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Measured and modeled variation in pollutant concentration near roadways

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A B S T R A C T

This paper presents a study of the evolution of particles and gases downwind of a highway, with a focus on the diurnal variation of pollutant gradients and its controlling variables. A mobile laboratory was used to measure the concentration gradients of ultra-fine particles (UFP), black carbon (BC), CO_2, NO, and NO_2 at varying distances up to 850 m from a major highway. The horizontal distributions of pollutants show a strong diurnal pattern. Results suggest that the horizontal gradients are predominantly influenced by traffic levels, friction velocity, and atmospheric stability. The results were compared to a dispersion model, which showed good agreement with the measurements and was able to qualitatively capture the observed diurnal cycles. Emission rates [g km^{-1}] calculated from the model fits are within 10% of the Mobile 6.2C inventory for CO_2 and demonstrate good agreement for NO_x, but are higher than the inventory by a factor between 2.0 and 5.9 for black carbon. Hourly NO_x emission rates correlate with the fraction of heavy-duty vehicles in the total fleet and agree with inventory values based on maximum vehicle emission rates.

1. Introduction

Many recent studies have linked traffic proximity to the increased risk of adverse health effects. These adverse health effects include reduced lung function (Brunekreef et al., 1997), adverse respiratory symptoms (van Vliet et al., 1997; Venn et al., 2001), asthma (Lin et al., 2002), heart failure (Brook et al., 2002), cancer and leukemia in children (Pearson et al., 2000), and mortality (Hoek et al., 2002). In a review of 8 studies, Brugge et al. (2007) found that the most important exposure zone for elevated cardiopulmonary health risks is within 30 m of a roadway, while van Vliet et al. (1997) and Venn et al. (2001) found the greatest risk of adverse respiratory symptoms is to those living within 300 m of a freeway. This affects a significant portion of the population. For example, 45% of the population in Toronto live within 500 m of an expressway or within 100 m of a major road (Health Effects Institute, 2010) and approximately 16% of American households are within 100 m of a highway having 4 or more lanes (US Census Bureau, 2008).

In a review of 41 road-side studies, Karner et al. (2010) found that pollutants, including elemental carbon, ultra-fine particles (UFP), NO_x, and CO, are reduced to background levels between 115 and 570 m from roads. In a review of 33 studies, Zhou and Levy (2007) found the distance to reach background concentrations ranged between 100 and 500 m. In the Karner et al. (2010) review, it was found that the relative decay of different pollutants was dependent on the method of normalization. If the concentrations were normalized by the background concentration (C_{bg}), CO, and UFP (between 3 and 100 nm) demonstrated the largest change in concentration with distance downwind, characterized by a >50% drop in concentration within 150 m. Elemental carbon (EC), NO, NO_2, NO_x, and O_3 demonstrated a less rapid decay with distance from the road. If the concentrations are normalized by the road-edge concentration (C_E) the rapid decay group includes CO, UFP, EC, NO and NO_x, while NO_2 demonstrates a less rapid decay. Karner et al. attributed the difference between normalization methods to under-predicted background levels. In general, these results are consistent with the findings of Zhou and Levy (2007), who reviewed 33 studies and concluded that inert pollutants with high background concentrations (CO, EC) have the largest spatial extent, while pollutants formed in near-source chemical reactions (NO_2) have a larger spatial extent than those depleted in near-source chemical reactions (NO) or removed through coagulation processes (UFP). Particle evaporation has also been shown to result in significant UFP removal (Kuhn et al., 2005).

By far the bulk of these studies were done during daytime and evening periods when traffic levels are highest, with few studies looking at the changes in the extent of pollutants throughout the

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day and night at the same location. Zhu et al. (2006) measured the variations of UFP concentration near a highway during several nights in February, 2005, which was compared to measurements made at the same location during seven days in May, June, and July, 2001 (Zhu et al., 2002). They found that the particle number concentration relative to the traffic flow was more than three times higher at night in the winter. Hu et al. (2009) measured UFP and NO concentrations near a highway during five mornings for 1–2 h before sunrise. In winter, they found that concentrations were above background levels up to approximately 2600 m downwind and 600 m upwind of the road, while in summer the extent was less. The high concentrations and large extent were associated with low wind speeds, high relative humidity, and the nocturnal surface temperature inversion. Durant et al. (2010) measured a number of pollutants near a highway, including UFP, NO_x, and CO_2, for one morning between 6:00 and 11:00. They found that the horizontal distributions of pollutant concentration were related to traffic flow, wind speed, and surface boundary-layer height, with concentration gradients decreasing with increasing wind speed and boundary-layer height. Janhäll et al. (2006) measured UFP, NO_x, and CO at a fixed location near a highway and found a similar association with pollutant concentrations and boundary-layer height.

Since traffic flow, wind speeds, boundary-layer heights, and atmospheric stability vary throughout the day, horizontally distributed concentration measurements made throughout the day for multiple days could determine the relative influence of different factors that influence the variation in the horizontal concentration gradients. Karner et al. (2010), citing the studies listed above, recommended that future work integrate nighttime and daytime measurements, due to a lack of nighttime studies. The present study attempts to accomplish this by measuring pollutant gradients before and after sunrise, as well as during afternoon/early-evening rush hours. A mobile laboratory (the Canadian Regional and Urban Investigation System for Environmental Research, CRUISER) was used to measure the change in concentration of various gases and particles with distance from the highway. To investigate the causes of diurnal variation in the pollutant gradients, and to identify controlling variables, the measurement results are compared to the results of a physically-based dispersion model (Venkatram, 2004; Venkatram et al., 2007), which is modified to include the effects of atmospheric stability. This study is the first test of this model using individual transect measurements throughout the day, as opposed to stationary point measurements (Venkatram et al., 2007). The model results also provide the opportunity to compare average traffic emission rates to emission inventories to evaluate the inventory values using realistic, in-situ measurements of a mixed traffic fleet.

2. Field study

The Fast Evolution of Vehicle Emissions from Roadways (FEVER, Gordon et al., in press; Liggio et al., 2012) project took place between Aug 16 and Sept 17, 2010. Measurements were made on a side road perpendicular to Hwy 400 (43.994 N, 79.583 W) north of Toronto, Ontario, Canada with a mobile lab (CRUISER). The mobile lab was used at this site to drive on the perpendicular side roads of the highway for approximately 1 km on each side. The highway at this location is 6 lanes wide (25 m across from the lane edges), with a 1 m high barrier at the centre, and drainage ditches (ca. 2 m deep) on either side. The side road ended in a turn-around near the highway, allowing measurements as close as 50 m from the highway centre (37 m from the lane edges). Highway traffic at this location was primarily due to commuters heading south into Toronto in the morning and returning in the evening, while on Fridays and weekends, the road is heavily traveled by those heading to cottage areas north of Toronto. Vegetation in the surrounding area is predominantly agricultural, with some trees lining the side roads (Gordon et al., in press). Mean daily temperatures during the study ranged from 12 °C to 27 °C, with an average of 18.4 °C.

A sonic anemometer (CSAT3) was mounted on a 3-m tower installed 9.5 m east of the Hwy edge (22 m from the Hwy centre). The anemometer measured mean wind speed (U_r) and direction (θ), friction velocity (u_*), and heat flux (H). Wind speeds were corrected for anemometer tilt by rotating the 30-min fluxes to align the horizontal wind speed (u) with the mean wind direction following Wilczak et al. (2001).A radiometer (LI-200, LI-COR) mounted at 3-m on the tower measured incoming short-wave radiation (S_{Rad}). A nearby traffic camera (Miovision) was used to determine traffic counts per minute (to give traffic flow, F) with counts classified as northbound or southbound passenger cars, medium sized, or heavy-duty vehicles, with >95% accuracy (Gordon et al., in press). Air-quality monitoring systems (Airpointer, Recordum) were installed at three fixed locations: 110 m west of the Hwy centre (Site A); 34 m east of the Hwy centre (Site B); and 300 m east of the Hwy centre (Site C). The monitoring systems measured 1-min averages of NO, NO_2, and NO_x concentrations, as well as wind speed (U_r) and direction (θ).

The mobile lab housed instrumentation to measure total particle concentration (UFP) for sizes 4.5 nm to >3 μm (CPC 5.4, GRIMM Aerosol Technik), CO_2 (LI-6200, LI-COR), NO_x (TECO 43C, Thermo Electron Co.) and black carbon (BC) mass (SP2, Droplet Measurement Technology). In addition to these instruments, the mobile lab was outfitted with high-frequency Global Positioning Sensors (GPS), inertial motion sensors, and 3D sonic anemometers (CSAT3, Campbell Scientific).

Driving transects were typically done for a few hours at a time, spanning peak traffic during the morning and/or the evening. More than 44 h of driving was done at this site over 17 days, comprising 8 morning sessions and 14 evening sessions. Data were filtered for winds within 45° of the highway normal, as measured by the monitoring system at Site B (34 m from the highway centre). Individual transects were defined as either an approach, driving into the wind towards the highway, or a return, driving with the wind away from the highway. The mobile lab was typically driven near 2 m s^{-1} during an approach (into the wind) and 6 m s^{-1} during a return (with the wind) in order to avoid sampling the exhaust of the mobile lab. The sonic anemometer on the mobile lab measured wind speed parallel to the vehicle (u) at a rate of 20 Hz. Any sampled data concurrent with $u < 0.2$ m s^{-1} (within each 1 s period) were removed from the analysis to avoid sampling the exhaust of the mobile lab during back-drafts. Interference in the transect measurements was observed due to occasional passing cars on the side road. The observation times of passing cars were recorded manually, and the concurrent spikes in the data were removed by visual inspection of the measurements, resulting in a removal of less than 5% of the data.

The mobile lab anemometer was used to calculate mean wind speed (U_r), friction velocity (u_*), and heat flux (H) for each transect. The mobile lab velocity was calculated from the high-frequency GPS measurements and was subtracted from the wind speeds measured by the anemometer. As with the road-side tower, wind speeds were corrected for anemometer tilt by rotating the 30-min fluxes to align the mean horizontal wind speed (u) with the mean wind direction following Wilczak et al. (2001). To determine the effect of vehicle vibration on the measurements, inertial motion sensors recorded the 3 components of acceleration at a rate of 40 Hz. Since horizontal vehicle motion was recorded by GPS (at 5 Hz), we are only concerned here with acceleration caused by vertical vibration (a_z). The vertical acceleration was integrated to give vertical velocity due to vehicle motion, $w_{z,a}$. During transect