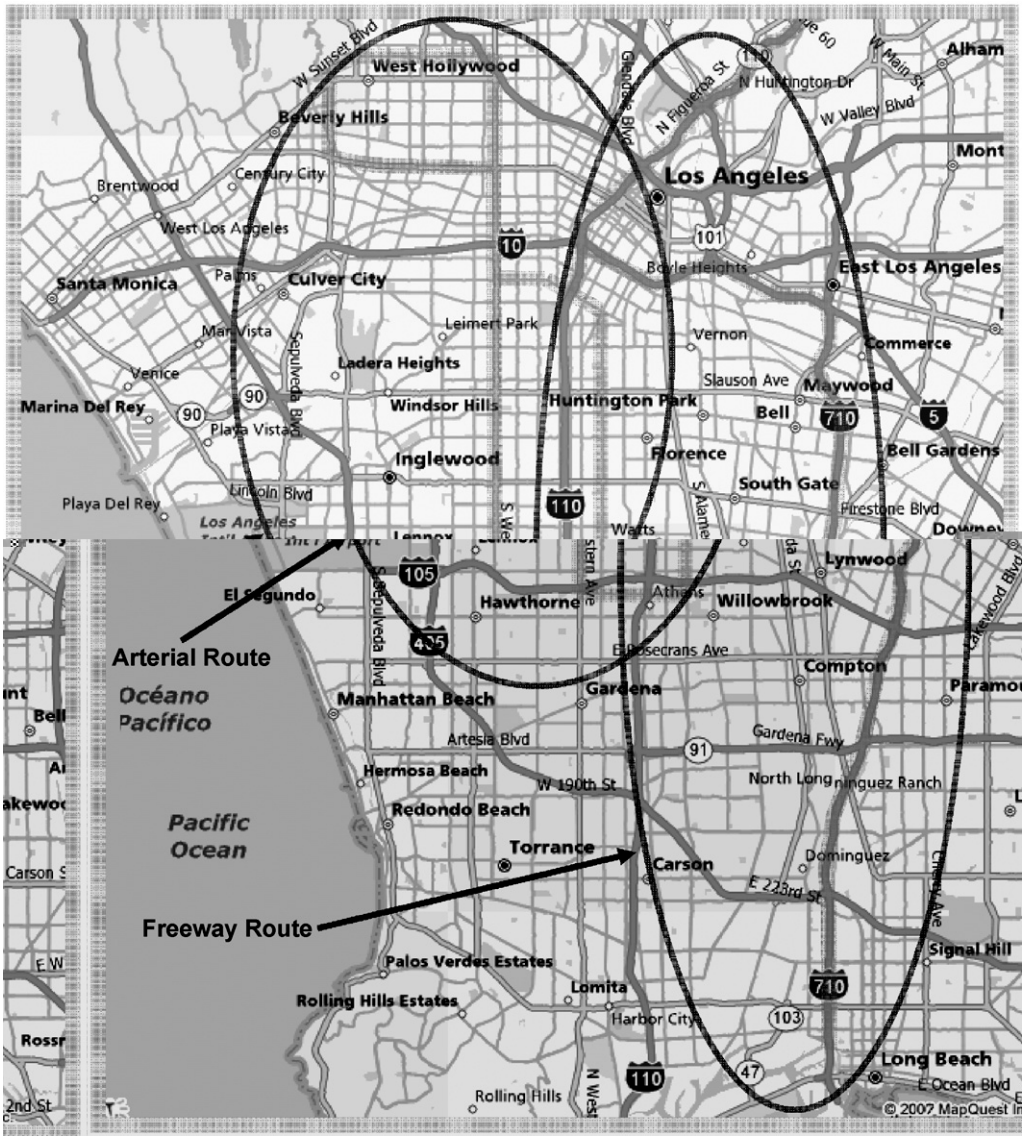

Fig. 1. Freeway route.

congestion. For freeway driving we recorded second-by-second observations of any changes in:

- Vehicle speed: parked, stopped, low (0–19 mph), medium (20–39 mph), high (40–60 mph), or very high (60 + mph).
- Acceleration: parked, steady state, acceleration, deceleration, or a mixture of acceleration and deceleration.
- Vehicle followed: NA (parked), none, passenger car, sport utility vehicle, two-axle diesel vehicle, and three- to five-axle diesel vehicle.
- Road type: parked (freeway shoulder or residential), arterial, arterial intersection, freeway, freeway entrance, or freeway exit.
- Road name and direction of travel.
- Surrounding traffic density: NA (if parked), none, low, medium (many vehicles but free flowing), or high (reduced speeds due to vehicle density).

The video analysis for the arterial route used similar descriptors, but an additional critical variable was the number of leading and surrounding