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Vertical variation of $PM_{2.5}$ mass and chemical composition, particle size distribution, NO_2, and BTEX at a high rise building☆

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ABSTRACT

Substantial efforts have been made in recent years to investigate the horizontal variability of air pollutants at regional and urban scales and epidemiological studies have taken advantage of resulting improvements in exposure assessment. On the contrary, only a few studies have investigated the vertical variability and their results are not consistent. In this study, a field experiment has been conducted to evaluate the variation of concentrations of different particle metrics and gaseous pollutants on the basis of floor height at a high rise building. Two 15-day monitoring campaigns were conducted in the urban area of Bologna, Northern Italy, one of the most polluted areas in Europe. Measurements sites were operated simultaneously at 2, 15, 26, 44 and 65 m a.g.l. Several particulate matter metrics including $PM_{2.5}$ mass and chemical composition, particle number concentration and size distribution were measured. Time integrated measurement of NO_2 and BTEX were also included in the monitoring campaigns. Measurements showed relevant vertical gradients for most traffic related pollutants. A monotonic gradient of $PM_{2.5}$ was found with ground-to-top differences of 4% during the warm period and 11% during the cold period. Larger gradients were found for UFP (~30% during both seasons) with a substantial loss of particles from ground to top in the sub-50 nm size range. The largest drops in concentrations for chemical components were found for Elemental Carbon (−27%), iron (−11%) and tin (−36%) during winter. The ground-to-top decline of concentrations for NO_2 and benzene during winter was equal to 74% and 35%, respectively. In conclusion, our findings emphasize the need to include vertical variations of urban air pollutants when evaluating population exposure and associated health effects, especially in relation to some traffic related pollutants and particle metrics.

1. Introduction

Throughout the last 30 years, an increasing body of research has consistently shown statistically significant positive associations between ambient $PM_{2.5}$ and mortality and various indices of morbidity (REVIHAAP, 2013). However it is still not clear which physical and/or chemical characteristics of particulate matter (PM) are most closely linked to health effects (REVIHAAP, 2013; Brunekreef and Holgate, 2002; Harrison and Yin, 2000) as well as the mechanisms of action. Recently, research has focused on ultrafine particles (UFP, particles with aerodynamic diameter <100 nm), because of their ability to affect deep airways (Peters et al., 2011; Hoek et al., 2010; von Klot et al., 2005), and to the

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specific chemical components of particles (Samoli et al., 2016; Basagaña et al., 2015).

Moreover, while research is generally more focused on various metrics of PM, some gaseous pollutants such as nitrogen dioxide (NO_2), benzene, toluene, ethylbenzene, xylenes (BTEX) are not only responsible for relevant health effects (Andersen et al., 2011; Gauderman et al., 2005) but are also used as air quality indicators, and markers of traffic-related pollution (Spinazzè et al., 2013; Janssen et al., 2001).

Substantial efforts have recently been made to investigate the horizontal variability of air pollutants at regional and urban scales (Eeftens et al., 2012). In this field, significant improvements have been made in the estimation of ambient air pollutant concentrations within urban areas by a mix of fixed site monitoring station data with satellite data, dispersion models and Land Use Regression models (LUR) which combine measured air pollution concentrations together with predictor variables such as various traffic-related variables, land use, and population density (Hoek et al., 2008). These studies have shown a limited spatial variability for $PM_{2.5}$ mass while more substantial differences at a suburban scale were found with respect to UFP concentrations (van Nunen et al., 2017), some gaseous pollutants such as NO_2 and BTEX (Wang et al., 2013) and some traffic-related chemical components such as iron and elemental carbon (de Hoogh et al., 2013). Marked spatial variability of some pollutants was also observed at the micro-scale near highways (He and Dhaniyala, 2012) or crossroads (Goel and Kumar, 2016) and at buildings which fronted onto a trafficked street (Zauli Sajani et al., 2016).

Therefore, in the last decade the horizontal variability of air pollutants on a regional and urban scale has been deeply investigated and their findings recently included in the most important epidemiological studies (Beelen et al., 2014; Raaschou-Nielsen et al., 2013). On the contrary, only a few studies have assessed the vertical variability of air pollutant concentrations within urban areas.

During the 90s some studies investigated the vertical variation of gaseous pollutants within street canyons, i.e. streets flanked by buildings on both sides creating a canyon-like environment. Several studies were published on CO and NO_2 (Qin and Kot, 1993; Zoumakis, 1995; Vakeva et al., 1999) and showed a marked decrease of concentration with increasing height.

Studies assessing the vertical profile of different particle metrics such as $PM_{2.5}$ mass and chemical composition, particle number concentration (PNC) and size distribution are more recent. As with gaseous pollutants, the studies were most frequently focused on street canyon configurations (Kumar et al., 2008; Weber et al., 2006; Li et al., 2007). To our knowledge only very few papers have assessed the vertical variation of air pollutants on isolated buildings or in non-canyon configurations. Table 1 gives an overview of these studies for the air pollutants and particle metrics considered in this study. It is clear from the table that very few studies are available in literature on this topic and most of them are from Asian countries. With regards to particle metrics only one study was carried out in Europe but not within an urban area (Kohler et al., 2005; Imhof et al., 2005). To author's knowledge no studies are available on vertical profiles of metals and ions.

In this paper we present the results of an experimental study aimed at assessing the vertical variation of air pollution at an isolated tall building located in a highly polluted urban area in Northern Italy. $PM_{2.5}$ mass and chemical composition, particle size distribution, NO_2 and BTEX concentrations were simultaneously measured at different heights during the warm and the cold season.

2. Methods

2.1. Study design and study area

The study area is the city of Bologna, Italy. This is an urban area of about 400,000 inhabitants located in the Po plain, a region surrounded by mountains and affected by high levels of air pollution. In the period 2013–2016 the city-average $PM_{2.5}$ annual mean concentration (derived from two fixed-site monitoring stations) was 17.9 $\mu g\, m^{-3}$ (EU limit 25 $\mu g\ m^{-3}$and WHO guideline value 10 $\mu g\, m^{-3}$) and the annual mean number of exceedances of the WHO $PM_{2.5}$ daily guideline value (25 $\mu g\, m^{-3}$) was 72. All parameters monitored in the study have traffic as the main source or one of the major ones (https://webbook.arpae.it/shared/documenti/ARIA/Emissioni/Ar_Emissioni_Rer_2010.xls). For some pollutants traffic is a primary source (BTEX) or is a major contributor (NO_2), while for $PM_{2.5}$ the contribution of traffic emissions to secondary formation is more relevant (Larsen et al., 2012).

The monitoring site is a tall (~70 m high) building located at 2 km from the historical center very close to a trafficked street (daily traffic volume of about 41000 vehicles per day with 3–4% heavy duty vehicles). Fig. 1 shows an overview of the monitoring sites and area. The building has a very regular shape and instruments were placed on balconies which fronted the busy street. Instruments were placed at the ground floor (at 2 m above ground, from now on Level 0), at the 2nd floor (15 m above ground – Level 1), at the 5th floor (26 m above ground – Level 2), at the 10th floor (44 m above ground – Level 3), at the 16th floor (65 m above ground – Level 4). $PM_{2.5}$, NO_2 and BTEX were measured at each level while chemical composition was analyzed only at level 0, 2 and 4 due to funding contraints. The availability of two spectrometers allowed the monitoring of particle size distribution at the ground and at the 16th floor.

Two monitoring campaigns were conducted in the period February-June 2015. Each monitoring campaign lasted for 15 days: 1st campaign from 20 February to 6 March and 2nd campaign from 28 May to 11 June. Supplemental campaigns were performed from 15 to 26 February 2016 for BTEX and from 22 September to 9 October 2016 for particle size distribution due to instrumental problems in the first and second campaign, respectively.

All the above mentioned parameters were measured simultaneously at the different floors.

2.2. Instrumentation and monitoring procedure

$PM_{2.5}$ concentrations were measured by means of five identical gravimetric samplers (Skypost PM, TCR TECORA Instruments, Corsico, Milan, Italy) operating at a flow rate of 2.3 $m^3\, h^{-1}$. The instruments were placed on balconies at Level 0,1,2,3,4 (see Fig. 1). Samples were weighed following the procedure defined in European Standard EN 12341:2014. $PM_{2.5}$ samples were collected on quartz fiber filters (Whatman, 47 mm diameter) and analyzed for various chemical species. Filters were changed daily at each measurement site, and chemical speciation was performed sequentially every three days for metals, ions, and carbon (Elemental Carbon - EC - and Organic Carbon - OC). During the first two campaigns EC and OC were measured on an 8 h basis in order to avoid an overload of the filters (Ricciardelli et al., 2017). In this paper we present the data of the chemical species having more than 50% of contemporary data above the limit of quantification (LOQ). LOQs for chemical components as well as methods and procedure for chemical speciation are reported in the Supplementary Material (Table 1_S).

Two Fast Mobility Particle Sizers (FMPS model 3091; TSI, Shoreview, MN, USA) were used to measure particle size distributions and to estimate UltraFine Particle (UFP) concentrations. The