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Rapid physical and chemical transformation of traffic–related atmospheric particles near a highway

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ABSTRACT

The health of a substantial portion of urban populations is potentially being impacted by exposure to traffic–related atmospheric pollutants. To better understand the rapid physical and chemical transformation of these pollutants, the number size distributions of non–volatile traffic–related particles were investigated at different distances from a major highway. Particle volatility measurements were performed upwind and downwind of the highway using a fast mobility particle sizing spectrometer with a thermodenuder on a mobile laboratory. The number concentration of non–denuded ultrafine particles decreased exponentially with distance from the highway, whereas a more gradual gradient was observed for non–volatile particles. The non–volatile number concentration at 27 m was higher than that at 280 m by a factor of approximately 3, and the concentration at 280 m was still higher than that upwind of the highway. The proportion of non–volatile particles increased away from the highway, representing 36% of the total particle number at 27 m, 62% at 280 m, and 81% at the upwind site. A slight decrease in the geometric mean diameter of the non–volatile particle size distributions from approximately 35 nm to 30 nm was found between 27 m and 280 m, in contrast to the growth of non–denuded particles with increasing distance from the highway. Single particle analysis results show that the contribution of elemental carbon (EC)–rich particle types at 27 m was higher than the contribution at 280 m by a factor of approximately 2. The findings suggest that people living or spending time near major roadways could be exposed to elevated number concentrations of nucleation–mode volatile particles (<30 nm), Aitken–mode non–volatile particles (30–100 nm), and EC–rich fine–mode particles (>100 nm). The impact of the highway emissions on air quality was observable up to 300 m.

Keywords: *Near–road, non–volatile particles, traffic–related air pollutant, spatial variation, single particle analysis*

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1. Introduction

Traffic–related atmospheric pollutants (TRAPs), including ultrafine particles (UFP, <100 nm in diameter), black carbon (BC), polycyclic aromatic hydrocarbons (PAHs), and volatile organic compounds (VOCs), are believed to adversely impact the health of populations living and working near roadways. Proximity to major roadways and exposure to TRAPs may be associated with a number of respiratory and cardiovascular issues including asthma, reduced lung function, adverse birth outcomes, cardiac effects, respiratory symptoms, premature mortality, and lung cancer (e.g., HEI, 2010). Most recently, diesel exhaust was officially classified as carcinogenic by the International Agency for Research on Cancer (IARC) of the World Health Organization (Straif et al., 2013). Gauderman et al. (2007) found that children who lived within 500 m of a freeway had substantial deficits in lung function, when compared to children who lived 1 500 m or more from a freeway. A cohort study in Toronto, Canada suggested that traffic proximity (residence within 50 m of a major road or 100 m of a highway) may be associated with an increase in circulatory mortality of over 40% (Jerrett et al., 2009). In Canada approximately 4 million and 10 million people live within 100 m and 250 m of a major road, respectively, based on 2006 Census data (Evans et al., 2011). Thus a high proportion of Canada's urban populations are potentially exposed to TRAPs. For example, in Toronto, the most populated city in Canada, ~13% and ~56% of the population lives within 50 m and 250 m of a major road, respectively.

Assessment of exposure to TRAPs can be challenging due to the intra–urban spatial variability of pollutants, such as UFP and BC. Near–road field studies have shown a large difference in the concentrations of traffic–related pollutants between near–road measurements and typical air quality monitoring stations (e.g., Riediker et al., 2003). A number of on–road measurements have shown that significantly higher concentrations of traffic–related pollutants can occur on roadways as compared to background levels measured 150–500 m away (Hitchins et al., 2000; Bukowiecki et al., 2002; Riediker et al., 2003; Kittelson et al., 2004; Pirjola et al., 2004; Kaur et al., 2005; Weimer et al., 2009; Hagler et al., 2010; Pirjola et al., 2012). Roadside field studies have also consistently found decreasing concentrations of UFP and NO_2 with increasing distance from a road, while there were inconsistent trends in PM_{10} and $PM_{2.5}$ (Roorda–Knape et al., 1998; Zhu et al., 2002a; Zhu et al., 2002b; Westerdahl et al., 2005; Beckerman et al., 2008; Hagler et al., 2009; Durant et al., 2010; Gordon et al., 2012; Salimi et al., 2013).

Additionally, there is a growing interest in understanding the spatial variability of particle chemical properties near busy roadways. In particular, particle volatility is an important property of UFP in terms of the potential health effects of particles as well as the spatial variation of particles near major roadways. Traffic–related UFP generally contain compounds that range in volatility, such as organics from unburned fuel, lubricating oil, and graphitic carbon (e.g., Kittelson, 1998; Sakurai et al., 2003). To characterize the volatility of traffic–related particles, different types of tandem differential mobility analyzers (TDMA) have been deployed near

major roadways (Wehner et al., 2004; Biswas et al., 2007; Tiitta et al., 2010). The TDMA system provides detailed information about the volatile fraction of an individual particle diameter, but no information about the volatile/non–volatile fractions of the entire size distribution. Little work has been done to differentiate between the entire volatile and non–volatile fraction of UFP near major roadways (Wehner et al., 2004; Pirjola et al., 2012). The spatial variation of the particle volatility in the entire size distribution can provide useful information about exposure to non–volatile particles as a function of distance from a roadway.

In this paper, the evolution of volatile and non–volatile UFP in a near–road environment was examined using a thermodenuder (TD) and a fast mobility particle sizing spectrometer (FMPS) at distances up to 280 m from a major highway. In addition, single particle analysis using an aerosol time–of–flight mass spectrometer (ATOFMS) provided insights into the size and composition of the accumulation mode particles (100–3 000 nm). The measurements of aerosol volatility and chemical composition provide an improved understanding of the evolution of traffic–related particles in the vicinity of major roadways. This information is potentially useful for understanding the associated exposure and health impacts.

2. Methods

2.1. Measurement design

The study was performed in the Greater Toronto Area, Ontario, Canada from August 17 to September 17, 2010 as a part of the Fast Evolution of Vehicle Emissions near Roadways (FEVER) campaign. The University of Toronto mobile laboratory (MAPLE) was deployed to measure the decay gradients of TRAPs near highway 400. This major highway has 6 lanes that, when combined, carry ~90 000 vehicles/day (Liggio et al., 2012). The mobile lab, MAPLE, was moved back and forth among sites 27 m (Site A) and 280 m (Site B) downwind of the highway, as well as at 90 m (Site C) upwind of the highway (Figure 1). During these mobile measurements, traffic–related pollutants were sampled at each site for at least 1 hour before MAPLE moved to the next site. Only those morning and evening rush–hour periods on August 19, 26, and 27 during which the wind was perpendicular to the highway (200–330) were chosen for UFP gradient analysis. Information on the mobile measurement days and times is listed in Table 1.

In addition to the mobile measurements, UFP was continuously measured at a stationary monitoring site (Site D) located 15 m from the highway. The site was surrounded by agricultural areas with wide open level terrain and there were no nearby significant point sources of ultrafine particles or other main roadways. In order to explore the influence of the highway traffic on particle number concentrations, the relationship between UFP concentration and wind direction was depicted by using a wind rose plot generated with 1 minute averaged data at Site D (see the Supporting Material, SM, Figure S1). The results show that higher number concentrations (>40 000 cm^{-3}) occurred when the wind was coming from the highway, further confirming that the highway was the only significant source of UFP present.

In order to evaluate the micro–scale gradient of UFP within 20 m, UFP number concentrations were simultaneously measured at 3 m (near the shoulder of the highway) and 18 m downwind of the highway on September 1. The FMPS was connected to a long piece of conductive tubing (~15 m) for sampling at the 3 m location, whereas a short piece (~30 cm) was used for the second FMPS at the 18 m sampling. The long sampling inlet was moved back to the 18 m location every hour to calculate the diffusional losses through the long sampling line. The wind speed and wind direction were relatively constant over the 8 h measurement period. To investigate the spatial transformation of individual plumes from heavy emitters, 37 high particle number events were selected by inspecting particle number spikes (>40 000 cm^{-3} at the 3 m location) and video observations.

From September 2 to September 17, MAPLE was parked at Site A or B to characterize individual ambient particles. To ensure that vehicle emissions were sampled, the analysis was limited to single particle mass spectral data measured during morning rush hours (5:30 am to 11:00 am) on the days (September 14 and 15) with winds between the southwest (200°) and the northwest (330°). It should be noted that data from low wind speed (<0.5 m s^{-1}) conditions were also excluded from the analysis.

Table 1 summarizes the weather conditions and traffic density on these sampling days. While wind direction was relatively constant for these selected days, average temperature and wind speed varied from 12 to 31 °C and from 2 to 8 m s^{-1}, respectively. The number density of light duty vehicles (i.e., cars) was relatively constant, although on August 27 (Friday) the traffic density was higher than on other weekdays. Heavy–duty diesel vehicles (i.e., heavy class) accounted for approximately 4% of the total vehicles on August 26 and 27. This traffic pattern has been described in detail elsewhere (Liggio et al., 2012).

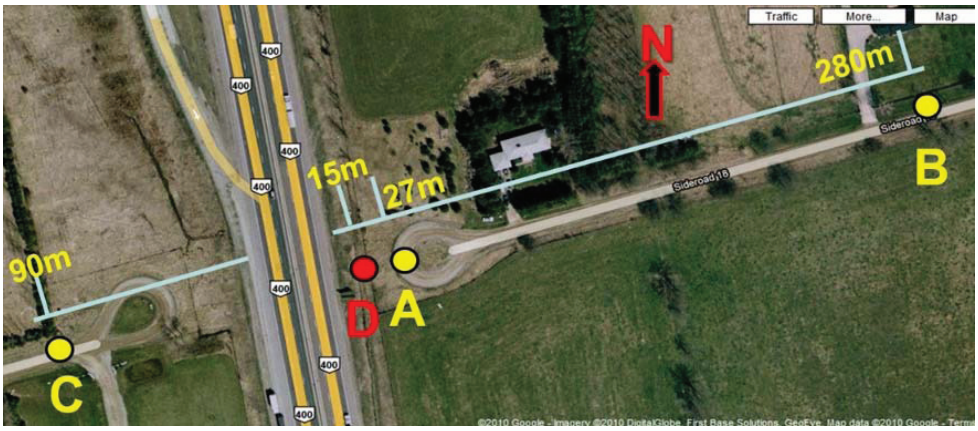

Figure 1. *Location of the measurement sites near Highway 400, Ontario, Canada: Site A (27 m from the highway), Site B (280 m from the highway), Site C (upwind 90 m from the highway), and Site D (stationary site 15 m from the highway) (created in Google Maps, maps.google.com).*