Science of the Total Environment 835 (2022) 155349

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Source apportionment of children daily exposure to particulate matter

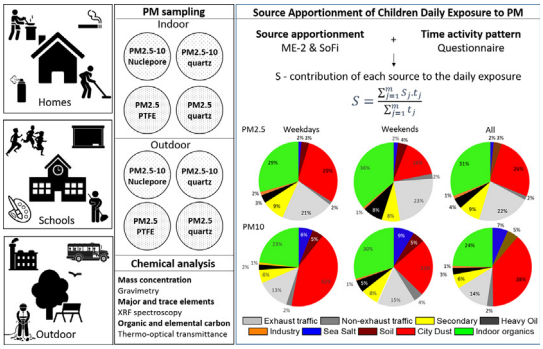
Susana Marta Almeida [a,*], Tiago Faria [a], Vânia Martins [a], Nuno Canha [a], Evangelia Diapouli [b], Konstantinos Eleftheriadis [b], Manousos Ioannis Manousakas [c]

[a] Centro de Ciências e Tecnologias Nucleares, Instituto Superior Técnico, Universidade de Lisboa, Estrada Nacional 10, 2695-066 Bobadela-LRS, Portugal
[b] Institute of Nuclear & Radiological Sciences and Technology, Energy & Safety, National Centre for Scientific Research "Demokritos", Agia Paraskevi, 15310 Athens, Greece
[c] Laboratory of Atmospheric Chemistry, Paul Scherrer Institute (PSI), 5232, Villigen PSI, Switzerland

HIGHLIGHTS

- Receptor models were used to estimate the contribution of sources to PM exposure.
- Nine outdoor and indoor sources were identified.
- Indoor organics was the main contributor for the integrated exposure to PM2.5.
- Dust resuspension was the main reason for the high PM10 concentrations in schools.
- Traffic contributed on average by 24% to the children's exposure to PM2.5.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Anastasia Paschalidou

Keywords:
Aerosol
Indoor
Outdoor
Homes
Schools
Receptor models

ABSTRACT

The present study aims to investigate the sources of particulate pollution in indoor and outdoor environments, with focus on determining their contribution to the exposure of children to airborne particulate matter (PM). To this end, parallel indoor and outdoor measurements were carried out for a selection of 40 homes and 5 schools between September 2017 and October 2018. PM2.5 and PM2.5–10 samples were collected during five days in each microenvironment (ME) and analysed by X-Ray Fluorescence (XRF), for the determination of elements, and by a thermal-optical technique, for the measurement of organic and elemental carbon. The source apportionment analysis of the PM composition data, by means of the receptor model SoFi (Source Finder) 8 Pro, resulted in the identification of nine sources: exhaust and non-exhaust emissions from traffic, secondary particles, heavy oil combustion, industry, sea salt, soil, city dust, and an indoor source characterized by high levels of organic carbon. Integrated daily exposure to PM2.5 was on average 21 μg/m^3. The organic matter, resulting from cleaning, cooking, smoking and biological material, was the major source contributing by 31% to the PM2.5 exposure. The source city dust, which was highly influenced by the resuspension of dust in classrooms, was the second main source (26%), followed by traffic (24%). The major sources affecting the integrated exposure to PM10, which was on average 33 μg/m^3, were the city dust (39%), indoor organics (24%) and traffic (16%). This study provides important information for the design of measures to reduce the exposure of children to PM.

1. Introduction

Air quality management policies, aiming at protecting the citizens' health, are based on ambient concentration levels, assessed by national air quality measurement networks, assuming that the corresponding monitoring stations are representative of the population exposure across a given

* Corresponding author.
E-mail address: smarta@ctn.tecnico.ulisboa.pt (S.M. Almeida).

http://dx.doi.org/10.1016/j.scitotenv.2022.155349
Received 3 February 2022; Received in revised form 13 April 2022; Accepted 13 April 2022
Available online 21 April 2022

city. However, ambient outdoor concentrations may lead to exposure misclassification, which is likely to be determined by the individual's time-activity profiles including time spent in indoor environments (Almeida et al., 2016; Faria et al., 2020; Faria et al., 2022). Since people spend about 90% of their time indoors, individual's exposure to air pollutants is dominated by indoor air quality, which is affected by both the building characteristics and indoor pollution sources (Morawska et al., 2013). Indoor air pollutants result from outdoor air and from indoor sources, including the presence of occupants, human activities, combustion processes, building materials, and consumer products (MacNeill et al., 2014; Morawska et al., 2017; Urso et al., 2015). Indoor chemistry is itself a source of chemicals that might otherwise not be present indoors (Abbatt and Wang, 2020). Indoor chemistry is governed by different factors and processes than those that control outdoor atmospheric chemistry for several reasons, such as absence of direct sunlight and rain, less extreme temperature fluctuations, much larger surface-to-volume ratios, and much higher concentrations of organic compounds (Farmer and Vance, 2019; Weschler and Carslaw, 2018). Therefore, highly variable exposure conditions among different buildings can result in large exposures and health risks for a subset of the population, which may not correspond only to people living in areas identified by the national air quality networks as heavily polluted.

Particulate matter (PM) is a key air pollutant in terms of adverse health effects. Epidemiological and clinical studies have demonstrated particles to be a risk factor for several respiratory disease, such as lung cancer, chronic obstructive pulmonary disease and asthma (Kulhánová et al., 2018; Lu et al., 2020; Mateen and Brook, 2011), along with numerous cardiovascular disease, such as stroke, coronary events, myocardial infarction, cerebrovascular events, atherosclerosis, and deep vein thrombosis (Gu et al., 2017; Mateen and Brook, 2011; Miller et al., 2007). In general, children are more susceptible to PM than adults, because their respiratory, immune, central nervous, digestive and reproductive systems are still evolving (Salvi, 2007; Suwanwaiphatthana et al., 2010). Moreover, children have higher inhalation rates per body weight and higher pulmonary specific surface area as compared to adults (Bateson and Schwartz, 2008). Deep breathing pulls PM faster and further into the lungs, bypassing initial areas of deposition (Zwozdziak et al., 2016). According to Ginsberg et al. (2005) the pulmonary region of the lung has slower clearance, therefore PM remain there longer, and consequently the particle dose can be two to four fold higher among young children.

Europe is moving towards the implementation of the Thematic Strategy on Air Pollution, where Member States are required to draw up plans and programs to guarantee compliance with legally binding limit values for air pollutants. However, the current policy efforts, at European and national level, have not fully delivered the expected results. In spite of the numerous efforts, 97% of the urban population in Europe is exposed to concentrations of fine particles (PM2.5) above the new World Health Organization (WHO) guideline level of 5 $\mu g/m^3$ (WHO, 2021) and serious air pollution impacts on health still persist. Prompt action through efficient air quality management, considering outdoor and indoor sources, is required not only to ensure that the outdoor air legal limits are not exceeded, but principally to guarantee that the consequences of poor air quality on human heath are controlled and minimized. Therefore, management of air quality requires quantitative estimates of population integrated exposure to pollutants, considering all the microenvironments (MEs) frequented by the individuals during their daily activities, as well as the influence of the different PM sources and factors affecting this exposure (Morawska et al., 2013).

While the major sources of indoor air PM have been identified, few studies have attempted to estimate the contribution of specific sources using receptor-modeling techniques. Receptor models are statistical procedures for identifying and quantifying the sources of air pollutants at a receptor location. Receptor models have been extensively used for outdoor air quality (Almeida et al., 2020; Amato et al., 2016; Karagulian et al., 2015) but few studies on indoor PM have been conducted (Amato et al., 2014; Kalaiarasan et al., 2017; Zhao et al., 2007), mainly because the presence of both indoor and outdoor sources, the building-related mechanisms (e.g. ventilation and infiltration), as well as the outdoor meteorology and long range transport of pollutants makes Source Apportionment (SA) challenging (Bekö et al., 2020).

This study aims at improving the understanding of the sources affecting the indoor and outdoor environments and how these sources contribute to the overall exposure, focusing on elementary school children. To the best of our knowledge, there is a limited number of SA studies at indoor school and home MEs, while the contribution of the different sources to the daily exposure of children is still far from being well understood.

2. Methodology

2.1. Assessment of time-microenvironment-activity pattern in Lisbon

A necessary step in measuring the extent of children's exposure to air pollutants and assessing the sources affecting this exposure is to evaluate where they spend their time. Therefore, data on children time-microenvironment-activity was collected using a questionnaire that was distributed in 26 schools and applied to children, aged between 5 and 10 years-old, living and studying in Lisbon. The questionnaire was developed in two main parts: the first part was dedicated to the characterization of the children and location of their houses and schools; and the second part focused on the characterization of the time activity pattern of children during the weekdays and weekends. The time was divided in periods of 30 min and 17 MEs were considered. The questionnaires were filled by the parents of the children. To increase the interest of the school community and the response rate, the teamwork put into action the awareness campaign “The air belongs to everyone” that aimed to aware students and teachers to the problem of air quality. 6096 questionnaires were delivered, 1251 of the parents returned the questionnaires, but only 1189 were completed, representing a response rate of 20%. The questionnaire and more details about its application and results can be consulted in LIFE Index-Air (2017) report. The results of the questionnaire were essential to identify the MEs where children spend more time and to select the sampling locations.

2.2. Air quality measurements

Air quality measurements were carried out indoors and outdoors of 40 homes and 5 primary schools located in Lisbon, between September 2017 and October 2018. Homes and schools were carefully selected in order to represent different zones of the city and different building characteristics (age, envelope, materials, etc.). The sampling was made in more homes than classrooms due to the higher variability of the conditions, sources and concentrations in dwellings and to the less number of works focusing the indoor air quality in this ME, which increases the need of more knowledge about processes and sources inside homes. The sampling at each home and school lasted for 5 days, during the occupancy hours, i.e. in schools, 8 h from Monday to Friday and in homes 15 h during the week (from 18:00 to 9:00) and 24 h during the weekend. Outdoor air measurements followed the same time schedule as indoor sampling.

PM was sampled concurrently indoors and outdoors using four medium volume samplers (MVS6, Leckel, Sven Leckel, Germany). The Leckel samplers were equipped with a sampling head for the simultaneous collection of PM2.5 and PM2.5–10 at a constant flow rate of 2.3 m^3 (Faria et al., 2020). Two samplers were installed indoors (in living-room or classroom) with air inlets at roughly 1 m above the floor level, corresponding to the breathing level of the children, and away from the door, thus avoiding disturbances resulting from air currents. The other two samplers were installed outdoors, in the balcony or playground. The use of two samplers in each site allowed the collection of PM at two different filter substrates and consequently the chemical analyses of filters by different techniques. One sampler collected PM2.5 and PM2.5–10 on Quartz fibre filters (Pall) and the other sampler collected PM2.5–10 in Nuclepore filters with 0.4 μm pore size (Whatman) and PM2.5 in polytetrafluoroethylene filters (PTFE) with 2 μm pore size (Whatman).

Filters were weighted before and after sampling in a laboratory with controlled temperature and humidity, using a microbalance (Sartorius R160P, Greifensee, Switzerland). PM mass concentrations were determined by dividing filter loads by the volume of air filtered. PM10 concentrations were obtained by summing PM2.5 and PM2.5–10 concentrations. PM samples collected on Nuclepore and PTFE filters were analysed by X-Ray Fluorescence (XRF), to determine the concentrations of Na, Mg, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Sr, Ba, and Pb, using an Energy Dispersive X-ray Spectrometer (ED-XRF) Laboratory Instrument (Epsilon 5, PANalytical, the Netherlands), following the methodology described at Manousakas et al. (2018). PM samples collected on Quartz filters were analysed by the Thermo-Optical Transmittance method for the determination of Organic Carbon (OC) and Elemental Carbon (EC) using the Lab OC-EC Aerosol Analyzer (Sunset Laboratory Inc., USA) and the EUSAAR2 protocol. More details about sampling, chemical analysis and quality assurance and control are described in Faria et al. (2020) and Faria et al. (2022).

2.3. Source apportionment

Source apportionment was implemented through the multilinear engine-2 (ME-2) solver and controlled via SoFi (Source Finder) 8 Pro, which allows for a comprehensive and systematic PMF analysis (Canonaco et al., 2013).

Source apportionment of PM was performed by receptor modeling that is based on the mass conservation principle:

$$xij = \sum_{k=1}^{p} gik\, fkj + eij \quad (1)$$

where *xij* is the concentration of the j^{th} species in the i^{th} sample, *gik* is the contribution of the k^{th} source in the i^{th} sample, *fkj* is the concentration of the j^{th} species in the k^{th} source k, and *eij* is the residual of each individual measurement result.

Data below the limit of detection (LOD) were substituted by half of the LOD and the uncertainties were set to 5/6 of the LOD. Missing data were substituted by the geometric mean of the measured concentrations and the corresponding uncertainties were set as 4 times these geometric mean (Polissar et al., 2001).

Chemical species with high noise were down-weighted based on their signal-to-noise (S/N) ratio to reduce the influence of poor variables on the analysis. Species with S/N lower than 0.5 were considered as bad variables and excluded from the analysis, and species with S/N between 0.5 and 1 were defined as weak variables and down-weighted by increasing the uncertainty.

ME-2 allows exploring the rotational space around the base solution by introducing limits into the Positive Matrix Factorization (PMF) model for deviation from predetermined values for *gik* and/or *fkj* for one or more factors (Paatero, 1999). This approach is referred to as constraining. In the current study a constraining technique that is called *a*-value approach was used. In this approach one or more output factor profiles are required to be within predefined limits of a reference profile, with the tightness of constraint defined by the scalar a ($0 \le a \le 1$). The scalar a defined the degree of freedom ($a = 0$ means 0% allowed deviation from the anchor profile, and $a = 1$ means 100% allowed deviation).

For identifying the indoor and outdoor sources, four different datasets were used: PM2.5 indoor and outdoor combining data from schools and houses in each case, as wells as PM10 indoor and outdoor combining data from schools and houses in each case. The total number of variables used was 22 (OC, EC, Na, Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Br, Sr, Ba, Pb). To estimate the factor contribution, PM was also used as species in the analysis with a 300% uncertainty assigned.

A novel methodology was used for the identification of indoor and outdoor sources, based on the *a*-value approach. First, the outdoor sources for both PM10 and PM2.5 datasets were identified by performing fully unconstrained analysis on the datasets. Then, the factor profiles that were obtained from the analysis of the outdoor data, were used to constrain the indoor data analysis. In particular, the indoor data source apportionment analysis was performed by constraining the outdoor profiles on the indoor runs with an *a*-value of 0.2 (all species except PM were constrained), and adding an additional unconstrained source. Since the indoor PM levels are the sum of outdoor contributions plus indoor contributions, by using this approach we were able to successfully characterize the influence of the outdoor sources in the indoors environment, as opposed to those that are exclusively indoor. The chemical profiles of the outdoor sources are not expected to remain unchanged in the indoor MEs, due to infiltration or/and other processes that can take place in an indoor environment; the selected *a*-value of 0.2 (20% change allowed) provides enough space to the model to adapt the outdoor source profiles to those changes. By following this approach two main goals are achieved: (i) the contribution/effect of outdoor sources to the indoor environment is accurately estimated, and (ii) the contribution of exclusively indoor sources is accurately estimated.

A sensitivity analysis was performed to determine the used *a*-value and the number of extra sources indoors in comparison to the outdoor ones. As stated before, an *a*-value equal to 0.2 and one extra indoor source provided the optimum results. Including more indoor sources did not lead to extra sources with reasonable source profiles (more OC sources with no other tracers were produced), while increasing the *a*-value did not lead to significant changes in the factor profiles. The small change in the Q values (<1%) before and after the application of the constraints, indicates that the solutions are mathematically equal and the model is not forced to deviate too much from the unconstrained solution.

Three main reasons are the cause of uncertainties in source apportionment analysis: random errors in data values, rotational ambiguity, and modeling errors. Modeling errors are identified by monitoring the residuals of the solution. The residuals were normally distributed, unstructured over time and variables throughout all ranges as suggested by Reff et al. (2007). The rotational ambiguity was investigated using the methodology described in Canonaco et al. (2021). This method is based on a combination of the *a*-value approach (information about the rotational ambiguity) and the classical bootstrapping approach (information about the statistical uncertainty and random errors) (Ulbrich et al., 2009). All mathematical indicators suggest source apportionment solution with low uncertainty. The optimum number of factors was decided by examining the mathematical diagnostics (Q/Q_{exp}, scaled residuals, structure of the residuals, unexplained variation), as well as the physical meaning of the factors.

The contribution of each source to the daily exposure of children (S) was calculated by integrating the time-activity data, provided from the time activity questionnaire, with the contribution of the source in the different MEs, according to Eq. (2):

$$S = \frac{\sum_{j=1}^{m} S_j \cdot t_j}{\sum_{j=1}^{m} t_j} \quad (2)$$

where S_j is the source contribution in the ME (j) and t_j is the time spent in the ME (j).

2.4. Statistical analysis

The non-parametric Wilcoxon Matched pairs test was applied to assess the differences between pairs of indoor and outdoor levels and source contributions and the Mann–Whitney *U* test was selected to evaluate the differences between schools and homes. Spearman's correlation coefficients were calculated to assess the contribution from infiltration and/or indoor sources to the observed chemical species concentrations. Statistical significance refers to $p < 0.05$. All statistical analysis were implemented in STATISTICA software.

3. Results and discussion

3.1. Children's daily time activity patterns

In this work a time activity pattern survey targeting children between 5 and 10 years old from Lisbon was performed based on a questionnaire.

Results showed that during the weekday, children spent 89% of their time indoors – 57% in home and 26% in classrooms and that only 10% of their time was spent outdoors. During the weekend, children spent 87% of their time indoors, increasing the time spent in home and leisure places, in comparison to the weekdays (Fig. 1). The analysis of the children's time activity pattern showed the importance of the indoor MEs for the children's daily exposure.

3.2. PM concentrations

Table 1 reports the average concentrations of indoor and outdoor PM total and chemical compounds mass, as well as the children integrated exposure levels calculated by integrating the time-activity data with the concentrations in the different MEs according to Eq. 3.

$$E = \frac{\sum_{j=1}^{m} C_j \cdot t_j}{\sum_{j=1}^{m} t_j} \quad (3)$$

where C_j is the PM concentration measured in the ME (j) and t_j is the time spent in the ME (j).

The concentrations and size distribution of the particles sampled in schools and homes differed significantly (Martins et al., 2020). The schools displayed higher concentrations (PM2.5: 35 μg/m^3 and 14 μg/m^3; PM10: 65 μg/m^3 and 18 μg/m^3, for school and homes, respectively) and I/O ratios (PM2.5: 2.4 and 1.4; PM10: 2.3 and 0.9, for school and homes, respectively), and lower PM2.5/PM10 ratios (schools: 0.57; homes: 0.78).

While the school environment normally lacks typical indoor PM sources such as cooking and heating, results indicated that the classrooms is a ME characterized by high concentration of PM, principally in coarse fraction. The reason for that can be attributed to insufficient ventilation in schools, unappropriated cleaning practices, a large number of students in a limited room area over a period of several hours (average density of 2 m^2 per student), a constant resuspension of particles from room surfaces, and the close proximity of the schools to busy roads. Faria et al. (2020) used a laser photometer (DustTrak, model 8533, TSI) in the same five schools of the present study to measure PM2.5 and PM10 in real-time providing information on the diurnal variability, as well as impact of the occupancy and emission sources. This work showed that there is an increase of PM concentrations before the beginning of the classes (9:00) due to the cleaning of the room, which takes place every day before the first class, and during the class-hours due to the presence of students in the room, leading to the generation and resuspension of particles.

Although students spent less time in classrooms (6.1 h) than in homes (13.2 h), exposure in schools led to a substantial contribution to their total daily exposure to PM, due to the higher concentrations measured in this ME. The personal exposure to PM2.5 and PM10, calculated based on the children time-microenvironment-activity pattern and the concentrations measured in homes, classrooms and outdoor, was 21 μg/m^3 and 33 μg/m^3, respectively. PM2.5 exposure exceeded the new 24 h WHO guideline of 15 μg/m^3 and the annual guideline of 5 μg/m^3 (WHO, 2021). PM10 exposure did not exceed the 24 h WHO guideline of 45 μg/m^3, but highly exceeded the annual guideline of 15 μg/m^3.

3.3. Composition of PM

The major compounds found in the PM were OC, EC, S, Ca, Na, Cl, Si, K and Fe. Although the other compounds did not contribute significantly to the PM mass, they provided valuable insights in the identification of PM sources and health impact assessment, as we will observe in the source apportionment section.

Spearman's correlation coefficients (Rs) were calculated to evaluate the contribution from infiltration and/or indoor sources to the observed chemical species concentrations. The highest correlations were calculated for those species of exclusively outdoor origin, such as traffic related pollutants (EC, 0.89–0.93) and secondary aerosol (S, 0.70–0.95).

OC average concentrations were significantly higher in homes (PM2.5: 6.2 μg/m^3; PM10: 7.5 μg/m^3) and schools (PM2.5: 13 μg/m^3; PM10: 21 μg/m^3) than in the respective outdoors, which indicates the importance of indoor sources of OC, such as cleaning, cooking, heating and smoking. In the homes of smokers, OC and K increased 4.7 and 5.9 times in PM2.5 and 4.0 and 5.1 times in PM10, respectively, indicating that these two compounds are good indicators of smoking, as already indicated by the study developed by Zhao et al. (2007).

In homes, the ratio I/O for the major part of the chemical compounds was higher in PM2.5 than in PM10, probably due to the contribution of the coarse fraction that has reduced penetration efficiency and faster settling times, as showed by Hussein et al. (2007) and Martins et al. (2020).

In schools the I/O ratios were higher than in homes, principally for mineral species, reflecting the importance of the particle resuspension by the children's activity, which is a significant source of indoor coarse particles in schools (Almeida et al., 2011; Fromme et al., 2008; Kim et al., 2017). According to Morawska et al. (2017) this source is likely the main reason for the elevated PM10 concentrations in this ME.

3.4. Source apportionment

The source apportionment analysis was conducted to resolve the sources and quantify their contribution to PM in homes, schools and ambient environment. The most reliable solution identified nine source factors. Fig. 2 shows that a very good agreement was obtained between the gravimetric and the sum of the source contributions ($0.77 < R^2 < 0.86$).

The nine sources were identified as exhaust traffic, non-exhaust traffic, secondary sulfate, heavy oil combustion, industry, sea salt, soil, city dust and indoor organics. Factor profiles are displayed in Tables S1 to S4 in

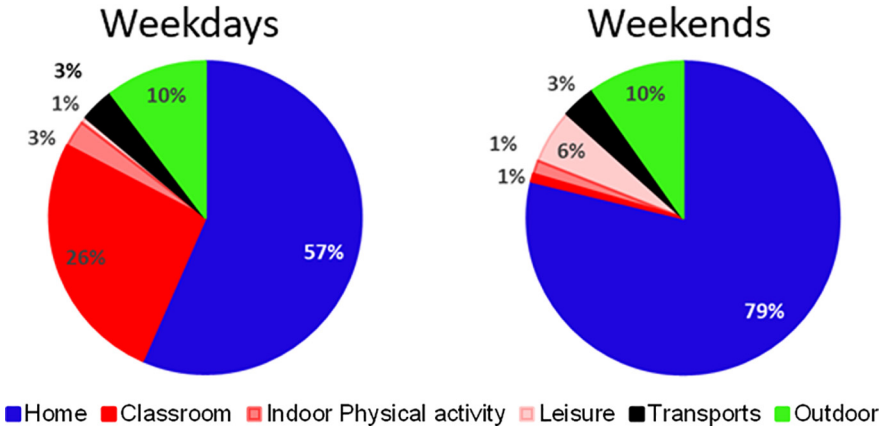


Fig. 1. Time activity pattern for children between 5 and 10 years old from Lisbon for weekdays and weekends.

Table 1
Average concentrations measured in the indoor and outdoor of homes and schools and average individual exposure. PM, OC and EC in μg/m^3 and elements in ng/m^3. Indoor/outdoor (I/O) ratios (significant differences at $p < 0.05$ in bold) and Spearman correlation (Rs) between indoor and outdoor concentrations (significant correlations at $p < 0.05$ in bold).

	PM2.5									PM10								
	Houses				Schools				Exposure	Houses				Schools				Exposure
	Indoor	Outdoor	I/O	Rs	Indoor	Outdoor	I/O	Rs		Indoor	Outdoor	I/O	Rs	Indoor	Outdoor	I/O	Rs	
PM	14	13	1.4	**0.63**	35	21	**2.4**	**0.47**	21	18	22	**0.9**	**0.57**	65	32	**2.3**	0.24	33
OC	6.2	3.0	**2.8**	**0.59**	13	5.1	**2.7**	0.34	7.9	7.5	3.8	**2.6**	**0.50**	21	6.7	**3.4**	0.25	11
EC	1.0	1.2	**0.9**	**0.92**	1.3	1.3	1.0	**0.92**	1.2	1.1	1.3	**1.0**	**0.89**	1.7	1.4	**1.3**	**0.93**	1.3
Al	56	69	**2.1**	**0.64**	410	170	**2.7**	0.16	170	87	130	**1.6**	**0.59**	660	310	**2.4**	**0.56**	280
Ba	1.5	1.7	0.9	0.20	6.9	15	0.5	−0.09	3.0	3.4	8.4	0.9	−0.11	17	27	1.2	−0.40	7.5
Br	1.9	2.4	**0.9**	**0.73**	5.8	2.5	1.0	−0.80	3.1	2.8	5.0	**0.8**	**0.32**	7.1	5.2	0.8	0.61	4.3
Ca	200	200	1.5	**0.59**	2500	720	**5.9**	**0.59**	918	380	560	**0.9**	**0.50**	5900	1500	**5.3**	0.30	2100
Cl	170	160	**2.6**	**0.77**	240	230	3.1	**0.81**	195	480	1000	**0.5**	**0.84**	670	1000	**0.9**	**0.87**	590
Cr	0.54	0.83	**1.0**	**0.46**	2.6	0.71	1.6	0.50	1.15	0.91	2.0	**0.8**	**0.56**	5.1	1.8	**3.3**	**0.53**	2.2
Cu	10	10	1.8	**0.26**	16	14	1.9	0.45	12	13	17	**1.4**	**0.35**	19	19	1.3	**0.49**	15
Fe	100	190	**0.7**	**0.83**	400	270	**1.4**	**0.81**	208	180	460	**0.4**	**0.75**	800	600	1.2	**0.80**	400
K	160	140	1.8	**0.82**	370	300	**1.3**	**0.76**	239	190	200	**1.1**	**0.76**	500	400	1.3	**0.73**	300
Mg	13	19	1.3	**0.63**	100	21	3.2	0.49	41	18	52	**0.8**	**0.52**	120	49	3.9	0.80	50
Mn	2.0	3.1	1.0	**0.60**	8.9	5.7	**1.6**	0.36	4.4	3.3	7.1	**0.7**	**0.47**	16	9.8	**1.9**	**0.46**	7.8
Na	200	240	**1.0**	**0.78**	300	240	1.3	**0.84**	234	290	440	**0.8**	**0.77**	450	440	1.4	**0.78**	360
Ni	0.94	0.90	**1.0**	**0.66**	0.78	0.56	1.4	−0.11	0.85	1.2	1.4	1.5	**0.56**	1.5	1.2	2.3	−0.30	1.3
Pb	3.6	4.2	1.0	**0.58**	13	11	**1.4**	0.45	7.1	5.4	6.5	1.2	**0.42**	18	14	2.5	0.37	9.9
S	480	540	**0.9**	**0.95**	720	550	1.2	**0.91**	557	530	640	**0.9**	**0.70**	810	660	1.2	**0.81**	620
Si	160	190	1.8	**0.54**	1100	480	**2.4**	**0.62**	453	240	320	**1.6**	**0.47**	1600	780	**2.4**	**0.63**	700
Sr	0.42	0.48	0.9	0.27	3.04	0.79	1.2	0.37	1.2	1.1	2.1	**0.7**	**0.39**	11	3.0	**3.7**	**0.65**	4.1
Ti	7.2	6.9	1.4	**0.54**	66	45	**3.3**	**0.59**	28	12	16	**1.2**	**0.52**	140	77	**3.7**	**0.75**	55
V	2.0	2.5	**1.1**	**0.74**	2.5	2.1	1.1	−0.02	2.1	2.2	2.9	1.1	**0.68**	2.7	2.3	2.0	−0.10	2.4
Zn	12	13	**1.3**	**0.78**	33	30	1.5	0.38	20	15	22	**1.0**	**0.69**	60	40	**2.1**	0.31	31

the supplementary material. Fig. 3 and Table S5 present the contribution of the identified sources to PM2.5 and PM10 sampled in the indoor and outdoor of homes and schools.

3.4.1. Traffic exhaust emissions

Traffic is a source of three different contributions to outdoor PM, comprising combustion emissions, mechanical/abrasive processes and resuspension. The exhaust emissions were traced by OC and EC. In this source, the ratio OC/EC was 0.98 for PM2.5 and 1.2 for PM10, which is comparable with the OC/EC ratio typically found in traffic monitoring sites, which varies between 1.6 and 1.7, while in remote sites varies between 12 and 15 (Querol et al., 2013).

Particles from vehicle exhaust are mainly in the fine fraction of PM, and are able to efficiently infiltrate in indoor environments. As expected the contribution of this source was higher in the outdoor than in the indoor and the correlations between indoor and outdoor levels were strong for both places (homes: Rs = 0.82; schools: Rs = 0.57) (Fig. 4 and Table S5), indicating the importance of the infiltration of traffic related pollutants. The average contribution of the traffic exhaust emissions to PM2.5 measured in homes and schools was 4.5 μg/m^3 (24%) and 7.3 μg/m^3 (16%), respectively.

The higher contribution of traffic in schools was partly caused by the fact that sampling was performed during the occupied periods, which means during the day in schools and mainly during the night and weekends in homes. These results agree with those from Amato et al. (2014) that estimated an average traffic exhaust contribution of 13% for PM2.5 measured inside schools in Barcelona.

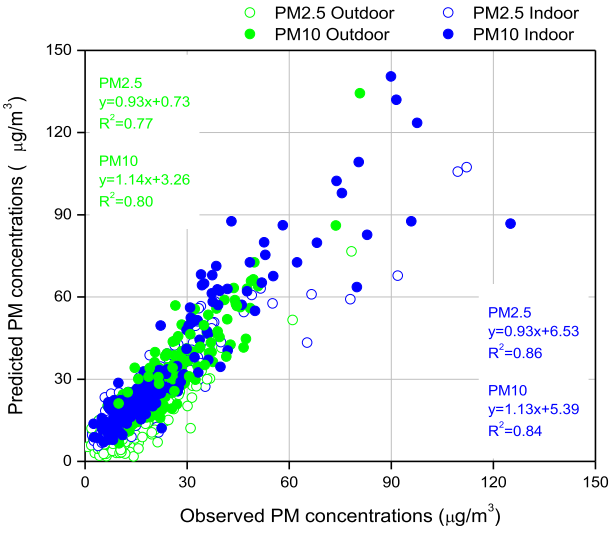


Fig. 2. Observed versus predicted PM2.5 and PM10 concentrations (values in μg/m^3).

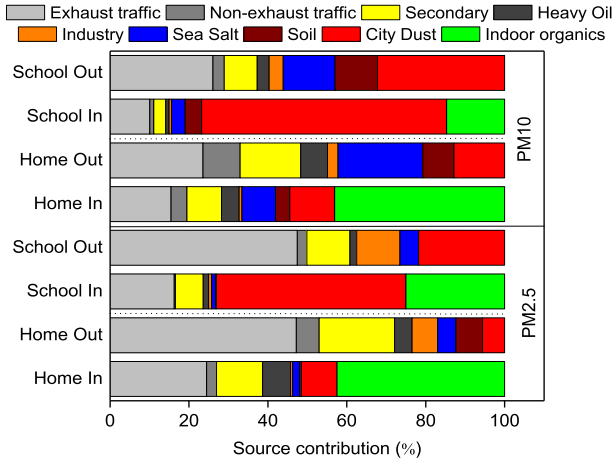


Fig. 3. Average relative source contribution for PM2.5 and PM10 sampled in the indoor and outdoor of schools and homes.

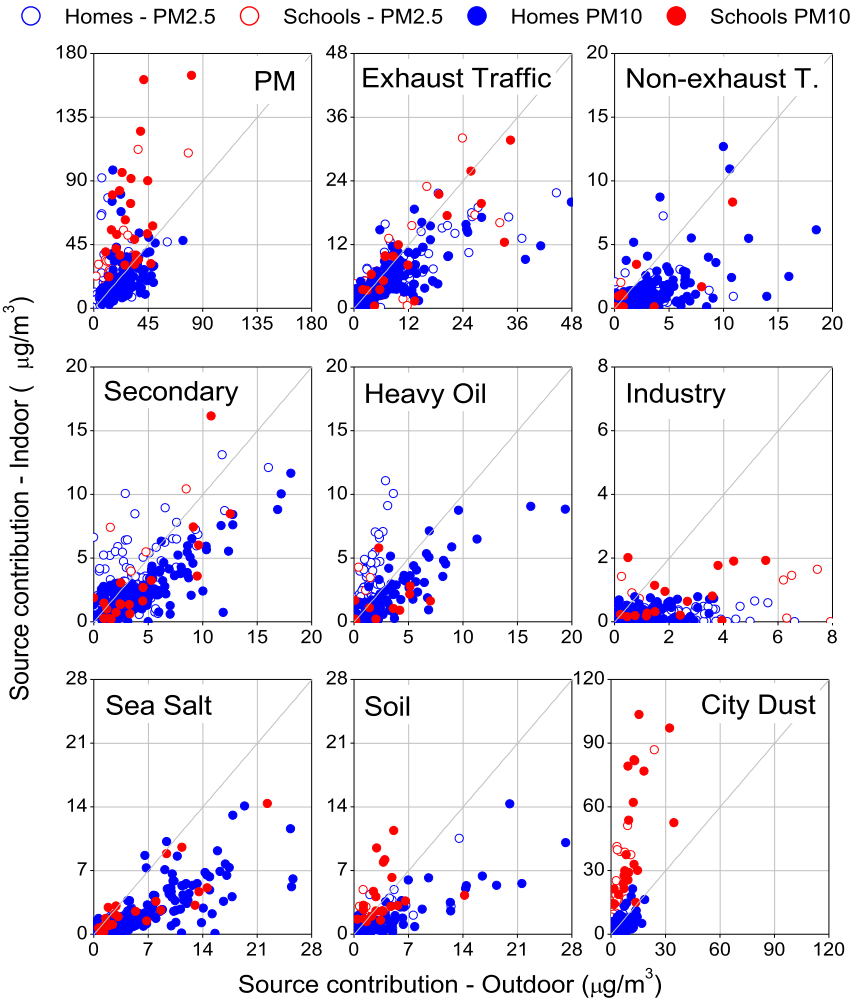


Fig. 4. Correlation between source contribution for indoor and outdoor PM2.5 and PM10 levels (values in ng/m^3).

3.4.2. Traffic non-exhaust emissions

As stringent policies and technological developments have led to sizeable reductions in exhaust emissions, the contribution of non-exhaust emissions is becoming more relevant (Padoan and Amato, 2018). Traffic non-exhaust emissions include (i) Fe, Ba, Sb, Sn, and Cu from brake wear (fraction of pad, disc, and clutch), (ii) carbonaceous, Al, Si, Zn, and S from tire wear, (iii) mineral aggregates bond by bitumen from road wear and (iv) a mix of exhaust emissions, road sanding and salting, and geogenic material carried onto the road from road dust resuspension (Padoan and Amato, 2018). In this study, the traffic non-exhaust emission factor explains most of the variance of Cu, Cr, Fe, Mn, Zn and EC, which result from break and tire wear, road wear and dust resuspension (Tables S1-S4).

The contribution of this source for PM2.5 and PM10 was 2.5% (0.47 μg/m^3) and 4.0% (1.0 μg/m^3) for homes and 0.32% (0.15 μg/m^3) and 1.0% (0.84 μg/m^3) for schools, respectively. The contribution of traffic non-exhaust emissions was higher outdoor than indoor and the contributions outdoors were less correlated with those indoors than in the exhaust emissions, probably because non-exhausts particles are mainly contained in the coarse fraction and the infiltration of coarse particles is lower (Bennett and Koutrakis, 2006). Some indoor sources of the tracers of the non-exhaust emissions can also contribute to these poor correlations. For instance, several studies have already reported that vacuum cleaners, air dryers and electrical heathers are important sources of Cu (Tofful et al., 2021).

3.4.3. Secondary sulfate

Secondary aerosols are not emitted directly into the atmosphere by a single source, they result from the atmospheric chemical transformations of gaseous precursors emitted by combustion processes, mostly in industry and transports, at shorter or longer timescales, often involving heterogeneous processes. In source apportionment studies, secondary aerosols are often not allocated to the primary source of their precursor, which might complicate the interpretation of results (Karagulian et al., 2015). The secondary sulfate factor was traced by S and to a lesser extent by As. Since the secondary aerosol is mainly in the fine fraction, the correlation between indoor and outdoor contributions was strong (Rs between 0.59 and 0.77) due to the high infiltration of fine particles. The contribution of this source to PM2.5 was 2.2 μg/m^3 (11%) and 3.1 μg/m^3 (6.8%) for homes and classrooms, respectively. In Barcelona schools, Amato et al. (2014) estimated a contribution of secondary sulfate of 11% to the indoor PM2.5 concentrations.

3.4.4. Heavy oil combustion

The Heavy Oil Combustion factor, mostly associated with combustion processes from industry and shipping, was traced by V and Ni and to a lesser extent by S and Br, as it has already been reported in other studies (Almeida et al., 2020). According to Amato et al. (2009), since industries and shipping typically burn high sulfur content residual oil, the sulfate related to V and Ni probably represents the direct SO_3 emission, condensed in particulate sulfate at the receptor site. The contribution of this source in homes

was 6.9% for PM2.5 and 4.5% for PM10 and in schools was 1.3% for PM2.5 and 0.80% for PM10.

3.4.5. Industrial emissions

Industrial emissions are a complex mixture of stationary and diffuse emissions associated with the main processes and operations (Almeida et al., 2015). While, the contribution from industry is incorporated in other factors such as secondary sulfate and fuel oil combustion, SoFI identified a factor related principally with Pb but also with Cu and Mn. The contribution of this source in homes and schools was 0.51% and 0.71% for PM.5 and 0.73% and 0.60% for PM10, respectively. The relative contribution of this source was higher for PM2.5 and outdoors (balcony: 6.5% and playground: 10%); however, the indoor and outdoor concentrations were uncorrelated (Rs between 0.27 and 0.42), which can indicate an additional source of Pb indoors. Old lead-based paint is the most significant source of Pb exposure indoors. Harmful exposures to Pb can be created when Pb-based paint is improperly removed from surfaces by dry scraping, sanding, or open-flame burning (EPA, 2022).

3.4.6. Sea salt

The Sea Salt factor was traced by Na and Cl and to a lesser extent by Br and Sr in PM10. The ratio Cl/Na in this factor was 2.0 (outdoor) and 1.7 (indoor) for PM2.5 and 1.4 (outdoor) and 1.5 (indoor) for PM10, which is comparable to the ratio of these elements for sea water (1.8). The contribution of this factor was similar in the two indoor MEs, 2.2 μg/m^3 and 3.0 μg/m^3 for the PM10 sampled in the indoor of homes and schools, respectively. The contribution of this source to the PM10 was significantly higher in the outdoor (homes: 5.9; schools: 5.6 μg/m^3) but the indoor and outdoor contributions were highly correlated (Rs = 0.84 for homes and Rs = 0.83 for schools), indicating the importance of the penetration from outdoor to indoor environments, even though sea salt particles are mainly contained in the coarse fraction. The contribution of the Sea Salt source was higher indoors than outdoors in 35% and 38% of the PM2.5 samples collected in homes and schools, respectively, probably because particles with high salt content found indoors can be formed by absorption of cleaning agents (Fromme et al., 2008; Zhao et al., 2007).

3.4.7. Soil

The Soil factor was identified by typical crustal species such as Al, Si and Ti, which result from the erosion of earth crust but also from the suspension from unpaved playgrounds. Consequently, the average contribution of this source for PM was higher in classrooms (PM2.5: 1.4 μg/m^3; PM10: 3.5 μg/m^3) than in homes (PM2.5: 0.50 μg/m^3; PM10: 0.92 μg/m^3). In homes the soil contribution was significantly higher in the outdoor (PM2.5: 0.85 μg/m^3; PM10: 2.2 μg/m^3) than in the indoor and the correlation between indoor and outdoor for fine fraction was strong (Rs = 0.77), indicating the importance of infiltration for this source. In schools, the soil contribution for PM10 was also higher in the outdoor (4.6 μg/m^3).

3.4.8. City dust

This source is characterized by abundance of Ca, Ti, Si, Fe and Sr and the absence of Al, which makes the main difference for not identifying this source as a mineral source.

This is the largest source in classrooms contributing with 22 μg/m^3 (47%) for PM2.5 and 52 μg/m^3 (62%) for PM10. In addition to the infiltration of dust from the outdoor, this source is related to the continuous resuspension of particles brought inside directly by the children on their shoes or clothing and with indoor sources of Ca-rich particles, such as chalk and building deterioration. This source also explains the high variation in Sr concentrations, due to its common substitution for Ca atoms in $CaCO_3$ used for chalk. In classrooms using chalk, PM2.5 and PM10 average concentrations doubled and the contribution of city dust increased to 55% for PM2.5 and 71% for PM10. In the playground, the contribution of city dust is still significant (20% for PM2.5 and 32% for PM10). However, as the I/O ratio of this source is 6.83 for PM2.5 and 3.87 for PM10, it is expected that the increase of ventilation will promote the dilution of the PM concentrations inside classrooms, once the effect of the outflow of internal pollutants will be larger than the inflow of external particles. In schools with mechanical ventilation, air handling units should be equipped with filters, with efficiency selected according to the concentration of PM outdoors, which will avoid the entrance of particles to the indoor.

In homes the contribution of this source to PM2.5 (1.7 μg/m^3, 8.8%) and PM10 (2.9 μg/m^3, 11%) was significantly lower than in schools. In Barcelona, Minguillón et al. (2012) also found a factor contributing to the indoor of homes characterized by abundance of Ca, Li, Ti and Sr and the absence of Al and attributed this source to cosmetics. Ti and Ca are included in the European inventory of cosmetics ingredients (Commission Decision, 2006/257/EC) and their presence in cosmetics has been documented by several studies (Di Maiuta and Schwarzentruber, 2011; Kaida et al., 2004). Moreover, sunscreen products may use nanoparticles of titanium dioxide as an active ingredient (Cengiz et al., 2006).

3.4.9. Organic matter

In indoor environments, a factor characterized by the high abundance of OC was identified. On average, this factor represented the most important contribution in homes, explaining 42% of the PM2.5 and 43% of the PM10. These results are in agreement with Tofful et al. (2021) who also identified the organic matter as the main contributor to PM2.5 in home (54%). The occupants in homes are considered one of the most important sources of coarse PM because they are responsible for the emission of primary biological material such as skin flakes, debris and hair (Fromme et al., 2008). In addition, the re-suspension of the deposited dust caused by the occupants movement and cleaning activities, such as vacuuming, dusting, house-keeping, also contribute to the organic carbon levels in the coarse fraction. Other domestic activities such as cooking, smoking and burning of candles and incenses, are also responsible for the emission of OC but in the fine and ultra-fine fraction of PM. According to Géhin et al. (2008), combustion generated particles are generally considerably smaller than 2.5 μm, often smaller than 300 nm, therefore the number concentration of ultrafine particles can be more relevant than PM2.5 mass to assess the impact of residential exposure to combustion-related particles (Isaxon et al., 2015). The fact that the indoor sources could not be separated further and appear as one source, is attributed to the lack of suitable tracers, and especially organic tracers.

In schools, the relative contribution of this source decreased to 24% for PM2.5 and 15% for PM10, respectively. However, the absolute contribution of this source in schools (PM2.5: 11 μg/m^3; PM10: 12 μg/m^3) was higher than in homes (PM2.5: 7.8 μg/m^3; PM10: 11 μg/m^3), probably due to the higher occupational density and dust resuspension. In schools, the sources of fine particles are less frequent and therefore the OC PM2.5/PM10 was 0.60, while in homes was 0.82. In Barcelona primary schools, Amato et al. (2014) estimated a contribution to PM2.5 of 45% (16 μg/m^3) for the factor associated with OC and also with Ca.

3.4.10. Contribution of the sources to the children exposure to particles

In order to consider the variability between the different days of the week, the contribution of the sources to total exposure was firstly calculated separately for weekdays and weekends. In the weekdays we considered the outdoor measured in the schools for the calculation of the source contribution to exposure, while in the weekends we used the outdoor measured in homes. For the indoor environments/activities that were not monitored (indoor physical activity, leisure and transports), which represented 7% and 10% of time during the week and weekend, respectively, we considered the concentrations measured in the schools. Afterwards, the average contribution of the sources was calculated by weighting the contribution of the weekdays (71%) and weekends (29%) during the week. The average contribution of the sources to the children's exposure to PM2.5 and PM10 is presented in Fig. 5 and Table S6.

Although children spent less time at school than at home, the contribution of the main sources in the classroom highly affected the integrated daily exposure. During the week the exposure was mainly affected by the factor indoor organics (29%, 8.1 μg/m^3) and city dust (29%, 8.0 μg/m^3),

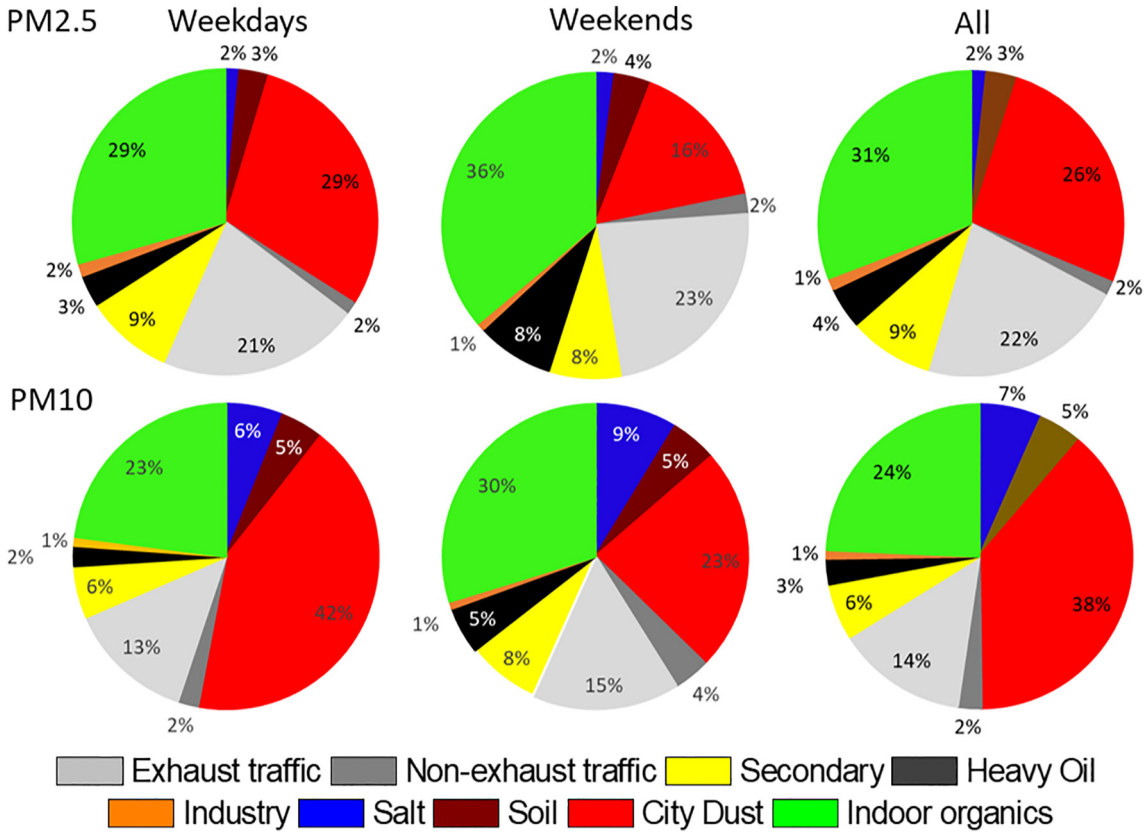


Fig. 5. Source contribution for the PM2.5 and PM10 daily exposure.

which is highly influenced by the resuspension of dust inside the classrooms. The contribution of vehicular traffic, both exhaust and non-exhaust, contributed by 23% (6.2 μg/m^3). As source apportionment inside transports was not carried out, the contribution of traffic related sources for the exposure should be underestimated. Although children only spent 3% of time in-transit, it is known that commuting lead to a substantial contribution to their total daily exposure (Cunha-Lopes et al., 2019; Faria et al., 2020), especially in high vehicle-density metropolitan areas, because transport MEs have higher air pollutant concentrations than other settings children occupy in their daily routines (Buitrago et al., 2021; Correia et al., 2020; Martins et al., 2021). The contribution of the secondary sulfate was 9.3% (2.6 μg/m^3) and the other sources contributed less than 3% to the exposure.

During the weekend, the exposure to PM2.5 decreased from 22 to 15 μg/m^3, because children were not exposed to high concentration of pollutants at the school, along with the decrease of the contribution of some outdoor sources, such as traffic exhaust, traffic non-exhaust, secondary sulfate and industry (−24%, −15%, −42% and − 27%, respectively). The contribution of the city dust during the weekend decreased to 3.0 μg/m^3 (16%) and the major sources were the indoor organics (36%, 6.9 μg/m^3) and traffic (25%, 4.9 μg/m^3).

In PM10, the major source during weekdays was city dust, accounting by 42% (19 μg/m^3). The indoor organics and traffic displayed a relative contribution of 23% and 15%, respectively. During the weekends, the exposure reduced from 34 to 22 μg/m^3, and the main source affecting the exposure was the indoor organics with 30% (8.6 μg/m^3).

The high exposure levels verified in this study do not necessarily result in higher risks to the children, since the composition of PM and sources are key factors for the health impacts.

Our results showed that city dust, indoor organics and traffic were the main sources affecting the children exposure to PM. On average city dust was responsible for 26% and 38% of the exposure to PM2.5 and PM10, respectively. This factor was mainly identified by mineral material and, epidemiological studies developed, by Hoek et al. (2002) and Janssen et al. (2005) support the hypothesis that PM derived from earth crust material have less health impacts than particles generated by combustion processes. Indoor organics accounted on average for 31% and 24% of the exposure to PM2.5 and PM10, respectively. This source was identified by OC that refers to the carbon found in the form of organic compounds, and comprises a complex mixture of different classes of organic compounds, some of them related to primary biological material but other resulting from the use of some cleaning products that can have carcinogenic, neurotoxic, immunogenic, and estrogenic activities (Gunathilake et al., 2021). Moreover, traffic contributed on average by 24% and 16% to the exposure to PM2.5 and PM10, respectively. Jung et al. (2015) showed that exposure to traffic-related air pollution may be associated with increased risk of asthma, allergic rhinitis, allergic sensitization, and with reduced lung function in schoolchildren. Traffic-related air pollution exposure was also associated with brain changes of a functional nature and slower cognitive growth (Pujol et al., 2016; Sunyer et al., 2017). Children from schools with higher traffic-related pollution showed lower functional integration and segregation in key brain networks (Pujol et al., 2016). The most recent research results show that non-exhaust particles can be more harmful than exhaust PM. Oxidative stress is one of the principal biological mechanisms causing toxicity and is frequently related to transition metals and/or redox active organics. Yanosky et al. (2012) showed that brake and tire wear particles have higher oxidative potential than other traffic-related sources and their effect is very local (50–100 m from the source), yielding more oxidant PM at road sites rather than at urban background sites.

Therefore, this study showed that emission sources contributing to children's exposure to PM can lead to major health problems and therefore it is urgent to clarify the toxicological and biological relevance of this exposure.

4. Conclusions

This work provides information on the sources contributing to children exposure to PM2.5 and PM10 concentration and supports the identification

of actions to reduce the concentration of PM in indoor and outdoor environments, and consequently reduce their exposure and health impacts.

Source apportionment showed that the major sources contributing to exposure are related with the emission of organic carbon in indoor environments, from cooking, cleaning, smoking, biomass burning, candles and occupation; resuspension of mineral particles in schools and traffic.

The results of this study highlights the importance of:

- Promoting a good ventilation in order to effectively dilute and remove contaminants emitted by occupants and indoor activities;
- Exhausting air in the vicinity of localized indoor sources such as cooking;
- Regulating measures that prohibit smoking inside and near the buildings;
- Avoiding the use of candles, incense, diffusers and air fresheners;
- Changing the cleaning routines to reduce the resuspension of mineral particles, selecting low emission products and vacuum cleaners or mops instead of brooms, and in the schools scheduling the cleaning operations in the afternoon after the school hours.
- Giving preference to the whiteboard, but if the classroom is equipped with a chalkboard cleaning it with a damp cloth, to avoid high concentrations of chalk particles suspended in the air.
- Developing sustainable transport solutions and urban planning to reduce the exposure of children to traffic related pollutants.

CRediT authorship contribution statement

Susana Marta Almeida: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Resources, Writing – original draft, Visualization, Supervision, Project administration, Funding acquisition. **Tiago Faria**: Conceptualization, Methodology, Investigation, Formal analysis, Writing - review & editing. **Vânia Martins**: Conceptualization, Methodology, Investigation, Writing - review & editing. **Nuno Canha**: Investigation. Evangelia Diapouli: Conceptualization, Methodology, Investigation, Writing - review & editing. **Konstantinos Eleftheriadis**: Conceptualization, Resources. **Manousos Ioannis Manousakas**: Conceptualization, Methodology, Investigation, Formal analysis, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was developed in the scope of the project LIFE Index-Air, funded by the EC LIFE Program (LIFE15 ENV/PT/000674), and the project ExpoLIS, funded by FEDER, through Programa Operacional Regional de Lisboa (LISBOA-01-0145-FEDER-032088), and by national funds (OE), through FCT - Fundação para a Ciência e Tecnologia, I.P. (PTDC/EAM-AMB/32088/2017). Authors also gratefully acknowledge the FCT support through the UIDB/04349/2020 project, the contract CEECIND/04228/2018 and the PhD grant SFRH/BD/129149/2017. This work reflects only the authors' view.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.155349.

References

Abbatt, J.P.D., Wang, C., 2020. The atmospheric chemistry of indoor environments. Environ. Sci.: Processes Impacts 22 (1), 25–48. https://doi.org/10.1039/C9EM00386J.

Almeida, S.M., Canha, N., Silva, A., Do Carmo Freitas, M., Pegas, P., Alves, C., Evtyugina, M., Pio, C.A., 2011. Children exposure to atmospheric particles in indoor of Lisbon primary schools. Atmos. Environ. 45 (40). https://doi.org/10.1016/j.atmosenv.2010.11.052.

Almeida, S.M., Lage, J., Fernández, B., Garcia, S., Reis, M.A., Chaves, P.C., 2015. Chemical characterization of atmospheric particles and source apportionment in the vicinity of a steelmaking industry. Sci. Total Environ. 521–522. https://doi.org/10.1016/j.scitotenv.2015.03.112.

Almeida, S.M., Ramos, C.A., Almeida-Silva, M., 2016. Exposure and inhaled dose of susceptible population to chemical elements in atmospheric particles. J. Radioanal. Nucl. Chem. 309 (1). https://doi.org/10.1007/s10967-015-4673-5.

Almeida, S.M., Manousakas, M., Diapouli, E., Kertesz, Z., Samek, L., Hristova, E., Šega, K., Alvarez, R.P., Belis, C.A., Eleftheriadis, K., Albania, Civici, N., Radic, R., Vukic, L., Hristova, E., Veleva, B., Šega, K., Bešlic, I., Davila, S., Karydas, A.G., 2020. Ambient particulate matter source apportionment using receptor modelling in European and Central Asia urban areas. Environ. Pollut. 266. https://doi.org/10.1016/j.envpol.2020.115199.

Amato, F., Pandolfi, M., Escrig, A., Querol, X., Alastuey, A., Pey, J., Perez, N., Hopke, P.K., 2009. Quantifying road dust resuspension in urban environment by multilinear engine: a comparison with PMF2. Atmos. Environ. 43 (17), 2770–2780. https://doi.org/10.1016/j.atmosenv.2009.02.039.

Amato, F., Rivas, I., Viana, M., Moreno, T., Bouso, L., Reche, C., Àlvarez-Pedrerol, M., Alastuey, A., Sunyer, J., Querol, X., 2014. Sources of indoor and outdoor PM2.5 concentrations in primary schools. Sci. Total Environ. 490, 757–765. https://doi.org/10.1016/j.scitotenv.2014.05.051.

Amato, Fulvio, Alastuey, A., Karanasiou, A., Lucarelli, F., Nava, S., Calzolai, G., Severi, M., Becagli, S., Gianelle, V.L., Colombi, C., Alves, C., Custódio, D., Nunes, T., Cerqueira, M., Pio, C., Eleftheriadis, K., Diapouli, E., Reche, C., Minguillón, M.C., Querol, X., 2016. AIRUSE-LIFE+: a harmonized PM speciation and source apportionment in five southern European cities. Atmos. Chem. Phys. 16 (5), 3289–3309. https://doi.org/10.5194/acp-16-3289-2016.

Bateson, T.F., Schwartz, J., 2008. Children's response to air pollutants. J. Toxicol. Environ. Health A 71 (3), 238–243. https://doi.org/10.1080/15287390701598234.

Bekö, G., Carslaw, N., Fauser, P., Kauneliene, V., Nehr, S., Phillips, G., Saraga, D., Schoemaecker, C., Wierzbicka, A., Querol, X., 2020. The past, present, and future of indoor air chemistry. Indoor Air 30 (3), 373–376. https://doi.org/10.1111/ina.12634.

Bennett, D., Koutrakis, P., 2006. Determining the infiltration of outdoor particles in the indoor environment using a dynamic model. J. Aerosol. Sci. 37, 766–785. https://doi.org/10.1016/j.jaerosci.2005.05.020.

Buitrago, N.D., Savdie, J., Almeida, S.M., Verde, S.C., 2021. Factors affecting the exposure to physicochemical and microbiological pollutants in vehicle cabins while commuting in Lisbon. Environ. Pollut. 270 (xxxx), 116062. https://doi.org/10.1016/j.envpol.2020.116062.

Canonaco, F., Crippa, M., Slowik, J.G., Baltensperger, U., Prévôt, A.S.H., 2013. SoFi, an IGOR-based interface for the efficient use of the generalized multilinear engine (ME-2) for the source apportionment: ME-2 application to aerosol mass spectrometer data. Atmos. Meas. Tech. 6 (12), 3649–3661. https://doi.org/10.5194/amt-6-3649-2013.

Canonaco, Francesco, Tobler, A., Chen, G., Sosedova, Y., Gates Slowik, J., Bozzetti, C., Rudolf Daellenbach, K., El Haddad, I., Crippa, M., Huang, R.J., Furger, M., Baltensperger, U., Prévôt, A.S.H., 2021. A new method for long-term source apportionment with time-dependent factor profiles and uncertainty assessment using SoFi pro: application to 1 year of organic aerosol data. Atmos. Meas. Tech. 14 (2), 923–943. https://doi.org/10.5194/amt-14-923-2021.

Cengiz, E., Wissing, S.A., Müller, R.H., Yazan, Y., 2006. Sunblocking efficiency of various TiO (2)-loaded solid lipid nanoparticle formulations(1). Int. J. Cosmet. Sci. 28 (5), 371–378. https://doi.org/10.1111/j.1467-2494.2006.00320.x.

Correia, C., Martins, V., Cunha-Lopes, I., Faria, T., Diapouli, E., Eleftheriadis, K., Almeida, S.M., 2020. Particle exposure and inhaled dose while commuting in Lisbon. Environ. Pollut. 257. https://doi.org/10.1016/j.envpol.2019.113547.

Cunha-Lopes, I., Martins, V., Faria, T., Correia, C., Almeida, S.M., 2019. Children's exposure to sized-fractioned particulate matter and black carbon in an urban environment. Build. Environ. 155. https://doi.org/10.1016/j.buildenv.2019.03.045.

Di Maiuta, N., Schwarzentruber, P., 2011. Molecular detection of bacteria in calcium carbonate powder used in cosmetic formulations. Int. J. Cosmet. Sci. 33 (5), 426–431. https://doi.org/10.1111/j.1468-2494.2011.00648.x.

EPA, 2022. Lead's Impact on Indoor Air Quality. (consulted on 15th January 2022) https://www.epa.gov/indoor-air-quality-iaq/indoor-pollutants-and-sources.

Faria, T., Martins, V., Correia, C., Canha, N., Diapouli, E., Manousakas, M., Eleftheriadis, K., Almeida, S.M., 2020. Children's exposure and dose assessment to particulate matter in Lisbon. Build. Environ. 171. https://doi.org/10.1016/j.buildenv.2020.106666.

Faria, T., Martins, V., Canha, N., Diapouli, E., Manousakas, M., Fetfatzis, P., Gini, M.I., Almeida, S.M., 2022. Assessment of children's exposure to carbonaceous matter and to PM major and trace elements. Sci. Total Environ. 807 (xxxx), 151021. https://doi.org/10.1016/j.scitotenv.2021.151021.

Farmer, D.K., Vance, M.E., 2019. Indoor air: sources{,} chemistry and health effects. Environ. Sci.: Processes Impacts 21 (8), 1227–1228. https://doi.org/10.1039/C9EM90035G.

Fromme, H., Diemer, J., Dietrich, S., Cyrys, J., Heinrich, J., Lang, W., Kiranoglu, M., Twardella, D., 2008. Chemical and morphological properties of particulate matter (PM10, PM2.5) in school classrooms and outdoor air. Atmos. Environ. 42 (27), 6597–6605. https://doi.org/10.1016/j.atmosenv.2008.04.047.

Géhin, E., Ramalho, O., Kirchner, S., 2008. Size distribution and emission rate measurement of fine and ultrafine particle from indoor human activities. Atmos. Environ. 42 (35), 8341–8352. https://doi.org/10.1016/j.atmosenv.2008.07.021.

Ginsberg, G.L., Foos, B.P., Firestone, M.P., 2005. Review and analysis of inhalation dosimetry methods for application to children's risk assessment. J. Toxicol. Environ. Health A 68 (8), 573–615. https://doi.org/10.1080/15287390590921793.

Gu, X.Yu, Chu, X., Zeng, X.L., Bao, H.R., Liu, X.J., 2017. Effects of PM2.5 exposure on the Notch signaling pathway and immune imbalance in chronic obstructive pulmonary disease. Environ. Pollut. 226, 163–173. https://doi.org/10.1016/j.envpol.2017.03.070.

Gunathilake, T.M.S.U., Ching, Y.C., Kadokami, K., 2021. An overview of organic contaminants in indoor dust, their health impact, geographical distribution and recent extraction/analysis methods. Environ. Geochem. Health https://doi.org/10.1007/s10653-021-01013-x.

Hoek, G., Brunekreef, B., Goldbohm, S., Fischer, P., van den Brandt, P.A., 2002. Association between mortality and indicators of traffic-related air pollution in the Netherlands: a cohort study. Lancet 360 (9341), 1203–1209. https://doi.org/10.1016/S0140-6736(02)11280-3.

Hussein, T., Kukkonen, J., Korhonen, H., Pohjola, M., Pirjola, L., Wraith, D., Härkönen, J., Teinilä, K., Koponen, I.~.K., Karppinen, A., Hillamo, R., Kulmala, M., 2007. Evaluation and modeling of the size fractionated aerosol particle number concentration measurements nearby a major road in Helsinki - Part II: aerosol measurements within the SAPPHIRE project. Atmos. Chem. Phys. 7 (15), 4081–4094. https://doi.org/10.5194/acp-7-4081-2007.

Isaxon, C., Gudmundsson, A., Nordin, E.Z., Lönnblad, L., Dahl, A., Wieslander, G., Bohgard, M., Wierzbicka, A., 2015. Contribution of indoor-generated particles to residential exposure. Atmos. Environ. 106, 458–466. https://doi.org/10.1016/j.atmosenv.2014.07.053.

Janssen, N.A.H., Lanki, T., Hoek, G., Vallius, M., de Hartog, J.J., Van Grieken, R., Pekkanen, J., Brunekreef, B., 2005. Associations between ambient, personal, and indoor exposure to fine particulate matter constituents in Dutch and Finnish panels of cardiovascular patients. Occup. Environ. Med. 62 (12), 868–877. https://doi.org/10.1136/oem.2004.016618.

Jung, D.-Y., Leem, J.-H., Kim, H.-C., Kim, J.-H., Hwang, S.-S., Lee, J.-Y., Kim, B.-J., Hong, Y.-C., Hong, S.-J., Kwon, H.-J., 2015. Effect of traffic-related air pollution on allergic disease: results of the children's health and environmental research. Allergy Asthma Immunol. Res. 7 (4), 359–366. https://doi.org/10.4168/aair.2015.7.4.359.

Kaida, T., Kobayashi, K., Adachi, M., Suzuki, F., 2004. Optical characteristics of titanium oxide interference film and the film laminated with oxides and their applications for cosmetics. J. Cosmet. Sci. 55 (2), 219–220. http://europepmc.org/abstract/MED/15190897.

Kalaiarasan, G., Balakrishnan, R.M., Sethunath, N.A., Manoharan, S., 2017. Source apportionment of PM2.5 particles: influence of outdoor particles on indoor environment of schools using chemical mass balance. Aerosol Air Qual. Res. 17, 616–625.

Karagulian, F., Belis, C.A., Dora, C.F.C., Prüss-Ustün, A.M., Bonjour, S., Adair-Rohani, H., Amann, M., 2015. Contributions to cities' ambient particulate matter (PM): a systematic review of local source contributions at global level. Atmos. Environ. 120, 475–483. https://doi.org/10.1016/j.atmosenv.2015.08.087.

Kim, J., Park, S., Kim, H., Yeo, M.S., 2017. Emission characterization of size-resolved particles in a pre-school classroom in relation to children's activities. Indoor Built Environ. 28 (5), 659–676. https://doi.org/10.1177/1420326X17707565.

Kulhánová, I., Morelli, X., Le Tertre, A., Loomis, D., Charbotel, B., Medina, S., Ormsby, J.N., Lepeule, J., Slama, R., Soerjomataram, I., 2018. The fraction of lung cancer incidence attributable to fine particulate air pollution in France: impact of spatial resolution of air pollution models. Environ. Int. 121 (September), 1079–1086. https://doi.org/10.1016/j.envint.2018.09.055.

LIFE Index-Air, 2017. Deliverable B2.1 - Technical Report on Time Activity Patterns. http://www.lifeindexair.net/sitelifeindexair/wp-content/uploads/2018/03/Annex-B2-1-Technical-report-on-time-activity-patterns.pdf.

Lu, P., Zhang, Y., Lin, J., Xia, G., Zhang, W., Knibbs, L.D., Morgan, G.G., Jalaludin, B., Marks, G., Abramson, M., Li, S., Guo, Y., 2020. Multi-city study on air pollution and hospital outpatient visits for asthma in China. Environ. Pollut. 257, 113638. https://doi.org/10.1016/j.envpol.2019.113638.

MacNeill, M., Kearney, J., Wallace, L., Gibson, M., Héroux, M.E., Kuchta, J., Guernsey, J.R., Wheeler, A.J., 2014. Quantifying the contribution of ambient and indoor-generated fine particles to indoor air in residential environments. Indoor Air 24 (4), 362–375. https://doi.org/10.1111/ina.12084.

Manousakas, M., Diapouli, E., Papaefthymiou, H., Kantarelou, V., Zarkadas, C., Kalogridis, A.-C., Karydas, A.-G., Eleftheriadis, K., 2018. XRF characterization and source apportionment of PM10 samples collected in a coastal city. X-Ray Spectrom. 47 (3), 190–200. https://doi.org/10.1002/xrs.2817.

Martins, V., Faria, T., Diapouli, E., Manousakas, M.I., Eleftheriadis, K., Viana, M., Almeida, S.M., 2020. Relationship between indoor and outdoor size-fractionated particulate matter in urban microenvironments: levels, chemical composition and sources. Environ. Res. 183. https://doi.org/10.1016/j.envres.2020.109203.

Martins, Vânia, Correia, C., Cunha-Lopes, I., Faria, T., Diapouli, E., Manousakas, M.I., Eleftheriadis, K., Almeida, S.M., 2021. Chemical characterisation of particulate matter in urban transport modes. J. Environ. Sci. 100, 51–61. https://doi.org/10.1016/j.jes.2020.07.008.

Mateen, F.J., Brook, R.D., 2011. Air pollution as an emerging global risk factor for stroke. JAMA - J. Am. Med. Assoc. 305 (12), 1240–1241. https://doi.org/10.1001/jama.2011.352.

Miller, K.A., Siscovick, D.S., Sheppard, L., Shepherd, K., Sullivan, J.H., Anderson, G.L., Kaufman, J.D., 2007. Long-term exposure to air pollution and incidence of cardiovascular events in women. N. Engl. J. Med. 356 (5), 447–458. https://doi.org/10.1056/NEJMoa054409.

Minguillón, M.C., Schembari, A., Triguero-Mas, M., de Nazelle, A., Dadvand, P., Figueras, F., Salvado, J.A., Grimalt, J.O., Nieuwenhuijsen, M., Querol, X., 2012. Source apportionment of indoor, outdoor and personal PM2.5 exposure of pregnant women in Barcelona,Spain. Atmos. Environ. 59, 426–436. https://doi.org/10.1016/j.atmosenv.2012.04.052.

Morawska, L., Afshari, A., Bae, G.N., Buonanno, G., Chao, C.Y.H., Hänninen, O., Hofmann, W., Isaxon, C., Jayaratne, E.R., Pasanen, P., Salthammer, T., Waring, M., Wierzbicka, A., 2013. Indoor aerosols: from personal exposure to risk assessment. Indoor Air 23 (6), 462–487. https://doi.org/10.1111/ina.12044.

Morawska, L., Ayoko, G.A., Bae, G.N., Buonanno, G., Chao, C.Y.H., Clifford, S., Fu, S.C., Hänninen, O., He, C., Isaxon, C., Mazaheri, M., Salthammer, T., Waring, M.S., Wierzbicka, A., 2017. Airborne particles in indoor environment of homes, schools, offices and aged care facilities: the main routes of exposure. Environ. Int. 108 (August), 75–83. https://doi.org/10.1016/j.envint.2017.07.025.

Paatero, P., 1999. The multilinear engine—a table-driven, least squares program for solving multilinear problems, including the n-way parallel factor analysis model. J. Comput. Graph. Stat. 8 (4), 854–888. https://doi.org/10.1080/10618600.1999.10474853.

Padoan, E., Amato, F., 2018. Chapter 2 - vehicle non-exhaust emissions: impact on air quality. In: Amato, F. (Ed.), Non-exhaust Emissions: An Urban Air Quality Problem for Public Health; Impact and Mitigation Measures. Academic Press, pp. 21–65 https://doi.org/10.1016/B978-0-12-811770-5.00002-9.

Polissar, A.V., Hopke, P.K., Poirot, R.L., 2001. Atmospheric aerosol over Vermont: chemical composition and sources. Environ. Sci. Technol. 35 (23), 4604–4621. https://doi.org/10.1021/es0105865.

Pujol, J., Martínez-Vilavella, G., Macià, D., Fenoll, R., Alvarez-Pedrerol, M., Rivas, I., Forns, J., Blanco-Hinojo, L., Capellades, J., Querol, X., Deus, J., Sunyer, J., 2016. Traffic pollution exposure is associated with altered brain connectivity in school children. NeuroImage 129, 175–184. https://doi.org/10.1016/j.neuroimage.2016.01.036.

Querol, X., Alastuey, A., Viana, M., Moreno, T., Reche, C., Minguillón, M.C., Ripoll, A., Pandolfi, M., Amato, F., Karanasiou, A., Pérez, N., Pey, J., Cusack, M., Vázquez, R., Plana, F., Dall'Osto, M., De La Rosa, J., Sánchez De La Campa, A., Fernández-Camacho, R., Fernández Patier, R., 2013. Variability of carbonaceous aerosols in remote, rural, urban and industrial environments in Spain: implications for air quality policy. Atmos. Chem. Phys. 13 (13), 6185–6206. https://doi.org/10.5194/acp-13-6185-2013.

Reff, A., Eberly, S.I., Bhave, P.V., 2007. Receptor modeling of ambient particulate matter data using positive matrix factorization: review of existing methods. J. Air Waste Manag. Assoc. 57 (2), 146–154. https://doi.org/10.1080/10473289.2007.10465319.

Salvi, S., 2007. Health effects of ambient air pollution in children. Paediatr. Respir. Rev. 8 (4), 275–280. https://doi.org/10.1016/j.prrv.2007.08.008.

Sunyer, J., Suades-González, E., García-Esteban, R., Rivas, I., Pujol, J., Alvarez-Pedrerol, M., Forns, J., Querol, X., Basagaña, X., 2017. Traffic-related air pollution and attention in primary school children. Epidemiology 28 (2), 181–189. https://doi.org/10.1097/EDE.0000000000000603.

Suwanwaiphatthana, W., Ruangdej, K., Turner-Henson, A., 2010. Outdoor air pollution and children's health. Pediatr. Nurs. 36 (1), 25–32.

Tofful, L., Canepari, S., Sargolini, T., Perrino, C., 2021. Indoor air quality in a domestic environment: combined contribution of indoor and outdoor PM sources. Build. Environ. 202 (March), 108050. https://doi.org/10.1016/j.buildenv.2021.108050.

Ulbrich, I.M., Canagaratna, M.R., Zhang, Q., Worsnop, D.R., Jimenez, J.L., 2009. Interpretation of organic components from positive matrix factorization of aerosol mass spectrometric data. Atmos. Chem. Phys. 9 (9), 2891–2918. https://doi.org/10.5194/acp-9-2891-2009.

Urso, P., Cattaneo, A., Garramone, G., Peruzzo, C., Cavallo, D.M., Carrer, P., 2015. Identification of particulate matter determinants in residential homes. Build. Environ. 86, 61–69. https://doi.org/10.1016/j.buildenv.2014.12.019.

Weschler, C.J., Carslaw, N., 2018. Indoor chemistry. Environ. Sci. Technol. 52 (5), 2419–2428. https://doi.org/10.1021/acs.est.7b06387.

WHO, 2021. WHO global air quality guidelines. WHO Global Air Quality Guidelines. Particulate Matter (PM2.5 and PM10), Ozone, Nitrogen Dioxide, Sulfur Dioxide and Carbon Monoxide. World Health Organization, Geneva.

Yanosky, J.D., Tonne, C.C., Beevers, S.D., Wilkinson, P., Kelly, F.J., 2012. Modeling exposures to the oxidative potential of PM10. Environ. Sci. Technol. 46 (14), 7612–7620. https://doi.org/10.1021/es3010305.

Zhao, W., Hopke, P.K., Gelfand, E.W., Rabinovitch, N., 2007. Use of an expanded receptor model for personal exposure analysis in schoolchildren with asthma. Atmos. Environ. 41, 4084–4096.

Zwozdziak, A., Sówka, I., Willak-Janc, E., Zwozdziak, J., Kwiecińska, K., Balińska-Miśkiewicz, W., 2016. Influence of PM1 and PM2.5 on lung function parameters in healthy schoolchildren-a panel study. Environ. Sci. Pollut. Res. Int. 23 (23), 23892–23901. https://doi.org/10.1007/s11356-016-7605-1.