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Factors influencing the outdoor concentration of carbonaceous aerosols at urban schools in Brisbane, Australia: Implications for children's exposure

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ABSTRACT

This comprehensive study aimed to determine the sources and driving factors of organic carbon (OC) and elemental carbon (EC) concentrations in ambient $PM_{2.5}$ in urban schools. Sampling was conducted outdoors at 25 schools in the Brisbane Metropolitan Area, Australia. Concentrations of primary and secondary OC were quantified using the EC tracer method, with secondary OC accounting for an average of 60%. Principal component analysis distinguished the contributing sources above the background and identified groups of schools with differing levels of primary and secondary carbonaceous aerosols. Overall, the results showed that vehicle emissions, local weather conditions and secondary organic aerosols (SOA) were the key factors influencing concentrations of carbonaceous component of $PM_{2.5}$ at these schools. These results provide insights into children's exposure to vehicle emissions and SOA at such urban schools.

1. Introduction

There is a growing body of evidence suggesting that children who are exposed to vehicle emissions experience a number of detrimental health effects. These include increases in wheezing for infants (Ryan et al., 2009), worsening of existing asthma (Trenga et al., 2006), and the development of asthma in children (Gehring et al., 2010). Vehicle emissions represent a significant proportion of urban airborne particles and are mainly carbonaceous in nature (Ancelet et al., 2011; Kleeman et al., 2000). The carbonaceous aerosol can be defined as either elemental carbon (EC) fractions (EC1, EC2, EC3) or organic carbon (OC) fractions (OC1, OC2, OC3, OC4 and pyrolized carbon (PC)) and the definition depends on the analysis method (Chow et al., 2001).

EC is soot like and originates almost exclusively from primary sources such as vehicle emissions. OC is a complex mixture of many organic compounds and the result of both primary particle emissions and secondary reactions of volatile organic compounds in the atmosphere (Cao et al., 2006). Therefore the ratio of OC to EC can indicate whether the source of the carbonaceous aerosols is primary or secondary, with vehicle emissions typically having a ratio of around 1 to 2 (Keywood et al., 2011 and references therein). Additional insight can be gained into the source of the carbonaceous aerosol by the analysis of the OC and EC fractions (Cao et al., 2006). The EC fraction can also be defined as char-EC (EC1-PC) and soot-EC (EC2+EC3) to differentiate between the sources of EC where char-EC and soot-EC are similar to wood charred materials and diesel exhaust particles, respectively (Han et al., 2007).

A school environment is a site that can account for a significant portion of children's exposure to vehicle emissions. However there is only limited literature on children's exposure at schools (Mejía et al., 2011). Schools within a city are expected to have different levels of pollutants that result from local urban characteristics. Recent studies have shown that school bus emissions can have a significant impact on the air quality in schools (Hochstetler et al., 2011; Li et al., 2009; Richmond-Bryant et al., 2011). However, these studies are less relevant to schools that are not serviced by school buses, and as such other factors may be determining children's exposure to vehicle emissions in the schools.

The current study aims to determine children's exposure to airborne particles by characterising the carbonaceous aerosols, defined as carbonaceous component of particles with diameters less than 2.5 μm ($PM_{2.5}$) at a number of urban schools. The

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concentrations of primary OC (POC) and secondary OC (SOC) at each school were used to ascertain the primary (e.g. vehicle emissions) and secondary sources contributing to the levels of carbonaceous aerosols. Principal Component Analysis (PCA) was applied to the measured levels of OC, EC and their fractions to reveal the schools that were most affected by primary and secondary sources.

2. Method

2.1. Sampling sites

This study was part of a larger project designed to study the effect of Ultrafine Particle from Traffic Emissions on Children's Health, known as UPTECH (https://www.qut.edu.au/research/research-projects/uptech). The randomly selected 25 schools in this study, which will be referred to as S01 to S25, are in different suburbs in the Brisbane Metropolitan area, Australia. The measurement portion of the study was conducted from October 2010 until August 2012. The selected schools were not near any infrastructure projects or any other major source of air pollution, other than local road traffic. At each school, a centrally located outdoor monitoring site within the school grounds that was assumed to give the best overall exposure was chosen to conduct the measurements and sampling at each school. Schematic maps showing the location of the sampling site (referred to as Site B by Salimi et al.) and surrounding roads at the schools can be found in Salimi et al. (2013). The inlets for the sampling were approximately 3 m off the ground and placed on the top of a trailer, which served to house all of the instruments at the site, including an automatic weather station (Monitor Sensors). In order to account for the impact of schools layout and buildings on the collected meteorological data at the monitoring site, they were augumented by the data from nearby weather stations from the Australian Bureau of Meteorology and the Queensland Department of Science, Information Technology, Innovation and the Arts. Traffic counts with 5 min intervals were taken on the busiest road next to the school, referred to as the main road throughout. The measured traffic counts comprised of the total traffic count with passenger cars, motorbikes and scooters classified as light vehicles. Light trucks with 2,3 and 4 axels were classified as medium vehicles while long articulated trucks were classified as heavy vehicles.

2.2. Sampling and analysis methodology

The $PM_{2.5}$ were sampled for EC and OC analyses. This was achieved using a cascade impactor Dekati (Kangasala, Finland) to remove particles larger than 2.5 μm. A critical orifice and needle valve was used to maintain the 20 L per minute flow rate required by the impactor. The particles were collected on a preloaded 37 mm quartz cassette filter (SKC). At each school, the filter sampling was conducted over a nine - hour period from 08:00 to 17:00, each day for one week with a total of five samples collected per school. The sampling days at each school were typical school days with no special activities that may affect the results. The selected sampling duration captured the times that students spent at the school and will be referred to as school hours in the rest of the paper, including the official schools teaching hours as well as the pre- and after-school care hours. The typical sampling volume for the filter over the 9 h period was 9.7 m^3. Once the sampling was completed the filters were wrapped in aluminium foil, placed in a ziplock bag and kept frozen until analysis. The analysis of OC and EC components of $PM_{2.5}$ on the collected filters was performed at Chester Labnet, Oregon, USA using the IMPROVE method on a thermal/optical transmittance carbon analyser (Sunset Laboratories), which included the OC and EC fractions as defined by the protocol (Chow et al., 2001).

2.3. Quality control

The sampling flow rate of the impactor was checked at the beginning of each filter sampling and was always set to within ±0.1 L per minute of the target flow rate. Field blanks were taken at each school. The EC and OC results were all field-blank corrected with an average of 7.4 ± 1.9 μg/filter for OC and 0 ± 1.6 μg/filter for EC. The minimum detection limit (MDL) for both OC and EC was determined from the standard deviations of instrument blanks to be 1.6 μg/filter.

2.4. Data analysis

To determine the levels of secondary and primary OC present at the schools, the EC tracer method was used (Castro et al., 1999) to establish a minimum OC/EC ratio (OC/EC_{min}) that was representative of the main primary source. The EC tracer method assumes that there is one primary source and has been shown to give accurate estimation of SOC levels, provided that the correct OC/EC_{min} is used (Zhang et al., 2005). Due to the school selection criteria employed in this study, vehicle emissions are expected to be the main primary source of air pollution, including carbonaceous aerosols, at the schools and this justifies the use of the EC tracer method. The OC and EC concentrations are plotted to determine the OC/EC_{min}, with points above this line taken to contain additional OC or SOC (Harrison and Yin, 2008). Samples affected by significant rainfall were removed from the analysis as rain would remove aged aerosols and so bias the analysis (Keywood et al., 2011). Two of the 119 samples were clear outliers with unusually low OC values and they were removed before the analysis.

PCA was performed using SIMCA-P (Version 10) and the measured variables: OC, EC, OC and EC fractions, soot-EC, char-EC, OC/EC and char-EC/soot-EC ratios, traffic and meteorological parameters. For the PCA, heavier duty vehicle (HDV) counts were defined as the sum of medium and heavy vehicle counts. These parameters were included to explore the relationship of the OC and EC components with the traffic and meteorological variables at the schools. ANOVA and correlations analysis was performed using SPSS v19. Rose plots of wind speed and direction were performed in IGOR Pro v6.22A.

3. Results and discussion

3.1. Meteorological conditions

The measured average temperature, relative humidity (RH), solar radiation and wind speed during the sampling period (i.e. school hours over the five days of sampling) school are given in Table S1 (Supporting Information). The wind direction data was determined with wind rose plots, given in the Supporting Information Fig. S1, and summarised in Table S1. Brisbane is a subtropical city and consequently the average temperature during these measurements varied from 16.9 to 29 °C. The cooler, drier and more stable weather occurs during May to October and is referred to as winter while the warmer months, November to April, are more humid with higher rainfall and are referred to as summer. These seasonal characteristics are observed throughout the project, as shown in Table S1.

3.2. Traffic characteristics of the schools

The schools selected were in a variety of urban locations.

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