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

Zauli Sajani, Stefano; Trentini, Arianna; Rovelli, Sabrina; Ricciardelli, Isabella; Marchesi, Stefano; MacCone, Claudio; Bacco, Dimitri; Ferrari, Silvia; Scotto, Fabiana; Zigola, Claudia; Cattaneo, Andrea; Cavallo, Domenico Maria; Lauriola, Paolo; Poluzzi, Vanes; Harrison, Roy

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1 **Is particulate air pollution at the front door a**  
2 **good proxy of residential exposure?**  
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7 **Stefano Zauli Sajani<sup>1</sup>□, Arianna Trentini<sup>2</sup>, Sabrina Rovelli,<sup>4</sup>**  
8 **Isabella Ricciardelli<sup>2</sup>, Stefano Marchesi,<sup>1</sup>**  
9 **Claudio Maccone<sup>2</sup>, Dimitri Bacco<sup>2</sup>**  
10 **Silvia Ferrari<sup>2</sup>, Fabiana Scotto<sup>2</sup>, Claudia Zigola<sup>3</sup>**  
11 **Andrea Cattaneo<sup>4</sup>, Domenico Maria Cavallo<sup>4</sup>,**  
12 **Paolo Lauriola<sup>1</sup>, Vanes Poluzzi<sup>2</sup> and Roy M. Harrison<sup>5</sup>**  
13

14  
15 **<sup>1</sup> Regional Centre for Environment and Health**  
16 **Arpae Emilia-Romagna, Via Begarelli, 13**  
17 **41121 Modena, Italy**  
18

19 **<sup>2</sup> Regional Centre for Urban Areas**  
20 **Arpae Emilia-Romagna, Via Rocchi, 19**  
21 **40138 Bologna, Italy**  
22

23 **<sup>3</sup> Provincial District of Ravenna**  
24 **Arpae Emilia-Romagna, Via Alberoni, 17/19**  
25 **48121 Ravenna, Italy**  
26

27 **<sup>4</sup> Department of Science and High Technology**  
28 **Università degli Studi dell'Insubria**  
29 **Via Valleggio 11**  
30 **22100 Como, Italy**  
31

32 **<sup>5</sup> Division of Environmental Health and Risk Management**  
33 **School of Geography, Earth & Environmental Sciences**  
34 **University of Birmingham**  
35 **Edgbaston, Birmingham, B15 2TT**  
36 **United Kingdom**

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□

1 **ABSTRACT**

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

23

1

2 **Capsule abstract:** Gradients in concentrations of several particle metrics and chemical components  
3 between the front and rear of a building are similar to those found between high and low traffic  
4 areas.

5

6 **Keywords:** Exposure, Traffic, Fine particles; Size distribution; Chemical components;  
7 Misclassification; Indoor

1       **1.       INTRODUCTION**

2       Air pollutants, and airborne particles in particular, pose significant risks to human health  
3       (REVIHAAP, 2013). A body of evidence has been accumulating over the last few decades on the  
4       effects of air pollution on cardiovascular and respiratory diseases, but there is still considerable  
5       uncertainty about the mechanisms of action linked to the health effects and about which physical  
6       and/or chemical characteristics of particulate matter (PM) are most important as determinants of  
7       health effects (Harrison and Yin, 2000; Kelly and Fussell, 2012).

8  
9       A key point in assessing the health effects of air pollution is contrasting exposure between people  
10      residing in different cities (Pope et al., 2009; Dockery et al., 1993) or different areas within the  
11      same city and its surroundings (Raaschou-Nielsen et al., 2013; Beelen et al. 2014). Differences in  
12      exposure for people residing in urban areas are mainly related to differences in proximity to traffic  
13      sources and the most recent and advanced epidemiological studies, especially those devoted to long  
14      term and traffic-related health effects (Hampel et al., 2015; Wang et al. 2014), assign exposure  
15      based on outdoor concentration of air pollutants measured or estimated at the front door. In this  
16      respect, Land Use Regression Models and Dispersion Models provide comparable performance (de  
17      Hoogh et al., 2014; Beelen et al., 2010) and have been demonstrated to be effective tools to improve  
18      exposure assessment compared to the use of data from fixed site monitoring stations. Nitrogen  
19      dioxide and particle concentration (usually ultrafine particle number or particles with aerodynamic  
20      diameter below 10  $\mu\text{m}$  –  $\text{PM}_{10}$  – or particles with aerodynamic diameter below 2.5  $\mu\text{m}$  –  $\text{PM}_{2.5}$ ) are  
21      the most common parameters used as air quality indicators.

22  
23      While a number of studies have investigated spatial variations of air pollutant concentrations  
24      between traffic and urban background sites (Boogard et al., 2011; Naser et al., 2008; Harrison et al.,  
25      2004) and in specific locations, such as building-free areas near highways (Patton et al., 2014; Zhu  
26      et al., 2002) or inside street canyons (Zhou et al. 2008), only very few studies have addressed

1 specifically the issue of the differences between the concentrations of air pollutants at the front and  
2 back of buildings next to busy streets (Weber et al., 2008; Hitchins et al., 2002). Weber et al. found  
3 differences in particle mass and number concentrations between a busy urban street canyon and an  
4 adjacent backyard using optical particle counters. Higher concentrations in the canyon of on  
5 average 30 % for  $PM_{10}$  and 22 % for  $PM_1$  were found within the street canyon. On the contrary  
6 Hitchins et al. found no significant gradients from the front to the rear of the building for  $PM_{2.5}$  and  
7 submicrometre particle number concentrations considering three low-rise buildings at a distance  
8 between 11 and 75 m from roads.

9  
10 The main goal of this paper is to investigate which particle metrics measured at the front door can  
11 be used as proxies of residential exposure. More specifically, we would like to investigate how large  
12 are the errors in assigning the same exposure to individuals residing in the same building near major  
13 roads. This is a key point in epidemiological studies because these individuals represent the very  
14 important subpopulation of highly exposed subjects. This work is part of a series of monitoring  
15 campaigns planned within the “Supersito” project (<http://www.arpa.emr.it/supersito>) aimed at  
16 assessing the variability of exposure within urban areas with a special emphasis on various PM  
17 metrics (Zauli Sajani et al., 2015). .

## 18 19 **2. METHODS**

### 20 **2.1 Study Design**

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

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

24 The second is the choice of monitoring uninhabited indoor environments. The main reason for this  
25 choice was related to the fact that many studies suggest that particles of outdoor and indoor origin  
26 have different physical and chemical characteristics (Brown et al., 2008) and probably can also

1 cause different health effects (Zhou et al., 2013; Ebel et al., 2005). In fact, indoor exposure to  
2 particulates comes from particles of outdoor origin on which additive contributions of indoor-  
3 generated particles arising from specific indoor sources superimpose (Urso et al., 2015, Fuller et al.,  
4 2013). It was assumed that mean within-city gradients of residential population exposure are  
5 primarily generated by exposure to air pollution of outdoor origin.

6  
7 We selected two indoor environments similar in terms of volume and building materials, with  
8 virtually identical Air Exchange Rates (AERs). AERs were controlled by installing in each indoor  
9 environment a mechanical system to force air to be exchanged between indoors and outdoors. The  
10 system consisted of a fan connected to an air pipe (length = 1.2 m) carrying the air to the centre of  
11 the room (at a height of 2 m). Increased indoor air pressure caused the flow to go outwards through  
12 a grid. The fan velocity in each room was regulated in order to obtain an estimated AER of  $0.4 \text{ h}^{-1}$  in  
13 each room, reflective of a typical level for residential environments (Cattaneo et al., 2011). Our  
14 earlier work has shown that this method is highly effective and does not cause significant loss of  
15 particles (Zauli Sajani et al., 2015).

16  
17 The measurements at the two sides of the building were conducted simultaneously indoors and  
18 outdoors (i.e. we had four simultaneous measurement sites). Figure 1 outlines the size of the  
19 building and the location of the monitoring sites. Figure 1S shows a map of the area and gives a  
20 bird's eye view of the surroundings of the monitoring sites. The study building was two-storeyed  
21 and located next to a street which surrounds the historical centre of Bologna, a 400,000-inhabitant  
22 city in northern Italy. Traffic and domestic heating during the cold season are the dominant air  
23 pollution sources in the area and cause high levels of air pollutants. In the period 2011-2013 the  
24 city-average annual concentration of  $\text{PM}_{2.5}$  was  $19.8 \mu\text{g}/\text{m}^3$  (average value derived from the two  
25 fixed site monitoring stations located in the urban area). The area near the monitoring sites carries a  
26 moderate volume of traffic, and the street next to the building is one of the busiest streets of the

1 entire municipal area with a traffic load of 31,000 vehicles (4–5% heavy duty vehicles) each  
2 working day. The building is located in a broad (20 m) two-way street canyon. No sources of  
3 particles were present in the inner courtyard.

4  
5 The indoor monitoring site at the front side (from now on “indoor front site”) was on the ground  
6 floor (street level). The volume of the room was 119 m<sup>3</sup> with a ceiling height of 3.5 m. The indoor  
7 environment at the back (from now on “indoor back site”) was on the ground floor as well. The  
8 volume of the room was 61 m<sup>3</sup> and the ceiling height was the same as that at the front side. The two  
9 indoor environments were completely separated with no common air exchange. For practical  
10 reasons the outdoor PM<sub>2.5</sub> monitoring site was located next to the building but at about 15 m from  
11 the indoor front site along the same street (Figure 1).

12  
13 Two monitoring campaigns were conducted in the period June–December 2013. Each monitoring  
14 campaign lasted 15 days: 1<sup>st</sup> campaign from 11 to 25 June (often referred to in the text as “hot  
15 period”), 2<sup>nd</sup> campaign (“cold period”) from 28 November to 13 December. Due to the availability  
16 of a unique filter for each day and measuring site, the chemical speciation was performed  
17 sequentially every three days for metals, ions, and carbon (Elemental Carbon and Organic Carbon).  
18 During cold periods elemental and organic carbon were measured on an 8 hour basis in order to  
19 avoid filter overload.

## 21 **2.2 Instrumentation and Monitoring Procedure**

22  
23 Four identical gravimetric samplers (Skypost PM, TCR TECORA Instruments, Corsico, Milan,  
24 Italy) were operated to measure indoor and outdoor daily PM<sub>2.5</sub> concentrations at the four sites  
25 (flow rate 2.3 m<sup>3</sup>h<sup>-1</sup>). Samples were collected on quartz fiber filters (Whatman, 47 mm diameter)  
26 and weighed following the procedure outlined in European Standard EN 12341:2014.



1  
2 PM<sub>2.5</sub> samples were analyzed for various chemical species. In this paper we present the findings of  
3 the chemical species having more than 50% of contemporary data above the limit of quantification  
4 (LOQ). LOQs for chemical components are reported in the Supplementary Information. Organic  
5 Carbon (OC) and Elemental Carbon (EC) were quantified by means of thermal-optical transmittance  
6 (Lab OC-EC Aerosol Analyzer, Sunset Laboratory Inc., Tigard, Oregon, USA) using the  
7 EUSAAR\_2 protocol (Cavalli et al., 2010). Inorganic ions were determined by extracting species in  
8 10 mL of ultrapure water. OC and EC values were summed to obtain TC. The extracts were filtered  
9 and analyzed by Ion Chromatography (Dionex ICS-1000 for anions and ICS-1100 for cations,  
10 Thermo Fischer Scientific Inc., USA). Metals were analyzed by Inductively Coupled Plasma –  
11 Mass Spectrometry (8800 ICP-MS, Agilent Technologies Inc., USA). Sample digestion with nitric  
12 acid and hydrogen peroxide in a microwave digestion apparatus and analysis were made according  
13 to European Standard EN 14902:2005, with a recovery efficiency over 85%  
14  
15 Two Fast Mobility Particle Sizers (FMPS model 3091; TSI, Shoreview, MN, USA) were used to  
16 measure particle size distributions and to estimate UltraFine Particle (UFP) concentrations. The  
17 FMPS was developed based on electrical aerosol spectrometer technology from Tartu University  
18 (Tammet et al. 2002). The FMPS spectrometer measured the size and number concentration of  
19 particles from 5.6 nm to 560 nm with 32 size bins every one second. Size bins below 13 nm were  
20 not included in the analysis because of the amount of data below the detection limit and also  
21 because of artifacts in the size distribution observed in other studies (Kaminski et al., 2013; Jeong et  
22 al., 2009). UFP concentrations were obtained summing the number of particles detected in the  
23 channels between 13 and 100 nm. Raw data were recorded every minute. Hourly and daily averages  
24 were derived from 1-minute data and used in the analyses.

25

1 Based on the availability of only two FMPS, indoor and outdoor size distributions were obtained  
2 by coupling each FMPS with a switching system (Mod 11sc200, Pneumoidraulica Engineering  
3 S.r.l., Vicenza, Italy). One FMPS-switching system couple was located at the indoor front site and  
4 the other at the indoor back site. The switching systems allowed for sampling from indoor and  
5 outdoor air, switching from one to the other within a time frame set by the user. A valve installed in  
6 the system could switch between sampling from the outdoor air, or from the indoor air. After the  
7 valve switched, there was a short time delay before the air from the sampled environment reached  
8 the instruments, which was the time the air travelled from the valve to the instruments. The system  
9 switched every 10 min between the indoor and outdoor measurements. In order to avoid the  
10 possibility of mixing of the outdoor and indoor air streams, the 2 min samples taken at the  
11 beginning of each 10 min period were deleted from the database. Particles were sampled indoor and  
12 outdoor through two 3/8 inches conductive silicon sampling tubes 2 m long. There was concern that  
13 this experimental arrangement for providing a forced input of aerosol might lead to depletion due to  
14 passage through the fan and pipe. This possibility was tested by experiments in which the particle  
15 number size distribution was measured at the inlet to the fan and outlet to the pipe by an FMPS  
16 system with rapid switching between the two sampling locations (upstream and downstream). The  
17 air inflow system was found to cause a minor loss of particles (Zauli Sajani et al., 2015).

18  
19 The Air Exchange Rate in each indoor site was estimated in two ways. Firstly, AERs were estimated  
20 based on the air inflow measured at the end of the pipes taking into account the volumes of the  
21 rooms. The air inflow was measured with a TESTO 417 Anemometer (Testo AG, Lenzkirch,  
22 Deutschland). The measurements were performed five times at each velocity of the fan (ten equally  
23 spaced values of velocity of fan in each room). The velocity of fan in each room was chosen in  
24 order to obtain an AER equal to  $0.4 \text{ h}^{-1}$ . Furthermore, in order to verify the estimated AERs,  
25 additional measurements based on the tracer gas-decay technique (ASTM Standard E741-95) were  
26 carried out. During these measurements, pure  $\text{CO}_2$  was released as tracer gas within each indoor

1 environment to obtain a uniform tracer concentration through the spaces being studied (ASHRAE,  
2 1997). The decay in CO<sub>2</sub> concentrations was continuously (sampling intervals of 1 min) monitored  
3 using non-dispersive infrared analyzers (GE sensing Telaire 7001, Goleta, CA, USA) provided with  
4 a battery-operated data logger (Hobo U12; Onset Computer Inc., Pocasset, MA, USA). A CO<sub>2</sub>  
5 analyzer was placed at each indoor and outdoor sampling site in order to monitor both the indoor  
6 gas decay - until the indoor baseline was reached - and the corresponding outdoor CO<sub>2</sub> levels.

7  
8 Meteorological outdoor data were obtained from the meteorological station belonging to the urban  
9 meteorological network of ARPAE Emilia-Romagna (“Bologna Urbana” station -  
10 [http://www.arpa.emr.it/sim/?osservazioni\\_e\\_dati/dati\\_stazioni\\_regionali](http://www.arpa.emr.it/sim/?osservazioni_e_dati/dati_stazioni_regionali)). The station is located  
11 close to the monitoring sites at an horizontal distance of about 30 m. The station is on the top of a  
12 building 25 metres above ground level and it measures hourly temperature, relative humidity, and  
13 wind speed and direction. Temperature is measured by means of a Pt100 RTD sensing element,  
14 while a capacitive sensor is used for humidity; wind speed and direction are measured using a three-  
15 cup anemometer and a wind vane respectively. Two digital thermo-hygrometers (Testo 175 H2,  
16 Testo AG, Lenzkirch, Deutschland) were used to measure temperature and relative humidity in the  
17 two indoor environments. Measurements were taken every hour.

## 20 **2.3 QA/QC and statistical analysis**

### 21 **2.3.1 QA/QC**

22 Agreements among gravimetric PM<sub>2.5</sub> measurements were checked in several intercomparison  
23 campaigns carried out in the years 2008-2012 before and during the “Supersito” project. Both  
24 correlation levels and tests for differences for slope and intercept of orthogonal regressions between  
25 co-located instruments were used (EC, 2010). Determination coefficients were always higher than  
26 0.972 (mean correlation 0.985). Typical errors (standard deviation of the differences between

1 samplers) were about  $2 \mu\text{g}/\text{m}^3$  and were quite similar among the various intercomparisons. The  
2 differences for slope (from identity) and intercepts (from zero) were usually not significant and not  
3 related to specific instruments. Consequently, no corrections were applied to  $\text{PM}_{2.5}$  data.

4 In the preliminary phase of the monitoring campaign we carried out four intercomparisons (one  
5 before and after each monitoring campaign - mean duration 2 days) between the two FMPS using  
6 the same methodology applied for  $\text{PM}_{2.5}$  samplers. No heteroscedasticity was detected and typical  
7 errors (standard deviation of the differences between UFP hourly data from the intercomparison  
8 campaigns) were  $580 \text{ \#/cm}^3$ . The determination coefficient was 0.976. No correction was applied to  
9 the data.

10

11 Quality control of  $\text{PM}_{2.5}$  mass and chemical composition data has been carried out based on  
12 residuals calculated by regression analysis between indoor and outdoor data. Data with residuals  
13 larger than three times the standard deviation of residuals were identified as anomalous. For FMPS  
14 data quality control the following procedure was used: a) applying a  $\log_{10}$  function on the UFP  
15 minute data; b) stratifying data in time slots of three hours (0-3, 3-6 etc) and calculating the  
16 summary statistics for each slot and campaign; c) classifying data as anomalous if they were higher  
17 than the mean plus three times the standard deviation for the corresponding campaign and time slot.  
18 Then we averaged non-anomalous data on an hourly and daily basis. Statistical data analysis was  
19 carried out using the R package (Version 3.0.1).

20

### 21 **2.3.2 Statistical analyses**

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

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### 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).

Figure 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<sup>-1</sup> in both indoor sites. These values were slightly lower than those estimated (0.4 h<sup>-1</sup>) based on air inflow at the end of the pipes and room volumes.

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## **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.

Figure 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 #/cm<sup>3</sup> while the highest value in the back site was 26,860 #/cm<sup>3</sup>. 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 (Diapouli et al., 2011) showing a spatial variability ranging from ratios of 1.8 to 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 km<sup>2</sup> 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).

1 The range of UFP indoor concentrations was much lower than outdoors, at both sites. As for  
2 outdoors, large gradients were found in the UFP indoor concentrations between the front and back  
3 (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  
4 season.

5  
6 The I/O ratio for UFP was higher in the back compared to the front of the building (0.5 vs 0.3) and  
7 remained almost constant over the two campaigns (Table 1). As shown in the following section,  
8 higher I/O ratio at the front side is due to the very effective loss of freshly nucleated particles in the  
9 indoor environment. The I/O ratios found in this study are in good agreement with that obtained in  
10 the previous SUPERSITO campaign (Zauli Sajani et al., 2015). Kearney et al. (2011) found median  
11 I/O ratios of hourly data ranging from 0.27 to 0.39. Diapouli et al. (2011) found an I/O ratio for  
12 particles in the 10-400 nm size range equal to 0.6 with AERs in the 0.5-1 h<sup>-1</sup> range, while I/O ratios  
13 between 0.3-0.4 were found in Erfurt (Germany) (Cyrus et al., 2004) and in other major European  
14 urban areas (Hoek et al., 2008). Meier et al. (2015) investigated I/O ratios for Particle Number  
15 Concentration (PNC) in four Swiss cities. The cities most similar to Bologna were Geneva and  
16 Lugano and the I/O ratios for homes without smoking influence were 0.52 (Geneva) and 0.70  
17 (Lugano). As expected, those values were a little bit higher than ours because of the presence of  
18 indoor sources.

19  
20 The Pearson correlation coefficient between daily outdoor UFP concentrations at the front and rear  
21 was equal to 0.84 (Figure 3). This value was slightly lower than in the previous SUPERSITO  
22 campaign and significantly higher than those reported in other studies focused on particle number  
23 concentrations (Puustinen et al., 2007). Correlations between indoor UFP concentrations were lower  
24 ( $R = 0.79$ ). Very similar correlation coefficients were found between indoor and outdoor UFP  
25 concentrations at the front and back sides of the building ( $R = 0.94$  and  $0.85$  respectively).

26

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

### 15 16 **3.2.2 Particle size distribution**

17 Figure 4 shows the mean indoor and outdoor size distributions at the four measuring sites for the  
18 two measurement periods. Multimodal distributions with sharp peaks at 20 and 30 nm in the  
19 concentrations were found at the outdoor front site. A second peak can occur at about 60-100 nm.  
20 This is typical of heavily trafficked sites, with the modes arising from the semi-volatile nucleation  
21 particles and solid graphitic particles respectively (Harrison et al., 2011).  
22 The presence of a bi (or tri)-modal distribution was also shown in previous studies (Morawska et  
23 al., 2008; Hussein et al., 2005) and is in line with knowledge of particle emissions and  
24 transformation. The 30 nm mode is due to the combination of freshly nucleated particles formed as  
25 the exhaust gases are diluted with ambient air and particles directly emitted by vehicles (Charron  
26 and Harrison, 2003). Nucleation mode particles are associated with the hot exhaust gases expelled  
27 from the tailpipe of a vehicle. These gases cool and condense to form large numbers of very small



1 particles in the air (Shi and Harrison, 1999). On-road dilution of the exhaust plume is very  
2 important in the generation of particles in the exhaust plume. These nucleation processes are  
3 favoured by low ambient temperatures and high relative humidity (Charron and Harrison, 2003)  
4 which are typical in the area during the cold season (Table 1S). In addition, the gaseous precursors  
5 condense or adsorb on to the surface of carbon particles in the accumulation mode.

6  
7 Upon entry into the building, not only would the nucleation mode fraction show a higher deposition  
8 velocity than the coarser graphitic mode (Riley et al., 2002), it would be subject in winter to  
9 evaporation at the higher indoor temperatures (Dall'Osto et al., 2011) and the hydrocarbon vapours  
10 released would tend to adsorb to indoor surfaces (Weschler and Nazaroff, 2008) and settled indoor  
11 dusts (Weschler and Nazaroff, 2010). Such processes would contribute to a relatively rapid loss of  
12 the nucleation mode of particles such that shown in the front site between outdoor and indoor and  
13 from front outdoor to back .

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

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

26

### 1 3.2.3 PM<sub>2.5</sub> mass

2 Indoor and outdoor PM<sub>2.5</sub> concentrations during the two monitoring campaigns are shown in Figure  
3 2 (b) and Table 1. Small but significant differences in PM<sub>2.5</sub> outdoor concentrations were found,  
4 with average PM<sub>2.5</sub> levels at the outdoor front site about 14% higher than those at the outdoor back  
5 site. The highest outdoor values for daily mean PM<sub>2.5</sub> concentrations were 68 µg/m<sup>3</sup> and 64 µg/m<sup>3</sup>  
6 for the front and back site, respectively. As expected, outdoor concentrations during the cold period  
7 were, on average, more than three times higher compared to summer, according to the typical PM  
8 seasonal trends in some European countries (Oeder et al., 2012).

9  
10 The outdoor PM<sub>2.5</sub> ratio between front and back levels was slightly higher than the ratio between  
11 traffic and residential sites found in similar monitoring campaigns conducted under the same  
12 SUPERSITO project (1.14 vs 1.06) (Zauli Sajani et al., 2015). The PM<sub>2.5</sub> spatial variability in our  
13 study was virtually equal to the mean within-city variability reported in the ESCAPE study, a very  
14 large epidemiological survey in Europe which included monitoring campaigns on air pollution  
15 spatial variability in urban areas (Eeftens et al., 2012). That survey reported a mean ratio between  
16 traffic sites and urban background sites equal to 1.14, with a quite broad range of values (0.96 –  
17 1.30). In particular, street and urban background ratios equal to 1.02 and 1.14 were found for  
18 Lugano and Turin, the closest ESCAPE cities to Bologna. Similar PM<sub>2.5</sub> gradients (from 1.0 to 1.3)  
19 were found in a study focused on the comparison between street and background locations within  
20 the same cities in the Netherlands (Boogaard et al., 2011).

21 Differently from UFPs, higher PM<sub>2.5</sub> ratios were found between front and back indoor  
22 concentrations (1.44 on average, 1.82 during the cold period and 1.31 during the hot period).

23 Mean indoor/outdoor (I/O) ratios for PM<sub>2.5</sub> during the cold period were equal to 0.57 at the front  
24 side and 0.46 at the rear. Differently from UFPs, higher values were found during the hot period  
25 with mean I/O ratios of 1.02 and 0.78 for the front and back side, respectively (Table 1). These  
26 results were in good agreement with previous studies in indoor settings (Chen et al., 2011;

1 Hanninen et al., 2004). Averaging I/O values for both building sides and measurements campaigns  
2 provided an I/O ratio equal to 0.71, a value within the range of the I/O ratios obtained in Swiss  
3 homes (Meier et al, 2015) . In particular, the homes without smoking influence provided median I/O  
4 ratios equal to 0.86 in Geneva and 0.65 in Lugano. Inter-campaign  $PM_{2.5}$  variations in the indoor  
5 environments were lower than outdoors. The ratios between the  $PM_{2.5}$  indoor concentrations  
6 measured during the cold and the hot period were 1.71 at the indoor front site and 2.38 at the  
7 indoor back site. The corresponding ratios for outdoor  $PM_{2.5}$  concentrations were markedly higher  
8 (3.02 and 3.91 at the front and back site, respectively).

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

#### 15 16 **3.2.4 Chemical composition of $PM_{2.5}$**

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

22  
23 Significant differences ( $p$ -value  $< 0.005$ ) were found between outdoor front and back average levels  
24 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).  
25 Large but not statistically significant were the outdoor trends for Fe and Sn (Table 2). The same  
26 patterns were confirmed indoors, with indoor differences always higher than outdoors. The largest

1  $I_{\text{front}}/I_{\text{back}}$  ratios were found for Fe and Sn, with values more than three times at the front door  
2 compared to the back side (Table 2).

3  
4 As reported in the literature, EC is an important component of diesel exhaust particulate matter  
5 (Suvendrini Lena et al., 2002; Shi et al., 2000). Mn and Sn are typical markers of vehicle emissions  
6 for fine PM (Marcazzan et al., 2001; Monaci et al., 2000). Fe, Mn and also Cu are related with  
7 brake wear emissions (Manoli et al., 2002; Gietl et al., 2010). Tyre wear, brake wear, vehicle  
8 component detachment and fluid leakage are known sources of Fe (Ball et al., 1991), as well as  
9 motor exhausts (Pant et al., 2013). Furthermore, traffic-induced road dust resuspension also plays  
10 an important role for some elements (Fe and Mn), including those originated from crustal sources.  
11 Although more marked for the coarse size fraction, this can be considered also a contributor to fine  
12 fraction aerosol (Manoli et al., 2002; Harrison et al., 2012). Götschi et al. (2005) found substantially  
13 elevated concentrations of Cu, Fe and Mn in PM<sub>2.5</sub> particles at a busy street location. A strong  
14 elevation in EC, organic compounds and Fe-rich dust was also reported elsewhere (Harrison et al.  
15 2004).

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

19 Large influence of traffic proximity on chemical composition of PM<sub>2.5</sub> was also found in the  
20 ESCAPE data, with relevant differences in concentrations between traffic and urban background  
21 sites especially for Fe and Zn (Tsai et al., 2015).

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

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

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

9  
10 Table 4 shows the Pearson correlation coefficients among the chemical components. Outdoor data  
11 were highly correlated. The correlation coefficients were all greater than 0.9 with the exception of  
12 Sn (0.70) and Fe (0.38), which further confirms in general the origin of these elements, closely  
13 related to the traffic source. The findings for OC, EC, ammonium, nitrates and sulfates were  
14 expected as high correlation levels between within-city outdoor concentrations of PM these  
15 constituents were found by Bell et al. (2011) and Naser et al. (2008).

16 Very high correlations were also found between indoor data, except for Sn (0.69). Correlation levels  
17 were also generally high between indoor and outdoor data. Low R values were found only for Fe  
18 (0.16) at the front side of the building and for Sn (0.45) at the rear. High correlations between  
19 indoor and outdoor levels of OC and EC were found by Sawant et al. (2004) in several schools in  
20 California . The I/O correlations for ammonium found by the same authors showed large variability  
21 in the different schools. Particles of outdoor origin can undergo substantial changes and may be lost  
22 to building walls during indoor penetration. A study investigating the transformation of ambient  
23 ammonium nitrate aerosols in indoor environments has shown that measured indoor concentrations  
24 were considerably lower than the values predicted based only on penetration and deposition losses  
25 (Lunden et al., 2003). This was attributed to the semi-volatility of ammonium nitrate, leading to

1 losses as nitric acid and ammonia vapours. This behavior was also highlighted in the previous  
2 SUPERSITO campaign (Zauli Sajani et al., 2015).

### 3 4 **3.3 Strengths and Weaknesses of the Study**

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

18 The choice of monitoring uninhabited indoor environments made possible the use of instruments  
19 such as instruments for PM<sub>2.5</sub> mass measurements using reference methods that are rarely used in  
20 inhabited house due to their size and noise emissions.

21 A possible weakness of the study is related to the choice of a unique building to be monitored .  
22 Other monitoring campaigns in different settings could be useful but we think that this study should  
23 be considered significantly more than a pilot study. As a matter of fact, the selected building is a  
24 quite common setting within urban areas even though pure canyon configuration with higher and  
25 continuous buildings could produce even higher front/back gradients compared to our findings. In  
26 conclusion we think that our study gives important insights about possible misclassification of

1 people residing next to busy streets. The choice of selecting AER to a typical value should be a plus  
2 with regards to the generalization of our findings.

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

8 It may also be questioned as to whether the air ventilation system could be a weakness of our study.  
9 This question was considered in detail in our earlier paper (Zauli Sajani et al., 2015). In brief, we  
10 think that the two identical simple systems installed to impose fixed and equal air exchange rates  
11 between sites should be considered a strength and not a weakness of the study. Firstly, this choice  
12 provided a good control on the air exchange rate in the two indoor environments. Secondly, the  
13 choice eliminated the effect of possibly different specific infiltration factors of each building  
14 envelope.

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

18

#### 19 **4. SUMMARY AND CONCLUSIONS**

20 This study focused on the variations of exposure to various particle metrics from the front to the  
21 back of a building located next to a trafficked street. In particular, both indoor and outdoor spatial  
22 gradients have been analysed in terms of PM<sub>2.5</sub> mass and chemical composition, size distribution  
23 and particle number concentrations. Large differences in the concentrations of UFP, tin, manganese,  
24 iron and elemental carbon were found both indoors and outdoors. Sizeable but less substantial were  
25 the spatial gradients for PM<sub>2.5</sub>. Significant differences were also found for the shape of particle size

1 distributions for outdoor particles, while indoor particles showed very similar distributions. Indoor  
2 front/back ratios are generally consistent in terms of direction but with remarkable differences in  
3 magnitudes between the different particle metrics. Indoor concentrations were much lower than  
4 outdoors for PM<sub>2.5</sub> mass and UFP. Taking into account the chemical components, the building  
5 environment was protective especially for nitrates, ammonium, potassium, sulfates, lead and  
6 cadmium underlining the different characteristics of indoor particulate matter compared to outdoor  
7 independently from the presence of indoor sources.

8 Our findings showed that the variability of exposure to air pollution of people living in the same  
9 building next to a busy street may be large i.e. some people could be erroneously classified as  
10 highly exposed. The front/back variability was comparable to that found in previous studies  
11 involving people residing in buildings in heavy and low traffic areas. Given that a common way to  
12 assess the risks due to exposure to air pollutants, and in particular to the risks associated with  
13 proximity to traffic sources, is by contrasting exposure levels within cities, and considering that the  
14 assessment of population exposure is usually based on residential outdoor concentrations estimated  
15 at the front-door, the impact of exposure misclassification could be important. Epidemiological  
16 studies should consider these findings when designing their strategy for exposure assessment in  
17 order to avoid or at least reduce potentially large overestimation (with regards to concentration of  
18 some metrics) and mis-estimation (with regards to physical and chemical characteristics of  
19 particles) of exposure to air pollutants for people living in the back of buildings close to heavy  
20 trafficked roads

21

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## 1 REFERENCES

- 2 ASHRAE, 1997. Evaluating Building IAQ and Ventilation with Indoor Carbon Dioxide. ASHRAE  
3 Transactions, Vol. 103, No. 2. Available online:  
4 <http://fire.nist.gov/bfrlpubs/build97/PDF/b97044.pdf>  
5
- 6 Ball, D., Hamilton, R., Harrison, R., 1991. The influence of highway-related pollutants on  
7 environmental quality. In: Hamilton R, Harrison R (Eds), Highway Pollution, Elsevier Science,  
8 New York, pp. 1-47.  
9
- 10 Beelen, R., Voogt, M., Duyzer, J., Zandveld, P., Hoek, G., 2010. Comparison of the performances  
11 of land use regression modelling and dispersion modelling in estimating small-scale variations in  
12 long-term air pollution concentrations in a Dutch urban area. *Atmospheric Environment* 44, 4614-  
13 4621.  
14
- 15 Beelen, R., Raaschou-Nielsen, O., Stafoggia, M., Andersen, Z.J., Weinmayr, G., Hoffmann, B.,  
16 Wolf, K., Samoli, E., Fischer, P., Nieuwenhuijsen, M., Vineis, P., Xun, W.W., Katsouyanni, K.,  
17 Dimakopoulou, K., Oudin, A., Forsberg, B., Modig, L., Havulinna, A.S., Lanki, T., Turunen, A.,  
18 Oftedal, B., Nystad, W., Nafstad, P., De Faire, U., Pedersen, N.L., Östenson, C.G., Fratiglioni, L.,  
19 Penell, J., Korek, M., Pershagen, G., Eriksen, K.T., Overvad, K., Ellermann, T., Eeftens, M.,  
20 Peeters, P.H., Meliefste, K., Wang, M., Bueno-de-Mesquita, B., Sugiri, D., Krämer, U., Heinrich, J.,  
21 de Hoogh, K., Key, T., Peters, A., Hampel, R., Concin, H., Nagel, G., Ineichen, A., Schaffner, E.,  
22 Probst-Hensch, N., Künzli, N., Schindler, C., Schikowski, T., Adam, M., Phuleria, H., Vilier, A.,  
23 Clavel-Chapelon, F., Declercq, C., Grioni, S., Krogh, V., Tsai, M.Y., Ricceri, F., Sacerdote, C.,  
24 Galassi, C., Migliore, E., Ranzi, A., Cesaroni, G., Badaloni, C., Forastiere, F., Tamayo, I., Amiano,  
25 P., Dorronsoro, M., Katsoulis, M., Trichopoulou, A., Brunekreef, B., Hoek, G., 2014. Effects of  
26 long-term exposure to air pollution on natural-cause mortality: an analysis of 22 European cohorts  
27 within the multicentre ESCAPE project. *Lancet*. 383, 785-795.  
28
- 29 Bell, M.L., Ebisu, K., Peng, R.D., 2011. Community-level spatial heterogeneity of chemical  
30 constituent levels of fine particulates and implications for epidemiological research. *Journal of*  
31 *Exposure Science and Environmental Epidemiology* 21, 372-384.  
32
- 33 Boogaard, H., Kos, G.P.A., Weijers E.P., Janssen, N.A.H., Fischer P.H., Van der Zee, S.C., De  
34 Hartog, J.J., Hoek, G., 2011. Contrast in air pollution components between major streets and  
35 background locations: Particulate matter mass, black carbon, elemental composition, nitrogen oxide  
36 and ultrafine particle number. *Atmospheric Environment* 45, 650-658.  
37
- 38 Brown KW, Sarnat JA, Suh HH, Coull BA, Spengler JD, Koutrakis P, 2008. Ambient site, home  
39 outdoor and home indoor particulate concentrations as proxies of personal exposures. *Journal of*  
40 *Environmental Monitoring* 10(9), 1041-1051.  
41
- 42 Cattaneo, A., Peruzzo, C., Garramone, G., Urso, P., Ruggeri, R., Carrer, P., Cavallo, D.M., 2011.  
43 Airborne particulate matter and gaseous air pollutants in residential structures in Lodi province,  
44 Italy. *Indoor Air* 21, 489-500.  
45
- 46 Cavalli, F., Viana, M., Yttri, K., Genberg, J., Putaud, J., 2010. Toward a standardised thermal-  
47 optical protocol for measuring atmospheric organic and elemental carbon: the EUSAAR protocol.  
48 *Atmospheric Measurement Techniques* 3, 79-89.  
49
- 50 Charron, A., Harrison, R.M., 2003. Primary particle formation from vehicle emissions during  
51 exhaust dilution in the roadside atmosphere. *Atmospheric Environment* 37, 4109-4119.

- 1  
2 Chen, C., Zhao, B., 2011. Review of relationship between indoor and outdoor particles: I/O ratio,  
3 infiltration factor and penetration factor [Rivista] // Atmospheric Environment 45, 275-288.  
4
- 5 Cyrus, J., Heinrich, J., Richter, K., Wölke, G., Wichmann, H.-E., 2000. Source and concentrations  
6 of indoor nitrogen dioxide in Hamburg (west Germany) and Erfurt (east Germany). The Science of  
7 the Total Environment 250, 51-62.  
8
- 9 Cyrus, J., Pitz, M., Bischof, W., Wichmann, H.E., Heinrich, J., 2004. Relationship between indoor  
10 and outdoor levels of fine particle mass, particle number concentrations and black smoke under  
11 different ventilation conditions. Journal of Exposure Analysis and Environmental Epidemiology  
12 14, 275-283.  
13
- 14 Dall'Osto, M., Thorpe, A., Beddows, D.C.S., Harrison, R.M., Barlow, J.F., Dunbar, T., Williams,  
15 P.I. and Coe, H., 2011. Remarkable dynamics of nanoparticles in the urban atmosphere.  
16 Atmospheric Chemistry and Physics 11, 6623-6637.  
17
- 18 Diapouli, E., Eleftheriadis, K., Karanasiou, A., Vratolis, S., 2011. Indoor and outdoor particle  
19 number and mass concentrations in Athens. Sources, sinks and variability of aerosol parameters.  
20 Aerosol and Air Quality Research 11, 632-642.  
21
- 22 Dockery, D.W., Pope III, A., Xu, X., Spengler, J.D., Ware, J.H., Fay, M.E., Ferris, B.G., Speizer,  
23 F.E., 1993. An association between air pollution and mortality in six U.S. cities. The New England  
24 Journal of Medicine 329, 1753-1759.  
25
- 26 Ebelt ST, Wilson WE, Brauer M., 2005. Exposure to ambient and nonambient components of  
27 particulate matter: a comparison of health effects. Epidemiology 16(3), 396-405.  
28
- 29 EC, 2010. Guide to the demonstration of equivalence of ambient air monitoring methods. Report  
30 by an EC Working Group on Guidance for the Demonstration of Equivalence. - 2010. EN 15267,  
31 n 3). URL: [http://europa.eu.int/comm/environment/air/pdf/equivalence\\_report3.pdf](http://europa.eu.int/comm/environment/air/pdf/equivalence_report3.pdf)  
32
- 33 Eeftens, M., Tsai, M.-Y., Ampe, C., Anwander, B., Beelen, R., Bellander, T., Cesaroni, G., Cirach,  
34 M., Cyrus, J., Hoogh, K. D., Nazelle, A. D., Vocht, F. D., Declercq, C., Dedele, A., Eriksen, K.,  
35 Galassi, C., Grazuleviciene, R., Grivas, G., Heinrich, J., Hoffmann, B., Iakovides, M., Ineichen, A.,  
36 Katsouyanni, K., Korek, M., Krämer, U., Kuhlbusch, T., Lanki, T., Madsen, C., Meliefste, K.,  
37 Mölter, A., Mosler, G., Nieuwenhuijsen, M., Oldenwening, M., Pennanen, A., Probst-Hensch, N.,  
38 Quass, U., Raaschou-Nielsen, O., Ranzi, A., Stephanou, E., Sugiri, D., Udvardy, O., Vaskövi, É.,  
39 Weinmayr, G., Brunekreef, B., and Hoek, G. , 2012. Spatial variation of PM<sub>2.5</sub>, PM<sub>10</sub>, PM<sub>2.5</sub>  
40 absorbance and PM<sub>coarse</sub> concentrations between and within 20 European study areas and the  
41 relationship with NO<sub>2</sub> - results of the ESCAPE project. Atmospheric Environment 62, 303-317.  
42
- 43 Fuller, W.A. 1987. Measurement Error Models. Wiley, New York.  
44
- 45 Fuller, C.H., Brugge, D., Williams, P.L., Mittleman, M.A., Lane, K., Durant, J.L., Spengler, J.D.,  
46 2013. Indoor and outdoor measurements of particle number concentration in near-highway homes.  
47 Journal of Exposure Science Environmental Epidemiology 23, 506-512.  
48
- 49 Gietl, J.K., Lawrence, R., Thorpe, A.J. and Harrison, R.M. Identification of brake wear particles  
50 and derivation of a quantitative tracer for brake dust at a major road. Atmos. Environ., 44, 141-146  
51 (2010).  
52

- 1 Götschi, T., Hazenkamp-von Arx, M.E., Heinrich, J., Bono, R., Burney, R., Forsberg, B., Jarvis, D.,  
2 Maldonadog, J., Norbäckh, D., Sterni, W.B., Sunyerj, J., Torénk, K., Verlatol, G., Villani, S.,  
3 Künzli, N., 2005. Elemental composition and reflectance of ambient fine particles at 21 European  
4 location. *Atmospheric Environment* 39, 5947-5958.
- 5
- 6 Hampel, R., Peters, A., Beelen, R., Brunekreef, B., Cyrys, J., de Faire, U., de Hoogh, K., Fuks, K.,  
7 Hoffmann, B., Hüls, A., Imboden, M., Jedynska, A., Kooter, I., Koenig, W., Künzli, N., Leander, K.,  
8 Magnusson, P., Männistö, S., Penell, J., Pershagen, G., Phuleria, H., Probst-Hensch, N., Pundt, N.,  
9 Schaffner, E., Schikowski, T., Sugiri, D., Tiittanen, P., Tsai, M.Y., Wang, M., Wolf, K., Lanki, T.,  
10 ESCAPE TRANSPHORM study groups. 2015. Long-term effects of elemental composition of  
11 particulate matter on inflammatory blood markers in European cohorts. *Environment International*  
12 82, 76-84.
- 13
- 14 Hanninen, O.O., Lebet, E., Ilacqua, V., Katsouyanni, K., Kunzli, N., Sram, R.J., Jantunen, M.,  
15 2004. Infiltration of ambient PM<sub>2.5</sub> and levels of indoor generated non-ETS PM<sub>2.5</sub> in residences of  
16 four European cities. *Atmospheric Environment* 38, 6411-6423.
- 17
- 18 Harrison, R.M., Jones, A., Gietl, J., Yin, J., Green, D., 2012. Estimation of the contribution of brake  
19 dust, tire wear and resuspension to nonexhaust traffic particles derived from atmospheric  
20 measurements. *Environmental Science & Technology* 46, 6523-6529.
- 21
- 22 Harrison, R.M., Beddows, D.C.S. and Dall'Osto, M., 2011. PMF analysis of wide-range particle  
23 size spectra collected on a major highway. *Environmental Science and Technology* 45, 5522-5528.
- 24
- 25 Harrison, R.M., Jones, A.M., Lawrence, R.G., 2004. Major component composition of PM<sub>10</sub> and  
26 PM<sub>2.5</sub> from roadside and urban background sites. *Atmospheric Environment* 38, 4531-4538.
- 27
- 28 Harrison, R.M., J. Yin, 2000. Particulate matter in the atmosphere: Which particle properties are  
29 important for its effects on health? *Science of the Total Environment* 249, 85-101.
- 30
- 31 Harrison, R.M., Leung, P.-L., Somervaille, L., Smith, R., Gilman, E., 1999. Analysis of incidence  
32 of childhood cancer in the West Midlands of the United Kingdom in relation to proximity to main  
33 roads and petrol stations. *Occupational & Environmental Medicine* 56, 774-780.
- 34
- 35 Hitchins, J., Morawska, L., Gilbert, D., Jamriska, M. 2002. Dispersion of particles from vehicle  
36 emissions around high- and low-rise buildings. *Indoor Air* 12: 64-71.
- 37
- 38 Hoek, G., Beelen, R., de Hoog, K., Viennau, D., Gulliver, J., Fisher, P., Briggs, D., 2008. A review  
39 of land-use regression models to asses spatial variation of outdoor air pollution. *Atmospheric*  
40 *Environment* 42, 7561-7578.
- 41
- 42 Hoek, G., Ko,s G., Harrison, R.M., de Hartog, J., Meliefste, K., ten Brink, H., Katsouyanni, K.,  
43 Karakatsani, A., Lianou, M., Kotronarou, A., Kavouras, I., Pekkanen, J., Vallius, M., Kulmala, M.,  
44 Puustine,n A., Thomas, S., Meddings, C., Ayres, J., van Wijnen, J., Hameri, K., 2008. Indoor-  
45 outdoor relationships of particle number and mass in four European cities. *Atmospheric*  
46 *Environment* 42, 156-169.
- 47
- 48 de Hoogh, K., Korek, M., Vienneau, D., Keuken, M., Kukkonen, J., Nieuwenhuijsen, M.J.,  
49 Badaloni, C., Beelen, R., Bolignano, A., Cesaroni, G., Pradas, M.C., Cyrys, J., Douros, J., Eeftens,  
50 M., Forastiere, F., Forsberg, B., Fuks, K., Gehring, U., Gryparis, A., Gulliver, J., Hansell, A.L.,  
51 Hoffmann, B., Johansson, C., Jonkers, S., Kangas, L., Katsouyanni, K., Künzli, N., Lanki, T.,  
52 Memmesheimer, M., Moussiopoulos, N., Modig, L., Pershagen, G., Probst-Hensch, N., Schindler,

- 1 C., Schikowski, T., Sugiri, D., Teixidó, O., Tsai, M.Y., Yli-Tuomi, T., Brunekreef, B., Hoek, G.,  
2 Bellander, T., 2014. Comparing land use regression and dispersion modelling to assess residential  
3 exposure to ambient air pollution for epidemiological studies. *Environment International* 73, 382-  
4 392.
- 5
- 6 Hussein, T., Hameri, K., Aalto, P., Asmi, A., Kakko, L., Kulmala, M., 2004. Particle size  
7 characterization and indoor-to-outdoor relationship of atmospheric aerosol in Helsinki.  
8 *Scandinavian Journal of Work, Environment and Health* 30, 54-62.
- 9
- 10 Hussein, T., Hameri, K., Aalto, P.P., Paatero, P., Kulmala, M., 2005. Modal structure and spatial-  
11 temporal variations of urban and suburban aerosol in Helsinki - Finland. *Atmospheric Environment*  
12 39, 1655-1668.
- 13
- 14 Jeong, C.H., Evans, G., 200. Inter-comparison of a fast mobility particle sizer and a scanning  
15 mobility particle sizer incorporating an ultrafine water-based condensation particle counter.  
16 *Aerosol Science and Technology* 43, 364-373.
- 17
- 18 Kaminski, H., Kuhlbusch, T.A.J., Rath, S., Gotz, U., Sprenger, M., Wels, D., Polloczek, J.,  
19 Bachmann, V., Dziurowitz, N., Kiesling, H.-J., Schwiegelshohn, A., Monz, C., Dahmann, D.,  
20 Asbach, C., 2013. Comparability of mobility particle sizers and diffusion chargers. *Journal of*  
21 *Aerosol Science* 57, 156-178.
- 22
- 23 Kearney, J., Wallace, L., MacNeill, M., Xu, X., VanRyswyk, K., You, H., Kulka, R., Wheeler A.J.,  
24 2011, Residential indoor and outdoor ultrafine particles in Windsor, Ontario. *Atmospheric*  
25 *Environment* 45, 7583-7593.
- 26
- 27 Kelly, F.J., Fussell, J.C., 2012. Size, source and chemical composition as determinants of toxicity  
28 attributable to ambient particulate matter. *Atmospheric Environment* 60, 504-526.
- 29
- 30 Lunden, M.M., Revzana, K.L., Fischer, M.L., Thatcher, T.L., Littlejohn, D., Hering, S.V., Brown,  
31 N.J., 2003, The transformation of outdoor ammonium nitrate aerosols in the indoor environment.  
32 *Atmospheric Environment* 37, 5633-5644.
- 33
- 34 Manoli, E., Voutsas, D., Samara, C., 2002. Chemical characterization and source identification/  
35 apportionment of fine and coarse air particles in Thessaloniki, Greece. *Atmospheric Environment*  
36 36, 949-961.
- 37
- 38 Marcazzan, G.M., Vaccaro, S., Valli, G., Vecchi, R., 2001. Characterization of PM<sub>10</sub> and PM<sub>2.5</sub>  
39 particulate matter in the ambient air of Milan (Italy). *Atmospheric Environment* 35, 4639-4650.
- 40
- 41 Meier, R., Eeftens, M., Phuleria, H.C., Ineichen, A., Corradi, E., Davey, M., Fierz, M., Ducret-Stich,  
42 R.E., Aguilera, I., Schindler, C., Rochat, T., Probst-Hensch, N., Tsai, M.Y., Künzli, N., 2015.  
43 Differences in indoor versus outdoor concentrations of ultrafine particles, PM<sub>2.5</sub>, PM<sub>absorbance</sub>  
44 and NO<sub>2</sub> in Swiss homes. *Journal of Exposure Science and Environmental Epidemiology* 25, 499-  
45 505.
- 46
- 47 Monaci, F., Moni, F., Lanciotti, E., Grechi, D., Bargagli, R., 2000. Biomonitoring of airborne  
48 metals in urban environments: new tracers of vehicle emission, in place of lead. *Environmental*  
49 *Pollution* 107, 321-327.
- 50

- 1 Moore, K., Krudysz, M., Pakbin, P. Hudda, N., Sioutas, C., 2009. Intra-community variability in  
2 total particulate number concentrations in the San Pedro Harbor area (Los Angeles, California).  
3 *Aerosol Science & Technology* 43, 587-603.
- 4
- 5 Morawska, L., Ristovski, Z., Jayaratne, E.R., Keogh, D.U., Ling, X., 2008. Ambient nano and  
6 ultrafine particles from motor vehicle emissions: Characteristics, ambient processing and  
7 implications on human exposure. *Atmospheric Environment* 42, 8113-8138.
- 8
- 9 Naser, T.M. Yoshimura, Y., Sekiguchi, K., Wang, Q., Sakamoto, K., 2008. Chemical composition of  
10 PM<sub>2.5</sub> and PM<sub>10</sub> and associated Polycyclic Aromatic Hydrocarbons at a roadside and urban  
11 background area in Saitama, Japan. *Asian Journal of Atmospheric Environment* 2, 99-101.
- 12
- 13 Oeder, S., Dietrich, S., Weichenmeir, I., Schober, W., Pusch, G., Jörres, R.A., Schierl, R., Nowak,  
14 D., Fromme, H., Behrendt, H., Buters, J.T.M., 2012. Toxicity and elemental composition of  
15 particulate matter from outdoor and indoor air of elementary schools in Munich, Germany. *Indoor*  
16 *Air* 22, 148-158.
- 17
- 18 Pant, P., Harrison, R.M., 2013. Estimation of the contribution of road traffic emissions to  
19 particulate matter concentrations from field measurements: a review. *Atmospheric Environment* 77,  
20 89-97.
- 21
- 22 Patton, A.P., Perkins, J., Zamore, W., Levy, J.I., Brugge, D., Durant, J.L., 2014. Spatial and  
23 temporal differences in traffic-related air pollution in three urban neighborhoods near an interstate  
24 highway. *Atmospheric Environment* 99, 309-321.
- 25
- 26 Pope CA 3rd, Ezzati M, Dockery DW., 2009. Fine-particulate air pollution and life expectancy in  
27 the United States. *New England Journal Medicine* 360:376-86.
- 28
- 29 Puustinen, A., Hämeri, K., Pekkanen, J., Kulmala, M., de Hartog, J., Meliefste, K., ten Brink, H.,  
30 Kos, G., Katsouyanni, K., Karakatsani, A., Kotronarou, A., Kavouras, I., Meddings, C., Thomas, S.,  
31 Harrison, R., Ayres, J., van der Zee, S., Hoek, G., 2007. Spatial variation of particle number and  
32 mass over four European cities. *Atmospheric Environment* 41, 6622-6636.
- 33
- 34 Ragettli, M.S., Ducret-Stich, R.E., Foraster, M., Morelli, X., Aguilera, I., Basagaña, X., Corradi, E.,  
35 Ineichen, A., Tsai, M.Y., Probst-Hensch, N., Rivera, M., Slama, R., Künzli, N., Phuleria, H.C. 2014.  
36 Spatio-temporal variation of urban ultrafine particle number concentrations. *Atmospheric*  
37 *Environment* 96, 275-283.
- 38
- 39
- 40 Raaschou-Nielsen O, Andersen ZJ, Beelen R, Samoli E, Stafoggia M, Weinmayr G, Hoffmann B,  
41 Fischer P, Nieuwenhuijsen MJ, Brunekreef B, Xun WW, Katsouyanni K, Dimakopoulou K,  
42 Sommar J, Forsberg B, Modig L, Oudin A, Oftedal B, Schwarze PE, Nafstad P, De Faire U,  
43 Pedersen NL, Ostenson CG, Fratiglioni L, Penell J, Korek M, Pershagen G, Eriksen KT, Sørensen  
44 M, Tjønneland A, Ellermann T, Eeftens M, Peeters PH, Meliefste K, Wang M, Bueno-de-Mesquita  
45 B, Key TJ, de Hoogh K, Concini H, Nagel G, Vilier A, Grioni S, Krogh V, Tsai MY, Ricceri F,  
46 Sacerdote C, Galassi C, Migliore E, Ranzi A, Cesaroni G, Badaloni C, Forastiere F, Tamayo I,  
47 Amiano P, Dorronsoro M, Trichopoulos A, Bamia C, Vineis P, Hoek G. 2013. Air pollution and  
48 lung cancer incidence in 17 European cohorts: prospective analyses from the European Study of  
49 Cohorts for Air Pollution Effects (ESCAPE). *Lancet Oncology* 14, 813-822.
- 50
- 51 REVIHAAP, 2013. Review of evidence on health aspects of air pollution - REVIHAAP Project.  
52 Technical Report, World Health Organization, Regional Office for Europe, Copenhagen.

1  
2 Riley, Q.J., Mckone, T.E., Lai, A.C.K., Nazaroff, W.W., 2002. Indoor particulate matter of outdoor  
3 origin: importance of size dependent removal mechanisms. *Environmental Science & Technology*,  
4 36, 200-207.  
5  
6 Rivera, M., Basagana, X., Aguilera, I., Agis, D., Bouso, L., Foraster, M., Medina-Ramón, M., Pey,  
7 J., Künzli, N., Hoek, G., 2012. Spatial distribution of ultrafine particles in urban settings: a land use  
8 regression model. *Atmospheric Environment* 54, 657-666.  
9  
10 Ruths, M., von Bismarck-Osten, C., Weber, S. 2014. Measuring and modelling the local-scale  
11 spatio-temporal variation of urban particle number size distributions and black carbon. *Atmospheric*  
12 *Environment* 96, 37-49.  
13  
14 Sawant, A.A., Na, K., Zhu, X., Cocker, K., Butt, S., Song, C., Cocker III, D.R., 2004.  
15 Characterization of PM<sub>2.5</sub> and selected gas-phase compounds at multiple indoor and outdoor sites in  
16 Mira Loma, California. *Atmospheric Environment* 38, 6269-6278.  
17  
18 Shi, J.P., Mark, D., Harrison, R.M., 2000. Characterization of particles from a current technology  
19 heavy-duty diesel engine. *Environmental Science & Technology* 34, 748-755.  
20  
21 Shi, J.P., Harrison R.M., 1999. Investigation of ultrafine particle formation during diesel exhaust  
22 dilution. *Environmental Science & Technology* 33, 3730-3736.  
23  
24 Suvendrini Lena, T., Ochieng, V., Carter, M., Holguin-Veras, J., Kinney, P.L., 2002. Elemental  
25 carbon and PM<sub>2.5</sub> levels in a urban community heavily impacted by truck traffic. *Environmental*  
26 *Health Perspectives* 110, 1009-1015.  
27  
28 Tammet, H., Mirme, A., Tamm, E., 2002. Electrical aerosol spectrometer of Tartu University.  
29 *Atmospheric Research* 62, 315-324.  
30  
31 Tsai, M.Y., Hoek, G., Eeftens, M., de Hoogh, K., Beelen, R., Beregszászi, T., Cesaroni, G., Cirach,  
32 M., Cyrus, J., De Nazelle, A., de Vocht, F., Ducret-Stich, R., Eriksen, K., Galassi, C.,  
33 Gražulevičienė, R., Gražulevičius, T., Grivas, G., Gryparis, A., Heinrich, J., Hoffmann, B.,  
34 Iakovides, M., Keuken, M., Krämer, U., Künzli, N., Lanki, T., Madsen, C., Meliefste, K., Merritt,  
35 A.S., Mölter, A., Mosler, G., Nieuwenhuijsen, M.J., Pershagen, G., Phuleria, H., Quass, U., Ranzi,  
36 A., Schaffner, E., Sokhi, R., Stempfelet, M., Stephanou, E., Sugiri, D., Taimisto, P., Tewis, M.,  
37 Udvardy, O., Wang, M., Brunekreef, B., 2015. Spatial variation of PM elemental composition  
38 between and within 20 European study areas--Results of the ESCAPE project. *Environment*  
39 *International* 84, 181-192.  
40  
41 Urso, P., Cattaneo, A., Garramone, G., Peruzzo, C., Cavallo, D.M., Carrer, P. Identification of  
42 particulate matter determinants in residential homes. *Building and Environment* 86, 61-69.  
43  
44 Wang, M., Beelen, R., Stafoggia, M., Raaschou-Nielsen, O., Andersen, Z.J., Hoffmann, B., Fischer,  
45 P., Houthuijs, D., Nieuwenhuijsen, M., Weinmayr, G., Vineis, P., Xun, W.W., Dimakopoulou, K.,  
46 Samoli, E., Laatikainen, T., Lanki, T., Turunen, A.W., Oftedal, B., Schwarze, P., Aamodt, G., Penell,  
47 J., De Faire, U., Korek, M., Leander, K., Pershagen, G., Pedersen, N.L., Östenson, C.G., Fratiglioni,  
48 L., Eriksen, K.T., Sørensen, M., Tjønneland, A., Bueno-de-Mesquita, B., Eeftens, M., Bots, M.L.,  
49 Meliefste, K., Krämer, U., Heinrich, J., Sugiri, D., Key, T., de Hoogh, K., Wolf, K., Peters, A.,  
50 Cyrus, J., Jaensch, A., Concin, H., Nagel, G., Tsai, M.Y., Phuleria, H., Ineichen, A., Künzli, N.,  
51 Probst-Hensch, N., Schaffner, E., Vilier, A., Clavel-Chapelon, F., Declerq, C., Ricceri, F., Sacerdote,  
52 C., Marcon, A., Galassi, C., Migliore, E., Ranzi, A., Cesaroni, G., Badaloni, C., Forastiere, F.,

1 Katsoulis, M., Trichopoulou, A., Keuken, M., Jedynska, A., Kooter, I.M., Kukkonen, J., Sokhi, R.S.,  
2 Brunekreef, B., Katsouyanni, K., Hoek, G., 2014. Long-term exposure to elemental constituents of  
3 particulate matter and cardiovascular mortality in 19 European cohorts: results from the ESCAPE  
4 and TRANSPHORM projects. *Environment International* 66, 97-106.  
5  
6 Weber, S., Weber, K., 2008. Coupling of urban street canyon and backyard particle concentrations.  
7 *Meteorologische Zeitschrift* 17, 251-261.  
8  
9 Weschler, C.J. and Nazaroff, W.W., 2008. Semivolatile organic compounds in indoor environments.  
10 *Atmospheric Environment* 42, 9018-9040.  
11  
12 Weschler, C.J. and Nazaroff, W.W., 2010. SVOC partitioning between the gas phase and settled  
13 dust indoors. *Atmospheric Environment* 44, 3609-3620.  
14  
15 Zauli Sajani, S., Ricciardelli, I., Trentini, A., Bacco, D., Maccone, C., Castellazzi, S., Lauriola, P.,  
16 Poluzzi, V., Harrison, R.M., 2015. Spatial and indoor/outdoor gradients in urban concentrations of  
17 ultrafine particles and PM<sub>2.5</sub> mass and chemical components. *Atmospheric Environment* 103, 307-  
18 320.  
19  
20 Zhu Y, Hinds WC, Kim S, Sioutas C., 2002. Concentration and size distribution of ultrafine  
21 particles near a major highway. *Journal of the Air and Waste Management Association*. 52, 1032-  
22 1042.  
23 Zhu, Y. , Hinds, W.C., Krudysz, M., Kuhn, T., Froines, J., Sioutas, C., 2005. Penetration of freeway  
24 ultrafine particles into indoor environments. *Journal of Aerosol Science* 36, 303–322.  
25  
26 Zhou, Y., Levy, J.I., 2008. The impact of urban street canyons on population exposure to traffic-  
27 related primary pollutants. *Atmospheric Environment* 42, 3087–3098.  
28  
29 Zhou, Y., Levy, J.I., 2013. Investigating the geographical heterogeneity in PM 10-mortality  
30 associations in the China Air Pollution and Health Effects Study (CAPES): A potential role of  
31 indoor exposure to PM 10 of outdoor origin. *Atmospheric Environment* 75, 217-223.  
32



1 **TABLE LEGENDS**

2

3 **Table 1:** Summary statistics of PM<sub>2.5</sub> mass ( $\mu\text{g m}^{-3}$ ) and UFP number ( $\text{cm}^{-3}$ ) during the  
4 monitoring campaigns.

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6 **Table 2:** Front-back ratio for different particle metrics considering both indoor and outdoor  
7 concentrations. Table is ordered by outdoor gradients. P-value from paired t-test are also  
8 reported.

9

10 **Table 3:** Descriptive statistics and Indoor/Outdoor ratio (I/O) for different chemical components.  
11 OC, EC, TC, NH<sub>4</sub>, NO<sub>3</sub>, SO<sub>4</sub>, K are expressed in  $\mu\text{g/m}^3$  while the other elements in  
12  $\text{ng/m}^3$ .

13

14 **Table 4:** Pearson correlation coefficients (R) and regression lines for different chemical species.  
15 Intercepts for OC, EC, TC, NH<sub>4</sub>, NO<sub>3</sub>, SO<sub>4</sub>, K are expressed in  $\mu\text{g/m}^3$ ; intercepts for the  
16 other elements are expressed in  $\text{ng/m}^3$ .

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22 **FIGURE LEGENDS**

23

24 **Figure 1:** Schematic diagram of the building and location of the sampling points.

25

26 **Figure 2:** Average value and standard error of PM<sub>2.5</sub> (a) and UFP (b) during the two monitoring  
27 campaigns.

28

29 **Figure 3:** Scatter plots for daily PM<sub>2.5</sub> mass and UFP number concentrations.

30

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

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Table 1. Summary statistics of PM<sub>2.5</sub> mass ( $\mu\text{g m}^{-3}$ ) and UFP number ( $\text{cm}^{-3}$ ) 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)	
<b>PM<sub>2.5</sub> (24-h data)</b>										
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
<b>UFP (1-h data)</b>										
All data	652	25,358 (2118 - 120931)	652	7,625 (1472 - 21142)	0.39	652	7,444 (647 - 26860)	652	3,544 (213 - 8854)	0.55
Hot period	353	15,502 (2118 - 38367)	353	5,109 (1472 - 10773)	0.39	353	6,169 (647 - 24308)	353	2,932 (213 - 7901)	0.52
Cold period	299	36,994 (3095 - 120931)	299	10,595 (2854 - 21142)	0.38	299	8,948 (1781 - 26860)	299	4,266 (1730 - 8854)	0.58

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

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Table 2. Gradients of concentrations for different pollutants comparing both indoor and outdoor concentrations. Table is ordered by outdoor gradients.

	Outdoor		Indoor	
	Front / Back	p-value	Front / Back	p-value
UFP	3.4	< 0.001*	2.2	< 0.001*
PM <sub>2.5</sub>	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 <sub>3</sub> )	1.1	0.073	1.3	0.086
Sulfates (SO <sub>4</sub> )	1.0	0.184	1.0	0.889
Ammonium (NH <sub>4</sub> )	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*

Table 3. Descriptive statistics and I/O ratio for different chemical components. OC, EC, TC, NH<sub>4</sub>, NO<sub>3</sub>, SO<sub>4</sub>, K are expressed in µg/m<sup>3</sup> while the other elements in ng/m<sup>3</sup>.

	Number of valid data *	Front			Back		
		Outdoor mean (min-max)	Indoor mean (min-max)	I/O mean	Outdoor mean	Indoor mean	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 <sub>4</sub> )	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 <sub>3</sub> )	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 <sub>4</sub> )	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

\* Number of data simultaneously above LOQ in all monitoring sites

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Table 4. Pearson correlation coefficients (R) and orthogonal regression lines for different chemical species. Intercepts for OC, EC, TC, NH<sub>4</sub>, NO<sub>3</sub>, SO<sub>4</sub>, K are expressed in µg/m<sup>3</sup>; intercepts for the other elements are expressed in ng/m<sup>3</sup>.

	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 <sub>4</sub> )	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 <sub>3</sub> )	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 <sub>4</sub> )	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	1.08	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

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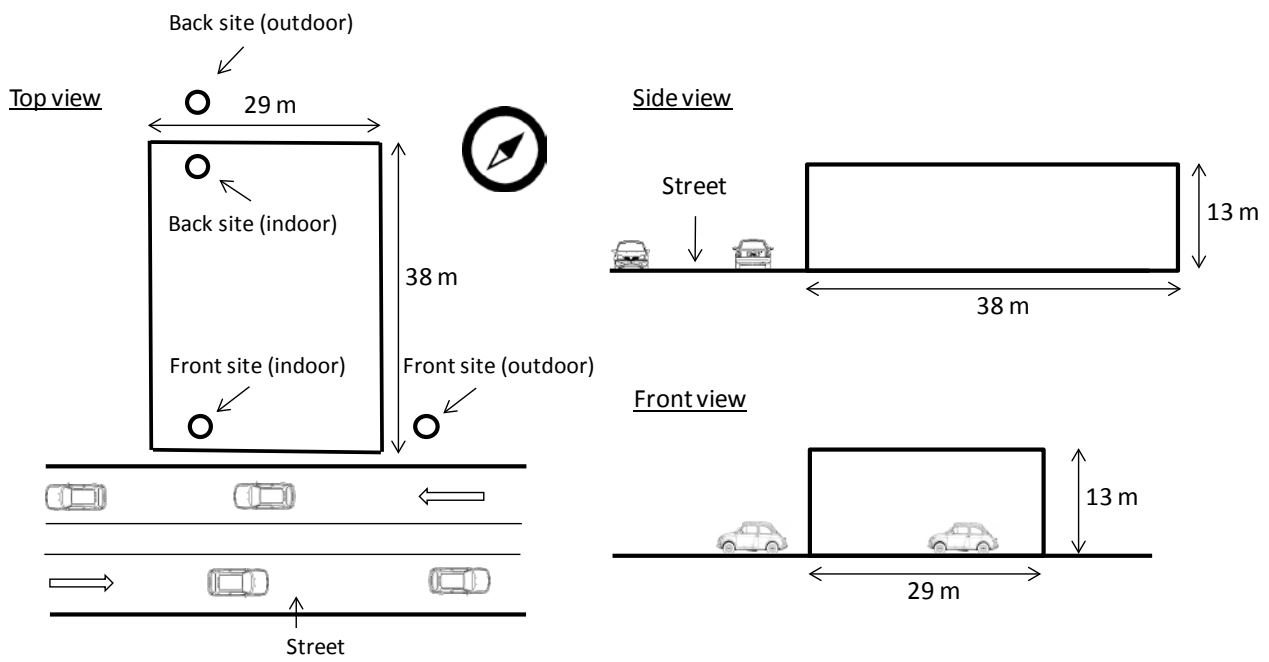
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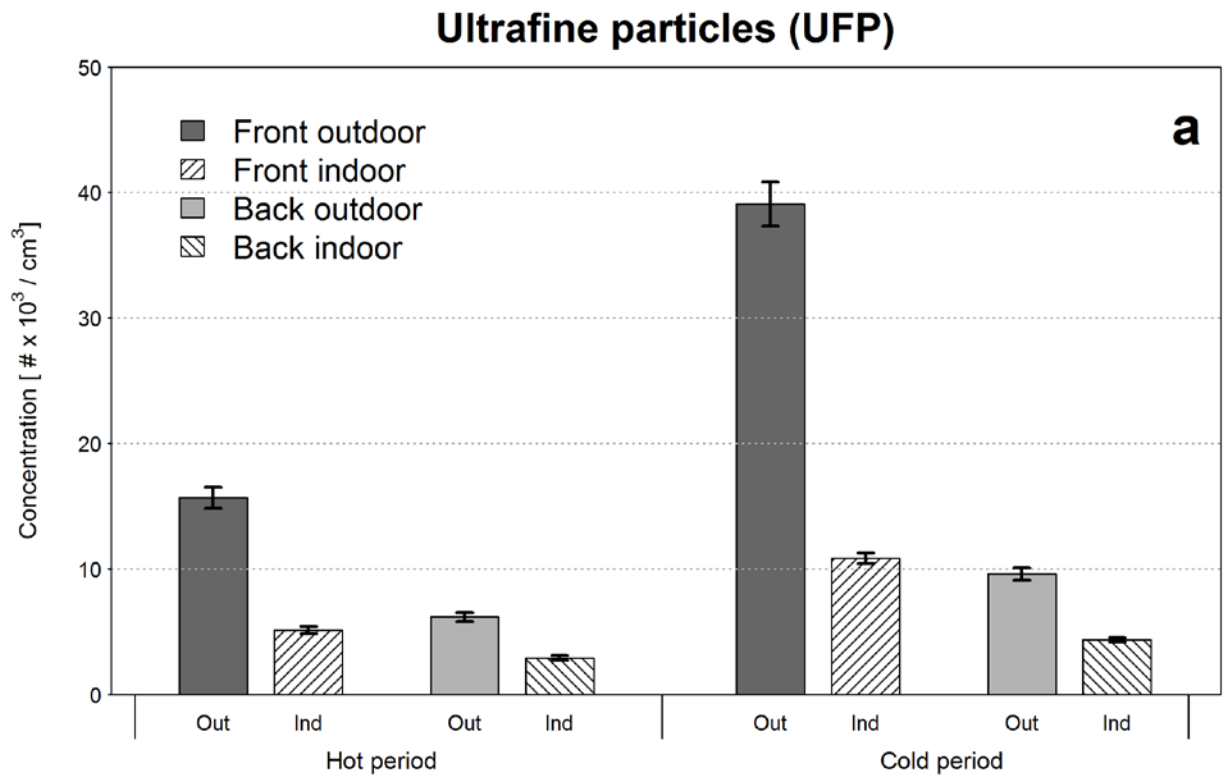


3

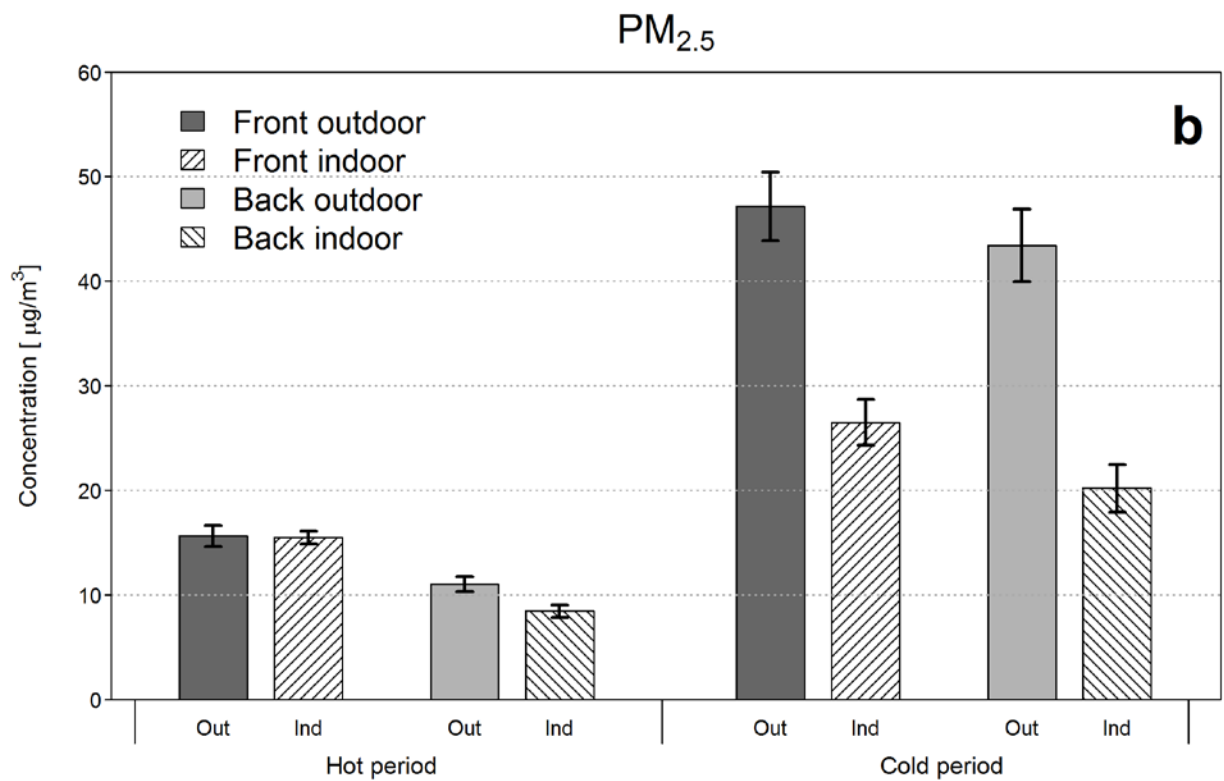
4 Figure 1. Schematic diagram of the building and location of the sampling points.

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3 Figure 2. Average value and standard error of PM<sub>2.5</sub> (a) and UFP (b) during the two monitoring  
 4 campaigns.

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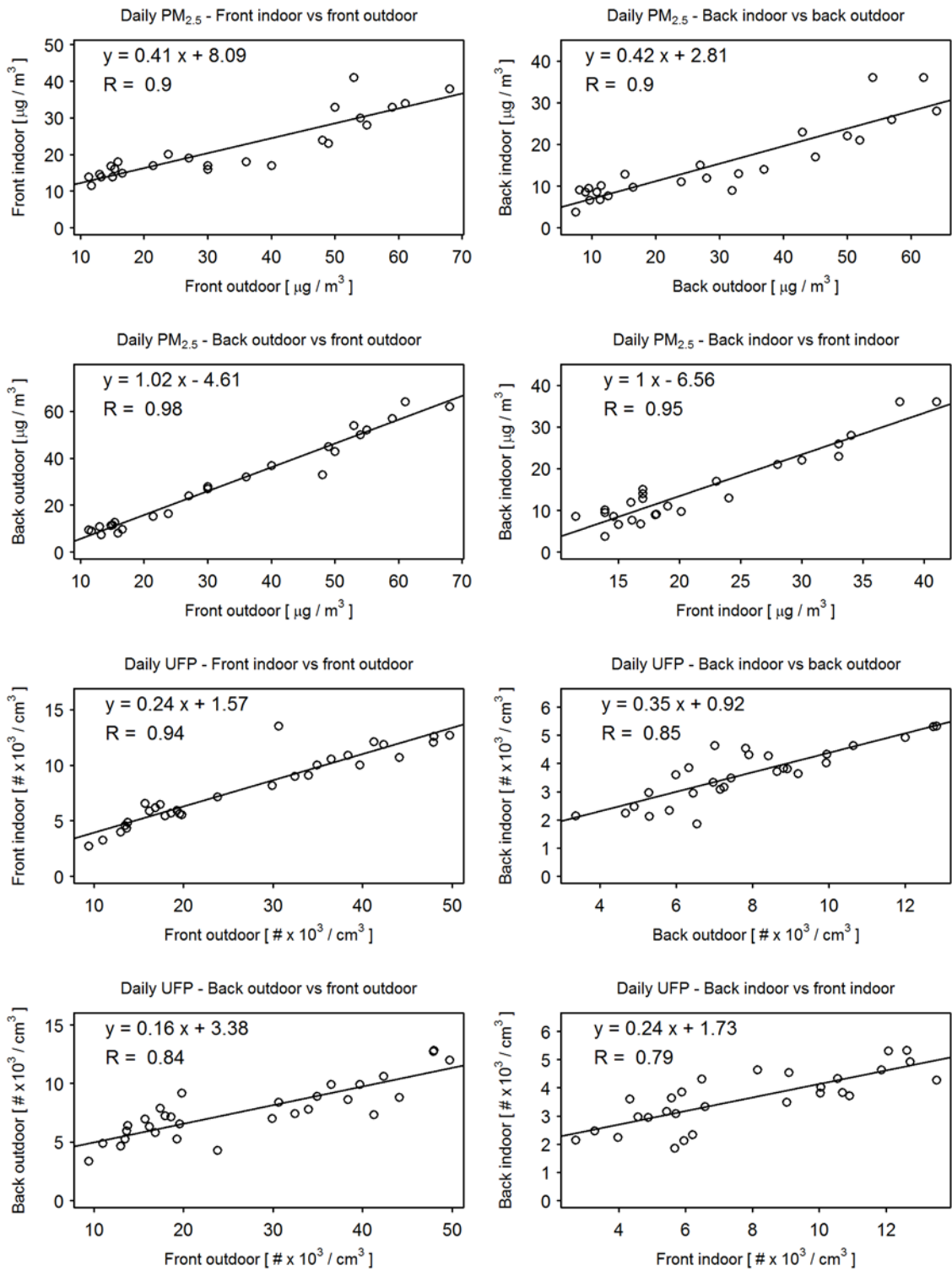
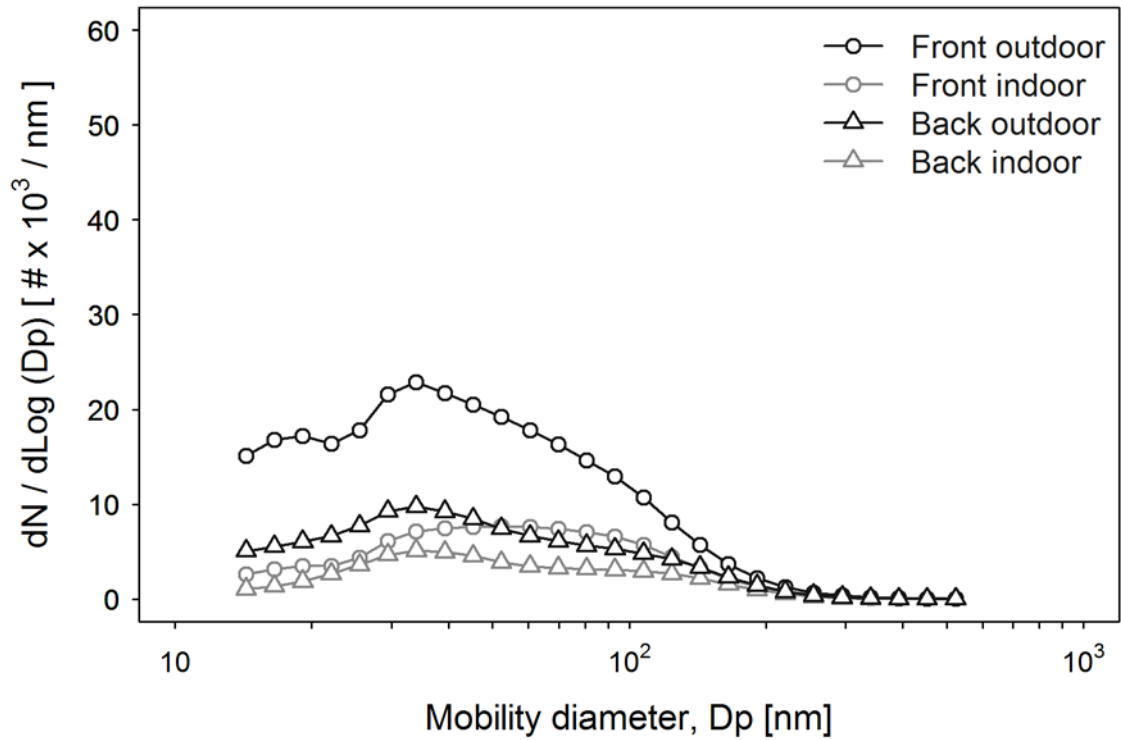


Figure 3. Scatter plots for daily PM<sub>2.5</sub> mass and UFP number concentrations.

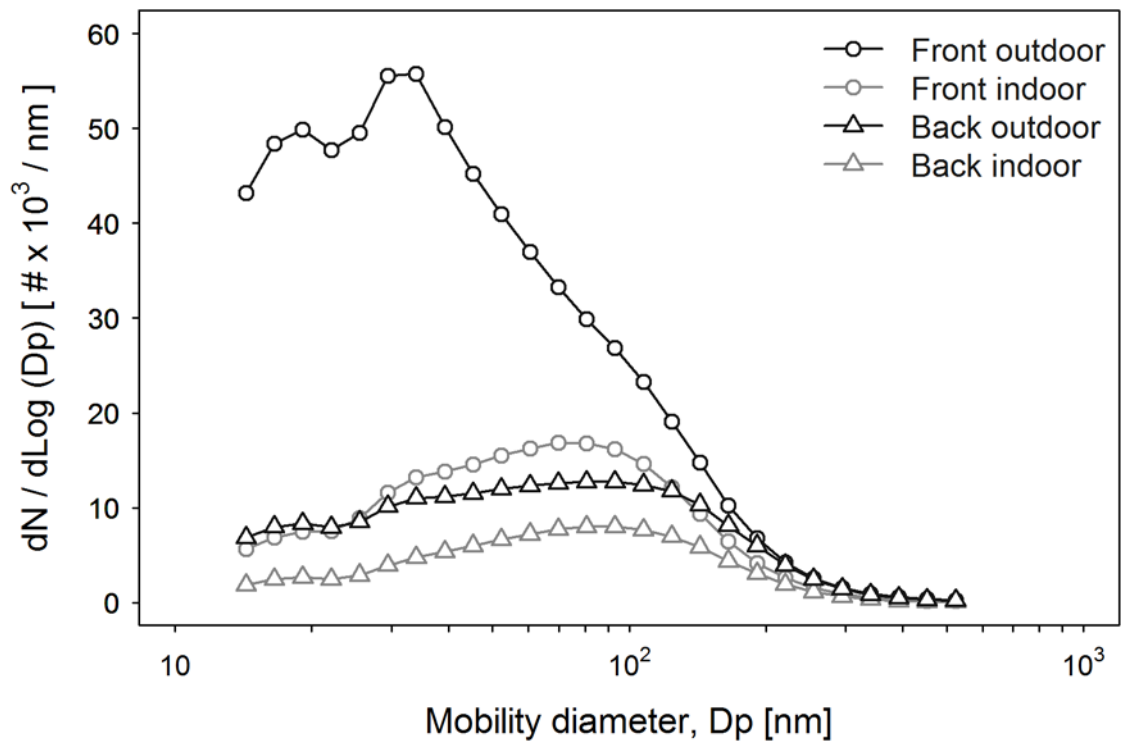
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## a - Hot period



## b - Cold period



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