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Sources of indoor and outdoor PM2.5 concentrations in primary schools

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HIGHLIGHTS

- We collected PM2.5 samples at indoor and outdoor environments of 39 primary schools.
- Seven outdoor sources and two children-activity-related sources were identified.
- In classrooms, 47% of PM2.5 was indoor generated, mostly organics.
- Unpaved playgrounds increased PM concentrations in classrooms by 5–6 μg/m^3.
- Traffic contributions were higher at classrooms with windows oriented directly to the street.

GRAPHICAL ABSTRACT

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ABSTRACT

Children spend a third of their day in the classroom, where air pollution levels may differ substantially from those outdoors due to specific indoor sources. Air pollution exposure assessments based on atmospheric particle mass measured outdoors may therefore have little to do with the daily PM dose received by school children. This study aims to investigate outdoor and indoor sources of PM2.5 measured at 39 primary schools in Barcelona during 2012. On average 47% of indoor PM2.5 measured concentrations was found to be generated indoors due to continuous resuspension of soil particles (13%) and a mixed source (34%) comprising organic (skin flakes, clothes fibers, possible condensation of VOCs) and Ca-rich particles (from chalk and building deterioration). Emissions from seven outdoor sources penetrated easily indoors being responsible for the remaining 53% of measured PM2.5 indoors. Unpaved playgrounds were found to increase mineral contributions in classrooms by 5–6 μg/m^3 on average with respect to schools with paved playgrounds. Weekday traffic contributions varied considerably across Barcelona within ranges of 1–14 μg/m^3 outdoor and 1–10 μg/m^3 indoor. Indoors, traffic contributions were significantly higher (more than twofold) for classrooms with windows oriented directly to the street, rather than to the interior of the block or to playgrounds. This highlights the importance of urban planning in order to reduce children's exposure to traffic emissions.

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1. Introduction

Recently the European Environmental Agency (EEA, 2013) reported that 20–31% of the urban population in the EU region is exposed to PM2.5 concentrations (2009–2011) above the EU reference level (25 μg/m^3) and 91–96% above the WHO PM2.5 guideline (10 μg/m^3). Long-term studies have already shown associations between PM concentrations and population mortality also at levels well below the WHO guideline level for PM2.5, without evidence of a threshold of exposure to PM below which no adverse health effects would be anticipated (WHO, 2006a,b, 2013).

Routine pollutants measurements, which are commonly used in epidemiological studies, may not provide a good characterization of personal exposure (Nerriere et al., 2005; Kornartit et al., 2010). Exposure studies should take into account that adults spend approximately 60–80% of their time indoors (Klepeis et al., 2001), and in the case of children at least a third of their day is spent in the classroom, where PM levels may vary substantially from outdoors due to building insulation, indoor sources and resuspension. In fact, indoor-generated PM may represent a significant fraction of total exposure. Health effects are expected to be much more marked in children, who inhale a higher normalized dose of airborne particles compared to adults, due to both their lung capacity and higher breathing rates for physical activity (Burtscher and Schüepp, 2012; Bateson and Schwartz, 2010; Pinkerton and Joad, 2006; Buonanno et al., 2011, 2012). PM exposures are linked to cognitive deficits, oxidative stress, neuroinflammation and neurodegeneration (Calderón-Garcidueñas et al., 2013). Several individual metals including aluminum, arsenic, cadmium, lead, manganese, and mercury have been demonstrated to affect the neurological system (Pohl et al., 2011), and general accumulation of metal ions in the brain contributes to heightened oxidative stress and neuronal damage (Zatta et al., 2008; Bolognin et al., 2009).

Investigating the levels and sources of atmospheric particulate matter (PM) pollution in primary schools and understanding their role in child health is therefore one of the challenges of our time. Only few studies reported inorganic chemical analysis of PM2.5 indoors and generally are based on few elements and the samples are collected on a few schools (Molnár et al., 2007; Zwoździak et al., 2013; Stranger et al., 2008). To our knowledge there are no studies of source apportionment at indoor and outdoor environment at schools. The BREATHE project (BRain dEvelopment and Air polluTion ultrafine particles in scHool childrEn), funded by the European Research Council (ERC Advanced Grant), aims at studying the impact of urban air pollution on the cognitive development of children. The project involves 2904 children attending second, third and fourth grades at 39 primary schools in Barcelona (NE of Spain) which are characterized by contrasting air pollution levels.

In the framework of the BREATHE project, this study aimed to identify major sources of PM2.5 and their contributions based on PM2.5 source apportionment analysis performed indoor (in classrooms with pupils), outdoor (in the playground) and at a reference background station in order to enable source-related studies on children health outcomes. This will supply the source contributions data needed for further epidemiological studies within BREATHE.

2. Methods

PM2.5 filter sampling was carried out over one year (February 2012–February 2013) at 39 primary schools in the Barcelona metropolitan area (identified with numbers from 1 to 40 in Fig. 1, Rivas et al., in press). Thirty-six schools were located in the city of Barcelona (1.6 million inhab.) and 3 in the municipality of Sant Cugat (89,000 inhab.), only 7 km away but separated from Barcelona by the Collserola range (512 m. a.s.l.). The sampling schedule was interrupted during summer school holidays: July and August 2012. The sampling instruments were installed simultaneously at two schools per week, two times during the school year. The indoor and outdoor samplers were always installed in a classroom and in the playground respectively. The orientation (courtyard- or street-facing) and altitude (street level to 4th or 5th storey) of both classroom and playground can also vary. Possible effects on source contributions due to these factors will be explored in this study. In addition 24 h PM2.5 samples were available from a fixed monitoring station (Palau Reial, the only monitoring site of the local network where PM2.5 samples are routinely analyzed) as a reference urban background site, collecting every 4th day during the whole study. Samples were collected onto Pallflex quartz fibre filters (15 cm diameter) by means of high volume (30 m^3/h) MCV samplers four times a week (Monday to Thursday schooldays, plus 10 holiday samples) for 8 h of each day (from 9 am to 5 pm, being children at classroom for about 80% of the time). Before sampling, filters were pre-baked at 205 °C during 5 h and conditioned for 48 h at 20 °C and 50% relative humidity. Weights of blank filters were measured three times every 24 h by means of a Sartorius LA 130 S-F microbalance (1 μg sensitivity). After sampling, filters were brought back to the laboratory to be weighed three more times every 24 h under the same temperature and relative humidity conditions of the first weighing. Once the weights of samples had been determined, filters were subjected to several analytical treatments. A quarter of each filter was acid digested (5 ml HF, 2.5 ml HNO_3, 2.5 ml $HClO_4$) for the determination of major and trace elements and analyzed by inductively coupled plasma mass spectrometry and atomic emission spectrometry, respectively (ICP-MS and ICP-AES) (Querol et al., 2001). A quarter of the filter was leached in 20 ml of bi-distilled water for the extraction of water-soluble ions and subsequent analysis by ion chromatography (IC) for sulfate, nitrate and chloride and by specific electrode for ammonium. Finally, a section of 1.5 cm^2 from the remaining half of the filter was used for the determination of organic carbon (OC) and elemental carbon (EC) by a thermal-optical transmission technique (Birch and Cary, 1996) using a Sunset Laboratory OCEC Analyzer with the NIOSH temperature program. Total carbon (TC) was determined as the sum of OC and EC. In every case blank concentrations were subtracted for determining final concentrations in samples.

A constrained Positive Matrix Factorization (PMF) model was used for source apportionment. PMF is a weighted least-squares method so that individual estimates of the uncertainty in each data value are needed. The uncertainty estimates were based on the approach by Amato et al. (2009) and provided a criterion to separate the species which retain a significant signal from the ones dominated by noise. This criterion is based on the signal-to-noise S/N ratio defined by Paatero and Hopke (2003). Species with S/N < 2 are generally defined as weak variables and downweighted by a factor of 3. Nevertheless, since S/N is very sensitive to sporadic values much higher than the level of noise, the percentage of data above detection limit was used as complementary criterion. The combination of both criteria permitted the selection of 31 strong species and 2 weak species (Ba and Sn) out of the 61 available species for the PMF analysis. After preliminary tests where PMF was performed separately for indoor and outdoor samples, the best solution was found when gathering indoor and outdoor PM samples from all schools into a single array. Also, the 24-h samples collected at the Palau Reial urban background reference station were included. This data assembling method showed the most satisfactory results for factor profiles since it allowed exploration of a larger area of the N-dimensional source contributions space. The data matrix was uncensored, i.e. negative, zero and below detection limit (BDL) values were included as such in the analyses to avoid a bias in the results (Paatero, 2007). A total of 683 samples were included in the PMF: 577 from schools (47 samples were lost due to power or instrument failures) and 106 from the Palau Reial station. PMF was run by means of the Multilinear Engine program which allowed the handling of *a priori* information such as the source profile of local road dust (average of city centre samples, as reported by Amato et al., 2009) and sea spray (SPECIATE database) in form of targets for pulling equations (Paatero and Hopke, 2009). A bootstrap technique was implemented in the script