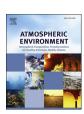


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Transition metals in coarse, fine, very fine and ultra-fine particles from an interstate highway transect near Detroit



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HIGHLIGHTS

- I-96 freeway PM_{2.5} mass emission rate per 5000 v/hr, $2.0 \pm 0.7 \mu g/m^3$, measured.
- I-96 freeway PM_{2.5} mass emission rate per 5000 v/hr, $1.6 \pm 0.5 \mu g/m^3$, modeled.
- Using European PM_{2.5} emission rates at the I-96 site, $3.1 \pm 1 \mu g/m^3$, modeled.
- Using California 1973 PM_{2,5} emission rates at I-96, $16 \pm 3 \mu g/m^3$, modeled.
- Very fine and ultra-fine transition metals were regional, not from the I-96 site.

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ABSTRACT

As one component of a study investigating the impact of vehicle emissions on near-road air quality, human exposures, and potential health effects, particles were measured from September 21 to October 30, 2010 on both sides of a major roadway (Interstate-96) in Detroit. Traffic moved freely on this 12 lane freeway with a mean velocity of 69 mi/hr. with little braking and acceleration. The UC Davis DELTA Group rotating drum (DRUM) impactors were used to collect particles in 8 size ranges at sites nominally 100 m south, 10 m north, 100 m north, and 300 m north of the highway. Ultra-fine particles were continuously collected at the 10 m north and 100 m north sites. Samples were analyzed every 3 h for mass (soft beta ray transmission), 42 elements (synchrotron-induced x-ray fluorescence) and optical attenuation (350 -800 nm spectroscopy). A three day period of steady southerly winds along the array allowed direct measurement of freeway emission rates for coarse (10 > D_p > 1.0 μm), PM_{2.5}, very fine $(0.26 > D_p > 0.09~\mu m)$, and ultra-fine $(D_p < 0.09~\mu m)$ particles. The PM_{2.5} mass concentrations were modeled using literature emission rates during the south to north wind periods, and averaged 1.6 \pm 0.5 $\mu g/m^3$, versus the measured value of $2.0 \pm 0.7 \mu g/m^3$. Using European freeway emission rates from 2010, and modeling them at the I-96 site, we would predict roughly 3.1 µg/m³ of PM_{2.5} particles, corrected from the 4.9 PM₁₀ value by their measured road dust contributions. Using California car and truck emission rates of 1973, this value would have been about $16 \mu g/m^3$, corrected down from the $19 \mu g/m^3 PM_{5.0}$ using measured roadway dust contributions. This would have included 2.7 µg/m³ of lead, versus the 0.0033 µg/ m³ measured. Very fine particles were distributed across the array with a relatively weak falloff versus distance. For the ultra-fine particles, emissions of soot and metals seen in vehicular braking studies correlated with traffic at the 10 m site, but only the soot was statistically significant at the 100 m north site. Otherwise, the 10 m north and 100 m north sites were essentially identical in mean concentration and highly correlated in time for most of the 5 week study. This result supports earlier publications showing the ability of very fine and ultra-fine particles to transport to sites well removed from the freeway sources. The concentrations of very fine and ultra-fine metals from brake wear and zinc in motor

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oil observed in Detroit have the potential of being a significant component in statistically established $PM_{2.5}$ mortality rates.

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1. Introduction

Enormous advances have been made in the past 40 years in reducing particulate pollutants from highway vehicles, including the elimination of lead from gasoline. Additionally, important advances have been made in reducing emissions from diesel trucks by improved engine design, low-sulfur fuels, and especially diesel particulate filters. However, studies show that highways continue to be a source of both coarse and fine particulate matter (PM) including known toxics such as diesel exhaust (Zhu et al., 2002; Karner et al., 2010; Cahill and Cahill, 2013). In addition, particles associated from vehicular wear such as from brake pads and drums are currently roughly equal to tailpipe emissions and are on track to exceed tail pipe emissions in the near future (Denier Van der Gon et al., 2013). Only limited data are available on wear particles by size and composition, but a major source is known to be ultra-fine metals from abrasion of brake pads and drums (Cahill et al., 2014).

In vitro laboratory studies of nanoparticles raise concerns about the health impacts of highway emissions, especially very fine and ultra-fine metals due to their propensity for penetrating to the deepest portions of the lung and diffusing into the circulatory system (Lewis et al., 2005; Chen and Lippmann, 2009; Lippmann, 2009; Oberdorster et al., 2007; Denier Van der Gon et al., 2013). Additionally, epidemiological health studies continue to show that highway emissions are damaging to both pulmonary and cardio-vascular systems of people living near highways (Cahill et al., 2011), with reduced lung function in children (Peters et al., 1999a,b); Gauderman et al., 2000; HEI, 2009; Lin and Peng, 2010; Karner et al., 2010). These data have been combined with data on the health impacts of roadway pollutants, including potential cancer impacts largely from diesel exhaust, and used to generate estimated health impacts in models such as Emfac2007 (ARB, 2007).

A recent European survey noted that wear particles were approaching and would soon surpass exhaust particles near roadways, with uncertain health impacts. Their consensus statement concludes, "In light of the continuous increase of the relative contribution of non-exhaust emission to ambient PM, where it is becoming the dominant emission process for urban transport, it is more than timely to devote greater efforts to properly quantifying non-exhaust emissions and assessing health relevance." (Denier Van der Gon et al., 2013).

The U.S. Environmental Protection Agency (EPA) and the University of Michigan conducted the Near-road Exposures to Urban Air Pollutants Study (NEXUS) — a research project to study the impact of vehicle emissions on near-road air quality, human exposures, and potential health effects. Components of NEXUS include but are not limited to human exposures in near-road residences (Vette et al., 2013), studies of near roadway allergic impacts in mice (McGee et al., 2015). The NEXUS aerosol component reported in this publication was designed and conducted to update our information on highway emissions and their potential human impact with a component on vehicular wear emissions.

1.1. Experimental methodologies

1.1.1. Sampling array

Particles were measured from September 21 to October 30, 2010

on both sides of the Interstate 96 freeway 9 miles west of downtown Detroit. The I-96 monitoring location was established as part of a collaborative research study conducted by the U.S. Environmental Protection Agency (EPA) and the U.S. Department of Transportation Federal Highway Administration (FHWA) (US EPA, 2013). The test section was a 12-lane section of I-96 with an atgrade roadway configuration aligned east to west. Obstructions from vegetation between the test array of four sites, which extended from 100 m south of the highway to 300 m north of the highway, were negligible, as seen in Fig. 1. As part of NEXUS, additional PM monitoring was conducted at these sites.

Particles were collected in 8 size ranges at sites 100 m south, 10 m north, 100 m north, and 300 m north of the highway, while ultra-fine particles were continuously collected at the 10 m north and 100 m north sites (Table 1). All sampling sites were on the top of trailers or one-story buildings with minimal obstructions (approximately 4 m from ground to inlets). The exception was the 100 m south site that had heavy trees south of the site, which did not obstruct the air flow from the highway.

1.1.2. Weather

Wind direction and wind speed were measured continuously at the 100 m north site (Fig. 2). During most of the study winds were weak, and extensive periods of calms occurred, especially at night. Wind directions were most frequent from the northwest (315°) through north northeast (35°). Given the southeast to northwest orientation of the sampling array, an upwind/downwind analysis of I-96 traffic emissions is best conducted when winds are from the southeast to southwest sector.

Thus, in order to address the prime goal of the study, a small fraction of the entire five-week period was chosen for which there was a clean upwind-downwind profile along the array. Since the goal was to quantify aerosol transport downwind of the freeway, results were limited to periods when the upwind-downwind trajectory was within $\pm 67^{\circ}$ of the north-south axis across the eastwest freeway with wind speeds >1 km/hr. Periods of extreme wind or rain events were excluded as well as wind trajectories pointing back to strong upwind sources such as the Monroe coal-fired power plant.

In terms of duration, while any 3 h period provides a valid signature in time, the ideal is several days in a row. The reasons include reducing the inherent 1½ hr uncertainty in the elemental data set by the width of the x-ray analysis microprobe to roughly 30 min, which allows better accord with meteorology and traffic patterns. It also allows better sensitivity by summing the periods for averaging actions such as size profiles. Limiting the intensive to week days avoided the complexity of highly variable weekend conditions, especially in terms of the truck traffic.

1.2. Experimental techniques

1.2.1. Sample collection

The primary aerosol collection instrument was the UC Davis DELTA Group rotating drum (DRUM) impactor (Cahill et al., 1985; Raabe et al., 1988) delivering aerosol samples in 8 size ranges: 10 to 5.0, 5.0 to 2.5, 2.5 to 1.15, 1.15 to 0.75, 0.75 to 0.56, 0.56 to 0.34, 0.34 to 0.26 and 0.29 to 0.09 μ m aerodynamic diameter. Samples

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