



Is particulate air pollution at the front door a good proxy of residential exposure?☆



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ABSTRACT

The most advanced epidemiological studies on health effects of air pollution assign exposure to individuals based on residential outdoor concentrations of air pollutants measured or estimated at the front-door. In order to assess to what extent this approach could cause misclassification, indoor measurements were carried out in unoccupied rooms at the front and back of a building which fronted onto a major urban road. Simultaneous measurements were also carried out at adjacent outdoor locations to the front and rear of the building. Two 15-day monitoring campaigns were conducted in the period June–December 2013 in a building located in the urban area of Bologna, Italy. Particulate matter metrics including PM_{2.5} mass and chemical composition, particle number concentration and size distribution were measured. Both outdoor and indoor concentrations at the front of the building substantially exceeded those at the rear. The highest front/back ratio was found for ultrafine particles with outdoor concentration at the front door 3.4 times higher than at the rear. A weak influence on front/back ratios was found for wind direction. Particle size distribution showed a substantial loss of particles within the sub-50 nm size range between the front and rear of the building and a further loss of this size range in the indoor data. The chemical speciation data showed relevant reductions for most constituents between the front and the rear, especially for traffic related elements such as Elemental Carbon, Iron, Manganese and Tin. The main conclusion of the study is that gradients in concentrations between the front and rear, both outside and inside the building, are relevant and comparable to those measured between buildings located in high and low traffic areas. These findings show high potential for misclassification in the epidemiological studies that assign exposure based on particle concentrations estimated or measured at subjects' home addresses.

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1. Introduction

Air pollutants, and airborne particles in particular, pose significant risks to human health (REVIHAAP, 2013). A body of evidence

has been accumulating over the last few decades on the effects of air pollution on cardiovascular and respiratory diseases, but there is still considerable uncertainty about the mechanisms of action linked to the health effects and about which physical and/or chemical characteristics of particulate matter (PM) are most important as determinants of health effects (Harrison and Yin, 2000; Kelly and Fussell, 2012).

A key point in assessing the health effects of air pollution is contrasting exposure between people residing in different cities

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(Pope et al., 2009; Dockery et al., 1993) or different areas within the same city and its surroundings (Raaschou-Nielsen et al., 2013; Beelen et al., 2014). Differences in exposure for people residing in urban areas are mainly related to differences in proximity to traffic sources and the most recent and advanced epidemiological studies, especially those devoted to long term and traffic-related health effects (Hampel et al., 2015; Wang et al., 2014), assign exposure based on outdoor concentration of air pollutants measured or estimated at the front door. In this respect, Land Use Regression Models and Dispersion Models provide comparable performance (de Hoogh et al., 2014; Beelen et al., 2010) and have been demonstrated to be effective tools to improve exposure assessment compared to the use of data from fixed site monitoring stations. Nitrogen dioxide and particle concentration (usually ultrafine particle number or particles with aerodynamic diameter below $10\text{ }\mu\text{m}$ – PM_{10} – or particles with aerodynamic diameter below $2.5\text{ }\mu\text{m}$ – $\text{PM}_{2.5}$) are the most common parameters used as air quality indicators.

While a number of studies have investigated spatial variations of air pollutant concentrations between traffic and urban background sites (Boogaard et al., 2011; Naser et al., 2008; Harrison et al., 2004) and in specific locations, such as building-free areas near highways (Patton et al., 2014; Zhu et al., 2002) or inside street canyons (Zhou and Levy, 2008), only very few studies have addressed specifically the issue of the differences between the concentrations of air pollutants at the front and back of buildings next to busy streets (Weber and Weber, 2008; Hitchins et al., 2002). Weber et al. found differences in particle mass and number concentrations between a busy urban street canyon and an adjacent backyard using optical particle counters. Higher concentrations in the canyon of on average 30% for PM_{10} and 22% for PM_1 were found within the street canyon. On the contrary Hitchins et al. found no significant gradients from the front to the rear of the building for $\text{PM}_{2.5}$ and sub-micrometer particle number concentrations considering three low-rise buildings at a distance between 11 and 75 m from roads.

The main goal of this paper is to investigate which particle metrics measured at the front door can be used as proxies of residential exposure. More specifically, we would like to investigate how large are the errors in assigning the same exposure to individuals residing in the same building near major roads. This is a key point in epidemiological studies because these individuals represent the very important subpopulation of highly exposed subjects. This work is part of a series of monitoring campaigns planned within the “Supersito” project (<http://www.arpa.emr.it/supersito>) aimed at assessing the variability of exposure within urban areas with a special emphasis on various PM metrics (Zauli Sajani et al., 2015).

2. Methods

2.1. Study design

To achieve the study aims, two main methodological options were selected a-priori.

The first is the choice of going beyond the mere comparison of front/back outdoor particle concentrations by including analyses indoors, where population exposure mostly occurs.

The second is the choice of monitoring uninhabited indoor environments. The main reason for this choice was related to the fact that many studies suggest that particles of outdoor and indoor origin have different physical and chemical characteristics (Brown et al., 2008) and probably can also cause different health effects (Zhou and Levy, 2013; Ebel et al., 2005). In fact, indoor exposure to particulates comes from particles of outdoor origin on which additive contributions of indoor-generated particles arising from

specific indoor sources superimpose (Urso et al., 2015; Fuller et al., 2013). It was assumed that mean within-city gradients of residential population exposure are primarily generated by exposure to air pollution of outdoor origin.

We selected two indoor environments similar in terms of volume and building materials, with virtually identical Air Exchange Rates (AERs). AERs were controlled by installing in each indoor environment a mechanical system to force air to be exchanged between indoors and outdoors. The system consisted of a fan connected to an air pipe (length = 1.2 m) carrying the air to the centre of the room (at a height of 2 m). Increased indoor air pressure caused the flow to go outwards through a grid. The fan velocity in each room was regulated in order to obtain an estimated AER of 0.4 h^{-1} in each room, reflective of a typical level for residential environments (Cattaneo et al., 2011). Our earlier work has shown that this method is highly effective and does not cause significant loss of particles (Zauli Sajani et al., 2015).

The measurements at the two sides of the building were conducted simultaneously indoors and outdoors (i.e. we had four simultaneous measurement sites). Fig. 1 outlines the size of the building and the location of the monitoring sites. Fig. 1S shows a map of the area and gives a bird's eye view of the surroundings of the monitoring sites. The study building was two-storeyed and located next to a street which surrounds the historical centre of Bologna, a 400,000-inhabitant city in northern Italy. Traffic and domestic heating during the cold season are the dominant air pollution sources in the area and cause high levels of air pollutants. In the period 2011–2013 the city-average annual concentration of $\text{PM}_{2.5}$ was $19.8\text{ }\mu\text{g}/\text{m}^3$ (average value derived from the two fixed site monitoring stations located in the urban area). The area near the monitoring sites carries a moderate volume of traffic, and the street next to the building is one of the busiest streets of the entire municipal area with a traffic load of 31,000 vehicles (4–5% heavy duty vehicles) each working day. The building is located in a broad (20 m) two-way street canyon. No sources of particles were present in the inner courtyard.

The indoor monitoring site at the front side (from now on “indoor front site”) was on the ground floor (street level). The volume of the room was 119 m^3 with a ceiling height of 3.5 m. The indoor environment at the back (from now on “indoor back site”) was on the ground floor as well. The volume of the room was 61 m^3 and the ceiling height was the same as that at the front side. The two indoor environments were completely separated with no common air exchange. For practical reasons the outdoor $\text{PM}_{2.5}$ monitoring site was located next to the building but at about 15 m from the indoor front site along the same street (Fig. 1).

Two monitoring campaigns were conducted in the period June–December 2013. Each monitoring campaign lasted 15 days: 1st campaign from 11 to 25 June (often referred to in the text as “hot period”), 2nd campaign (“cold period”) from 28 November to 13 December. Due to the availability of a unique filter for each day and measuring site, the chemical speciation was performed sequentially every three days for metals, ions, and carbon (Elemental Carbon and Organic Carbon). During cold periods elemental and organic carbon were measured on an 8 h basis in order to avoid filter overload.

2.2. Instrumentation and monitoring procedure

Four identical gravimetric samplers (Skypost PM, TCR TECORA Instruments, Corsico, Milan, Italy) were operated to measure indoor and outdoor daily $\text{PM}_{2.5}$ concentrations at the four sites (flow rate $2.3\text{ m}^3\text{ h}^{-1}$). Samples were collected on quartz fibre filters (Whatman, 47 mm diameter) and weighed following the procedure outlined in European Standard EN 12341:2014.

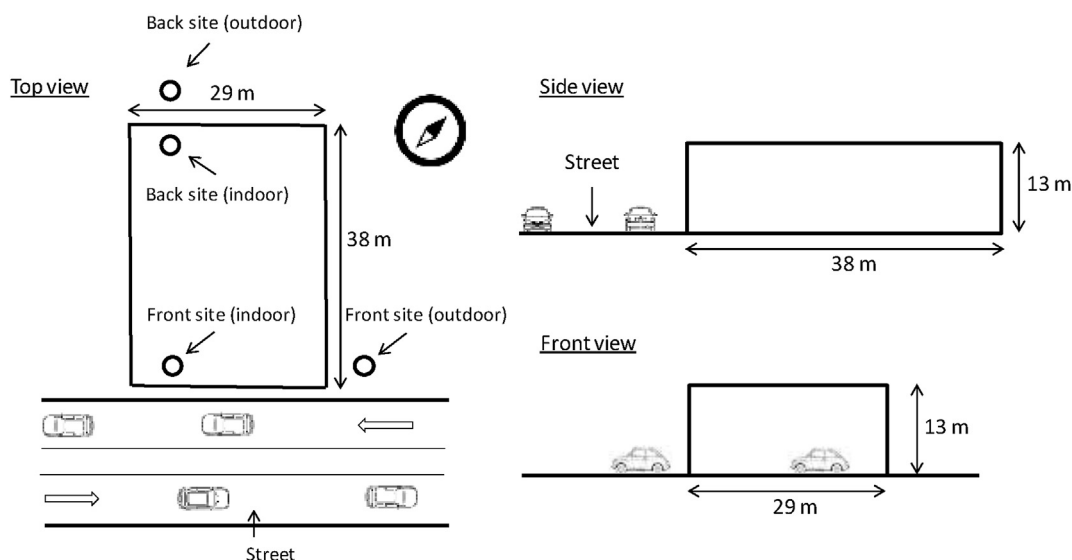


Fig. 1. Schematic diagram of the building and location of the sampling points.

PM_{2.5} samples were analysed for various chemical species. In this paper we present the findings of the chemical species having more than 50% of contemporary data above the limit of quantification (LOQ). LOQs for chemical components are reported in the Supplementary Information. Organic Carbon (OC) and Elemental Carbon (EC) were quantified by means of thermal-optical transmittance (Lab OC-EC Aerosol Analyzer, Sunset Laboratory Inc., Tigard, Oregon, USA) using the EUSAAR_2 protocol (Cavalli et al., 2010). Inorganic ions were determined by extracting species in 10 mL of ultrapure water. OC and EC values were summed to obtain TC. The extracts were filtered and analysed by Ion Chromatography (Dionex ICS-1000 for anions and ICS-1100 for cations, Thermo Fischer Scientific Inc., USA). Metals were analysed by Inductively Coupled Plasma – Mass Spectrometry (8800 ICP-MS, Agilent Technologies Inc., USA). Sample digestion with nitric acid and hydrogen peroxide in a microwave digestion apparatus and analysis were made according to European Standard EN 14902:2005, with a recovery efficiency over 85%.

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 FMPS was developed based on electrical aerosol spectrometer technology from Tartu University (Tammet et al., 2002). The FMPS spectrometer measured the size and number concentration of particles from 5.6 nm to 560 nm with 32 size bins every 1 s. Size bins below 13 nm were not included in the analysis because of the amount of data below the detection limit and also because of artifacts in the size distribution observed in other studies (Kaminski et al., 2013; Jeong and Evans, 2009). UFP concentrations were obtained summing the number of particles detected in the channels between 13 and 100 nm. Raw data were recorded every minute. Hourly and daily averages were derived from 1-min data and used in the analyses.

Based on the availability of only two FMPS, indoor and outdoor size distributions were obtained by coupling each FMPS with a switching system (Mod 11sc200, Pneumoidraulica Engineering S.r.l., Vicenza, Italy). One FMPS-switching system couple was located at the indoor front site and the other at the indoor back site. The switching systems allowed for sampling from indoor and outdoor air, switching from one to the other within a time frame set by the user. A valve installed in the system could switch between

sampling from the outdoor air, or from the indoor air. After the valve switched, there was a short time delay before the air from the sampled environment reached the instruments, which was the time the air travelled from the valve to the instruments. The system switched every 10 min between the indoor and outdoor measurements. In order to avoid the possibility of mixing of the outdoor and indoor air streams, the 2 min samples taken at the beginning of each 10 min period were deleted from the database. Particles were sampled indoor and outdoor through two 3/8 inches conductive silicon sampling tubes 2 m long. There was concern that this experimental arrangement for providing a forced input of aerosol might lead to depletion due to passage through the fan and pipe. This possibility was tested by experiments in which the particle number size distribution was measured at the inlet to the fan and outlet to the pipe by an FMPS system with rapid switching between the two sampling locations (upstream and downstream). The air inflow system was found to cause a minor loss of particles (Zauli Sajani et al., 2015).

The Air Exchange Rate in each indoor site was estimated in two ways. Firstly, AERs were estimated based on the air inflow measured at the end of the pipes taking into account the volumes of the rooms. The air inflow was measured with a TESTO 417 Anemometer (Testo AG, Lenzkirch, Deutschland). The measurements were performed five times at each velocity of the fan (ten equally spaced values of velocity of fan in each room). The velocity of fan in each room was chosen in order to obtain an AER equal to 0.4 h⁻¹. Furthermore, in order to verify the estimated AERs, additional measurements based on the tracer gas-decay technique (ASTM Standard E741-95) were carried out. During these measurements, pure CO₂ was released as tracer gas within each indoor environment to obtain a uniform tracer concentration through the spaces being studied (ASHRAE, 1997). The decay in CO₂ concentrations was continuously (sampling intervals of 1 min) monitored using non-dispersive infrared analysers (GE sensing Telaire 7001, Goleta, CA, USA) provided with a battery-operated data logger (Hobo U12; Onset Computer Inc., Pocasset, MA, USA). A CO₂ analyser was placed at each indoor and outdoor sampling site in order to monitor both the indoor gas decay – until the indoor baseline was reached – and the corresponding outdoor CO₂ levels.

Meteorological outdoor data were obtained from the meteorological station belonging to the urban meteorological network of

ARPAE Emilia-Romagna (“Bologna Urbana” station - http://www.arpa.emr.it/sim/?osservazioni_e_dati/dati_stazioni_regionali). The station is located close to the monitoring sites at an horizontal distance of about 30 m. The station is on the top of a building 25 m above ground level and it measures hourly temperature, relative humidity, and wind speed and direction. Temperature is measured by means of a Pt100 RTD sensing element, while a capacitive sensor is used for humidity; wind speed and direction are measured using a three-cup anemometer and a wind vane respectively. Two digital thermo-hygrometers (Testo 175 H2, Testo AG, Lenzkirch, Deutschland) were used to measure temperature and relative humidity in the two indoor environments. Measurements were taken very hour.

2.3. QA/QC and statistical analysis

2.3.1. QA/QC

Agreements among gravimetric PM_{2.5} measurements were checked in several intercomparison campaigns carried out in the years 2008–2012 before and during the “Supersito” project. Both correlation levels and tests for differences for slope and intercept of orthogonal regressions between co-located instruments were used (EC, 2010). Determination coefficients were always higher than 0.972 (mean correlation 0.985). Typical errors (standard deviation of the differences between samplers) were about 2 µg/m³ and were quite similar among the various intercomparisons. The differences for slope (from identity) and intercepts (from zero) were usually not significant and not related to specific instruments. Consequently, no corrections were applied to PM_{2.5} data.

In the preliminary phase of the monitoring campaign we carried out four intercomparisons (one before and after each monitoring campaign – mean duration 2 days) between the two FMPS using the same methodology applied for PM_{2.5} samplers. No heteroscedasticity was detected and typical errors (standard deviation of the differences between UFP hourly data from the intercomparison campaigns) were 580 #/cm³. The determination coefficient was 0.976. No correction was applied to the data.

Quality control of PM_{2.5} mass and chemical composition data has been carried out based on residuals calculated by regression analysis between indoor and outdoor data. Data with residuals larger than three times the standard deviation of residuals were identified as anomalous. For FMPS data quality control the following procedure was used: a) applying a log10 function on the UFP minute data; b) stratifying data in time slots of 3 h (0–3, 3–6 etc) and calculating the summary statistics for each slot and campaign; c) classifying data as anomalous if they were higher than the mean plus three times the standard deviation for the corresponding campaign and time slot. Then we averaged non-anomalous data on an hourly and daily basis. Statistical data analysis was carried out using the R package (Version 3.0.1).

2.3.2. Statistical analyses

Summary statistics and paired t-test have been used to investigate differences between series of measurements. Pearson correlation coefficients and regression analysis have been used to address linear relationships between data. An orthogonal regression approach (Fuller, 1987), which is the most suitable when both dependent and independent variable are affected by errors and are not related by a causal relationship, has been adopted.

3. Results and discussion

3.1. Meteorological conditions and air exchange rates

The meteorological conditions during the study periods are

summarized in Table 1S. The sampling periods were quite representative of the typical annual variability in the area (Table 2S). The mean outdoor temperature measured in the nearby meteorological monitoring station during the first campaign (hot period) was 25.2 °C with hourly values ranging from 15.6 °C to 34 °C. Temperatures were much lower during the cold period, as expected, with a mean value of 4.8 °C and minimum and maximum equal to −1.4 and 11.7 °C, respectively. The area is characterized by low wind intensities and this was a common characteristic of the two monitoring campaigns (mean wind intensities from 1.6 m/s during the second campaign to 2.5 m/s during the first campaign). During the sampling periods precipitation events were rare: in fact, only one rainy day during the hot period was recorded with a total of 5.6 mm.

The temperatures measured at the two indoor monitoring sites showed very similar values and temporal patterns. Indoor seasonal differences were clearly reduced compared to those outdoors, especially due to the higher minimum values. Mean indoor temperatures during the hot period were 29.4 °C and 29.9 °C for the front and back site, respectively. In the cold season, indoor temperatures reached mean values of 16.1 °C (front site) and 17.7 °C (back site).

Fig. 2S shows the wind rose for the campaign periods in 2013 compared to the overall average for the years 2012–2014. It is seen that the campaign period is broadly representative of the longer term average wind rose.

AERs estimated with the tracer gas-decay technique were between 0.2 and 0.3 h^{−1} in both indoor sites. These values were slightly lower than those estimated (0.4 h^{−1}) based on air inflow at the end of the pipes and room volumes.

3.2. Comparison of front and back monitoring sites

3.2.1. Ultrafine particle number concentrations

Based on the procedure outlined in the methods section, 1.2% of the raw data from the FMPS located at the front of the building (and measuring alternatively indoor and outdoor) were classified as outliers and removed from the database. A similar percentage of outliers (2%) were found for the FMPS located at the back side. The completeness of hourly data was above 94% at all measurement sites.

Fig. 2(a) and Table 1 give an overview of the UFP number concentrations during the monitoring campaigns. Mean UFP concentrations at the outdoor front site were 3.5 times higher than at the outdoor back site with higher front/back ratios during the cold period (4.2) compared to the hot season (2.5). The highest outdoor hourly value at the front site was 120,900 #/cm³ while the highest value in the back site was 26,860 #/cm³. Our findings were similar although with gradients generally a little higher than those found in other studies comparing traffic with background sites (Zauli Sajani et al., 2015; Patton et al., 2014; Boogaard et al., 2011; Moore et al., 2009; Rivera et al., 2012). Similar results were also found in a study in Athens (Diapoulis et al., 2011) showing a spatial variability ranging from ratios of 1.8–2.6 depending on the season. A difference of mean concentrations of UFP between traffic and urban-mean locations estimated with short-term measurements at 60 locations within the Basel urban area, showed a mean ratio of 1.6 (Ragettli et al., 2014). A study on local-scale spatio-temporal variation of particle number in a 1 km² area in Braunschweig, Germany, showed during the winter season UFP number concentrations almost double at the roadside sites compared to residential and backyard sites. Reduced gradients were found during the summer season (Ruths et al., 2014). Similar results were also found in Budapest (Salma et al., 2014).

The range of UFP indoor concentrations was much lower than

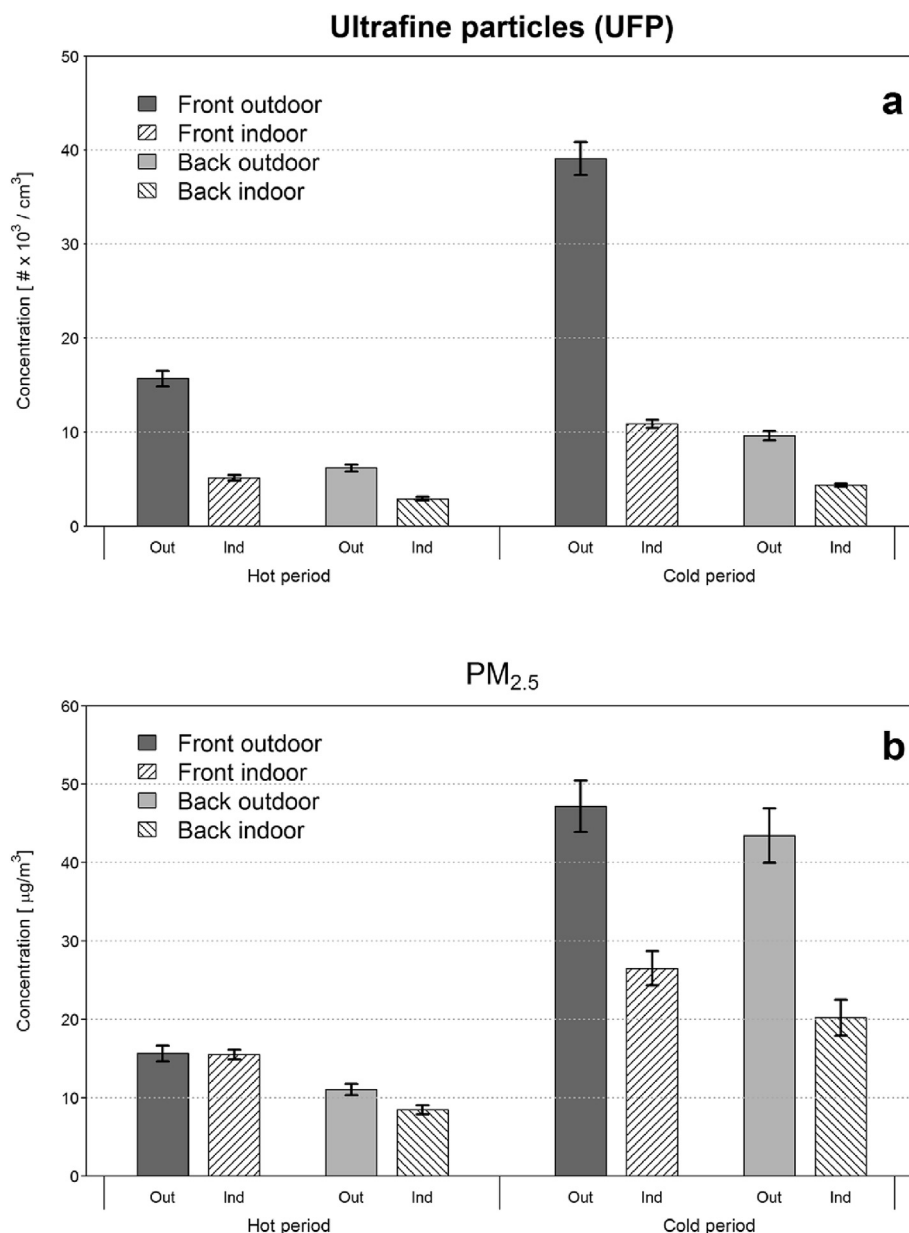


Fig. 2. Average value and standard error of PM_{2.5} (a) and UFP (b) during the two monitoring campaigns.

outdoors, at both sites. As for outdoors, large gradients were found in the UFP indoor concentrations between the front and back (ratio 2.2), with mean $I_{\text{front}}/I_{\text{back}}$ ratios ranging from 1.7 during the hot period to 2.5 during the cold season.

The I/O ratio for UFP was higher in the back compared to the front of the building (0.5 vs 0.3) and remained almost constant over the two campaigns (Table 1). As shown in the following section, higher I/O ratio at the front side is due to the very effective loss of freshly nucleated particles in the indoor environment. The I/O ratios found in this study are in good agreement with that obtained in the previous SUPERSITO campaign (Zauli Sajani et al., 2015). Kearney et al. (2011) found median I/O ratios of hourly data ranging from 0.27 to 0.39. Diapouli et al. (2011) found an I/O ratio for particles in the 10–400 nm size range equal to 0.6 with AERs in the 0.5–1 h⁻¹ range, while I/O ratios between 0.3 and 0.4 were found in Erfurt (Germany) (Cyrus et al., 2004) and in other major European urban areas (Hoek et al., 2008a,b). Meier et al. (2015) investigated I/

O ratios for Particle Number Concentration (PNC) in four Swiss cities. The cities most similar to Bologna were Geneva and Lugano and the I/O ratios for homes without smoking influence were 0.52 (Geneva) and 0.70 (Lugano). As expected, those values were a little bit higher than ours because of the presence of indoor sources.

The Pearson correlation coefficient between daily outdoor UFP concentrations at the front and rear was equal to 0.84 (Fig. 3). This value was slightly lower than in the previous SUPERSITO campaign and significantly higher than those reported in other studies focused on particle number concentrations (Puustinen et al., 2007). Correlations between indoor UFP concentrations were lower ($R = 0.79$). Very similar correlation coefficients were found between indoor and outdoor UFP concentrations at the front and back sides of the building ($R = 0.94$ and 0.85 respectively).

It was expected that wind direction would influence the relationship between the outdoor concentrations at the front and back of the building and thereby also influence the indoor

Table 1
Summary statistics of PM_{2.5} mass ($\mu\text{g m}^{-3}$) and UFP number (cm^{-3}) during the monitoring campaigns.

	Front					Back				
	Outdoor		Indoor		I/O mean	Outdoor		Indoor		I/O mean
	Number of valid data	Mean (min–max)	Number of valid data	Mean (min–max)		Number of valid data	Outdoor mean (min–max)	Number of valid data	Indoor mean (min–max)	
PM2.5 (24-h data)										
All data	30	33.3 (11.3–68)	30	21.7 (11.5–41)	0.76	30	29.2 (7.5–64)	30	15.1 (3.8–36)	0.60
Hot period	11	15.6 (11.3–23.8)	15	15.5 (11.5–20.1)	1.02	15	11.1 (7.5–16.5)	15	8.5 (3.8–12.8)	0.78
Cold period	15	47.1 (27–68)	14	26.5 (16–41)	0.57	15	43.4 (24–64)	14	20.2 (9–36)	0.46
UFP (1-h data)										
All data	652	25,358 (2118–120931)	652	7625 (1472–21142)	0.39	652	7444 (647–26860)	652	3544 (213–8854)	0.55
Hot period	353	15,502 (2118–38367)	353	5109 (1472–10773)	0.39	353	6169 (647–24308)	353	2932 (213–7901)	0.52
Cold period	299	36,994 (3095–120931)	299	10,595 (2854–21142)	0.38	299	8948 (1781–26860)	299	4266 (1730–8854)	0.58

Sampling periods: hot period from 11 to 25 June 2013; cold period from 28 November to 13 December 2013.

concentrations. Both indoor and outdoor concentrations of UFP at the front of the building showed no appreciable sensitivity to wind direction (see Figs. 3S and 4S). At the back of the building, both outdoor and indoor UFP concentrations showed a similar wind speed and direction dependence with highest concentrations on stronger winds in the easterly sector. This is probably due to the fact that easterly winds tend to move primary or freshly nucleated particles from the street to the back of the building. The directional pattern of concentration ratios between the front and back of the building for UFP (Figs. 3S and 4S) was broadly similar, with the highest ratios occurring in the sector between north and west. Ratios between the front and back of the building outdoors showed a range from 2 to 4, while those indoors showed a smaller range from 2 to 2.5. Irrespective of wind direction, concentrations were always higher at the front than the rear of the building (Fig. 3S) and the influence of wind direction on the pattern of concentrations was not high. Therefore all the collected data have been presented together without disaggregation according to wind direction.

3.2.2. Particle size distribution

Fig. 4 shows the mean indoor and outdoor size distributions at the four measuring sites for the two measurement periods. Multi-modal distributions with sharp peaks at 20 and 30 nm in the concentrations were found at the outdoor front site. A second peak can occur at about 60–100 nm. This is typical of heavily trafficked sites, with the modes arising from the semi-volatile nucleation particles and solid graphitic particles respectively (Harrison et al., 2011).

The presence of a bi (or tri)-modal distribution was also shown in previous studies (Morawska et al., 2008; Hussein et al., 2005) and is in line with knowledge of particle emissions and transformation. The 30 nm mode is due to the combination of freshly nucleated particles formed as the exhaust gases are diluted with ambient air and particles directly emitted by vehicles (Charron and Harrison, 2003). Nucleation mode particles are associated with the hot exhaust gases expelled from the tailpipe of a vehicle. These gases cool and condense to form large numbers of very small particles in the air (Shi and Harrison, 1999). On-road dilution of the exhaust plume is very important in the generation of particles in the exhaust plume. These nucleation processes are favoured by low ambient temperatures and high relative humidity (Charron and Harrison, 2003) which are typical in the area during the cold season (Table 1S). In addition, the gaseous precursors condense or adsorb on to the surface of carbon particles in the accumulation mode.

Upon entry into the building, not only would the nucleation mode fraction show a higher deposition velocity than the coarser graphitic mode (Riley et al., 2002), it would be subject in winter to

evaporation at the higher indoor temperatures (Dall'Osto et al., 2011) and the hydrocarbon vapours released would tend to adsorb to indoor surfaces (Weschler and Nazaroff, 2008) and settled indoor dusts (Weschler and Nazaroff, 2010). Such processes would contribute to a relatively rapid loss of the nucleation mode of particles such that shown in the front site between outdoor and indoor and from front outdoor to back.

Indoor size distributions were very similar to the findings of the previous work comparing traffic and residential sites (Zauli Sajani et al., 2015). Similar shapes and differences between sites were also found by Ruths et al. (2014). Much lower relative weight of the nucleation mode compared to the accumulation mode was found indoors compared to the outdoor size distributions as was also found in other previous studies (Hussein et al., 2004; Diapouli et al., 2011).

Relevant differences in the shape of the size distributions were found between the cold and the hot period. The front outdoor site showed a much higher relative weight of small particles during the cold period. On the contrary, all other measuring sites showed a shift of the principal mode from about 30 nm towards 60–80 nm. The striking difference in the front outdoor size distributions between the seasons is likely to be due to the rapid loss of the semi-volatile constituents of the particles within the street canyon due to evaporation at the high temperatures occurred during the hot period (Table 1).

3.2.3. PM_{2.5} mass

Indoor and outdoor PM_{2.5} concentrations during the two monitoring campaigns are shown in Fig. 2(b) and Table 1. Small but significant differences in PM_{2.5} outdoor concentrations were found, with average PM_{2.5} levels at the outdoor front site about 14% higher than those at the outdoor back site. The highest outdoor values for daily mean PM_{2.5} concentrations were 68 $\mu\text{g/m}^3$ and 64 $\mu\text{g/m}^3$ for the front and back site, respectively. As expected, outdoor concentrations during the cold period were, on average, more than three times higher compared to summer, according to the typical PM seasonal trends in some European countries (Oeder et al., 2012).

The outdoor PM_{2.5} ratio between front and back levels was slightly higher than the ratio between traffic and residential sites found in similar monitoring campaigns conducted under the same SUPERSITO project (1.14 vs 1.06) (Zauli Sajani et al., 2015). The PM_{2.5} spatial variability in our study was virtually equal to the mean within-city variability reported in the ESCAPE study, a very large epidemiological survey in Europe which included monitoring campaigns on air pollution spatial variability in urban areas (Eeftens et al., 2012). That survey reported a mean ratio between traffic sites and urban background sites equal to 1.14, with a quite broad range of values (0.96–1.30). In particular, street and urban

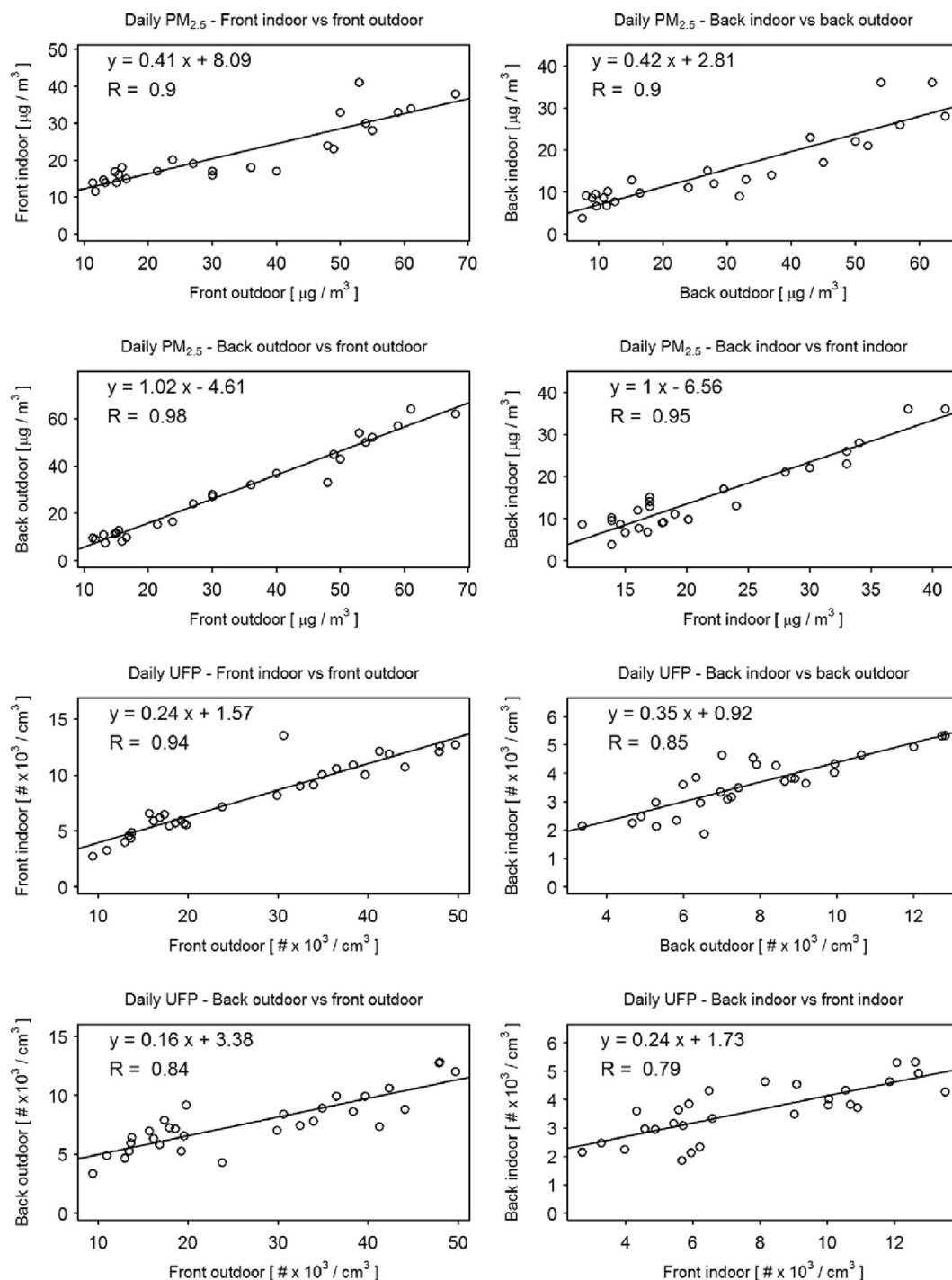


Fig. 3. Scatter plots for daily PM_{2.5} mass and UFP number concentrations.

background ratios equal to 1.02 and 1.14 were found for Lugano and Turin, the closest ESCAPE cities to Bologna. Similar PM_{2.5} gradients (from 1.0 to 1.3) were found in a study focused on the comparison between street and background locations within the same cities in the Netherlands (Boogaard et al., 2011).

Differently from UFPs, higher PM_{2.5} ratios were found between front and back indoor concentrations (1.44 on average, 1.82 during the cold period and 1.31 during the hot period).

Mean indoor/outdoor (I/O) ratios for PM_{2.5} during the cold period were equal to 0.57 at the front side and 0.46 at the rear.

Differently from UFPs, higher values were found during the hot period with mean I/O ratios of 1.02 and 0.78 for the front and back side, respectively (Table 1). These results were in good agreement with previous studies in indoor settings (Chen and Zhao, 2011; Hanninen et al., 2004). Averaging I/O values for both building sides and measurements campaigns provided an I/O ratio equal to 0.71, a value within the range of the I/O ratios obtained in Swiss homes (Meier et al., 2015). In particular, the homes without smoking influence provided median I/O ratios equal to 0.86 in Geneva and 0.65 in Lugano. Inter-campaign PM_{2.5} variations in the

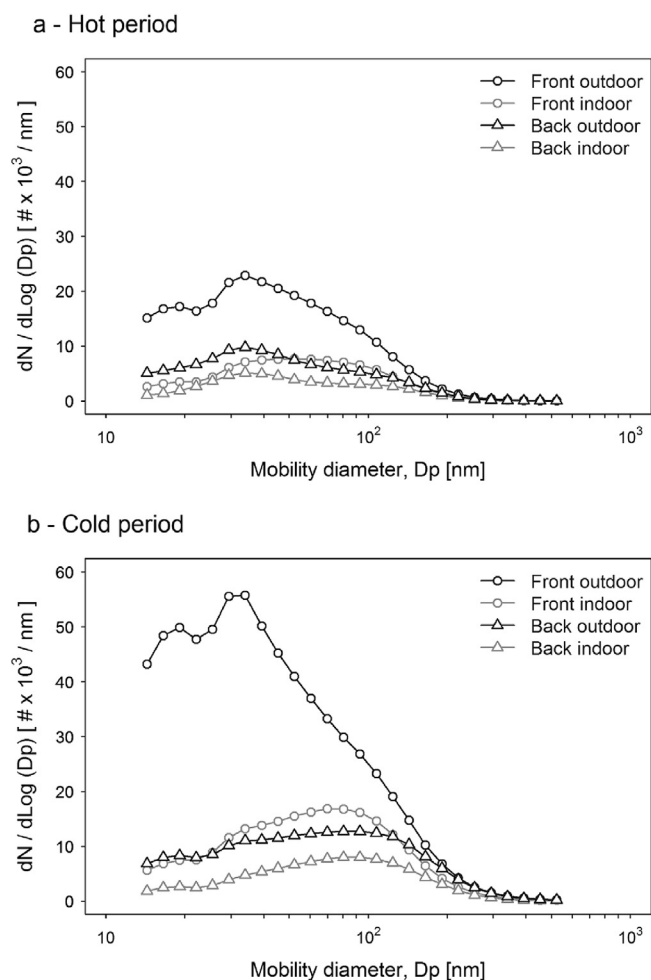


Fig. 4. Mean particle size distribution of hourly data for the hot (a) and the cold period (b). The x-axis the Aerodynamic Diameter (D_p) is reported on a logarithmic scale. On the y-axis $dN/d\log(D_p)$ represents the number of particles per unit increment of D_p on a logarithmic scale.

indoor environments were lower than outdoors. The ratios between the $PM_{2.5}$ indoor concentrations measured during the cold and the hot period were 1.71 at the indoor front site and 2.38 at the indoor back site. The corresponding ratios for outdoor $PM_{2.5}$ concentrations were markedly higher (3.02 and 3.91 at the front and back site, respectively).

A high correlation ($R = 0.98$) between $PM_{2.5}$ concentrations at the two outdoor sites was found (Fig. 3). Indoor $PM_{2.5}$ concentrations were highly correlated as well ($R = 0.95$). Somewhat lower correlations were found between indoor and outdoor concentrations with I/O correlation coefficients at the front and back of the building equal to 0.91 and 0.90, respectively. These findings are similar to those found in other studies (Hanninen et al., 2004) and the high I/O and I/I correlations is probably related to the absence of indoor sources.

3.2.4. Chemical composition of $PM_{2.5}$

Table 3 shows descriptive statistics for the $PM_{2.5}$ chemical components. Organic carbon (OC) and nitrates were found to be the largest contributors to $PM_{2.5}$ mass at the two outdoor monitoring sites, followed by elemental carbon (EC), sulfates and ammonium. Indoor data confirmed the primary role of OC and showed a pronounced decrease of nitrates. The contribution of ammonium was more than three times higher at the outdoor sites compared to

indoor.

Significant differences (p -value < 0.005) were found between outdoor front and back average levels for EC (3.96 and $2.25 \mu\text{g}/\text{m}^3$, respectively) and Mn (7.25 and $5.54 \text{ ng}/\text{m}^3$, respectively) (Table 2). Large but not statistically significant were the outdoor trends for Fe and Sn (Table 2). The same patterns were confirmed indoors, with indoor differences always higher than outdoors. The largest $I_{\text{front}}/I_{\text{back}}$ ratios were found for Fe and Sn, with values more than three times at the front door compared to the back side (Table 2).

As reported in the literature, EC is an important component of diesel exhaust particulate matter (Suvendrini Lena et al., 2002; Shi et al., 2000). Mn and Sn are typical markers of vehicle emissions for fine PM (Marcazzan et al., 2001; Monaci et al., 2000). Fe, Mn and also Cu are related with brake wear emissions (Manoli et al., 2002; Gietl et al., 2010). Tyre wear, brake wear, vehicle component detachment and fluid leakage are known sources of Fe (Ball et al., 1991), as well as motor exhausts (Pant and Harrison, 2013). Furthermore, traffic-induced road dust resuspension also plays an important role for some elements (Fe and Mn), including those originated from crustal sources. Although more marked for the coarse size fraction, this can be considered also a contributor to fine fraction aerosol (Manoli et al., 2002; Harrison et al., 2012). Götschi et al. (2005) found substantially elevated concentrations of Cu, Fe and Mn in $PM_{2.5}$ particles at a busy street location. A strong elevation in EC, organic compounds and Fe-rich dust was also reported elsewhere (Harrison et al., 2004).

Modestly increased Zn concentrations at the street site (1.11) were found in our survey. This could be due to the fact that Zn may derive from tyre wear (Manoli et al., 2002) but also from a large number of other atmospheric sources (Thorpe and Harrison, 2008).

Large influence of traffic proximity on chemical composition of $PM_{2.5}$ was also found in the ESCAPE data, with relevant differences in concentrations between traffic and urban background sites especially for Fe and Zn (Tsai et al., 2015).

I/O ratios were < 1 for all the chemical species at both sites, with the exception of strong traffic markers (EC, V, Sn, Sb and Fe) at the front side (Table 3). This could be due to the location of the outdoor sampling site (Fig. 1), which was placed in a more open space at about 15 m from the indoor environment, and it is likely that slightly higher concentrations of traffic-related pollutants would

Table 2

Front-back ratio for different particle metrics considering both indoor and outdoor concentrations. Table is ordered by outdoor gradients. P-value from paired t-test are also reported.

	Outdoor		Indoor	
	Front/back	p-value	Front/back	p-value
UFP	3.4	$< 0.001^{**}$	2.2	$< 0.001^{**}$
$PM_{2.5}$	1.1	$< 0.001^{**}$	1.4	$< 0.001^{**}$
Elemental carbon (EC)	1.8	$< 0.001^{**}$	2.5	$< 0.001^{**}$
Iron (Fe)	1.7	0.122	3.7	0.001**
Manganese (Mn)	1.3	0.003**	1.5	0.002**
Tin (Sn)	1.3	0.246	3.0	$< 0.001^{**}$
Total carbon (TC)	1.2	0.016*	1.5	$< 0.001^{**}$
Organic carbon (OC)	1.1	0.038*	1.1	0.008**
Zinc (Zn)	1.1	0.053	1.2	0.008**
Nitrates (NO_3)	1.1	0.073	1.3	0.086
Sulfates (SO_4)	1.0	0.184	1.0	0.889
Ammonium (NH_4)	1.0	0.456	1.1	0.129
Vanadium (V)	1.0	0.741	1.4	0.002**
Arsenic (As)	1.0	0.966	1.1	0.171
Cadmium (Cd)	1.0	0.802	1.1	0.866
Lead (Pb)	1.0	0.782	1.0	0.686
Potassium (K)	1.0	0.012*	1.0	0.279
Antimony (Sb)	0.9	0.591	2.3	0.002**

* p-value < 0.05 ; ** p-value < 0.01 .

have been found if the outdoor sampling location was in front of the corresponding indoor sampling site. Low I/O ratios (<0.7) were found for chemical species of typical outdoor origin including nitrates (0.15), ammonium (0.29) and sulfates (0.65) at both sites (Table 3). The very low I/O ratios for nitrate are due to the evaporation of ammonium nitrate indoors, due to deposition of ammonia and nitric acid vapour on indoor surfaces causing destabilisation (see below).

The outdoor and indoor EC/TC ratios were respectively 0.31 and 0.39 at the front sites and 0.21 and 0.22 at the back sites. These values were similar to those reported by Naser et al. (2008) for outdoor urban data.

Table 4 shows the Pearson correlation coefficients among the chemical components. Outdoor data were highly correlated. The correlation coefficients were all greater than 0.9 with the exception of Sn (0.70) and Fe (0.38), which further confirms in general the origin of these elements, closely related to the traffic source. The findings for OC, EC, ammonium, nitrates and sulfates were expected as high correlation levels between within-city outdoor concentrations of PM these constituents were found by Bell et al. (2011) and Naser et al. (2008).

Very high correlations were also found between indoor data, except for Sn (0.69). Correlation levels were also generally high between indoor and outdoor data. Low R values were found only for Fe (0.16) at the front side of the building and for Sn (0.45) at the rear. High correlations between indoor and outdoor levels of OC and EC were found by Sawant et al. (2004) in several schools in California. The I/O correlations for ammonium found by the same authors showed large variability in the different schools. Particles of outdoor origin can undergo substantial changes and may be lost to building walls during indoor penetration. A study investigating the transformation of ambient ammonium nitrate aerosols in indoor environments has shown that measured indoor concentrations were considerably lower than the values predicted based only on penetration and deposition losses (Lunden et al., 2003). This was attributed to the semi-volatility of ammonium nitrate, leading to losses as nitric acid and ammonia vapours. This behaviour was also highlighted in the previous SUPERSITO campaign (Zauli Sajani et al., 2015).

3.3. Strengths and weaknesses of the study

A major strength of the study was the simultaneous measurement of a number of particulate characteristics (mass, size

distribution, chemical composition) to assess the variability of exposure between front and back of a building next to a busy street. The simultaneous indoor and outdoor measurements were also a key strength of the study because of the role of the indoor environment in determining exposure. To our knowledge, this is the first study comparing simultaneous indoor and outdoor measurements on the front and back side of the same building taking into account both size distribution and chemical components of particulate matter. The findings of our study suggest the importance of improving the characterization of exposure of people living in buildings close to trafficked streets, in particular for epidemiological studies aiming at assessing the health risks associated with residential proximity to traffic source. A possible way to reduce misclassification could be to try to enrol as “highly exposed” only people residing in apartments facing trafficked streets. An alternative approach could be to assign exposure based on air pollutant concentrations estimated at the building centroids rather than at front door.

The choice of monitoring uninhabited indoor environments made possible the use of instruments such as instruments for PM_{2.5} mass measurements using reference methods that are rarely used in inhabited house due to their size and noise emissions.

A possible weakness of the study is related to the choice of a unique building to be monitored. Other monitoring campaigns in different settings could be useful but we think that this study should be considered significantly more than a pilot study. As a matter of fact, the selected building is a quite common setting within urban areas even though pure canyon configuration with higher and continuous buildings could produce even higher front/back gradients compared to our findings. In conclusion we think that our study gives important insights about possible misclassification of people residing next to busy streets. The choice of selecting AER to a typical value should be a plus with regards to the generalization of our findings.

Field campaigns in inhabited houses could give supplemental information but would not separate outdoor and indoor origins of particles. Moreover such an approach would suffer from the tremendous variability of indoor characteristics and personal habits of the population. This would represent a substantial problem in being able to identify gradients in population exposure due to particles of ambient origin.

It may also be questioned as to whether the air ventilation system could be a weakness of our study. This question was considered in detail in our earlier paper (Zauli Sajani et al., 2015). In

Table 3

Descriptive statistics and Indoor/Outdoor ratio (I/O) for different chemical components. OC, EC, TC, NH₄, NO₃, SO₄, K are expressed in $\mu\text{g}/\text{m}^3$ while the other elements in ng/m^3 .

	Number of valid data ^a	Front			Back		
		Outdoor mean (min–max)	Indoor mean (min–max)	I/O mean	Outdoor mean (min–max)	Indoor mean (min–max)	I/O mean
Organic carbon (OC)	9	8.84 (3.33–18.89)	6.82 (2.77–13.08)	0.77	7.84 (2.91–15.55)	5.98 (2.5–11.44)	0.76
Elemental carbon (EC)	9	3.96 (1.22–7.65)	4.3 (2.01–7.59)	1.09	2.25 (0.77–4.95)	1.7 (0.5–4.03)	0.76
Total carbon (TC)	9	12.8 (5.02–26.54)	11.12 (5.1–20.67)	0.87	10.64 (3.68–20.67)	7.68 (3–15.47)	0.72
Ammonium (NH ₄)	9	2.92 (0.86–5.96)	0.88 (0.44–1.86)	0.3	2.85 (0.92–6.19)	0.77 (0.37–1.33)	0.27
Nitrates (NO ₃)	9	8.2 (0.23–20.41)	1.28 (0.16–3.54)	0.16	7.76 (0.18–19.32)	1.02 (0.15–2.37)	0.13
Sulfates (SO ₄)	9	3.32 (1.57–5.06)	2.13 (1.01–3.1)	0.64	3.19 (1.46–5.05)	2.11 (1.07–3.41)	0.66
Potassium (K)	5	0.44 (0.24–0.59)	0.36 (0.18–0.49)	0.82	0.45 (0.24–0.6)	0.35 (0.19–0.47)	0.78
Vanadium (V)	8	1.06 (0.33–2.29)	1.24 (0.72–2.1)	1.17	1.05 (0.33–2.29)	0.92 (0.34–1.5)	0.88
Arsenic (As)	8	0.6 (0.21–1.12)	0.48 (0.22–0.88)	0.8	0.6 (0.19–1.1)	0.45 (0.12–0.85)	0.75
Cadmium (Cd)	7	0.19 (0.07–0.33)	0.15 (0.06–0.25)	0.79	0.19 (0.05–0.31)	0.14 (0.04–0.24)	0.74
Tin (Sn)	8	3.18 (0.9–6.41)	3.87 (1.39–5.52)	1.22	2.44 (0.62–5.04)	1.28 (0.56–2.67)	0.52
Antimony (Sb)	8	1.35 (0.36–4.14)	1.44 (0.28–3.19)	1.07	1.53 (0.24–6.21)	0.64 (0.13–1.44)	0.42
Lead (Pb)	8	7.14 (1.57–15.14)	5.29 (1.05–11.36)	0.74	7.21 (1.6–14.62)	5.22 (1.01–11.72)	0.72
Iron (Fe)	6	212.16 (86.48–395.53)	251.19 (141.35–350.87)	1.18	122.74 (84.9–203.99)	68.59 (29.95–127.45)	0.56
Zinc (Zn)	6	38.84 (17.18–52.12)	29.12 (13.5–40.96)	0.75	35.05 (20.62–46.8)	24.2 (13.14–33.77)	0.69
Manganese (Mn)	5	7.25 (5.07–10.33)	5.54 (4.12–7.76)	0.76	5.55 (4.31–8.32)	3.69 (2.83–5.86)	0.66

^a Number of data simultaneously above LOQ in all monitoring sites.

Table 4
Pearson correlation coefficients (R) and regression lines for different chemical species. Intercepts for OC, EC, TC, NH₄, NO₃, SO₄, K are expressed in µg/m³; intercepts for the other elements are expressed in ng/m³.

	Front outdoor vs back outdoor			Front indoor vs back indoor			Front indoor vs front outdoor			Back indoor vs back outdoor		
	R	Slope	Intercept	R	Slope	Intercept	R	Slope	Intercept	R	Slope	Intercept
Organic carbon (OC)	0.99	1.23	−0.79	0.99	1.17	−0.2	0.98	0.65	1.06	1	0.69	0.61
Elemental carbon (EC)	0.97	1.4	0.81	0.97	1.7	1.41	0.96	0.99	0.37	1	0.82	−0.14
Total carbon (TC)	0.97	1.23	−0.32	1	1	0	0.98	0.74	1.63	1	0.92	1.38
Ammonium (NH ₄)	0.99	0.98	0.12	0.94	1.5	−0.28	0.68	0.16	0.41	0.53	0.08	0.53
Nitrates (NO ₃)	1	1.02	0.32	0.98	1.41	−0.16	0.97	0.14	0.1	0.97	0.1	0.21
Sulfates (SO ₄)	0.98	0.99	0.14	0.85	1.01	0	0.89	0.6	0.16	0.94	0.6	0.19
Potassium (K)	1	0.99	0	1	1.07	−0.02	0.99	0.85	−0.02	1	0.79	0
Vanadium (V)	0.99	1	0.01	0.93	1.23	0.1	0.99	0.66	0.54	0.9	0.51	0.38
Arsenic (As)	0.98	1.02	−0.01	0.97	0.87	0.09	0.99	0.73	0.04	0.99	0.86	−0.07
Cadmium (Cd)	0.98	1	0	0.97	1.03	0	0.99	0.71	0.01	0.96	0.68	0.01
Tin (Sn)	0.7	1.81	−1.25	0.69	2.53	0.63	0.51	0.48	2.34	0.45	0.29	0.56
Antimony (Sb)	0.92	0.64	0.37	0.93	1.77	0.31	0.83	0.65	0.56	0.8	0.22	0.3
Lead (Pb)	0.99	1.03	−0.26	0.99	0.99	0.1	1	0.76	−0.11	1	0.78	−0.4
Iron (Fe)	0.38	5.97	−521.02	0.94	2.55	76.13	0.16	0.23	201.42	0.94	0.75	−23.75
Zinc (Zn)	0.98	1.31	−7	0.98	1.3	−2.42	0.99	0.72	1.02	0.97	0.72	−1.14
Manganese (Mn)	0.97	1.23	0.41	0.95	1.23	1	0.97	0.77	−0.03	0.99	0.78	−0.62

brief, we think that the two identical simple systems installed to impose fixed and equal air exchange rates between sites should be considered a strength and not a weakness of the study. Firstly, this choice provided a good control on the air exchange rate in the two indoor environments. Secondly, the choice eliminated the effect of possibly different specific infiltration factors of each building envelope.

This work didn't consider apartments facing both to the front and back of a building. In this situation front-back difference in air pollutant concentrations between rooms are expected to remain present but probably lower.

4. Summary and conclusions

This study focused on the variations of exposure to various particle metrics from the front to the back of a building located next to a trafficked street. In particular, both indoor and outdoor spatial gradients have been analysed in terms of PM_{2.5} mass and chemical composition, size distribution and particle number concentrations. Large differences in the concentrations of UFP, tin, manganese, iron and elemental carbon were found both indoors and outdoors. Sizeable but less substantial were the spatial gradients for PM_{2.5}. Significant differences were also found for the shape of particle size distributions for outdoor particles, while indoor particles showed very similar distributions. Indoor front/back ratios are generally consistent in terms of direction but with remarkable differences in magnitudes between the different particle metrics. Indoor concentrations were much lower than outdoors for PM_{2.5} mass and UFP. Taking into account the chemical components, the building environment was protective especially for nitrates, ammonium, potassium, sulfates, lead and cadmium underlining the different characteristics of indoor particulate matter compared to outdoor independently from the presence of indoor sources.

Our findings showed that the variability of exposure to air pollution of people living in the same building next to a busy street may be large i.e. some people could be erroneously classified as highly exposed. The front/back variability was comparable to that found in previous studies involving people residing in buildings in heavy and low traffic areas. Given that a common way to assess the risks due to exposure to air pollutants, and in particular to the risks associated with proximity to traffic sources, is by contrasting exposure levels within cities, and considering that the assessment of population exposure is usually based on residential outdoor concentrations estimated at the front-door, the impact of exposure

misclassification could be important. Epidemiological studies should consider these findings when designing their strategy for exposure assessment in order to avoid or at least reduce potentially large overestimation (with regards to concentration of some metrics) and mis-estimation (with regards to physical and chemical characteristics of particles) of exposure to air pollutants for people living in the back of buildings close to heavy trafficked roads.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.envpol.2016.02.033>.

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