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Children exposure to atmospheric particles in indoor of Lisbon primary schools

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ABSTRACT

Evidence continues to emerge showing that poor Indoor Air Quality (IAQ) can cause illness requiring absence from schools, and can cause acute health symptoms that decrease students' performance. Since children spend on average 7–11 h per weekday at school, the IAQ in classrooms is expected to play a key role in the assessment of the effects of their personal exposure to air pollution. Within this context the present study was conducted in order to fulfill three primary objectives 1) to measure the levels and the element composition of $PM_{2.5}$ and $PM_{2.5-10}$, in three primary schools placed in Lisbon, in order to assess the children exposure to these pollutants; 2) to study the relationship between indoor and outdoor atmospheric particles concentrations and 3) to investigate the sources of high aerosols concentrations in classrooms. In the studied classrooms, the concentrations of coarse particles significantly exceeded the ambient levels. Element concentrations suggested that the physical activity of students highly contributed to the re-suspension of sedimented particles. The high levels of CO_2 indicated that in these schools the ventilation was inadequate. This fact contributed to the establishment of poor IAQ.

1. Introduction

Due to the increasing of industrial emissions and traffic, outdoor air quality has become of growing concern during the past decades. Nevertheless, evidence has been made that children spend most of their time in indoor environments and therefore are more exposed to pollution indoors than outdoors. In their critical review, Mendell and Health (2005) concluded that poor indoor environment quality in schools has a great influence on the performance and attendance of students. The exposure to contaminants in such indoor environments may lead children to develop potential health consequences as they are more susceptible to air pollutants than adults because they breath higher volumes of air relative to their body weights and their tissue and organs are growing (Mendell and Health, 2005).

Poor indoor environments in schools may be attributed to three primary causes: i) inexistence or inadequate operation and maintenance of ventilation systems, ii) infrequent and unthoroughly cleaned indoor surfaces, and iii) a large number of students in relation to room area and volume, with constant re-suspension of particles from room surfaces.

Epidemiological studies have consistently shown an association between atmospheric particles pollution and the number of deaths from cancer and cardiovascular and respiratory diseases (Pope et al., 2002). There is also evidence linking particulate air pollution and increases in hospital admissions for respiratory and cardiovascular diseases (Zanobetti and Schwartz, 2005; Wellenius et al., 2006; Middleton et al., 2008). Evidence has pointed towards fine particles, which usually contain hazardous substances and are able to penetrate deep into the human lung provoking inflammation. Reports about measurements of particles in schools have been recently published (Blondeau et al., 2004; Fromme et al., 2007; Goyal and Khare, 2009; Tippayawong et al., 2009). These works showed that there is a growing evidence of comparatively high concentrations of atmospheric particles in classrooms. However, these high concentrations do not necessarily result in higher health risks to students, because the sources and the composition of atmospheric particles in indoor air may differ from those of outdoor air (e.g. Fromme et al., 2008). Therefore, assessing children's exposure to atmospheric particles and the associated health risks requires the knowledge of outdoor and indoor particle composition. In spite of the various studies performed worldwide to assess the students' exposure to particles, only few works were devoted to their element characterization (Molnár et al., 2007;

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Stranger et al., 2008; Fromme et al., 2008; Avigo et al., 2008). Therefore, the composition of indoor atmospheric particles and its sources still need to be clarified.

This study was developed within the project "Impact of Indoor on Human Health". In this project a number of indoor air parameters have been measured in 14 Lisbon Primary Schools including: 1) chemical pollutants (volatile organic compounds, carbon dioxide, particles, nitrogen dioxide); 2) biological pollutants (fungi and bacteria) and 3) physical parameters (temperature and humidity) (Pegas et al., 2009, 2010a,b,c). In this project, element characterization of particles sampled inside schools was done by the first time in Portugal.

The aim of this study was to provide data on compared outdoor and indoor atmospheric particles concentrations levels in school buildings, as well as information on the sources influencing the relationship between outdoor and indoor air quality. Improving the understanding of the sources of the high atmospheric particles in schools, this study will contribute to identify what sort of additional actions should be taken to enforce an effective improvement of IAQ in schools.

2. Materials and methods

2.1. Sampling site and schools description

This study was carried out in Lisbon, which is the largest city of Portugal and the westernmost capital in mainland Europe. Lisbon is located in the west of Portugal, on the Atlantic Ocean coast at the point where the river Tagus flows into the Atlantic. Lisbon has a population of about 500,000 inhabitants in 84.8 km^2 while the metropolitan area of 2870 km^2 has around 2.8 million of inhabitants.

Some source apportionment studies performed in Lisbon showed that the main source of pollutants in this city is the traffic (Almeida et al., 2009a,b). Moreover, Lisbon has an important input of marine aerosol. This is due to the geographic position of Portugal and the dominant western wind regime, influenced by the presence of the semi-permanent Azores high-pressure and the Icelandic low-pressure systems over the North Atlantic Ocean.

Fig. 1 shows the localization of the schools. School 1 is placed in a residential area in a low traffic zone. School 2 is located in the center of the city near a major road. School 3 is in the center of the city but far from a main road. Information concerning the school building characteristics and the number of students attending the lessons were assessed and used in the interpretation of the data. Table 1 presents an overview of these data. All the schools have natural ventilation, which means that there is no forced ventilation or air conditioning system in use; ventilation is done by opening doors and single glazing windows.

2.2. Sampling and chemical analysis

Six Gent samplers were used to collect simultaneously atmospheric particles in three classrooms and corresponding playgrounds in Lisbon. Sampling was performed in two campaigns (May 2009 and November 2009) with two weeks each. Sampling periods started Monday morning and finished Friday afternoon. In order to prevent overloading of the filters, timers were used to turn the pump on (during the first 20 min of each hour) and turn the pump off (during the last 40 min of each hour). Therefore, measurements represent all the day and not only the periods with students' occupation.

The sampling position in classrooms was opposite to the blackboard, about 1 m above the floor level, the level at which the students would normally inhale, and away from the door, thus avoiding disturbances resulting from air currents.

The Gent samplers were equipped with a stacked filter unit, which carried, in two sequential stages, 47 mm Nuclepore polycarbonate filters, with 8 and 0.4 μm pore size. Upstream of the coarse filter a pre-impactor stage was located. The air was sampled at a rate of 15–16 L min^{-1}, which allowed the collection of coarse particles with aerodynamic diameters (AD) between 10 and 2.5 μm, in the first stage, and fine particles with AD $<$ 2.5 μm in the second stage (Maenhaut, 1992).

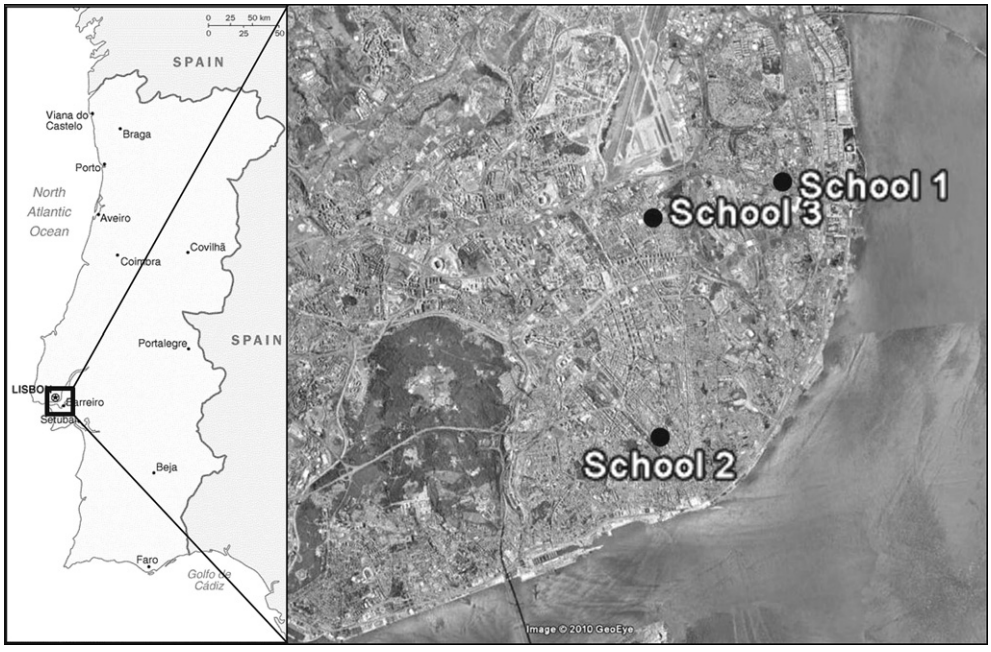

Fig. 1. Spatial distribution of the 3 schools in Lisbon, Portugal.

Table 1
Overview of the school characteristics.

	School 1	School 2	School 3
Area (m^2) × height (m)	65 × 3.7	50 × 3.5	46 × 3.5
Number of students	23	21	24
Ventilation	Natural	Natural	Natural
Windows	Single glazing	Single glazing	Single glazing
Board	Blackboard/ chalk	Blackboard/ chalk	Blackboard/ chalk
Floor covering	Brick	Brick	Brick
Floor	Ground	Ground	Ground
Direct outdoor door	Y	N	N
Classroom adjacent to playground	Y	Y	Y
Classroom adjacent to street	N	N	N
Year of construction	1970	1840	1961

The filter loads were measured by gravimetry in a controlled clean room (class 10,000) with a semi-micro balance. Filter mass before and after sampling was obtained as the average of three measurements, when observed variations were less than 5%.

The exposed filters were analyzed by Instrumental Neutron Activation Analysis (INAA) (Cornelis et al., 1976) with the k_0 methodology (De Corte, 1987).

Previous to the sampling campaign, tests of reproducibility within the filters and between filters were taken, using parallel sampling with two similar sampling units and measuring the particle species by INAA. Results were reproducible to within 5–15%, providing strong support for the validity of the analytical techniques. The details of sampling and analytical control tests are given in Almeida et al. (2003a,b). The accuracy of analytical methods was evaluated with NIST filter standards, revealing results with an agreement of ±10% (Almeida et al., 2006a).

Blank Nuclepore filters were treated in the same way as regular samples. All measured species were very homogeneously distributed; therefore concentrations were corrected by subtracting the filter blank contents.

In order to evaluate the occupancy and the ventilation efficiency, CO_2 concentrations were measured in the campaign performed in May with an automatic portable Indoor Air IQ-610 Quality Probe (GrayWolf monitor).

2.3. Statistical analysis

Statistical calculations were performed using STATISCA software. Wilcoxon Matched pairs and Mann–Whitney *U* were used. These tests are non-parametric – hence they do not consider any assumptions related to the distribution – and basically are the same in that they compare between two medians to suggest whether both samples come from the same population or not. When both of the samples were not entirely independent of each other and had some factor in common, the Wilcoxon Matched pairs test was applied (differences between pairs of indoor and outdoor levels). When the samples were independent Mann–Whitney *U* test was applied (differences between schools and sampling periods). Statistical significance refers to $p < 0.05$.

3. Results and discussion

3.1. $PM_{2.5}$ and $PM_{2.5–10}$ concentrations

Fig. 2 summarizes the indoor and outdoor total $PM_{2.5}$ and $PM_{2.5–10}$ mass concentrations obtained in the two sampling campaigns.

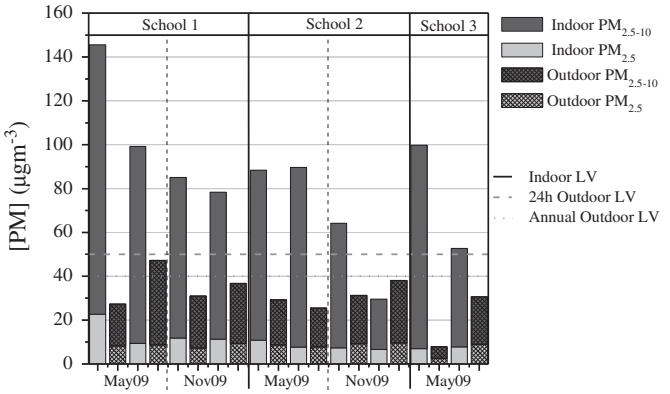

Fig. 2. $PM_{2.5}$ and $PM_{2.5–10}$ mass concentration measured in the indoors and outdoors (values in μg m^{-3}; PM_{10} Limit Value – LV).

The average indoor $PM_{2.5}$ and $PM_{2.5–10}$ total mass concentration was 10 μg m^{-3} and 73 μg m^{-3}, respectively. PM_{10} indoor concentrations varied between 30 μg m^{-3} and 146 μg m^{-3}.

PM_{10} concentrations did not exceed the limit value of 150 μg m^{-3} established by the Portuguese legislation for indoor air (Decreto Lei no. 79, 2006). However, as this study included time periods, in which no students were in class, atmospheric particles concentrations might be underestimating the children exposure to particles. In an investigation in the US, after the modification of the sampling procedure from 24 to 8 h measurements, the mean PM_{10} values were twice as high as before (Yip et al., 2004). Stranger et al. (2008) showed, in a pilot study made in Belgium classrooms, that 12 h $PM_{2.5}$ concentrations exceeded those of 24 h by 40%. In Netherlands, in three schools where PM_{10} measurements were carried out, concentrations during students occupancy were about twice as high as 24 h average classroom concentrations (Janssen et al., 1999). Besides the differences between sampling periods, previous studies have shown that personal exposure often is higher than indoor levels (Molnár et al., 2007).

3.2. Indoor and outdoor concentrations

PM_{10} and $PM_{2.5}$ outdoor concentrations ranged from 8 to 47 μg m^{-3} and from 3 to 10 μg m^{-3}, respectively (Fig. 2). There were no statistically significant differences between $PM_{2.5}$ measured indoors and outdoors ($p = 0.60$). However, statistical analysis showed that $PM_{2.5–10}$ concentrations were significantly higher indoor ($p = 0.00$), indicating that coarse particles measured in classrooms have major sources other than outdoor particles. Our results are in line with findings of previous studies on atmospheric particles levels in indoor air of schools. Blondeau et al. (2004) studied the particle number (within 15 size intervals ranging from 0.3 to 15 μm) in eight schools and observed that indoor concentrations of the finest particles closely track the outdoor ones, while the apparent correlation is far less obvious for larger particles. Occupancy, through re-suspension of previously deposited particles and possible particle generation, strongly influences the indoor concentration level of airborne particles. However, this influence decreases with particle size, reflecting the way deposition velocities vary as a function of size. Fromme et al. (2007) observed increased atmospheric particles concentrations in low level classes and in rooms with high number of pupils, which suggest that the physical activity of pupils (assumed to be more intense in younger children), contributes to a constant process of re-suspension of sedimented particles. Analysis with Scanning Electron Microscopy performed by Fromme et al. (2008) showed that besides particles

Table 2
Indoor and outdoor $PM_{2.5}$ and $PM_{2.5-10}$ element concentrations.

		$PM_{2.5}$		$PM_{2.5-10}$	
		Indoor ($N = 8$)	Outdoor ($N = 8$)	Indoor ($N = 8$)	Outdoor ($N = 8$)
As	ng m^{-3}	0.262 ± 0.104	0.253 ± 0.094	0.802 ± 0.202	0.242 ± 0.141
Ba	ng m^{-3}	4.237 ± 5.489	5.449 ± 4.362	32.721 ± 17.760	19.847 ± 22.762
Br	ng m^{-3}	(a)	(a)	6.322 ± 1.811	6.418 ± 2.821
Ca	μg m^{-3}	4.658 ± 5.258	2.596 ± 2.360	10.005 ± 3.701	2.271 ± 1.495
Co	ng m^{-3}	0.145 ± 0.216	0.206 ± 0.292	0.561 ± 0.352	0.213 ± 0.125
Cr	ng m^{-3}	(a)	(a)	10.810 ± 2.855	6.004 ± 3.332
Fe	μg m^{-3}	0.140 ± 0.075	0.110 ± 0.023	0.856 ± 0.422	0.307 ± 0.106
Hf	ng m^{-3}	0.031 ± 0.034	0.006 ± 0.001	0.109 ± 0.054	0.045 ± 0.014
K	μg m^{-3}	0.115 ± 0.048	0.085 ± 0.060	1.066 ± 0.419	0.188 ± 0.077
La	ng m^{-3}	0.058 ± 0.061	0.006 ± 0.002	0.764 ± 0.234	0.212 ± 0.129
Na	μg m^{-3}	0.212 ± 0.111	0.555 ± 0.202	1.537 ± 0.608	1.846 ± 0.686
Sb	ng m^{-3}	0.778 ± 0.580	1.075 ± 0.850	3.884 ± 2.889	1.328 ± 0.390
Sc	ng m^{-3}	0.023 ± 0.019	0.043 ± 0.074	0.161 ± 0.110	0.055 ± 0.039
Se	ng m^{-3}	0.732 ± 0.935	0.402 ± 0.244	0.487 ± 0.185	0.197 ± 0.079
Sm	ng m^{-3}	0.019 ± 0.012	0.050 ± 0.071	0.144 ± 0.070	0.040 ± 0.027
Zn	ng m^{-3}	8.203 ± 3.648	5.905 ± 2.709	85.951 ± 32.121	17.104 ± 4.730

(a) Below the detection limit.

with mineral origin (resulting from re-suspension) additional coarse particles arise from semi-transparent skin flakes.

For smaller particles, other processes such as homogeneous reactions between ozone and terpenes (coming from cleaning products, perfumes, personal care products), may also contribute to indoor particles (Sarwar et al., 2003; Weschler and Shields, 2003).

Significant high concentrations of particles were registered in the classroom from school 1. In this school, the doors of the classrooms open directly to the playground, whereas in the others schools, the doors of the classrooms open to the interior of the building. This fact can contribute to increase the transport of dust from the playground to the interior of the classroom from school 1.

3.3. Element concentrations

The average of the indoor and outdoor element concentrations measured both inside and outside of the schools is presented in Table 2. In the fine fraction, indoor and outdoor concentrations were not significantly different, except for Na, which originates essentially from the sea. In the coarse fraction, the indoor concentrations of Fe, La, Sc, Sm, K, Ca, Cr, Se and Zn were found to be enriched in comparison with the outdoor concentrations.

The crustal enrichment factor method has been used as an attempt to evaluate the strength of the crustal and non-crustal origin of the elements. Enrichment factors, using Sc as a crustal reference element (EF_{Sc}) were calculated based on Eq. (1) and using Mason and Moore (1982) soil composition (Fig. 3):

$$EF_{sc} = \frac{\left(\frac{[x]}{[Sc]}\right)_{PM}}{\left(\frac{[x]}{[Sc]}\right)_{crust}} \quad (1)$$

Sc was used as a reference element because previous studies performed in Lisbon showed that this element has a crustal origin with a negligible anthropogenic source to the atmosphere (Almeida et al., 2005). Given the local variation in soil composition, $EF_{Sc} > 10$ suggests that a significant fraction of the element is contributed by non-crustal sources.

EF_{Sc} indicated that Co, Fe, La, Sc, Sm and K were associated with soil emissions. These results agreed with source apportionment studies performed in the Lisbon region (Almeida et al., 2005, 2006b). Indoor concentrations for these elements in $PM_{2.5-10}$ were significantly higher than in outdoors. This fact indicates that the most probable cause of increased classroom coarse particle concentrations was the penetration of mineral dust through the windows and the re-suspension of settled dust or suspension of soil material brought in by the children's shoes.

For the strongly enriched elements ($EF_{Sc} > 10$) like As, Ba, Br, Ca, Cr, Sb, Se and Zn, an anthropogenic origin can be suggested, such as traffic and industrial emissions. In the fine fraction, the indoor and outdoor concentrations for these elements (except for Cr, which was below the detection limit) were not significantly different, suggesting that air infiltration is the main source for them. In the coarse fraction significant high concentrations were found for Ca, Cr, Sb, Se and Zn indicating the existence of an indoor source for these elements.

At both indoor and outdoor particles, Ca presented $EF_{Sc} > 10$ indicating the existence of non-mineral sources associated with this element. In the coarse fraction, Ca was detected at very high concentrations (10,000 ng m^{-3}). This lead to the idea that Ca could be attributed to a real indoor source, probably originating from the chalk (mainly $CaSO_4$), used in the blackboards, and/or the gypsum walls and plasters used as building materials.

In $PM_{2.5-10}$, Cr, Sb and Se presented significant high concentrations in the classrooms. However, the indoor EF_{Sc} for these

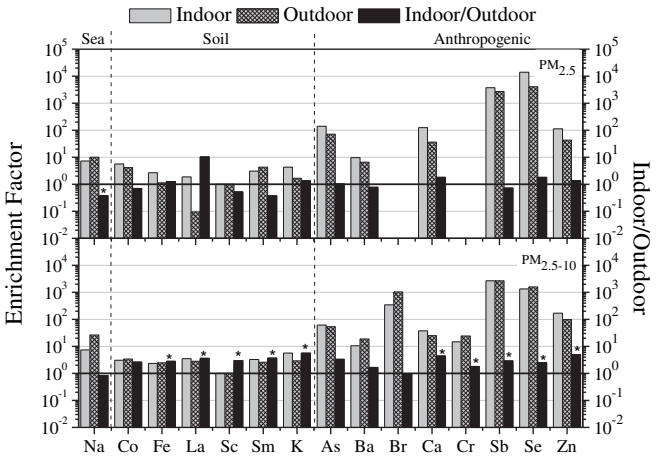

Fig. 3. Enrichment factor using Sc as a reference element and Mason and Moore (1982) soil composition. Average Indoor/Outdoor ratios for the element mass concentration (* – indoor and outdoor concentrations which are significantly different).

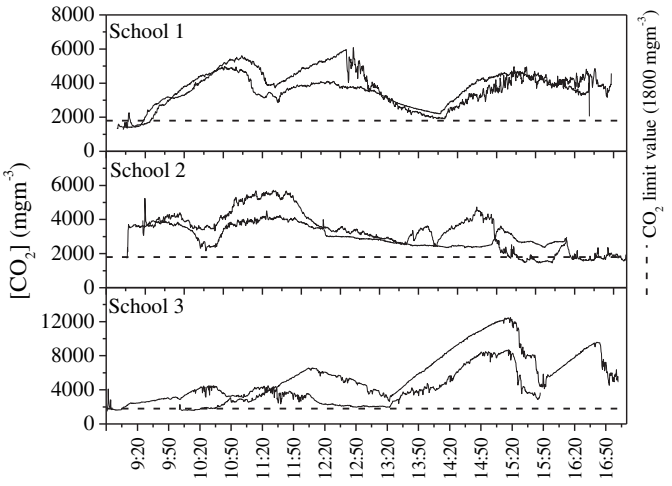

Fig. 4. CO_2 concentrations measured during two working days at the three schools (values in mg m^{-3}).

elements were lower, indicating the existence of a mixture of anthropogenic elements with dust, which was re-suspended.

Inside the classrooms significant high concentrations of Zn were measured for the coarse fraction (for $PM_{2.5}$ a *p*-value of 0.063 by Wilcoxon Matched Pair Test was found). This fact has already been observed by Avigo Jr. et al. (2008) in Brazilian elementary schools and indicates the existence of a Zn indoor source. In the market, several products using Zn, to be applied indoors to protect steel, walls, wood surfaces, doors and windows, are sold.

The outdoor concentrations of Na were found to be enriched in comparison with the indoor concentrations. This fact was expected as this element has marine origin.

Our results can support the idea posed by other authors (Fromme et al., 2008) that indoor-generated particles may have lower health impacts compared to outdoor particles. In one hand, indoor-generated particles are more associated with the coarse fraction, and in the other hand these particles have principally a crustal origin. This would be in line with results obtained by Köenig et al. (2005) that shown that indoor-generated component of $PM_{2.5}$ is less associated with inflammation markers in asthmatic children than the ambient-generated component. Laden et al. (2000) showed that crustal particles in the fine fraction are not associated with increased mortality.

3.4. CO_2 concentrations

CO_2 concentrations are often used as an indicator of ventilation efficiency and excess of occupancy. Fig. 4 depicts the variation of indoor CO_2 concentrations measured during two working days at the three schools, showing strong correlation of the CO_2 level with occupancy. Higher CO_2 concentrations were observed when students started physical activities inside the classrooms and lower concentrations were associated with the beginning of the day and lunch time. The observed values highly exceeded the CO_2 limit value (1800 mg m^{-3}) established by the Portuguese Legislation for IAQ (Decreto Lei no. 79, 2006). The continuously high CO_2 values indicated insufficient ventilation in schools. This lack of ventilation may inhibit the transport and removal of larger particles from indoor to outdoors.

4. Conclusion

This study provides information on magnitude and element characterization of particles as well as relationships between size-dependent indoor and outdoor concentrations. Besides characterizing the particles in classrooms and identifying possible sources, these findings may be used as a valuable contribution to improve the IAQ and to suggest effective mitigation strategies.

The element composition of the classrooms' particles suggested that earth crustal materials, detritions of the building materials and chalk have an important role in the particles levels and characteristics. Physical activity of the pupils led to re-suspension of mainly coarse particles and largely contributed to increased $PM_{2.5-10}$ concentration in classrooms. Assuming that crustal materials and combustion-related particles vary in toxicity, our findings on concentrations of particle components support the hypothesis that indoor-generated APM may be less toxic compared to APM in ambient air.

Results showed that local measurements of outdoor air concentrations do not provide an accurate estimation of the children's personal exposure to particles and specific elements because they spend most of their time inside the buildings. Therefore, the composition of atmospheric particles indoors is a concern and needs further investigation.

Acknowledgements

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