Atmospheric Environment 83 (2014) 211–219

Contents lists available at ScienceDirect

Atmospheric Environment

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

Spatial variation in particulate matter components over a large urban area

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HIGHLIGHTS

- Size-resolved PM mass, carbon components and NO_x measured spatially.
- Local traffic impacts observed for elemental and organic carbon and NO_x but not PM.
- Traffic impacts were relatively larger in cleaner communities.
- No PM components were better markers of traffic than NO_x.

ARTICLE INFO

Article history:
Received 20 May 2013
Received in revised form
15 October 2013
Accepted 29 October 2013

Keywords:
Air pollution
Particulate matter
Traffic emissions
Spatial variability

ABSTRACT

To characterize exposures to particulate matter (PM) and its components, we performed a large sampling study of small-scale spatial variation in size-resolved particle mass and composition. PM was collected in size ranges of <0.2, 0.2-to-2.5, and 2.5-to-10 μm on a scale of 100s to 1000s of meters to capture local sources. Within each of eight Southern California communities, up to 29 locations were sampled for rotating, month-long integrated periods at two different times of the year, six months apart, from Nov 2008 through Dec 2009. Additional sampling was conducted at each community's regional monitoring station to provide temporal coverage over the sampling campaign duration. Residential sampling locations were selected based on a novel design stratified by high- and low-predicted traffic emissions and locations over- and under-predicted from previous dispersion model and sampling comparisons. Primary vehicle emissions constituents, such as elemental carbon (EC), showed much stronger patterns of association with traffic than pollutants with significant secondary formation, such as $PM_{2.5}$ or water soluble organic carbon. Associations were also stronger during cooler times of the year (Oct through Mar). Primary pollutants also showed greater within-community spatial variation compared to pollutants with secondary formation contributions. For example, the average cool-season community mean and standard deviation (SD) for EC were 1.1 and 0.17 μg m^{-3}, respectively, giving a coefficient of variation (CV) of 18%. For $PM_{2.5}$, average mean and SD were 14 and 1.3 μg m^{-3}, respectively, with a CV of 9%. We conclude that within-community spatial differences are important for accurate exposure assessment of traffic-related pollutants.

1. Introduction

To evaluate the potential health effects in children of long-term exposures to poor air quality, the Southern California Children's Health Study (CHS) was launched in 1992 (Peters et al., 1999). CHS research has shown that regional levels of ambient air pollution are associated with reduced rates of lung function growth (Gauderman et al., 2004). At a finer spatial scale, statistically significant associations have been observed between residential proximity to busy

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http://dx.doi.org/10.1016/j.atmosenv.2013.10.063

roads (<75 m) and asthma prevalence (Gauderman et al., 2005; McConnell et al., 2006), as well as between residential proximity to freeways (<500 m) and both asthma (Gauderman et al., 2005) and reduced rates of lung function growth (Gauderman et al., 2007). These findings complement emerging evidence suggesting residential, near-road traffic-related pollutant (TRP) exposures are linked to respiratory infections and allergy (Brauer et al., 2002; Janssen et al., 2003), asthma and wheeze (Venn et al., 2001), and other health outcomes (Wjst et al., 1993; van Vliet et al., 1997; English et al., 1999; Venn et al., 2000; Nicolai et al., 2003; Kim et al., 2004; Zmirou et al., 2004; Gauderman et al., 2005). However, the reported associations between residential proximity to busy roads and childhood asthma are inconsistent (HEI, 2010), suggesting roadway proximity may not be a sufficiently adequate proxy for TRP exposure.

To more accurately estimate TRP exposures, fine-scale spatial variability of traffic-related air pollutants (TRPs) must be better understood. This is challenging because TRP concentration gradients are often steep, with several-fold concentration differences observed in less than 100 m (Rodes and Holland, 1981; Zhu et al., 2002). Spatially dense measurements are therefore necessary, and historically have been only achieved using low-cost, passive samplers for NO_x. However, NO_x may be only a surrogate for certain TRPs and not the broader range of TRPs that may be driving the adverse health effects linked to living near traffic, such as diesel particulate matter (black carbon) (Janssen et al., 2011) and ultrafine particles (Delfino et al., 2005). Furthermore, these TRPs may have different spatial patterns than those readily captured by NO_x.

This study compared within-community variation in size-resolved PM and PM components at "middle scale" (100–500 m) and "neighborhood scale" (500 m–4 km) to between-community differences over "urban scales" of 4–100 km, these scales being defined by USEPA to characterize areas of influence of various sources of primary and secondary PM (Watson et al., 1997).

By measuring specific components of PM in several size fractions, our goal was to develop a database suitable for estimation of exposure to different components of traffic-related PM in each of 8 CHS communities and to develop transferrable models for use in other locations. This database could then be utilized to reduce current exposure assignment uncertainties presently encountered using distance or reactive gases as proxies for TRP. The long-term goal for this research effort is to accurately quantify exposure to the TRP PM components most responsible for the adverse health effects identified in the CHS.

We hypothesized that primary PM components emitted directly from vehicles, such as EC or organic carbon (OC), have: 1) greater within-community variability compared to secondary PM components such as $PM_{2.5}$ or water-soluble OC (WSOC); 2) higher concentrations and greater roadway impacts during the cooler time periods of the year (due to reduced meteorological mixing); 3) larger freeway impacts compared to arterial roads (due to greater source strength); and 4) greater relative localized impacts from traffic in communities with lower overall pollution levels. We also hypothesized that these differences are more pronounced for the smaller PM sizes (0.2 μm compared to 2.5 μm), due to their shorter atmospheric lifetime, and that sub-0.2 μm-sized TRP PM components would be better markers of traffic than passive-sampler measurements of NO_x compounds, also due to shorter lifetimes and sharper concentration gradients.

2. Methods

2.1. Sampling instrumentation

Two-week time-integrated size-resolved PM samples were collected with a Harvard Cascade Impactor (CI) (Lee et al., 2006), modified to include an additional collection stage with a 0.2 μm cut-point. Additional CI stages were operated at 0.5, 2.5, and 10 μm to capture accumulation mode fine ($PM_{0.2-2.5}$) and coarse $PM_{2.5-10}$ (CPM) fractions. Poly-urethane foam (PUF) was used as the collection media in all CI upper stages to minimize particle bounce and allow larger mass accumulation than filter-based substrates (Kavouras and Koutrakis, 2001). Teflon filters (37 mm, 2 μm pore size) or quartz filters (Tissuquartz 37 mm) were used on the final impactor stage to collect $PM_{0.2}$. For comparability to other studies, in this study we report the combined <0.2 μm and 0.2-to-2.5 μm stages as $PM_{2.5}$.

At each sampling location, a sound-insulated pump box with programmable timer, elapsed time meter, multiple sampling lines leading to a single pump, and a weather-protective sun shield was deployed. Three of the four sampling lines were connected to individual cascade impactors, operated at 5 L per minute, while a fourth sampling line was used to collect $EC_{2.5}$ and $OC_{2.5}$ samples with a Harvard $PM_{2.5}$ single-stage impactor, operating at 1.8 L per minute, using pre-baked 37 mm quartz filters. NO_x and NO_2 concentrations were measured using passive samplers (Ogawa & Co USA Inc., FL, USA) at a subset of locations. HOBO temperature and relative humidity data-logging samplers (Onset Computer Corporation, MA, USA) were also deployed at several locations per community.

2.2. Sampling location selection and schedule

Fig. 1 shows the location of the communities sampled. These were initially selected for the CHS to represent different combinations of high and low regional ambient pollutant mixtures across Southern California (Peters et al., 1999). Santa Barbara, a coastal community northwest of Los Angeles and outside of the South Coast Air Basin, represents a lower-pollution contrast to the other communities sampled. Long Beach, a coastal community in the southwestern region of the Los Angeles Air Basin, has high volumes of container ship and container-hauling truck sources and locomotive diesel emissions from the Ports of Los Angeles and Long Beach, in addition to several petroleum refineries and operating oil fields. The remaining communities, in addition to local traffic pollution sources, generally show increasing regional pollution levels from west to east, in the direction of the prevailing daytime on-shore winds. Similarly, secondary aerosol formation from daytime and nighttime chemistry increases from west to east across the basin.

To estimate annual average concentrations, we sampled for a month-long integrated period at two different times of the year, similar to the analyses reported by Xu et al. (2007). Field sampling was conducted in a simultaneous, rotating, community-paired cycle, from December 2008 to December 2009, as shown in Table 1. To provide a better estimate of annual average exposure in each community, an additional two waves of four-week sampling were conducted at community central air monitoring stations at off-cycle intervals. Community monitors were operated by the South Coast Management District as part of their standard federal monitoring requirements to test compliance with USEPA National Ambient Air Quality Standards. Due to a wildfire in Santa Barbara during sampling wave B, an additional sampling wave (E) was conducted in Santa Barbara to replace B.

In each of the eight CHS communities, 26–29 sampling sites were selected. These included CHS students' homes, community regional air monitoring site locations, and neighborhood elementary schools attended by CHS participants. CHS participants' homes were stratified into the highest one-third and lowest two-thirds of dispersion-model-predicted annual average TRP concentrations, using CALINE-4 (Benson and Pinkerman, 1984).