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# Child exposure to indoor and outdoor air pollutants in schools in Barcelona, Spain

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ABSTRACT

Proximity to road traffic involves higher health risks because of atmospheric pollutants. In addition to outdoor air, indoor air quality contributes to overall exposure. In the framework of the BREATHE study, indoor and outdoor air pollution was assessed in 39 schools in Barcelona. The study quantifies indoor and outdoor air quality during school hours of the BREATHE schools. High levels of fine particles ($PM_{2.5}$), nitrogen dioxide ($NO_2$), equivalent black carbon (EBC), ultrafine particle (UFP) number concentration and road traffic related trace metals were detected in school playgrounds and indoor environments. $PM_{2.5}$ almost doubled (factor of 1.7) the usual urban background (UB) levels reported for Barcelona owing to high school-sourced $PM_{2.5}$ contributions: [1] an indoor-generated source characterised mainly by organic carbon (OC) from organic textile fibres, cooking and other organic emissions, and by calcium and strontium (chalk dust) and; [2] mineral elements from sand-filled playgrounds, detected both indoors and outdoors. The levels of mineral elements are unusually high in $PM_{2.5}$ because of the breakdown of mineral particles during playground activities. Moreover, anthropogenic PM components (such as OC and arsenic) are dry/wet deposited in this mineral matter. Therefore, $PM_{2.5}$ cannot be considered a good tracer of traffic emissions in schools despite being influenced by them. On the other hand, outdoor $NO_2$, EBC, UFP, and antimony appear to be good indicators of traffic emissions. The concentrations of $NO_2$ are 1.2 times higher at schools than UB, suggesting the proximity of some schools to road traffic. Indoor levels of these traffic-sourced pollutants are very similar to those detected outdoors, indicating easy penetration of atmospheric pollutants. Spatial variation shows higher levels of EBC, $NO_2$, UFP and, partially, $PM_{2.5}$ in schools in the centre than in the outskirts of Barcelona, highlighting the influence of traffic emissions. Mean child exposure to pollutants in schools in Barcelona attains intermediate levels between UB and traffic stations.



## 1. Introduction

Some of the health effects of exposure to air pollution, such as the impact on the respiratory and cardiovascular systems, have been extensively studied. Although it is well-known that exposure to air pollutants leads to an increase in mortality and morbidity rates of the population (e.g. Baccarelli et al., 2008; Künzli et al., 2000, 2004; Pope et al., 2002; WHO, 2005), few studies have focused on the role of air pollution on brain development. Evidence obtained from experimental studies in animals suggests that outdoor air pollution may play a major role in the inflammation of the central nervous system during sensitive periods (such as childhood) and consequently in behaviour and school performance (Block et al., 2012). A growing body of research, albeit limited, from epidemiological studies indicates that exposure to air pollution may be associated with an increased risk of neurodevelopmental disorders and cognitive impairments (Guxens and Sunyer, 2012).

Many epidemiological studies relate $PM_{2.5}$ (particles with and aerodynamic diameter <2.5 μm) to negative health outcomes (Dockery et al., 1993; Jerrett et al., 2005; Krewski et al., 2009; Laden et al., 2006; Lepeule et al., 2012; Pope et al., 2002). However, owing to the small size of ultrafine particles (UFP, particles <100 nm) that can translocate

*Abbreviations:* UB, urban background; UB-PR, urban background reference station of Palau Reial in Barcelona; SC, sampling campaign; UFP, ultrafine particles; LDSA, lung-deposited surface area; EC, elemental carbon; BC, black carbon; EBC, equivalent black carbon; OC, organic carbon; OM, organic matter.

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from the lung to the blood circulatory system or be taken up directly into the brain through the olfactory epithelium (Chen et al., 2006; Nemmar, 2002; Oberdörster et al., 2004), UFP arise as a potential $PM_{2.5}$ constituent to have large health effects (Knol et al., 2009) even though the evidence is still limited (Rückerl et al., 2011). The negative health effects of the proximity to road traffic might be more related to the exposure to UFP, black carbon (BC) and total PM counts since Zhu et al. (2002) found that they decreased rapidly in the first 150 m away from the traffic line and then levelled off, whereas $PM_{2.5}$ was found to be elevated only moderately.

Mediterranean cities are characterised by high densities of population and motor vehicles: there are about 5800 cars·km$^{-2}$ in Barcelona and about 4500 cars·km$^{-2}$ in Turin and Naples whereas these densities fall to 1000–1500 cars·km$^{-2}$ in northern and central European cities such as Budapest, Amsterdam or Berlin (Ajuntament de Barcelona, 2013). Hence, people living in more densely populated cities are closer to traffic and are more exposed to vehicle exhaust and non-exhaust emissions. In fact, recent studies have shown that cities in southern Europe have higher levels of $PM_{2.5-10}$ than those in northern and central Europe owing to the high vehicle density and drier weather (Eeftens et al., 2012; Querol et al., 2004a). The contribution from road dust (non-exhaust emissions from pavement, tyre and brake wear and re-suspension of the material deposited on the road) to PM levels is also higher in Mediterranean cities (Pant and Harrison, 2013). Some vehicle wear abrasion particles have a mode below $PM_{2.5}$ and another mode above this diameter that would only be present in $PM_{2.5-10}$; e.g. the mass modes of Fe, Cu, Ba and Sb are between 1.2 and 7.2 μm aerodynamic diameter (Gietl et al., 2010); a bimodal structure for Sb has been determined with a mode at 3.6–5.2 μm from brake wear and tear (Ijima et al., 2009).

There is increasing evidence that indoor air quality exposure is also responsible for a rise in mortality and morbidity (Sundell, 2004). However, little is known about air quality in indoor environments, where children spend most of the day (approximately 90%; Buonanno et al., 2012; US-EPA, 2008). Moreover, children constitute a particularly vulnerable population because of their physiological and behavioural characteristics. They have higher ventilation rates and higher levels of physical activity (Trasande and Thurston, 2005) with the result that they are more exposed to air pollutants than adults. Children spend a large part of their time at school both indoors and outdoors. In Spain, the school year lasts about 180 days and an average of 25 h per week at primary level (INCA, 2013). Although the association between air pollution exposure at schools and the impact on health has been the subject of more than 70 epidemiological publications (see Mejía et al., 2011), neurodevelopment has, by contrast, been poorly documented. Newman et al. (2013) found an association between elemental carbon (EC) from traffic and higher hyperactivity scores in children. In a study of one school in a highly polluted area and in another school in an area of low pollution in Quanzhou, China, Wang et al. (2009) found that neuropsychological functions, such as attention, were impaired in the former school with respect to the latter. The mechanisms responsible for the initiation of neuroinflammation in response to air pollution are poorly understood and may be exposure-specific (Block et al., 2012).

At schools, indoor concentrations of particulate matter have been shown to be highly correlated with outdoor levels, suggesting that indoor particles are largely of outdoor origin (Raysoni et al., 2011). However, this indoor penetration of outdoor particles depends not only on the physical barriers of the building and ventilation (natural or mechanical), but also on particle physico-chemical properties (Viana et al., 2011) and size (Tippayawong et al., 2009; Zhu et al., 2002).

It may well be that epidemiological studies are considerably influenced by the methods employed for the collection of air quality data, such as the instrumentation used, the sampling location, the pollutants and parameters monitored and the sampling period (Mejía et al., 2011) since the methodology selected could result in over/under-estimation of exposure. Mejía et al. (2011) have also highlighted the importance of the spatial unit of analysis. In epidemiological studies the nearest air quality monitoring station is generally used to represent the air quality in schools (sometimes using raw data from the station and other times estimating levels at schools). However, measuring in-situ at schools yields more accurate information about the exposure although Buonanno et al. (2013), Janssen et al. (2001) and Salimi et al. (2013) have reported some spatial variation in the concentration of some air pollutants within the school. Therefore, personal exposure monitoring is the most accurate methodology to assess the exposure to air pollutants (Buonanno et al., 2012).

The BREATHE (BRain dEvelopment and Air polluTion ultrafine particles in scHool childrEn) ERC Advanced Grant project seeks to determine whether traffic-related air pollutants have an adverse effect on neuropsychological development, exacerbating cognitive and neurobehavioral disorders. The aim of the present study is to characterise indoor and outdoor air quality and its variability, especially the parameters that are most influenced by traffic emissions at the schools participating in the BREATHE study.

## 2. Materials and methods

### 2.1. Study area

The study was carried out in the city of Barcelona (Spain; 15,993 inhabitants·km$^{-2}$) and in the adjacent municipality of Sant Cugat del Vallès (1761 inhabitants·km$^{-2}$; IDESCAT, 2012; Fig. 1). Both cities are located in the NE of the Iberian Peninsula and have a Mediterranean climate. Barcelona has one of the highest vehicle densities in Europe (Ajuntament de Barcelona, 2012). The urban traffic fleet is characterised by a large number of cars (60.6%, of which, since 2003, more than 60% of the new car registrations are diesel; DGT, 2011); motorcycles (30.2%), heavy duty vehicles (2.9%). Furthermore, Barcelona is one of the most important ports in the Mediterranean, and receives the highest number of cruise ships in Spain. This constitutes an additional source of atmospheric pollutants that are very often transported across the city by the sea breeze during the day. Owing to the topography of the area, the transport and dispersion of atmospheric pollutants within Barcelona are largely controlled by fluctuating coastal winds which blow in from the sea during the day (diurnal breeze), and, to a lesser extent, by winds from the land at night (night breeze, Jorba et al., 2013). In the city centre, the predominance of narrow streets (street canyons) and a dearth of green areas hinder the dispersion of pollutants. Moreover, the city is not infrequently affected by North African air mass transport (NAF), which contributes significantly to mineral $PM_{2.5}$.

On the other hand, Sant Cugat lies in the Vallès Depression away from the coast and is bounded by the Littoral mountain range to the southeast and by the Pre-Littoral mountain range to the northwest. Although these ranges shield the city from coastal pollutant intrusions (Fig. 1), the Llobregat Valley offers an atmospheric corridor into the Vallès Depression for air pollutants carried from the urban and industrial zones that surround the river. Once in this Depression, the pollutants accumulate due to the poor dispersion conditions.

### 2.2. Monitoring sites: schools and reference urban background station

Two sampling campaigns were carried out in 36 schools in Barcelona and 3 in Sant Cugat, from 27 January until 22 June 2012 (SC1; sampling campaign 1) and from 14 September 2012 until 22 February 2013 (SC2; sampling campaign 2). Traffic intensity and typology of the fleet around the schools is shown in Table S1. The sampling was performed simultaneously indoors (in a classroom with pupils) and outdoors (in the playground) at two schools per week; of this pair of schools, one was located in an urban background (UB) area, whereas the other one was situated near traffic. Indoor devices were placed where possible next to the wall opposite the blackboard (to avoid direct exposure to chalk or board marker emissions) and away from the windows (to avoid direct

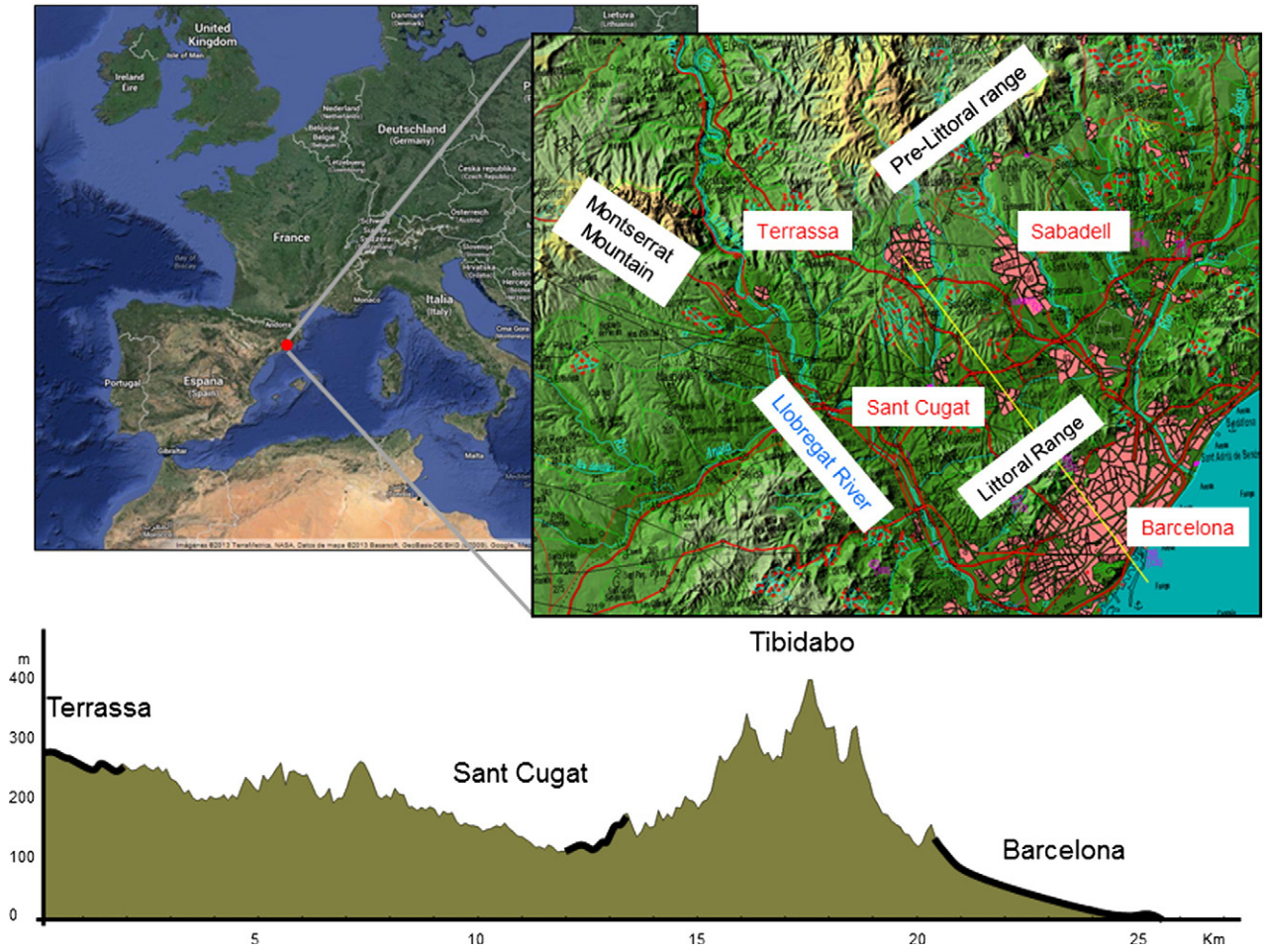



**Fig. 1.** Location and topographical profile of the study area. The black lines in the topographical profile indicate the location of the cities of Barcelona and Sant Cugat.

influence from outdoor levels and disturbances resulting from air currents). These conditions could not be met in all cases because of concerns for child safety. Air samples were collected at a height between 0.7 and 1.5 m above floor level, which is the height at which the pupils aged 7–9 would usually inhale.

Given that not all the schools were monitored simultaneously, data should be deseasonalised to remove temporal fluctuations when comparing the levels of pollutants between the schools. The data obtained at schools were deseasonalised with reference to the "Palau Reial" UB station (termed UB-PR), where the same pollutants were monitored throughout the sampling period. This station is located in the garden of the IDAEA-CSIC building (41°23′14″ N, 02°06′56″E, 78 m.a.s.l) and even though it represents UB conditions, it is exposed to road traffic emissions from the Diagonal Avenue (200 m away), one of the largest thoroughfares in Barcelona (100,000 cars·day$^{-1}$).

### 2.3. Sampling and analysis

Air quality in the schools was monitored for four days (from Monday morning to Friday morning), with a minimum of three days. No data are available for Fridays since this was the day when the monitoring instruments were moved from one school to the other. $PM_{2.5}$ samples were obtained by means of a high volume sampler MCV CAV-A/mb (30 m$^3$·h$^{-1}$) with an inlet with a specific nozzle plate for $PM_{2.5}$ (MCV). $PM_{2.5}$ was collected on Pallflex quartz fibre filters (PALL 2500 QAT-UP 150 mm) to obtain mass concentration and a complete chemical characterisation, resulting in a total of 553 8 h-daily samples (SC1: 140 indoors, 136 outdoors; SC2: 143 indoors, 134 outdoors). At schools, sampling duration was 8 h per day/filter during school hours (from 9 to 17 h, typical school hours in Barcelona). However, in UB-PR the sampling schedule was a 24 h sample every third day since this sampling is part of a long temporal series programme.

Once the gravimetric determination of the $PM_{2.5}$ mass concentration was performed, a complete chemical characterisation of the PM collected on the filters was carried out following the methodology of Querol et al. (2001) with a relative analytical error between 3 and 10% for the elements under study (Viana et al., 2006). A 1/4 fraction of each filter was bulk acid digested ($HNO_3$:HF:$HClO_4$) for the determination of the major elements by Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES; IRIS Advantage TJA Solutions, Thermo) and trace elements by Inductively Coupled Plasma-Mass Spectrometry (ICP-MS; X Series II, Thermo). Another 1/4 fraction of each filter was employed to determine water-soluble ions, $SO_4^{2-}$, $NO_3^-$ and $Cl^-$ by means of ion chromatography (ICHPLC) and $NH_4^+$ by means of a selective electrode. A 1.5 cm$^2$ filter punch was used for organic (OC) and elemental carbon (EC) determination by a thermal–optical transmission technique with a Sunset Laboratory OCEC Analyser with the NIOSH temperature programme (Birch and Cary, 1996). OC was converted into organic matter (OM) by a factor of 1.6, which according to Turpin and Lim (2001) accounts for the heteroatoms (O, H, N) present in the OM. Finally, contents of $SiO_2$ and $CO_3^{2-}$ were indirectly calculated using well-known experimental equations ($SiO_2 = 3 \cdot Al_2O_3$ and $CO_3^{2-} = 1.5 \cdot Ca$; Dulac et al., 1992; Molinaroli et al., 1993; Querol et al., 2001).

In addition, real-time concentrations of UFP (DiSCmini), mean UFP size (DiSCmini), lung deposited surface area in the alveolar region (LDSA; DiSCmini) and, BC (MicroAeth AE51) were recorded on a 5 or 10 minute basis. Weekly-averaged $NO_2$ concentrations with Gradko Environmental passive dosimeters were also obtained. All the DiSCmini devices employed were compared and corrected by correlation to a DiSCmini of reference to minimise any measuring differences (prior and post each sampling campaign). DiSCmini measures the number of UFP in the range 10–700 nm (accuracy: $\pm 500$ #·cm$^{-3}$). By contrast, at the UB-PR, the number of UFP was measured with a 3785 TSI CPC, measuring all the particles from 5 to 1000 nm. Therefore we are not comparing exactly the same parameter when measuring UFP at schools and at UB-PR. In fact, the slope and the $R^2$ of the equations obtained with a pre and post campaign intercomparison between the reference DiSCmini and the CPC were UFP[DiSCmini] = 1.62 · UFP[CPC] for the SC1 ($R^2 = 0.57$) and UFP[DiSCmini] = 1.40 · UFP[CPC] for the SC2 ($R^2 = 0.69$). This measurement difference has to be taken into account when comparing UFP at schools and UB-PR. LDSA in the alveolar region at UB-PR was recorded by a Nanoparticle Surface Area Monitor (3550 TSI NSAM).

The MicroAeth AE51 provides data of BC (in μg·m$^{-3}$) derived from absorption values. Eight hour-averages were cross correlated with EC concentrations from filter samples simultaneously collected in situ (at schools; including both campaigns), obtaining the equation $BC_{AE51} = 0.5436 \cdot EC$ ($R^2 = 0.88$). The results were converted to equivalent black carbon (EBC; in μg·m$^{-3}$). In the case of UB-PR, absorption values obtained with a Multi Angle Absorption Photometer (MAAP Thermo ESM Andersen Instruments) were converted into EBC in μg·m$^{-3}$ by an experimental Absorption/EC factor of 9.2 previously determined by Reche et al. (2011).

For this study, only mean values for school hours from Monday to Thursday were used (for both $PM_{2.5}$ filter samples and real-time measurements of BC and UFP) in order to take into account child exposure to air pollution during the time spent at school. In fact, this time restriction is a major advantage of our research given that PM concentrations (and the other pollutants) would be underestimated if considering 24 h means (Yip et al., 2004). The only exception is $NO_2$ concentration, which is 4 days averaged and includes the periods when children were not at schools.

### 2.4. Data deseasonalisation

To compare schools, seasonal adjustment of the daily values was conducted. This procedure, usually called (back-)extrapolation in time, is sometimes used in epidemiological studies when past values of pollutant levels are required to calculate the exposure of a given population using Land-Use Regression (Chen et al., 2010; Gehring et al., 2011; Mölter et al., 2010). Deseasonalised data were only employed for the spatial variation analysis. The remaining results were obtained from the data that were not deseasonalised.

To this end, an adjustment of all pollutant concentrations (both indoor and outdoor) was carried out using data from the UB-PR. The adjusted concentration $(C_i^j)_k^*$ of the *ith* pollutant for the *kth* day at the school *jth* was calculated as shown in Eq. (1):

$$\left(C_i^j\right)_k^* = \left(C_i^j\right)_k / \left[\left(C_i^{PR}\right)_k / \left(\overline{C_i^{PR}}\right)\right] \quad (1)$$

where $(C_i^j)_k$ is the concentration measured at the school, $C_i^{PR}$ and $\left(\overline{C_i^{PR}}\right)$ are the 8 h average corresponding to day *kth* and campaign averages at UB-PR, respectively.

To apply this method, the reference station must cover all the period and have at least 75% of valid data for each pollutant. These two conditions were met for most of the parameters analysed. Nevertheless, missing data of some pollutants were estimated by correlation with existing data of other pollutants at UB-PR.

## 3. Results

### 3.1. Levels of air pollutants

Table 1 shows the mean indoor and outdoor concentrations of $PM_{2.5}$, $NO_2$, UFP, UFP size, LDSA and EBC for BREATHE schools and for the UB-PR site. Averages were calculated for school hours except for $NO_2$ for which the daily means over 4 days were obtained.

Average outdoor levels are higher than indoors for $NO_2$, UFP, LDSA and EBC by a factor of 1.6, 1.5, 1.2 and 1.1 respectively (Table 1). Only average $PM_{2.5}$ levels and mean UFP size are higher indoors, with an indoor/outdoor ratio of 1.3 and 1.1 respectively. Mean outdoor concentrations are higher at schools than at UB-PR with school/UB-PR ratios of 1.7, 1.6 and 1.2 for $PM_{2.5}$, UFP and $NO_2$, respectively. However, levels are similar for EBC and LDSA. Concerning UFP levels it has to be taken into account the different measuring instruments (with different size range) at schools and the UB-PR site. Intercomparison exercises (see Section 2.3) showed that DiSCmini (at schools) measured between 40 and 60% more UFP than CPC (at UB-PR). Therefore, levels of UFP in UB-PR are probably lower than at schools but to a lesser extent than the factor 1.6.

The results obtained in this study are compared in Table 2 with those by Fromme et al. (2007), Molnár et al. (2007), Stranger et al. (2008), Wichmann et al. (2010) and Zwoździak et al. (2013). Other studies (e.g. Crilley et al., 2013; Pegas et al., 2012) are available, but not directly comparable since they focus on other pollutants.

Fig. 2a shows the spatial distribution of average outdoor $PM_{2.5}$, $NO_2$, EBC and UFP concentrations at the different schools. These maps were made with seasonally adjusted data (see Section 2.4). Two perimeters based on the highest (red) and lowest tercile (green) of EBC outdoor concentrations were drawn in all the maps to facilitate comparison with respect to the other pollutants.

EBC concentrations are markedly lower in the outskirts of Barcelona and Sant Cugat, especially in the area of the Coastal Range (NW of the city, Fig. 2a). These levels display an ascending gradient when approaching the city centre, which is severely affected by high vehicle density and characterised by a street architecture that hinders the dispersion of pollutants. $NO_2$ concentrations follow a similar spatial pattern: almost all the $NO_2$ green and red dots fall within the

**Table 1**
$PM_{2.5}$, $NO_2$, UFP, and EBC concentrations, UFP size, and LDSA for school hours (except for $NO_2$) of the 39 schools (indoor and outdoor), and of UB-PR. Highest mean values in bold.

| | INDOOR | | | | | OUTDOOR | | | | | UB-PR | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Mean | Min | Median | Max | SD | Mean | Min | Median | Max | SD | Mean | Min | Median | Max | SD |
| $PM_{2.5}$ (μg·m$^{-3}$) | **37** | 13 | 33 | 84 | 13 | 29 | 10 | 23 | 111 | 20 | 17 | 10 | 15 | 38 | 7 |
| $NO_2$ (μg·m$^{-3}$) | 30 | 5.1 | 30 | 69 | 12 | **47** | 14 | 46 | 98 | 17 | 41 | 23 | 38 | 97 | 15 |
| UFP (#·cm$^{-3}$) | 15,577 | 3584 | 15,376 | 30,932 | 6586 | **23,396** | 9868 | 20,955 | 55,804 | 9986 | 14,665 | 6335 | 13,286 | 32,654 | 5452 |
| Size mode (nm) | **42** | 30 | 41 | 57 | 5.5 | 39 | 27 | 36 | 65 | 7.0 | | | | | |
| LDSA (μm$^2$·cm$^{-3}$) | 34 | 8.6 | 31 | 71 | 14 | **42** | 15 | 40 | 83 | 15 | 42 | 21 | 37 | 86 | 14 |
| EBC (μg·m$^{-3}$) | 1.3 | 0.39 | 1.2 | 2.7 | 0.56 | **1.4** | 0.38 | 1.2 | 2.6 | 0.57 | 1.3 | 0.55 | 1.2 | 2.7 | 0.6 |

**Table 2**
Indoor and outdoor average concentrations of $PM_{2.5}$ ($\mu g \cdot m^{-3}$), UFP (#·cm-3) and EBC ($\mu g \cdot m^{-3}$) for various schools in Europe (w = winter samples; s = summer samples).

| Source | Location | N schools | Sampling time | Season | Indoor concentration (BREATHE/other ratio)[a] | | | | Outdoor concentration (BREATHE/other ratio)[a] | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | $PM_{2.5}$ | $NO_2$ | UFP | EBC | $PM_{2.5}$ | $NO_2$ | UFP | EBC |
| Wichmann et al. (2010) | Stockholm, Sweden | 6 | 8–16 h | | 8 (4.5) | 17 (1.7) | | 0.67[b] (1.9) | 10 (3.0) | 21 (2.3) | | 1.1[b] (1.2) |
| Fromme et al. (2007) | Münich, Germany | 64 | Teaching hours (5 h) | S | 22 (1.5) | | 6509[c] (2.9) | | | | | |
| | | | | W | 39 (1.0) | | | | | | | |
| Zwoździak et al. (2013) | Wroclaw, Poland | 1 | 24 h | S | 14 (2.5) | | | | 16 (1.8) | | | |
| | | | | W | 60 (0.6) | | | | 49 (0.6) | | | |
| Stranger et al. (2008) | Antwerp, Belgium | 15 (urban) | 8–20 h | S | 61 (0.6) | 113[d] (0.3) | | 2.0[e] (0.8) | 72 (0.4) | 97[d] (0.5) | | 2.0 (0.8)[e] |
| | | | | W | 57 (0.7) | 33[d] (0.9) | | 1.5[e] (0.8) | 53 (0.6) | 53[d] (0.9) | | 2.0 (0.6)[e] |
| This study | Barcelona, Spain | 39 | 9–17 h | | 37 | 30[d] | 15,577 | 1.3 | 29 | 47[d] | 23,396 | 1.4 |
| | | | | S | 34 | 32[d] | 18,848 | 1.5 | 29 | 48[d] | 23,144 | 1.5 |
| | | | | W | 38 | 29[d] | 13,656 | 1.2 | 30 | 47[d] | 23,547 | 1.3 |

[a] Ratio between the concentration of the considered element in our study (BREATHE) and the concentration found in the study to which it is compared.
[b] Soot.
[c] Different particle size range than BREATHE.
[d] 24 h average.
[e] BS has been converted to EBC.

corresponding EBC perimeter. Only some schools adjacent to the perimeter undergo a change in their category. Spatial patterns for UFP are also similar with one important exception: one school close to the harbour on the coastal side of the Montjuïc headland (S of the city) has high UFP levels albeit with low EBC levels, probably because of the influence from nearby shipping emissions and/or the ring road (Ronda Litoral). The spatial gradient for $PM_{2.5}$ also shows a similar pattern although with several exceptions, with high $PM_{2.5}$ levels in many low EBC schools. However, it should be noted that most of the $PM_{2.5}$ red spots are located close to the city centre. Fig. 2b shows the spatial distribution of indoor concentrations of pollutants under study. Indoor spatial patterns are very similar to those explained for outdoors, which suggests a significant infiltration of outdoor pollutants into indoor environments.

The same maps were created with data that were not seasonally adjusted. Fig. S1a shows how the increasing gradient of EBC and $NO_2$ outdoor concentrations towards the centre of Barcelona become blurred with respect to the adjusted data. This is why there are green dots in the centre of Barcelona. On the other hand, UFP gradient is more marked than when using deseasonalised data. $PM_{2.5}$ gradient is similar regardless of deseasonalisation. As regards indoor concentrations (Fig. S1b), the distribution of the school terciles is very similar to the scenario without deseasonalisation.

### 3.2. $PM_{2.5}$ components

The mean indoor and outdoor concentrations for different $PM_{2.5}$ components found at BREATHE schools and at UB-PR station are shown in Table 3. It should be noted that UB-PR concentrations are mean values for 24 h of $PM_{2.5}$ collection, whereas in schools they account for only 8 h. Fig. 3 summarizes the chemical profile of $PM_{2.5}$ at schools in indoor and outdoor environments. Mineral matter was estimated by the sum of the typical mineral compounds ($CO_3^{2-}$, $SiO_2$, $Al_2O_3$, Ca, Mg, Fe, K), and sea salt by Na and Cl.

Data show higher mean concentrations of most of the $PM_{2.5}$ components in outdoor than in indoor environments (Table 3). Exceptions are OC, Ca, $CO_3^{2-}$ and Sr, which attain higher indoor than outdoor concentrations. OC is, moreover, the PM component with the highest contribution to indoor $PM_{2.5}$ mass concentration, 33% of the total indoor $PM_{2.5}$ mass (reaching 44% when considering OM). It accounts for 22% (31% OM) of outdoor $PM_{2.5}$, being the second most important group after mineral compounds (Fig. 3).

A high variability between schools was observed for all the pollutants (high values of standard deviation). In most cases, the range of concentrations and standard deviation is higher in outdoor environments. This is especially true for $NO_3^-$ and mineral components ($SiO_2$, $Al_2O_3$, Fe, Ti, Mn, among others) because the levels of crustal elements are very dependent on the presence/absence of sand-filled playgrounds. In fact, the mineral components in schools with sand-filled playgrounds account for 16 $\mu g \cdot m^{-3}$ outdoors and 13 $\mu g \cdot m^{-3}$ indoors whereas in those with paved playgrounds it descends to 4 $\mu g \cdot m^{-3}$ outdoors and 8 $\mu g \cdot m^{-3}$ indoors.

Fig. 4a and b show the relative abundance of EBC, mineral matter and OC (after normalisation of their levels) at each school outdoors and indoors, respectively. This enables us to divide the schools into two groups:

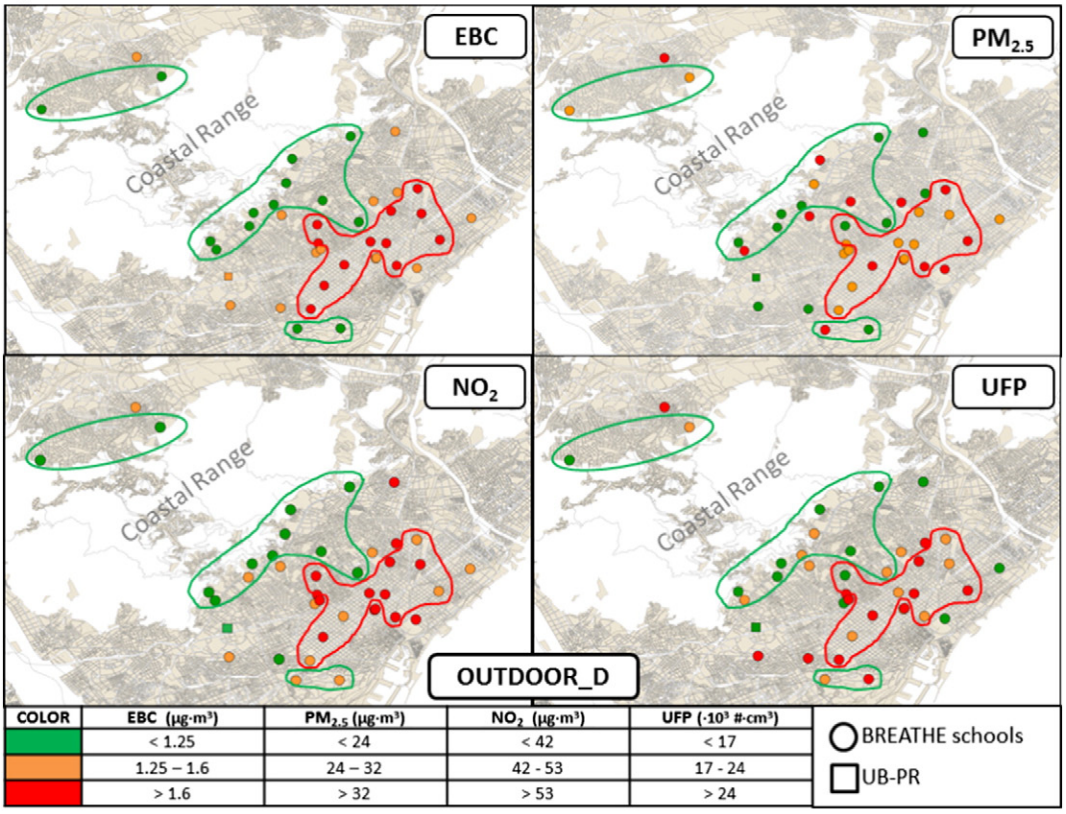
- *Group 1*: schools are defined by high relative normalised ternary proportions of mineral matter (>45%) and low influence of EBC (<25%). These schools are characterised by having sand-filled playgrounds and by being located in the outskirts of Barcelona or in Sant Cugat.
- *Group 2*: schools show low relative normalised abundance of mineral matter (<30%) and high levels of EBC (>35%). These schools are mainly located in the centre of Barcelona and have paved playgrounds.

Note that the indoor range of relative normalised levels of OC is narrower (25–45%) than the outdoor range (15–50%).

Eight hour concentrations (except the 4-day averaged $NO_2$) of all pollutants measured were plotted versus 8 h means of $Al_2O_3$ and EBC concentrations as tracers of mineral matter and traffic matter emissions, respectively (Fig. 5a and b, for outdoor and indoor, respectively). Based on this cross correlation analysis of the correlation coefficients (r) and on the PM source apportionment studies performed in the outdoor UB of Barcelona by Amato et al. (2009), specific pollutants of outdoor environments were selected as tracers of the following sources:

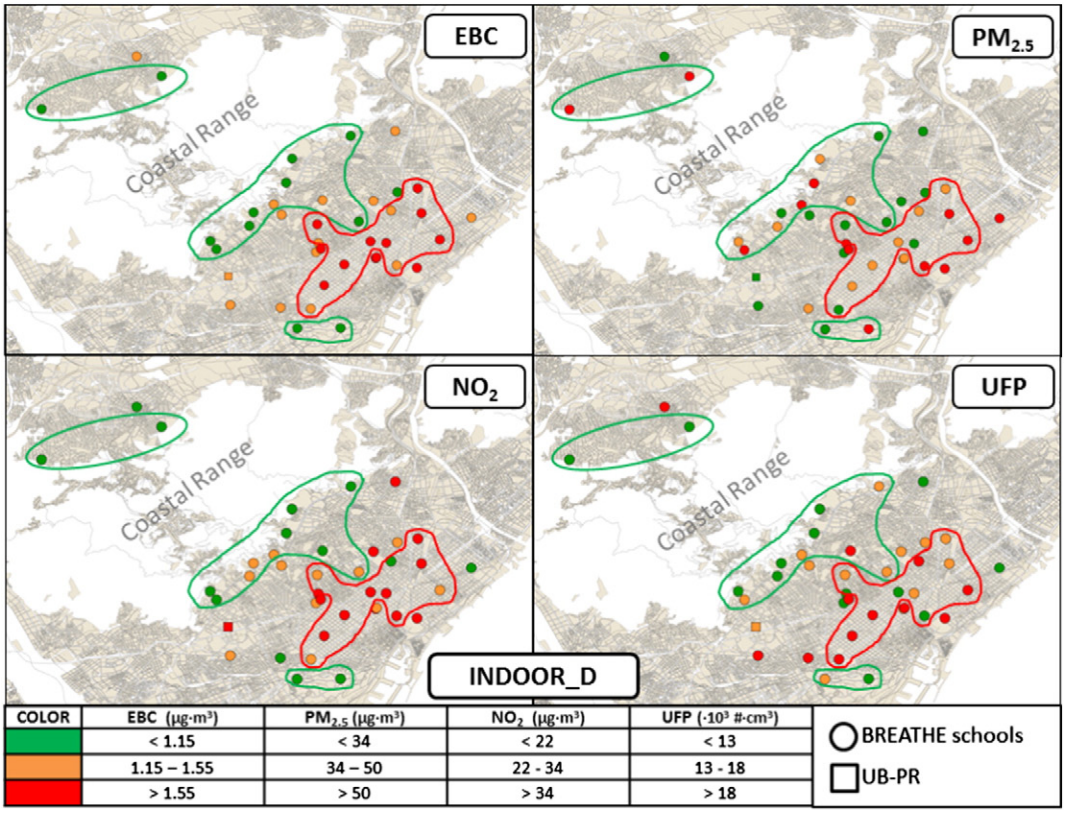
- Mineral components ($SiO_2$, Ca, Fe, K, Mg, Ti, Mn, Sr and rare Earth elements): include elements highly correlated with $Al_2O_3$ but not with EBC. This group consists of elements of mineral origin and is the most significant in the outdoor environment, and second in importance indoors (Fig. 3). Although $PM_{2.5}$ is outside this group, it is also highly correlated with $Al_2O_3$, whereas the correlation with EBC is low, especially for the indoor environment ($r = -0.03$ indoors; $r = 0.29$ outdoors, Table S2). Mineral elements have higher levels outdoors with the exception of Ca and Sr (Table 3).
- Traffic-related components ($NO_2$, Sb, UFP and LDSA): include the elements that correlate highly with EBC (traffic tracer) and do not correlate with $Al_2O_3$ in either environment. Sb and Cu levels (brake pad abrasion tracers, Sternbeck et al., 2002) are higher in outdoor environments than indoors by a factor of 1.3 and 1.1, respectively.

a)




| COLOR | EBC (μg·m³) | $PM_{2.5}$ (μg·m³) | $NO_2$ (μg·m³) | UFP (·10³ #·cm³) |
|---|---|---|---|---|
| | < 1.25 | < 24 | < 42 | < 17 |
| | 1.25 – 1.6 | 24 – 32 | 42 - 53 | 17 - 24 |
| | > 1.6 | > 32 | > 53 | > 24 |

b)




| COLOR | EBC (μg·m³) | $PM_{2.5}$ (μg·m³) | $NO_2$ (μg·m³) | UFP (·10³ #·cm³) |
|---|---|---|---|---|
| | < 1.15 | < 34 | < 22 | < 13 |
| | 1.15 – 1.55 | 34 – 50 | 22 - 34 | 13 - 18 |
| | > 1.55 | > 50 | > 34 | > 18 |

**Fig. 2.** a. Spatial distribution of seasonally adjusted outdoor levels of $NO_2$, $PM_{2.5}$, EBC and UFP in the BREATHE school. Perimeters are based on the outdoor EBC highest tercile (red) and the lowest tercile (green). b. Spatial distribution of seasonally adjusted indoor levels of $NO_2$, $PM_{2.5}$, EBC and UFP in the BREATHE schools. Perimeters are based the on outdoor EBC highest tercile (red) and the lowest tercile (green). UB-PR category refers to outdoor levels.

**Table 3**
$PM_{2.5}$ components concentrations for the 39 schools (indoor and outdoor) for school hours (8 h) and for the UB-PR (24 h) in Barcelona. Highest mean values in bold. N = number of schools in the first campaign plus schools in the second campaign, (mean of 3.5 samples per school and campaign); n = total number of samples; MM = mineral matter.

| | Indoor (N = 77) | | | | | Outdoor (N = 75) | | | | | UB-PR (n = 26) | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Mean | Min | Median | Max | SD | Mean | Min | Median | Max | SD | Mean | Min | Median | Max | SD |
| *(μg·m$^{-3}$)* | | | | | | | | | | | | | | | |
| OC | 10 | 3.5 | 9.6 | 19 | 3.2 | 5.5 | 2.1 | 4.7 | 15 | 2.5 | 2.1 | 0.78 | 1.7 | 5.4 | 0.94 |
| EC | 1.3 | 0.25 | 1.2 | 2.9 | 0.64 | 1.3 | 0.23 | 1.3 | 3.3 | 0.60 | 0.94 | 0.15 | 0.68 | 2.2 | 0.47 |
| $CO_3^{2-}$ | 2.3 | 0.31 | 2.1 | 7.6 | 1.5 | 1.2 | <0.1 | 0.81 | 6.3 | 1.3 | 0.13 | <0.1 | <0.1 | 0.3 | 0.07 |
| $SiO_2$ | 3.7 | 0.34 | 2.6 | 17 | 3.3 | 4.4 | 0.24 | 1.8 | 34 | 6.7 | 0.32 | <0.1 | 0.21 | 1.1 | 0.26 |
| $Al_2O_3$ | 1.2 | 0.11 | 0.86 | 5.6 | 1.1 | 1.5 | <0.1 | 0.60 | 11 | 2.2 | 0.11 | <0.1 | <0.1 | 0.37 | 0.09 |
| Ca | 1.6 | 0.21 | 1.4 | 5.1 | 0.97 | 0.82 | <0.1 | 0.54 | 4.2 | 0.86 | <0.1 | <0.1 | <0.1 | 0.19 | 0.04 |
| Fe | 0.42 | <0.1 | 0.29 | 1.6 | 0.32 | 0.58 | <0.1 | 0.28 | 3.8 | 0.76 | 0.14 | <0.1 | 0.10 | 0.30 | 0.06 |
| K | 0.37 | <0.1 | 0.31 | 1.2 | 0.24 | 0.40 | <0.1 | 0.23 | 2.2 | 0.43 | 0.11 | <0.1 | <0.1 | 0.26 | 0.07 |
| Na | 0.34 | <0.1 | 0.28 | 1.3 | 0.22 | 0.34 | <0.1 | 0.25 | 1.4 | 0.29 | 0.18 | <0.1 | 0.20 | 0.56 | 0.13 |
| Mg | 0.16 | <0.1 | 0.13 | 0.68 | 0.12 | 0.19 | <0.1 | 0.09 | 1.2 | 0.26 | <0.1 | <0.1 | <0.1 | 0.09 | 0.02 |
| $SO_4^{2-}$ | 1.4 | <0.1 | 1.2 | 4.3 | 1.1 | 1.6 | 0.21 | 1.2 | 4.8 | 1.1 | 1.9 | 0.41 | 1.2 | 7.3 | 1.8 |
| $NO_3^-$ | 0.72 | <0.1 | 0.61 | 4.4 | 0.69 | 1.8 | 0.21 | 1.0 | 11 | 2.1 | 1.0 | <0.1 | 0.41 | 13 | 2.1 |
| $NH_4^+$ | 0.48 | <0.1 | 0.34 | 1.8 | 0.46 | 0.86 | <0.1 | 0.56 | 3.7 | 0.73 | 0.79 | <0.1 | 0.30 | 5.7 | 1.1 |
| MM | 10 | 1.4 | 7.7 | 37 | 7.1 | 9.1 | 0.86 | 4.0 | 64 | 12 | 0.91 | 0.15 | 0.65 | 2.2 | 0.51 |
| *(ng·m$^{-3}$)* | | | | | | | | | | | | | | | |
| Li | 0.55 | <0.1 | 0.32 | 2.2 | 0.51 | 0.86 | <0.1 | 0.25 | 13 | 1.8 | <0.1 | <0.1 | <0.1 | 0.34 | 0.06 |
| Ti | 55 | 5.5 | 45 | 233 | 44 | 59 | 2.0 | 22 | 502 | 92 | 3.8 | <0.1 | 2.7 | 10 | 2.9 |
| V | 4.8 | 0.67 | 3.8 | 17 | 3.4 | 5.9 | 1.1 | 4.6 | 22 | 4.3 | 4.2 | 0.70 | 2.5 | 19 | 5.0 |
| Cr | 3.8 | <0.1 | 3.0 | 14 | 2.9 | 3.4 | <0.1 | 2.8 | 8.6[a] | 3.3 | 1.5 | <0.1 | 0.92 | 7.0 | 1.5 |
| Mn | 12 | 2.4 | 9.4 | 41 | 8.2 | 16 | 2.1 | 9.2 | 85 | 17 | 4.3 | 0.74 | 3.3 | 8.8 | 2.0 |
| Co | 0.21 | <0.1 | 0.16 | 0.81 | 0.17 | 0.22 | <0.1 | 0.16 | 1.3 | 0.25 | <0.1 | <0.1 | <0.1 | 0.26 | 0.06 |
| Ni | 2.6 | <0.1 | 2.3 | 10 | 1.8 | 3.3 | <0.1 | 2.8 | 7.5[b] | 3.1 | 1.8 | <0.1 | 1.0 | 6.5 | 1.7 |
| Cu | 8.2 | 3.4 | 7.6 | 15 | 2.7 | 8.8 | 3.0 | 7.9 | 21 | 3.9 | 6.7 | 1.7 | 5.3 | 14 | 2.7 |
| Zn | 52 | 19 | 47 | 147 | 24 | 55 | 14.4 | 48 | 181 | 28 | 41 | 5.2 | 31 | 92 | 25 |
| As | 0.46 | <0.1 | 0.43 | 1.4 | 0.21 | 0.50 | 0.19 | 0.41 | 2.0 | 0.27 | 0.30 | <0.1 | 0.27 | 0.88 | 0.17 |
| Se | 0.33 | <0.1 | 0.31 | 1.0 | 0.22 | 0.37 | <0.1 | 0.33 | 0.98 | 0.22 | 0.25 | <0.1 | 0.23 | 0.60 | 0.16 |
| Sr | 4.6 | 0.59 | 4.1 | 15 | 2.6 | 2.8 | <0.1 | 1.6 | 16 | 3.0 | 0.48 | 0.11 | 0.35 | 1.3 | 0.31 |
| Cd | 0.14 | <0.1 | 0.12 | 0.61 | 0.09 | 0.17 | <0.1 | 0.14 | 0.64 | 0.13 | 0.11 | <0.1 | 0.11 | 0.34 | 0.07 |
| Sn | 3.0 | 0.88 | 2.5 | 12 | 1.8 | 3.3 | 0.52 | 2.6 | 14 | 2.3 | 2.3 | 0.62 | 1.6 | 9.4 | 1.5 |
| Sb | 0.83 | 0.13 | 0.83 | 1.6 | 0.35 | 1.1 | 0.12 | 1.0 | 3.6 | 0.58 | 1.0 | 0.28 | 0.72 | 3.5 | 0.63 |
| Ba | 19 | <0.1 | 13 | 133 | 20 | 20 | <0.1 | 12 | 241 | 31 | 3.7 | <0.1 | 2.1 | 37 | 5.9 |
| La | 0.43 | <0.1 | 0.34 | 1.6 | 0.32 | 0.55 | <0.1 | 0.25 | 3.6 | 0.72 | <0.1 | <0.1 | <0.1 | 0.26 | 0.06 |
| Pb | 7.3 | 1.9 | 6.9 | 19 | 3.1 | 8.1 | 3.1 | 6.9 | 24 | 4.3 | 6.0 | 0.23 | 5.6 | 14 | 3.7 |

[a] The highest value after the maximum of 26 ng·m$^{-3}$, which might be considered an outlier due to an extremely polluted week.
[b] The highest value after two maximums of 21 and 17 ng·m$^{-3}$, which might be considered an outlier due to an extremely polluted week.

- Secondary inorganic aerosol (SIA: $SO_4^{2-}$, $NO_3^-$ and $NH_4^+$): these are urban-regional pollution tracers with the result that they are moderately correlated with EBC but not with $Al_2O_3$. Together, the secondary inorganic aerosols constitute the third most important group in the two environments (Fig. 3). The levels are similar in both environments but always higher in the playgrounds, and markedly higher outdoors for $NO_3^-$.
- Industrial elements (Cd) and fuel-oil combustion (Ni, V): these metals have a low correlation (r<0.5) or do not correlate (r<0.2) with EBC and with $Al_2O_3$. These elements are typically emitted by industrial processes (Cd) and heavy fuel combustion (Ni and V, mostly shipping in Barcelona, Amato et al., 2009). They are also correlated with one another (Table S2). Higher levels of tracers of heavy oil combustion and metallurgical activities (Zn, As, Cd, Cr, Co, Se, V, Ni and Pb, Querol et al., 2007) have been found in playgrounds than in indoor environments.
- OC and other pollutants associated with mineral matter resuspension (as in both environments, Co, Pb): elements which have a moderate correlation (0.2 < r < 0.6) with $Al_2O_3$ and a low correlation (r < 0.25) indoors or moderate correlation (0.2 < r < 0.6) outdoors with EBC.
- Sea salt (Na): there is no correlation between Na and EBC but Na is moderately correlated with $Al_2O_3$, suggesting an additional source (mineral) of Na. Its contribution is equal in the two environments.

When comparing outdoor school concentrations of $PM_{2.5}$ components with those measured in UB-PR, outdoor levels at schools are:

- Much higher: almost all the crustal elements (Ti, $Al_2O_3$, $SiO_2$, Li, Ca, Sr, La, Ba, Fe, Mn, K) are higher at schools than in UB-PR with a school/UB-PR ratio between 3.6 and 15.4; OC and Cr are 2.6 and 2.2 times higher, respectively (Table 3).
- Higher: Mg is 1.9 times higher in playgrounds than in UB-PR. Na (from sea salt) has a playground/UB-PR ratio of 1.9 and Ni and V (shipping emission tracers) ratios are 1.8 and 1.4, respectively. The different sampling periods (24 h vs 8 h) could play a major role in this component, since sea breezes usually occur at midday. Traffic related components such as Sn (1.5), Cu (1.3) and $NO_3^-$ (1.7) are also higher in playgrounds. The same pattern is observed for industrial elements such as Zn, As, Cd, and Pb (by factors from 1.3 to 1.7).
- Similar: Sb (1.04) and the secondary components $NH_4^+$ (1.09), and $SO_4^{2-}$ (0.84) have similar levels in the schools and at UB-PR.

## 4. Discussion

The levels of $PM_{2.5}$, $NO_2$, and UFP found at schools in Barcelona in both indoor and outdoor environments are higher than expected since $PM_{2.5}$ and $NO_2$ concentrations are 1.7 and 1.2 times higher than those found in the UB-PR station. Outdoor levels of $NO_2$ at BREATHE schools can be considered to be representative of all schools in Barcelona considering that they agree with modelled data employing Land Use Regression from the ESCAPE project for all the schools in Barcelona (Cyrys et al., 2012; De Nazelle et al., 2013). The modelled data yielded an average of 50 μg·m$^{-3}$, which is practically the same as the value obtained with measurements at the 39 BREATHE schools, and higher than the value at the reference station of UB-PR (41 μg·m$^{-3}$).

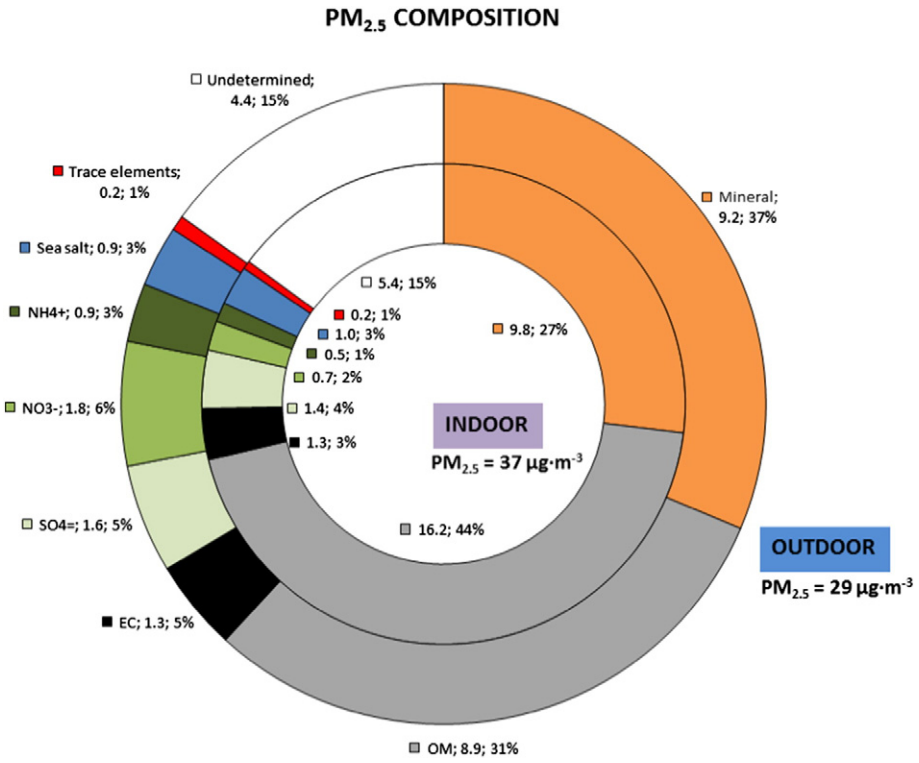



**Fig. 3.** Average indoor and outdoor $PM_{2.5}$ chemical composition in BREATHE schools.

Outdoor levels are higher than indoors for $NO_2$ and UFP as expected because the main sources (mostly road traffic) of these pollutants are located outside the buildings (e.g. Dall'Osto et al., 2011; Pey, 2007; Table 1). However, EBC levels are almost the same in both environments, thus indicating a high indoor penetration of this pollutant (in fact, the indoor–outdoor correlation for most of the schools was better for EBC than UFP). Fresh exhaust emissions from traffic are very fine (20–30 nm prevailing mode, Dall'Osto et al., 2011), and this primary UFP may increase their size mode when affected by different processes such as coagulation and condensation that modify their size distribution and, therefore finding slightly finer outdoor particle size with respect to that found indoors. High UFP concentrations combined with a finer particle size leads to higher LDSA values outdoors owing to traffic proximity. By contrast, markedly higher levels of $PM_{2.5}$ are found in classrooms than outdoors, suggesting an important indoor source affecting $PM_{2.5}$ levels.

As expected, most of the $PM_{2.5}$ components have higher concentrations outdoors, except OC (with an important contribution to $PM_{2.5}$), Ca, $CO_3^{2-}$ and Sr. The relatively high indoor levels (close to outdoors in many cases) of externally emitted traffic-related components ($NO_2$, Sb, UFP and LDSA), SIA, industrial elements (Cd) and elements derived from fuel oil combustion (Ni, V) suggest significant infiltration rates from the outdoor environment. The similarity of the relationship of Sb levels between schools playgrounds and UB-PR agrees with the one discussed for EBC, and, together with the higher levels of Sn, Cu and $NO_3^-$, corroborates our observation that the schools are exposed to slightly higher traffic emissions than UB in Barcelona. On the other hand, Na (marine in a relevant proportion), Ni and V (mostly from shipping emissions, Amato et al., 2009) have considerable higher levels at schools than at UB-PR, and, apart from the different sampling periods, it should also be noted that the schools are on average closer to the coast than UB-PR. Also higher levels at schools are observed for industrial elements such as Zn, As, Cd, and Pb. This may be attributed to the fact that the trajectories of the air masses from the Vallès Depression (location of industry) do not especially affect the zone where UB-PR is located whereas, some of the schools are directly exposed to industrial metals (Minguillón et al., 2014).

Mineral matter is usually characterised by a coarse grain size owing to its mechanical origin. Thus, in urban areas, levels of mineral matter in $PM_{2.5}$ are lower than 2 $\mu g \cdot m^{-3}$ (in UB-PR, 0.91 $\mu g \cdot m^{-3}$). However, in the present study unusually high levels of mineral matter in $PM_{2.5}$ were found, especially in the schools with sand-filled playgrounds. In fact, mineral matter is the most significant group in the outdoor environment, and second in importance in the indoors (Fig. 3). The activity in the playgrounds probably contributed to the breakdown of mineral particles. These fine mineral particles are continuously resuspended because of the very low precipitation in Barcelona (Querol et al., 2007). In Fig. 5a and b it can be observed that mineral matter correlates with OC and other pollutants such as As, Co and Pb, suggesting that mineral matter was polluted owing to the dry and wet deposition of these elements in the playgrounds and to their possible retention by adsorption on crustal elements. Therefore pollutants are resuspended at the same time as the mineral matter. This seems to be particularly true for As, which is present in both indoor and outdoor groups. It should be noted that outdoors OC is also correlated with EBC, thus suggesting that OC is mostly generated by or at the same time as traffic emissions (Minguillón et al., 2011) whereas in the indoor environment, OC is strongly influenced by local indoor sources since it has no relationship with EBC.

An important source of OC in indoor environments could be cotton fibres from clothes, skin cells, other organic emissions from children (as evidenced in studies at schools, Braniš and Šafránek, 2011; Fromme et al., 2008), cooking emissions (Abdullahi et al., 2013; Brunekreef et al., 2005; Lanki et al., 2007) and also condensation/nucleation of semi-volatile organic compounds (SVOCs, Weschler and Shields, 1999). The chalk for the blackboards might be responsible for the higher levels of Ca, $CO_3^{2-}$ and Sr indoors. X-ray diffraction analysis revealed that it was composed of calcite ($CaCO_3$). Since the atomic radius of Sr is similar to that of Ca, this element often replaces Ca in calcite and gypsum ($CaSO_4 \cdot 2H_2O$), accounting for the higher indoor levels of Sr.

All these elements have also been compared with the Spanish urban concentration ranges defined by Querol et al. (2007, 2008) (Figs. S2a

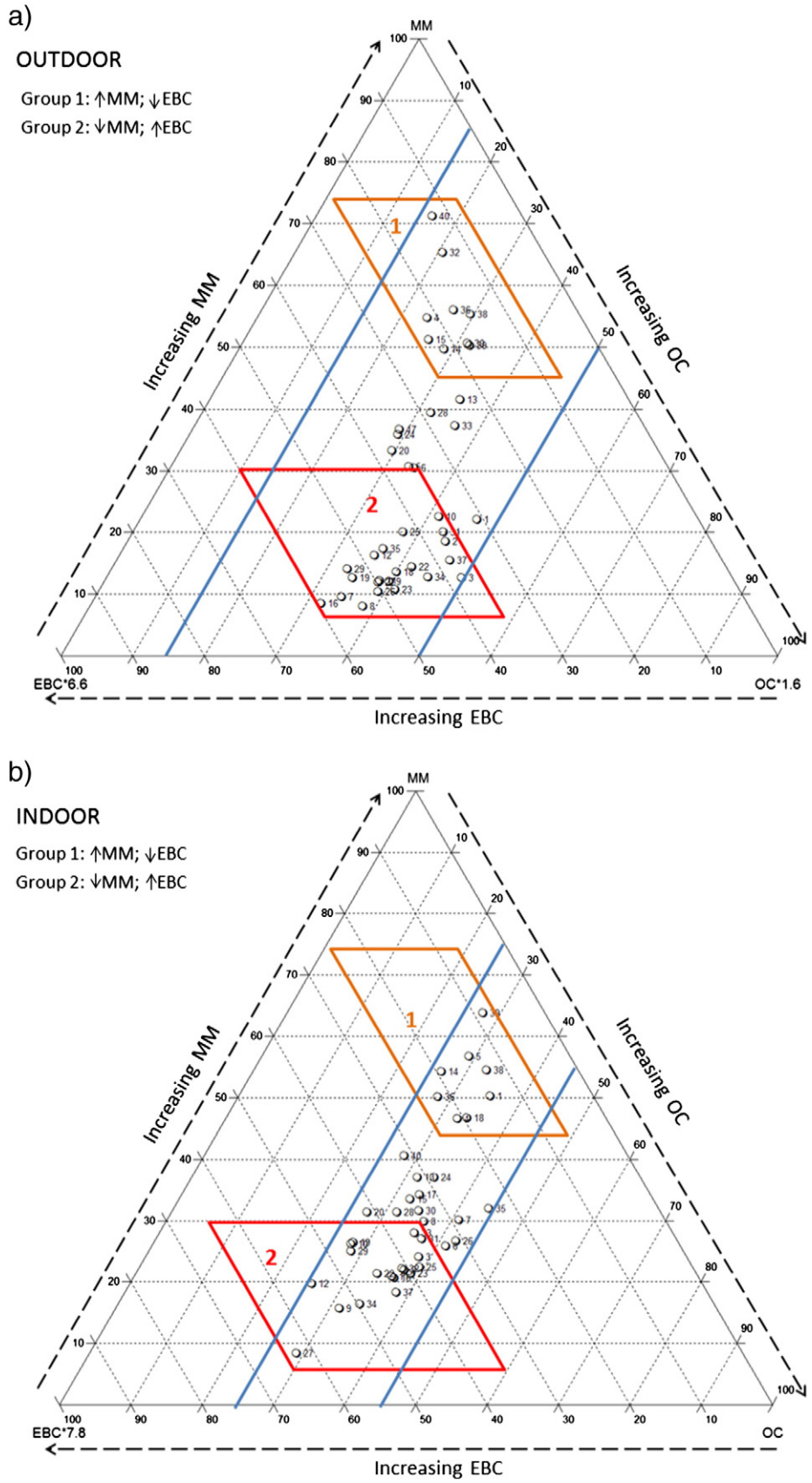



**Fig. 4.** a. Ternary plot showing the relationship between normalised outdoor EBC, mineral matter and OC for each school. MM = mineral matter. b. Ternary plot showing the relationship between normalised indoor EBC, mineral matter and OC for each school. MM = mineral matter.

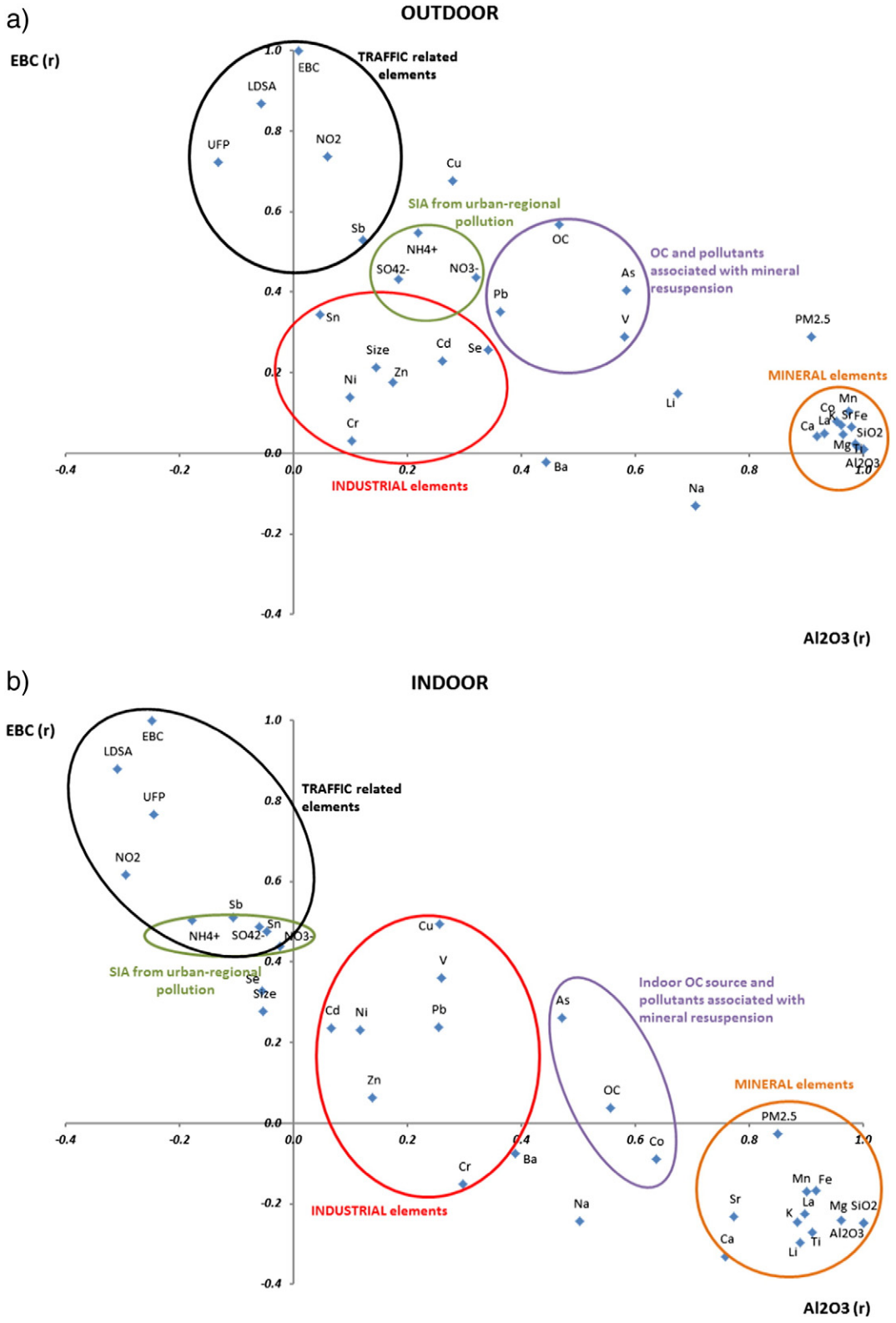



**Fig. 5.** a. Dispersion plot of the correlation coefficients (r) between EBC and $Al_2O_3$ and the other pollutants considered for the outdoor environment. b. Dispersion plot of the correlation coefficients (r) between EBC and $Al_2O_3$ and the other pollutants considered for the indoor environment.

and S2b). OC and mineral components (Ca, $Al_2O_3$, Fe, K, Sr, Ti, Li, etc.) are the only ones that attain higher concentrations in the school playgrounds than the aforementioned ranges (and are the main reason for the higher $PM_{2.5}$ levels at schools than at UB-PR), whereas the remaining PM components are within these ranges.

High variability among schools is observed for all the pollutants, highlighting the wide range of concentrations children are exposed to. It is especially true for mineral components (related to the presence/absence of sand-filled playgrounds) and for $NO_3^-$. As regards $NO_3^-$, the variation between schools could also be due to different environmental conditions (T and RH) throughout the campaign. Harrison et al.

(1994), Wakamatsu et al. (1996) and the references therein reported that $NH_4NO_3$ (the most common $NO_3^-$-bearing species in PM) is very unstable under typical summer temperatures in Barcelona, resulting in a wide disparity in $NO_3^-$ concentration between colder (February) and warmer months (June and September). This instability is also the cause of the lower levels of $NO_3^-$ found indoors, where the temperature is usually higher than outdoors (in particular during winter). In most cases, the range of concentrations and standard deviations is higher in outdoor environments. This indicates that indoor sources might be similar in schools and that physical barriers (buildings materials, windows, etc.) hinder, albeit slightly, the entry of outdoor pollutants into indoor environments (infiltration rates may vary widely from school to school, Adgate et al., 2003; Dockery and Spengler, 1981). As a representative indoor sourced component, note that the indoor range of relative normalised ternary (OC–MM–EBC) abundance of OC is narrower (25–45%) than the one outdoors (15–50%). By contrast, higher outdoor levels of OC are found in those schools with higher levels of EBC, showing that OC and EBC have a common outdoor source (traffic) as already stated before.

$PM_{2.5}$ levels from BREATHE schools are 4.5 (indoor) and 3.0 (outdoor) times higher to the six schools in Stockholm studied by Wichmann et al. (2010) where only school hours were considered (Table 2). Since $PM_{2.5}$ annual mean levels obtained within the framework of the ESCAPE project in Stockholm County were half those found in Barcelona (8.5 $\mu g \cdot m^{-3}$ vs 16.3 $\mu g \cdot m^{-3}$, respectively; Eeftens et al., 2012), the higher levels in Barcelona are due to a local source of $PM_{2.5}$ at schools (activity of children and the existence of sand-filled playgrounds) plus the higher $PM_{2.5}$ levels usually found in this city. This agrees to what is observed when comparing to Munich schools during summer (1.5 times higher indoor levels in BREATHE schools, Fromme et al., 2007), where the difference at schools cannot be only explained by the typical levels found in both cities (annual $PM_{2.5}$ mean for Munich 14.6 $\mu g \cdot m^{-3}$, similar to the 16.3 $\mu g \cdot m^{-3}$ in Barcelona, Eeftens et al., 2012). Coal for domestic heating in Poland may be the cause for the higher levels of $PM_{2.5}$ found in a school in Wroclaw (on a 24 h average basis in this case, Zwoździak et al., 2013) than in BREATHE schools during winter (BREATHE/Wroclaw ratios of 0.6 for both indoor and outdoor), whereas the opposite is found during summer (being 2.5 and 1.8 times higher in BREATHE, indoor and outdoor respectively). Unexpectedly, mean $PM_{2.5}$ concentrations were higher in five schools in Antwerp (Belgium, Stranger et al., 2008) than in BREATHE (BREATHE/Antwerp ratio of 0.6 for indoor and 0.5 for outdoor). Shifting to $NO_2$, the annual median concentration in Stockholm County was 14.9 $\mu g \cdot m^{-3}$ whereas in Barcelona it was 54.7 $\mu g \cdot m^{-3}$ (Cyrys et al., 2012), indicating a relationship similar to that of the schools (BREATHE/Stockholm ratio of 1.7 and 2.3 for indoor and outdoor, respectively). In Antwerp, $NO_2$ concentrations are also higher than in BREATHE (BREATHE/Antwerp ratio of 0.9 for both environments in winter and, 0.3 indoors and 0.5 outdoors in summer). The levels in the Belgian schools are surprisingly high since the ESCAPE annual $NO_2$ median was 30 $\mu g \cdot m^{-3}$ (Cyrys et al., 2012) which is much lower than the 97 and 53 $\mu g \cdot m^{-3}$ at Antwerp in summer and in winter, respectively. Moreover, EBC (measured in Barcelona) and soot (measured in Stockholm) are not directly comparable. However, higher levels are found in Barcelona (Stockholm vs BREATHE; 0.7 vs 1.3 $\mu g \cdot m^{-3}$ indoors; 1.1 vs 1.4 $\mu g \cdot m^{-3}$ outdoors), especially indoors, probably because of a higher traffic density and a higher diesel vehicle proportion in the fleet. In summer, the levels of UFP in Munich schools are 2.9 times lower than those at BREATHE schools. However, these levels are not comparable because the different particle size cut-off (DiSCmini were employed for this study whereas a SMPS system was used in Munich) for UFP measurement is very important when comparing absolute number concentrations.

Bringing the attention to $PM_{2.5}$ components, the concentrations of K, Ca, Fe, V, Cr, Mn, Ni, Cu, Zn, Pb were compared to those in Stockholm (Molnár et al., 2007, Table S3) with the conclusion that higher levels are found in BREATHE schools. This is particularly evident for mineral tracers owing to higher resuspension rates because of the low rainfall in southern Europe (Querol et al., 2004b). In similarity with our results, Ca levels in Stockholm were also higher in indoors than outdoors. Moreover, Molnár et al. (2007) concluded that the infiltration of outdoor particles during winter was relatively low owing to the tightly insulated buildings in cold climates. No comparison can be made with our study because no seasonally segregated data is shown. However, it may be assumed that buildings in Barcelona are less well insulated since the Cu indoor/outdoor ratio is 0.9 for Barcelona whilst it is 0.5 for schools in Stockholm. When comparing to Wroclaw (Zwoździak et al., 2013), indoor and outdoor levels of K, Ca, Fe, Cr, Ni, Cu, Zn, Pb and As in Barcelona are lower than in Wroclaw with the exception of mineral tracers in summer owing to the previously stated low rainfall and high resuspension in southern Europe (Querol et al., 2004b, Table S3). A similar pattern is found when comparing to Antwerp for K, Ca, Fe, V, Cr, Ni, Cu, Zn and Pb. Therefore, indoor and outdoor pollutants are higher in schools in Barcelona than in schools in Stockholm and Munich, but lower than those in Wroclaw and in Antwerp. Mineral components are an exception because of the higher resuspension rates found in southern Europe (Querol et al., 2004b) and the higher frequency of sand playgrounds in the schools of Barcelona.

Regarding the spatial variation of air pollutants, an increasing gradient of the outdoor concentrations towards the city centre has been observed for EBC, $NO_2$ and, with one important exception, also UFP (Fig. 2a). Therefore, it may be concluded that the three pollutants have a similar source and spatial dispersion in Barcelona. On the other hand, $PM_{2.5}$ also follows similar patterns although with several exceptions, with high $PM_{2.5}$ levels in many low EBC schools. This again suggests the significance of local school sources of $PM_{2.5}$ in specific cases. However, since most of the schools in the highest tercile of $PM_{2.5}$ concentrations are located close to the city centre, it may also be concluded that despite the school sources that prevent $PM_{2.5}$ from being a good indicator of traffic emissions, road traffic derived particles also contribute to $PM_{2.5}$. The similarities between the patterns of indoor and outdoor suggest a significant infiltration of outdoor pollutants into indoor environments.

The map created with data not seasonally adjusted (Fig. S1) demonstrate that deseasonalisation for EBC and $NO_2$ is fairly reliable, since the application of this methodology refines the expected ascendant gradient of concentrations towards the center of the city. However, this is unsuitable for UFP since its variability is influenced by complex photochemical processes in addition to road traffic emissions (Dall'Osto et al., 2011; Reche et al., 2011). Deseasonalisation could also be inappropriate for $PM_{2.5}$ because it is influenced by local sources at schools.

Since deseasonalisation was carried out using outdoor levels of the UB-PR station, this technique is probably not suitable for indoor levels. Indoor concentration depends on infiltration of outdoor particles and gaseous pollutants (that might differ in every school because of different building characteristics; Adgate et al., 2003; Dockery and Spengler, 1981) and on indoor-sourced PM (which might also be different in schools depending on the activities that take place; Kopperud et al., 2004; Long et al., 2000). Therefore, owing to the added complexity on the indoor environments, outdoor pollutants concentration should not be used to deseasonalise indoor concentrations.

## 5. Conclusions

In Barcelona and Sant Cugat, higher levels of air pollutants are found in school playgrounds and classrooms when simultaneously compared with local UB air quality monitoring site. Outdoor $PM_{2.5}$ levels at schools almost double the usual background levels reported for Barcelona, mainly because of the high $PM_{2.5}$ contribution of local school sources of mineral dust and indoor OC (outdoor OC is mainly the sum of local traffic and city-scale traffic emissions). Outdoor levels are higher than indoors for $NO_2$, UFP, Cu, Sn, among others with typical outdoor sources.

Nevertheless, indoor levels are very close to those found outdoors, indicating a fairly easy penetration of outdoor air pollutants into indoor environments. Moreover, levels of mineral matter components (except Ca) are also higher outdoors and seem to be very dependent on the presence of sand-filled playgrounds (also observed indoors). The unusually high levels of mineral matter found in $PM_{2.5}$ are very significant and suggest the breakdown of mineral particles due to playground activities under the specific Barcelona's climate, Thus, this fine mineral matter is easily resuspended alongside with anthropogenic pollutants (e.g. OC and a few metals) previously deposited in the sand.

By contrast, $PM_{2.5}$ levels are markedly higher indoors, which indicates that a significant fraction of $PM_{2.5}$ mass arises from indoor activities and is characterised mainly by OC (from clothes fibres, organic emissions from children and cooking), but also by Ca, $CO_3^{2-}$ and Sr (from chalk). Since $PM_{2.5}$ is more influenced by indoor sources and mineral matter than traffic emissions or other urban pollutants, $PM_{2.5}$ mass is not a good indicator of traffic pollution in indoor and outdoor environments. However, the levels of $PM_{2.5}$ are also affected by traffic and most of central schools recorded high $PM_{2.5}$ levels. Conversely, levels of EBC, $NO_2$, UFP and specific metals such as Sb were found to be very good indicators of road traffic emissions in the two environments.

Deseasonalisation of the data based on an UB reference station is feasible for outdoor $NO_2$ and EBC, but it is unsuitable for outdoor UFP and $PM_{2.5}$, as well as for indoor concentrations.

Since some traffic tracers such as $NO_2$ are 1.2 times higher in the playgrounds when compared with the local UB reference station, school children in Barcelona are about 20% more exposed to traffic-related pollutants. The levels of pollutants assessed in our study are between those measured at UB and at traffic stations in Barcelona. These stations should, therefore, be considered when characterising child exposure and, possibly, exposure of the general population to air pollutants.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.envint.2014.04.009.

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