

Indoor PM₁₀ in university classrooms: Chemical composition and source behaviour

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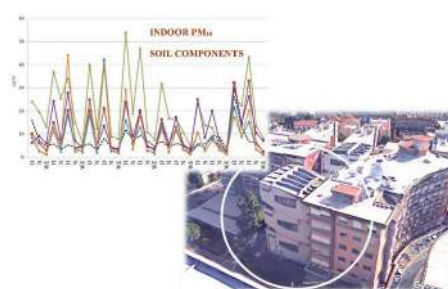
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HIGHLIGHTS

- PM₁₀ was determined in five university environments in real-world conditions.
- The main sources of PM and their ability to infiltrate were studied.
- The chemical composition of indoor and outdoor PM₁₀ was very different.
- During classes, indoor PM₁₀ concentration was higher than outdoors.
- Indoor concentration increase was due to soil components and to organic species.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:

Indoor pollution
I/O concentration ratio
PM composition
Source tracers
Mass closure

ABSTRACT

The indoor atmosphere of five university environments, ranging from a research laboratory to a vast lecture hall, was studied during a seven-week measurement campaign. The study was carried out in real-world conditions and was designed to distinguish periods when the environments were occupied from periods when they were empty. A comprehensive chemical characterisation of PM₁₀ was carried out (elements as total, extractable and residual fractions, ions, elemental carbon, organic carbon), which allowed the study of the main PM sources (soil, sea, secondary inorganic species, traffic emission, organics). Other sources (heavy oil combustion, biomass burning and non-exhausts traffic emission) were well traced by the extractable or residual fractions of some elements (V, Rb, Mo).

During classes, indoor PM₁₀ concentration exceeded outdoor values. The main differences between indoor and outdoor chemical composition were due to soil components and to the organics. Soil particles were carried in by the students and re-suspended by their movements. The organics were mainly due to bioparticles released by the occupants. An increase inside classrooms was also observed for sulphate and attributed to the used of mineral gypsum as blackboard chalk. Sea-salt particles and ammonium nitrate, instead, showed a sharp decrease when entering the indoor environments, due to their large size and a shift in its thermodynamic equilibrium, respectively.

The concentration of particles released by anthropogenic combustion sources (exhaust and non-exhaust traffic emission, domestic heating) was generally lower than outdoors, and the degree of their infiltration was determined by their dimensions.

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<https://doi.org/10.1016/j.atmosenv.2022.119260>

Received 22 February 2022; Received in revised form 27 May 2022; Accepted 28 June 2022

Available online 1 July 2022

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1. Introduction

During the last ten years, increasing awareness of the relevance of air quality in indoor environments has been recorded in both the scientific community and the general public. Many recent scientific studies have addressed the characterisation of indoor pollutants, the identification of their sources and their relative weight, and the study of the time changes in their concentrations (Perrino et al., 2016; Huang et al., 2018; Sánchez-Soberón et al., 2019; Martins et al., 2020; Heo et al., 2021, among others), also taking advantage of new techniques to monitor concentrations in real-time (Marques et al., 2018; Saini et al., 2020; Wang et al., 2020). In parallel, abatement systems able to reduce and control the concentration of harmful chemicals in indoor environments have been developed (Gonzalez-Martin et al., 2021).

Such studies have been carried out in various urban environments where citizens live: homes, offices, schools, means of transport, stores, buildings used for sports, entertainment, etc. These environments have some sources in common and other sources specific to the place or dependent on the type of people living there. The infiltration of pollutants from the outside air and the release of pollutants due to the presence of people are among the ubiquitous sources, while the use of cooking, pellet stoves, candles or incense are examples of sources that characterise peculiar environments. Cigarette smoke and the use of personal care products (perfumes, hairspray, deodorants, etc.) are examples of pollution sources that depend on people's habits (Amato et al., 2014; Drago et al., 2018; Morawska et al., 2017; Martins et al., 2020; Tofful et al., 2020).

Among these environments, schools proved to be very suitable to study the role played by people in generating pollutants in the particulate phase. As the combustive sources of dust (kitchen, stoves, cigarette smoke) are missing and there is no use of chemicals except, in some cases, blackboard chalk, PM sources in classrooms are substantially reduced to the infiltration from outside and the presence of people. Many scientific papers have addressed these environments, discussing the importance of PM infiltration from outdoors and soil dust re-suspension (Zwoździak et al., 2013; Amato et al., 2014; Fuoco et al., 2015; Tofful and Perrino, 2015; Carrion-Matta et al., 2019; Oliveira et al., 2019; Leppänen et al., 2020; Alameddine et al., 2022). In particular, Viana et al. (2015) investigated the size distribution pattern of elements in 39 primary schools in Barcelona, discussing the emission sources in each particle size range.

University environments, which are of interest for the simultaneous presence of tens to hundreds of students in a single room, are much less investigated, and the studies are limited to the determination of PM mass concentration (Braniš et al., 2005; Gaidajis and Angelakoglou, 2009) or a few elements (Sulaiman et al., 2017).

During 2017–2019, academic environments have been extensively studied by the VIEPI project (Integrated evaluation of the exposure to indoor particulate matter). The project's main objectives were: i) the study of the relationships between meteorological parameters, indoor airflow and PM concentration, ii) the chemical and biological characterisation of indoor PM, and iii) the investigation of infiltration/exfiltration mechanisms (Pelliccioni et al., 2020). In the framework of this project, PM composition was studied during three intensive campaigns and twelve long-duration (1-month) sampling periods, all carried out inside a building in the Sapienza University of Rome (Italy). The results of the long-duration study gave insight into the seasonal variations in the chemical composition of outdoor PM and the penetration of the various PM components into the study environments (Tofful et al., 2021).

This work reports and discusses the results obtained during one of the three intensive measurement campaigns, performed in five classrooms of the Sapienza University of Rome during the winter of 2017. This activity aimed to reconstruct the whole PM mass from the body of the analytical determinations (mass closure) and to evaluate the behaviour of natural (soil, sea) and anthropogenic (exhaust and non-

exhaust traffic, biomass burning, secondary inorganic species), sources, both outdoor and indoor, with particular attention to PM components related to the presence of the individuals. This experimental study had the following strengths: the simultaneous monitoring of PM composition outdoors and in five different indoor environments, ranging from a small research laboratory to a vast lecture hall; - the execution of the measurements in real-life conditions (during classes), which was made possible by the use of specifically developed very quiet samplers; - a comprehensive chemical characterisation of PM components: compared with other similar works, the added value lies in the chemical fractionation of the elements into two solubility fractions, which allows a significant increase in their selectivity as source tracers.

2. Experimental

2.1. Sampling sites, periods, and equipment

The study was carried out in a five-storey building inside the campus of the University of Rome (Physics Department, building Enrico Fermi; Google coordinates: 41°54'06" N; 12°30'57" E). The campus is about 50 m from the nearest high-traffic road; a limited number of cars can circulate inside. PM sampling was carried out outdoors, at the ground floor, and indoors, in five different environments: a Lecture Hall (LH, at the ground floor, 1150 m³), a computer room (CR, at the 2nd floor, 450 m³), two identical classrooms (A4 at the 2nd floor, A7 at the 4th floor, 570 m³), and a physics laboratory (SL, at the 4th floor, about 300 m³). A detailed description of the sampling location and the classrooms is reported in Pelliccioni et al. (2020). The campus position, a picture of the building, the maps of the floors with the indication of the sampling sites are shown in Fig. 1.

The intensive campaign lasted from November 1st to December 17th, 2017. The sampling schedule included five samples a week: two samples (D) during the lesson times (Monday + Tuesday, and Wednesday + Thursday + Friday, from 09:00 to 18:00); two samples (N) during the evenings/nights (Monday + Tuesday, and Wednesday + Thursday + Friday, from 18:00 to 9:00 of the following day), and one sample (WE) during the weekend (from Saturday at 9:00 to Monday at 9:00). Thirty-three day-night-weekend (D/N/WE) samples were obtained in each environment during the winter period, for a total of 165 samples. The list of the samples is reported in Supplementary Material Table S1.

To carry out PM measurements in real-world conditions without disturbing the classes, we used very quiet samplers (<35 dB) specifically designed to be operated in indoor environments (Silent Sequential Sampler, FAI Instruments, Fonte Nuova, Rome, Italy). Three samplers were placed in each indoor environment and outdoors. They were equipped with 47 mm Teflon (TEFLO, 2.0 µm pore size, PALL), quartz (TISSUQUARTZ 2500QAT, PALL) and polycarbonate filters (0.8 µm pore size, MILLIPORE, Merk Life Science), respectively. The samplers operated at the flow rate of 10 L min⁻¹ and were provided with sequential systems able to run four sampling lines without attendance.

Outdoors, in addition to the Silent Samplers, we also collected 24-h PM₁₀ samples (from midnight to midnight) using a beta attenuation monitor operating at the flow rate of 2.3 m³/h (SWAM 5aDual Channel Monitor, FAI Instruments, Fonte Nuova, Rome, Italy), equipped with Teflon and quartz filters.

The mixing properties of the lower atmosphere during the campaign were traced using a natural radioactivity monitor (PBL Mixing Monitor, FAI Instruments, Fonte Nuova, Rome, Italy). The monitor determines the natural radioactivity of short-lived Radon progeny and operates at 1-h time resolution. The ability of Radon progeny to give information about the dispersion conditions in the planetary boundary layer has been widely reported in the literature (Porstendorfer, 1994; Perrino et al., 2001, 2008; Griffith et al., 2013; Vecchi et al., 2019, among others). Briefly, this technique takes advantage of the constant emanation rate of the Radon gas from soil and rocks. Being radioactive decay into daughter radionuclides its only removal mechanism, the monitoring

of Radon and Radon progeny can give robust information about the dilution properties of the lower boundary layer, that is the layer where pollutants are emitted and pollution phenomena are observed. The value of natural radioactivity increases when atmospheric stability increases and decreases in the case of efficient atmospheric mixing or advection.

2.2. Analytical procedure

Samples collected on Teflon filters were weighted and then analysed for their inorganic content (elements, ions). The analysis of elemental and organic carbon was carried out on quartz filters, while polycarbonate filters were devoted to determining the bioaerosol content of PM (not discussed in this paper; [Marcovecchio and Perrino, 2021a,b](#)). The overall analytical procedure has been widely described and validated previously ([Perrino et al., 2008, 2014](#); [Canepari et al., 2009a, 2014](#)).

Briefly, samples collected on Teflon filters were weighted before and after PM collection using a microbalance (ME5, Sartorius AG, Goettingen, Germany) after conditioning at 50% r. h. and 20 °C for 48 h. In the case of the 24-h samples, the mass concentration of PM₁₀ was automatically determined by the beta attenuation method. After weighting, the filters were analysed for their elemental content (Si, Al, Fe, Na, K, Mg, Ca and minor elements) by energy-dispersion X-ray fluorescence (XRF) (XEPOS, Spectro Analytical Instruments, Kleve, Germany). Subsequently, the Teflon filters were extracted for 20 + 20 min under sonication in deionised water and filtered using cellulose nitrate filters (0.45 µm pore size). The solutions were analysed for their ionic content (chloride, nitrate, sulphate, sodium, potassium, ammonium, magnesium, calcium) by ion chromatography (IC) (ICS1000, Dionex Co., Sunnyvale, CA, USA) and for the soluble fraction of elements (As, Ba, Cd, Ce, Co, Cr, Cs, Cu, Fe, Li, K, Mg, Mn, Mo, Na, Ni, Pb, Rb, Sb, Sn, Sr, Ti, Tl, U, V, W) by Inductively Coupled Plasma Optical Emission Spectrometry with mass detection (ICP-MS) (Brucker 820-MS, Billerica, MA, USA). The residual (insoluble) fraction on both the sampling membrane and the filtration membrane was subjected to microwave-assisted acid

digestion in HNO₃:H₂O₂ (2:1), filtered again at 0.45 µm, and analysed by ICP-MS for the same elements. This procedure allows a complete characterisation of the inorganic fraction of PM on a single collection filter. Determining the soluble and residual fraction of each element provides insight into the chemical form in which the element is released and valuable information for estimating its elemental bio-accessibility and identifying PM sources ([Canepari et al., 2009b](#)).

Quartz filters were analysed for elemental carbon (EC) and organic carbon (OC) by thermo-optical analysis (TOA) (OCEC Carbon Aerosol Analyser, Sunset Laboratory, Tigard, OR, USA) using the NIOSH-QUARTZ temperature protocol.

The limits of detection (LODs) and the mean repeatability of each analyte are reported in [Supplementary Material Table S2](#).

3. Results and discussion

3.1. Mass closure and self-consistency

As a general quality control of the results, the samples collected both outdoors and in the five indoor environments were checked for mass closure, i.e., the correspondence between the PM₁₀ mass determined by gravimetry and the sum of the measured components. To account for non-measured atoms (oxygen and hydrogen), elements were considered as oxides ([Marcazzan et al., 2001](#)), and organic carbon was multiplied by a conversion factor α (average organic molecular weight per carbon weight) to obtain organic matter ([Turpin and Lim, 2001](#); [Viidanoja et al., 2002](#)). The value of α was set to 1.8 and 1.4 in the case of outdoor and indoor environments, respectively ([Pelliccioni et al., 2020](#), [Marcovecchio and Perrino, 2021a,b](#)). The difference between the two α values is justified by the different chemical compositions of the organic molecules in the two environments: more aged and oxidised outdoors, relatively non-polar, due to the presence of compounds rich in aliphatic CH, indoors. However, it is worth noting that the speciation of organics in indoor PM is still under study and identifying a suitable conversion factor is a matter of debate ([Chow et al., 2015](#); [Reff et al., 2007](#)). The α

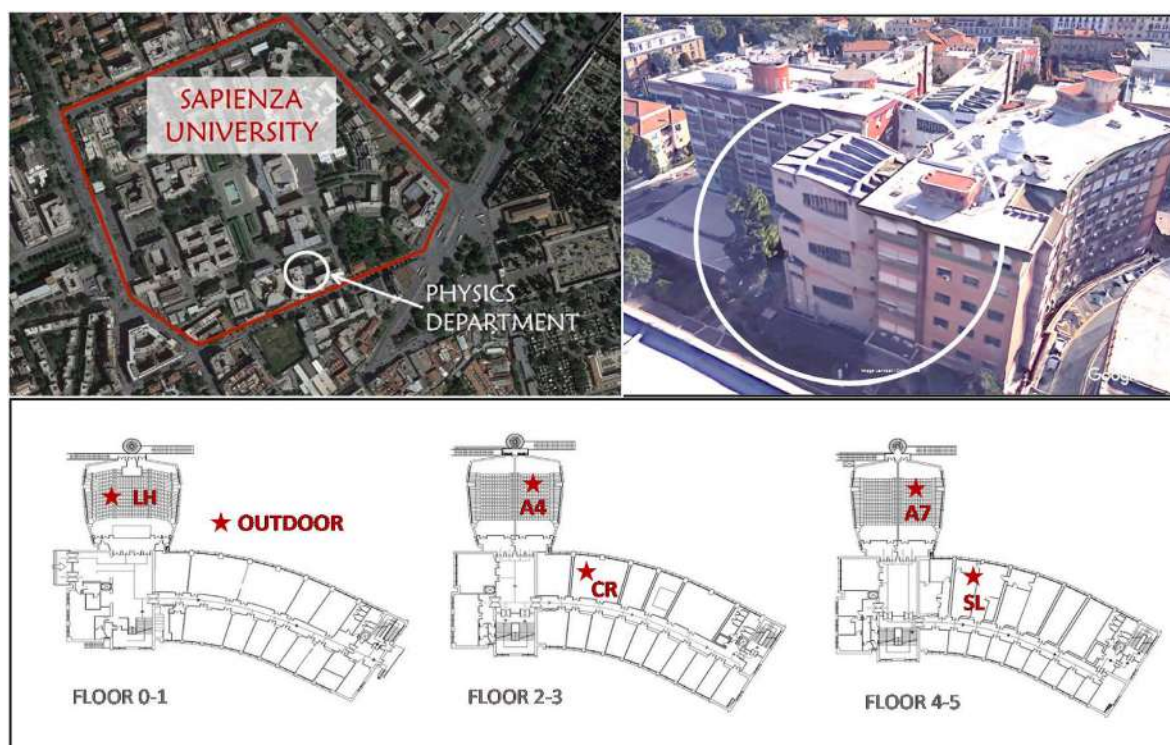


Fig. 1. Position of the Sapienza University campus (upper left panel) and of the Enrico Fermi building (upper right panel); maps of the sampling sites (lower panel).

value used in this work for indoor environments, suggested by Reff et al. (2007), also allowed the best fit in our mass reconstruction. Carbonate ion was calculated considering the stoichiometric ratios in CaCO_3 and MgCO_3 (1.5 and 2.5, respectively).

A satisfactory mass closure was obtained for all D/N/WE samples, with a mean reconstruction percentage varying from 96.3% (A4) to 108% (SL) and R^2 values in the range 0.93–0.97. In the case of the 24-h outdoor samples, the analytical determinations accounted for 98.9% of the PM_{10} mass determined by beta attenuation, and the R^2 value was 0.98.

As a further quality check, the results of the 24-h outdoor samples were grouped and compared with the corresponding D/N/WE samples (in this case, day and night concentration values were added). The results are shown in Fig. 2. Although there was a difference in the start time of the measurements (00:00 in the case of the 24-h samples, 9:00 in the case of D/N/WE), the self-consistency was satisfactory: in the regression equation, the slope was 1.0, the intercept was $1.2 \mu\text{g}/\text{m}^3$ and the value of R^2 was 0.90. These results indicate that the analytical quality was good and that the performance of the Silent samplers, operating on fractionated periods at the flow rate of 10 l min^{-1} , were comparable to those of the traditional 24-h samplers operating at the flow rate of $2.3 \text{ m}^3/\text{h}$.

3.2. PM_{10} concentration

Fig. 3 (left panel) shows the mean PM_{10} concentrations recorded during all the D, N and WE periods of the campaign in the five indoor environments and outdoors. The same data are detailed in the right panel as individual concentrations during each D, N, and WE period.

Outdoors, the mean PM_{10} concentrations recorded during the D and N periods were comparable, and they were both higher than during the WE. This can be explained considering that the campus was much less attended on Saturday and Sunday, and there were fewer people also in the surrounding area. In the five indoor environments, instead, the concentrations followed the pattern $\text{D} > \text{N} > \text{WE}$, a first indication of the presence of one or more indoor PM sources during the periods when the students attended the classrooms. The individual concentrations show that a substantial increase with respect to outdoor values was recorded

during all the individual D periods, except those including the midweek holidays of November 1st and December 8th (black arrows in Fig. 3) and the D period on December 11th–12th (red arrow in Fig. 3).

3.3. Macro-sources

To understand which sources were responsible for the increase observed in the D samples collected in the indoor environments, we calculated the concentration of five groups of components (macro-sources), which represent the main, ubiquitous sources of PM_{10} : *soil*, *sea-salt*, *secondary inorganic species*, *traffic emission*, and *biosphere (organics)*. This approach can be suitable for studying datasets that include many samples collected in different environments and during different periods (D, N, WE), which are not necessarily equivalent in terms of source contributions. Details about the algorithms are reported and discussed in Perrino et al. (2014) and Chow et al. (2015) and compared with the output of PMF analysis in Farao et al. (2014).

Briefly, *soil* was calculated as the sum of the concentration of the main crustal elements, considered as oxides: Al, Si, Fe, the insoluble fractions of K, Mg, and Ca (calculated as the difference between XRF and IC determinations), calcium and magnesium carbonate. *Sea salt* was calculated from the sum of Na^+ and Cl^- , multiplied by 1.176 to take minor sea-water components into account. Ammonium, nitrate and non-sea-salt sulphate constituted the *secondary inorganics*. *Traffic emission* was estimated by adding the concentration of elemental carbon and an equivalent amount of organic carbon. The *organics* were calculated as the remaining organic carbon multiplied by α to obtain the organic matter.

The same type of graphs shown in Fig. 3 for the mass concentration is reported in Figs. 4 and 5 for each group of macro-sources. In the case of *secondary inorganics*, the pattern of nitrate and sulphate are shown individually, as their behaviour in indoor environments was very different from each other.

The data in Fig. 4 show that the increase in PM_{10} mass concentration observed during the D period was mainly due to the *soil* components and the *organics*. For these two macro-sources, the concentration pattern was $\text{D} > \text{N} > \text{WE}$ in all the indoor environments.

Quantitatively, the highest increase with respect to outdoors was

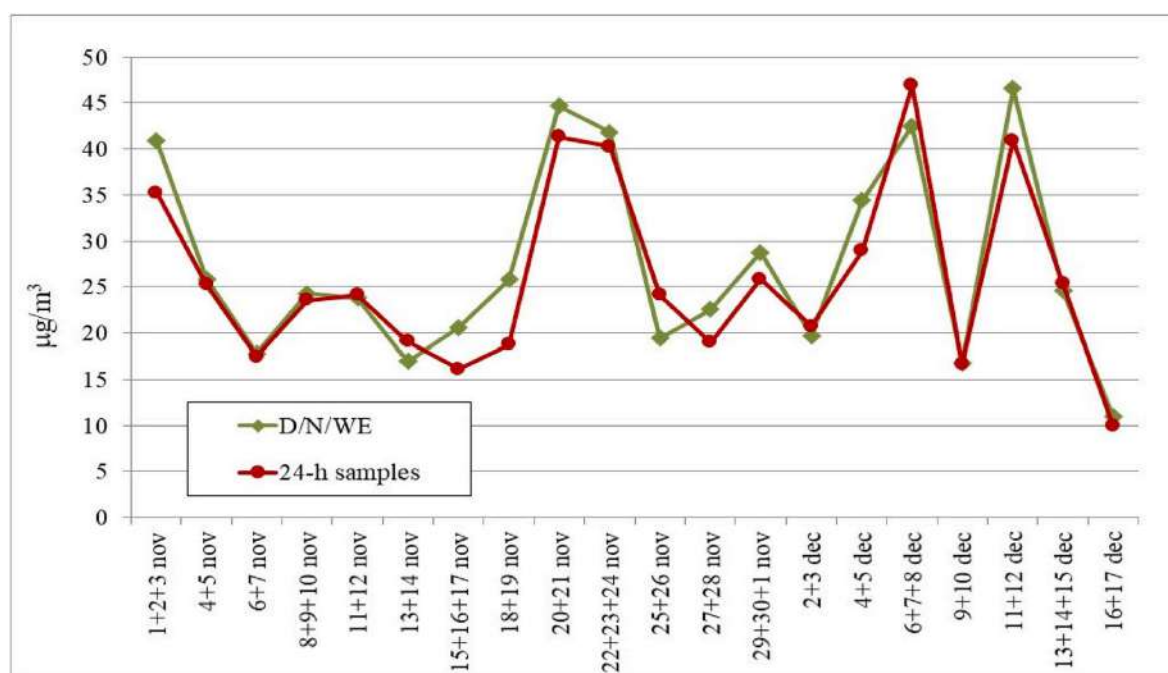


Fig. 2. Comparison of the outdoor PM_{10} mass concentration obtained by the 24-h and the D/N/WE measurements.

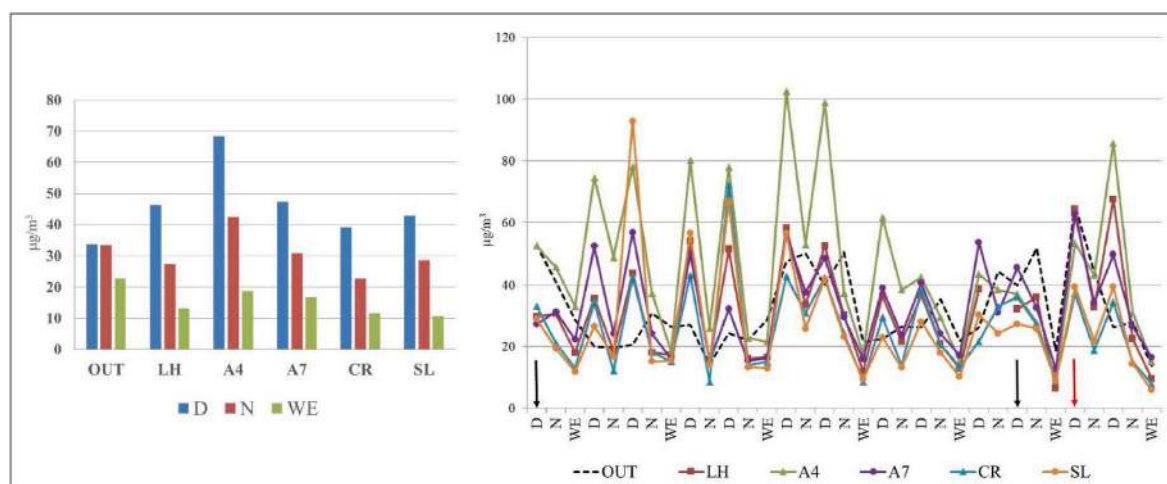


Fig. 3. Mean PM₁₀ concentration at the six environments during the day (D), night (N), and weekend (WE) periods (left panel); time pattern of individual concentration during each D, N and WE period (right panel).

shown by *soil* components: the mean indoor/outdoor ratio (I/O) during the D periods varied between 4.3 in A4 and 2.0 in CR. This increase may be attributed to the transport of dust from outdoors on shoes and clothes, and the re-suspension of deposited dust operated by the students' movements (Wang et al., 2021). The difference in the values recorded in A4 and A7 (A4 > A7) might be partially explained considering that the two identical classrooms are located at the 2nd and 4th floor, respectively: the additional flights of stairs can justify a lower amount of dust stuck to shoes.

The indoor increase in *soil* components is all the more important considering that the particles released by this source belong to the coarser fraction of PM₁₀, characterised by a low infiltration (Bennett and Koutrakis, 2006; Chen and Zhao, 2011). During the summer period, when the building was closed, the I/O value was in the range of 0.3–0.4 (Tofful et al., 2021). During the winter intensive campaign, so low I/O values were reached only at CR and SL during the weekend (I/O = 0.5) and, to a lesser extent, during the night periods (I/O = 0.6 and 0.7, respectively). In the three classrooms (LH, A4 and A7) instead, higher I/O values were recorded during both N and WE periods, particularly at A4 (Supplementary material Table S3). Plausible reasons for this increase in *soil* dust during the N and WE periods are the sweeping and cleaning of the classrooms, carried out each working day before 9:00, and the presence of some students after the end of the classes and, occasionally, on Saturday (never in CR and SL). Also, the slow sedimentation of the particles may have been responsible for a still high concentration during the first hours after the end of the classes.

Interestingly, during the D period indicated by the red arrow (December 11th - 12th) the *soil* concentration was similar in all the indoor environments and close to the outdoor value. During those two days, Central Italy was affected by a Saharan dust transport event, which had its maximum intensity during the morning of December 12th (Supplementary material Figure S1). These long-range transported particles were probably smaller and more easily infiltrating than those of local origin and were responsible for the high values of *soil* components recorded during the D and N periods outdoors (26.3 µg/m³) and in all the indoor environments. During this event, the outdoor concentration of PM₁₀ reached its maximum value (65 µg/m³, Fig. 3).

A considerable contribution to indoor PM₁₀ was due to the *organics* (Fig. 4, middle panels). Although they constitute a significant fraction of indoor PM, the literature reports that only a very small fraction of the organic species (10–30%) can be identified at molecular level, and little information is available about the possible sampling artefacts in indoor environments and the specific sources of these species (Polidori et al., 2006). The indoor formation of secondary organic aerosols as a product

of terpene-ozone reactions is a possible pathway for the contribution of people to organic particles (Rösch et al., 2017). Terpenes (limonene, α-pinene), in fact, in the form of scented agents, are commonly used in cleaning and personal hygiene products. It is also known that humans emit bioparticles (Bhangar et al., 2014), mainly in the form of skin fragments (Marcovecchio and Perrino, 2021a,b). A bioaerosol contribution of 25% to the OM mass and 10% to the PM₁₀ mass was detected during the present experimental study by Marcovecchio and Perrino (2021b). Fig. 4 shows that indoor daytime concentration of the *organics* was generally higher than outdoors and higher than those recorded during the N and WE periods. As in the case of *soil*, the concentration of the *organics* was the result of at least two sources: penetration from outdoor and indoor production. Since the latter source produce flat and light particles, they tend to sediment very slowly and were still present in the N samples.

The concentration of outdoor sulphate did not show important variations among the D, N and WE periods (Fig. 4, lower panels). In all indoor environments, during N and WE period the I/O ratio was about 0.8, in agreement with the behaviour of a species that is mainly in the fine fraction of PM₁₀ and easily penetrates indoors. Instead, a source of sulphate was detected in A4, SL and, occasionally, CR during the D periods. This source may be identified in the modern type of blackboard chalk, which is made of mineral gypsum (calcium sulphate).

Interestingly, during the Saharan dust transport event of December 12th (red arrow in Fig. 4) the outdoor concentration of sulphate increased remarkably. The back-trajectory show that the air masses came from the industrial harbour of Sfax, in Tunisia (Supplementary Material Figure S2). The increase in the sulphate content of PM in Rome during a very similar long-range transport episode had been already recorded by Perrino et al. (2010).

The strength of the indoor sources affecting *soil* components, *organics* and sulphate varied from period to period and from one environment to another. Although it was not possible to have a complete recording of the number of students attending each lesson, the occupancy of the environments (number of classes per day, number of students per class) was undoubtedly a determinant for the concentration increase. The lowest increase in *soil* and *organics* was recorded at CR, the less crowded environment, with a minimum allowed space of 11 m³/person; the highest ones were recorded in the three classrooms (LH, A4, A7, about 4 m³/person).

Fig. 5 reports the time pattern of three component groups that had no relevant indoor sources. In the case of nitrate, the concentrations recorded in all the indoor environments were much lower than outdoors. This difference was due to the shift in the ammonium nitrate

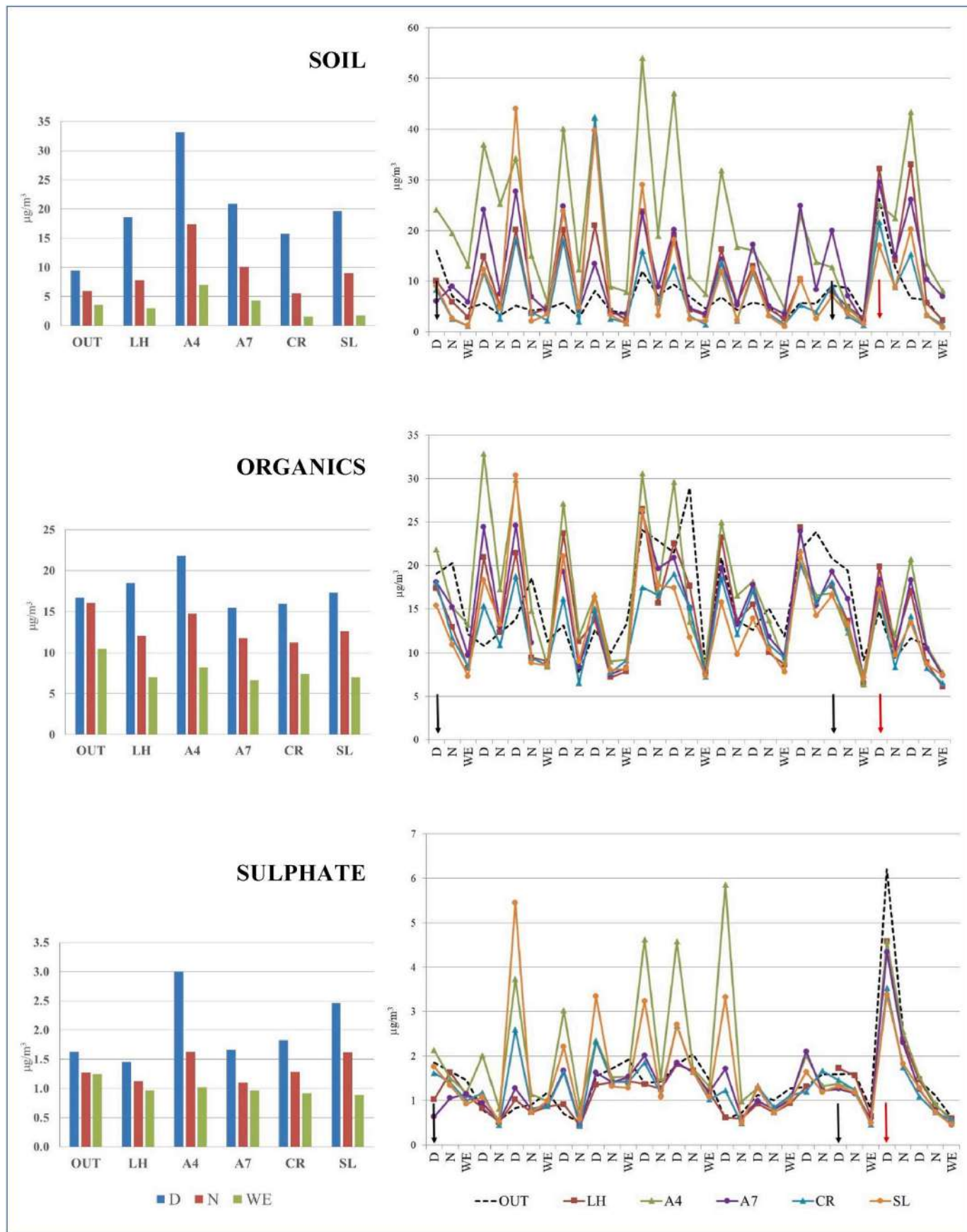


Fig. 4. Mean concentration of the macro-sources soil and organics and of sulphate during the day (D), night (N), and weekend (WE) periods (left panels); individual concentration during each D, N and WE period (right panels).

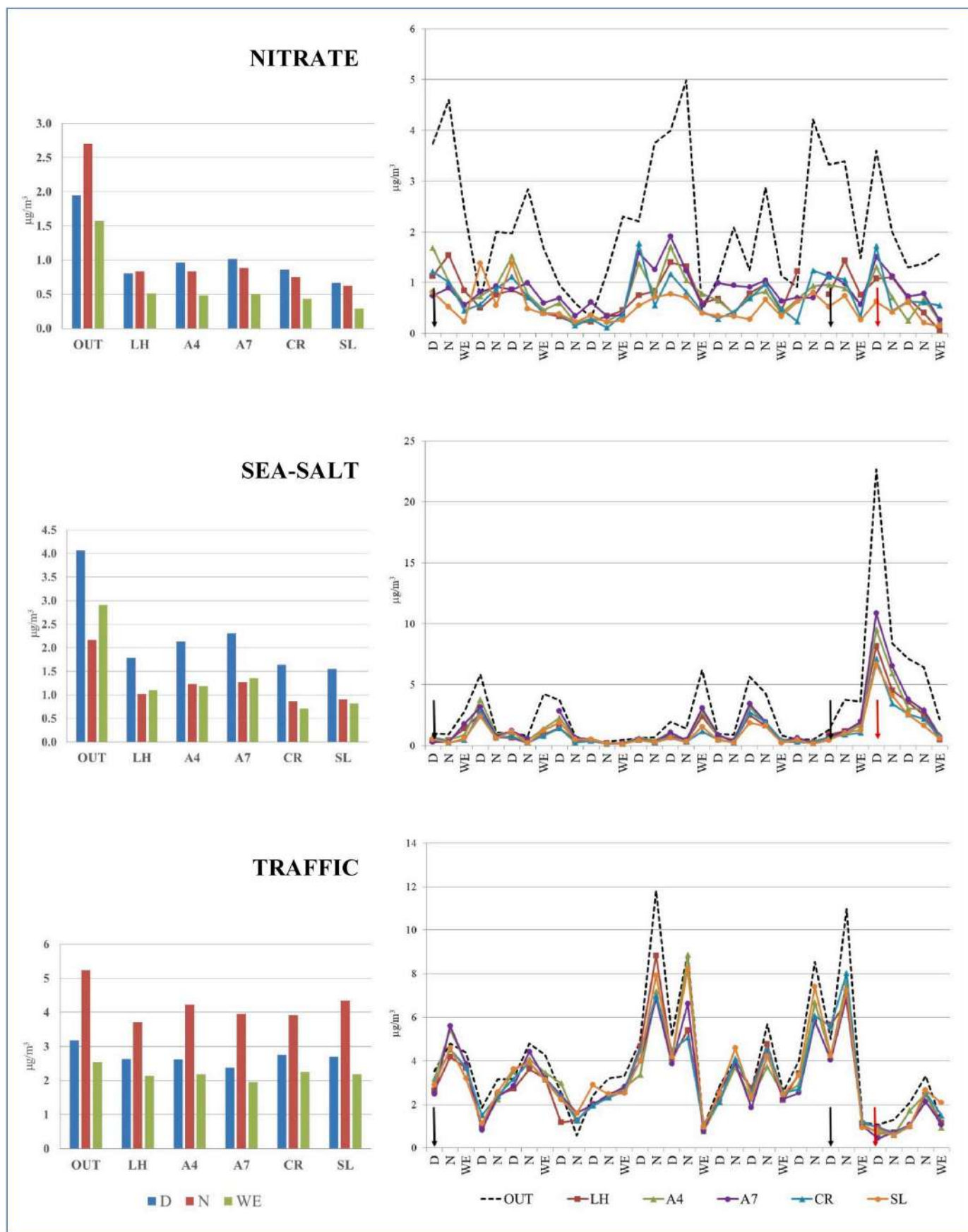


Fig. 5. Mean concentration of nitrate and of the macro-sources sea-salt and traffic emission during the day (D), night (N), and weekend (WE) periods (left panels); individual concentration during each D, N and WE period (right panels).

equilibrium towards the gas phase caused by the different thermodynamic conditions occurring in the classrooms (Keck and Wittmaack, 2005; Perrino et al., 2012). This behaviour has been frequently observed in indoor environments (Lunden et al., 2003, among others).

In the case of *sea-salt* (Fig. 5, middle panels), indoor concentrations kept the same time pattern as outdoors, but they were always much lower, in agreement with the absence of indoor sources and the low penetration of these particles, which are in the coarse fraction of PM₁₀. In all the indoor environments, I/O was 0.4 during the N and WE periods. During the D periods, I/O raised to 0.6 in CR, LAB and LH, and 0.8 in A4 e A7, due to the significant air exchanges through windows and doors during lesson time.

The lower panels in Fig. 5 refer to *traffic emission* and, in particular, the direct emission from exhausts. In this case, again, indoor values followed the time pattern of the outdoor concentration. Being exhaust particles in the fine and ultra-fine dimensional range and thus able to efficiently infiltrate in indoor environments, the I/O values were in all cases between 0.8 and 0.9.

It is worth noting that PM concentration due to traffic emission was higher during the N periods because the two rush hours in Rome occur during the time spans 7:00–9:00 and 18:00–20:00, both included in N. Moreover, the highest nighttime concentrations occurred during the N periods of the third and fifth weeks of the campaign. This was due to the more stable conditions of the atmosphere, which are highlighted in the natural radioactivity pattern reported in Fig. 6. During the cold season, periods of severe atmospheric stability are characterised by sequences of high natural radioactivity values during the night and, especially, during daylight hours, which are due to a very weak or negligible mixing of the lower atmospheric layers even during the sunniest hours. These circumstances mainly occurred during the weekdays from November 20th to 24th and December 4th to 8th, with very stable conditions during the daylight hours of November 21st and December 7th, respectively. The N periods that included the nights immediately following these two very stable days showed the maximum build-up of pollutants and thus the maximum concentration of the species emitted by vehicular traffic.

The comparison between the time pattern of natural radioactivity (Fig. 6) and those of the macro-sources in the outdoor environment (dotted black line in Figs. 4 and 5) clearly shows that the mixing properties of the lower atmosphere mainly influenced the organic components and ammonium nitrate, as well as direct traffic emission. A net

increase in the concentration of PM generated by these sources was, in fact, observed during the weekdays from November 20th to 24th and December 4th to 8th, when the pattern of natural radioactivity shows high nighttime values and, especially, high daylight minima. Conversely, the increase in natural PM sources (*sea-salt* and soil) was mostly observed during advection periods (last week of the campaign), characterised by constantly low values of natural radioactivity.

3.4. Sources tracers

Fig. 7 reports the time pattern of the extracted or residual fraction of three elements that are reliable tracers of sources that cannot be easily described using the main PM components.

The extractable fraction of V (V_{ext}) is a well-known tracer of heavy oil and coke combustion for power generation, industry and shipping (Mazzei et al., 2021; Bai et al., 2021). The upper panels in Fig. 7 show that minor differences between D, N and WE periods were observed outdoors. The increase in the mean concentration during the WE was due to the *sea-salt* episode that occurred on November 25th–26th, when the wind blew from the coast towards the city (see also the middle panel in Fig. 5).

In general, indoor concentrations followed the pattern of outdoor V_{ext} with I/O values close to one. However, during some D periods, indicated in Fig. 7 by an asterisk, a concentration increase of unknown origin was detected in some indoor environments (A4, SL and, to a lesser extent, A7). During the Saharan dust event over Central Italy (red arrow), when the air masses passed over the Tyrrhenian sea, an increase in V_{ext} concentration was observed both indoors and outdoors. It is interesting to note that V_{ext} and *sea-salt* (Fig. 5) show a similar increase in outdoor concentration but a different infiltration in indoor environments due to the different dimensions of the typical particles produced by combustion (V_{ext} , fine) and natural sources (*sea-salt*, coarse). Besides *sea-salt*, during this episode the pattern of V_{ext} was very similar also to sulphate (Fig. 4), which probably shares the same industrial and shipping sources (Cusack et al., 2012): during the last week of the campaign the correlation was very good ($R^2 = 0.91$), while it was poor ($R^2 = 0.21$) during the rest of the period).

The extractable fraction of Rb (Rb_{ext}) is an excellent tracer of biomass burning (Massimi et al., 2020). It has been reported that in Rome the biomass burning for domestic heating is responsible for a non-negligible

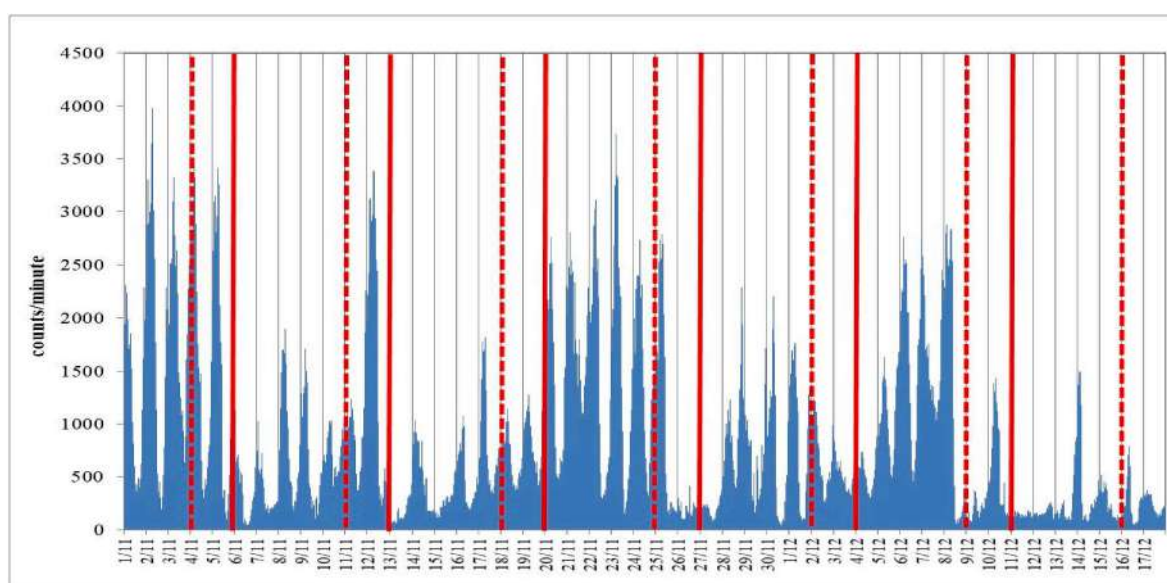


Fig. 6. 1-h time pattern of natural radioactivity during the campaign. The dotted red lines indicate the beginning of weekends, the solid lines indicate the beginning of each weekday period. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

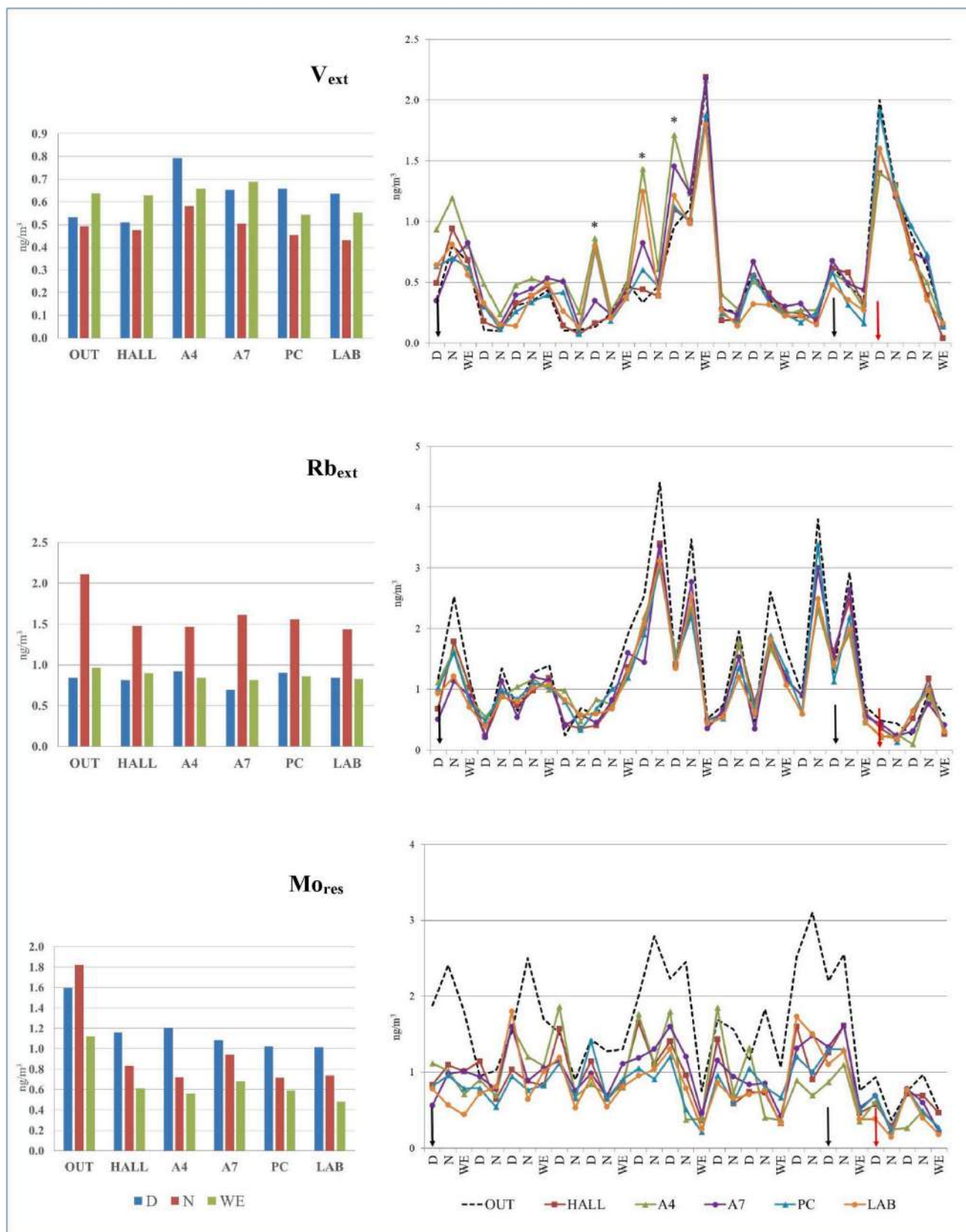


Fig. 7. Mean concentration of the extractable fraction of Rb and V and the residual fraction of Mo during the day (D), night (N), and weekend (WE) periods of the winter campaign (left panels); individual concentration during each D, N and WE period (right panels).

fraction of PM₁₀ (4.5–9.2%), due to both the transport from the countryside surrounding the city and the contribution of local heating appliances (Perrino et al., 2019; Massimi et al., 2022). As expected, the concentration of the tracer of this nighttime source was much higher during the N than during the D periods (Fig. 7, middle panels). For the same reasons discussed for traffic emission, also the time pattern of Rb_{ext} showed its maximum values during the periods of atmospheric stability.

Indoors, the time patterns of Rb_{ext} in all the environments well followed the outdoor variations. Being combustion particles in the fine and ultra-fine dimensional range, during the N and WE the I/O ratios were high (0.7–0.9). The I/O values were even higher during the D periods, in the range 1.0–1.1. Besides the more frequent air exchanges occurring during lesson time, the reason for these results stays in the incomplete ability of the extractable and residual fraction of Rb to trace different PM sources. It has been shown that Rb_{ext} traces PM from biomass burning, while Rb_{res} traces particles from the soil (Massimi et al., 2020). However, the split between the two sources is not clear-cut, and during the D periods, when biomass burning is minimal, soil re-suspension inside the classrooms may be responsible for a slight increase also in the concentration of the extractable fraction of Rb.

A further relevant source of PM in urban areas is the so-called non-exhaust traffic emission, which includes abrasion of brakes and other mechanical parts, tires and asphalt, and re-suspension of the deposited road dust (Charron et al., 2019). In the literature, a number of elements (Cu, Fe, Mn, Mo, Sb, Sn) have been indicated as tracers of non-exhaust emission (Thorpe and Harrison, 2008). Among these, the residual fraction of Sb, contained in brake pads, had shown excellent performances (Canepari et al., 2008). However, some of the above elements (Fe, Mn) are not selective tracers because they are also contained in the soil, and Cu and Sb_{res} have now become weaker tracers due to changes in the chemical composition of modern brake pads. Mo e Sn, and particularly their residual fraction, are now the most selective tracers of non-exhaust traffic emission (Massimi et al., 2022).

The lower panels in Fig. 7 report the results obtained for Mo_{res}, which are very similar to those obtained for Sn_{res}. The pattern of the outdoor concentration shows a general increase during the N periods, when traffic intensifies, as observed for the *traffic emission* source (lower panels in Fig. 5). However, the nighttime increase was lower, with an N/D ratio of 1.6 in the case of exhaust emission and 1.2 in the case of non-exhausts. A possible reason for this behaviour is that coarse particles, including those released by mechanical abrasion and re-suspension, tend to sediment more quickly than fine and ultra-fine particles emitted by combustion processes. Another possible explanation is the weaker re-suspension occurring during the night, when the wind speed is also lower. It is worth noting that during the first two WE periods of the campaign, and, to a lesser extent, during the third one, despite the low traffic intensity in the University area, the outdoor values were much higher than during the following WE periods. This was due to the stability conditions that occurred on November 5th and 12th, highlighted by the high natural radioactivity values in Fig. 6.

Indoors, the concentration of the particles released by non-exhaust traffic emission was much lower than outdoors, particularly during the N and WE periods. Being these particles in the coarse fraction of PM, their I/O ratio was in the range of 0.4–0.5. During the D period, the I/O ratio raised to 0.6–0.7 due to the presence of some particles released by this source in the dust transported from outdoors by the students.

4. Conclusions

The choice of indoor environments attended by people only during well-defined time intervals and characterised by a few PM sources allowed a fine description of the behaviour of atmospheric particles. Thanks to an extended chemical characterisation of the PM samples, it was possible to study the behaviour of particles from different emission sources when they enter the indoor environments (soil, sea, atmospheric reactions, biomass burning, heavy oil combustion, exhaust and non-

exhaust traffic emission, organics).

Substantial differences in PM concentration and composition between attended and unattended periods were highlighted and attributed to the presence/absence of the students. PM components mainly produced by outdoor sources were similar in all the classrooms, while remarkable variations were observed when considering PM coming from indoor sources (blackboard chalk and soil re-suspension).

The chemical composition of indoor PM₁₀ was very different from outdoors and determined by the infiltration, which is different for different particle size ranges, and by the presence of people. The latter had an influence mainly on soil components and organics. Soil particles were carried inside by the students' stepping and suspended and re-suspended by their movements. Organics were constituted by the bio-aerosol (skin flakes) and other particles, probably of secondary origin. The use of blackboard chalk constituted an additional typical source in the classrooms. In general, fine combustion particles penetrate more easily into the indoor environments, while coarse mechanically-generated particles show lower indoor/outdoor ratios.

The influence of the mixing properties of the lower atmosphere on the concentration and composition of outdoor PM was reflected in the behaviour of indoor particles, particularly in the case of anthropogenic sources (domestic heating, traffic emission).

Further research is needed to study the specific sources of PM, especially of organic species, in more complex indoor environments.

Funding

This research was funded by INAIL in the frame of its scientific research programs (2016–2018).

CRedit authorship contribution statement

C. Perrino: Conceptualization, Methodology, Validation, Formal analysis, Writing – original draft, Writing – review & editing, Visualization, Supervision. **A. Pelliccioni:** Project administration, Supervision. **L. Tofful:** Investigation, Formal analysis, Data curation, Resources. **S. Canepari:** Conceptualization, Methodology, Validation, Visualization, 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

The authors are indebted to F. Marcovecchio, S. Dalla Torre, M. Giusto, S. Pareti, E. Rantica and T. Sargolini (CNR-IIA), M.L. Astolfi, E. Marconi, M. Ristorini, M.A. Frezzini and L. Massimi (Sapienza University) to for their contribution to the sampling and analytical phases of the project, and to T. Davanzo and A. Paglialonga (CNR-IIA) for their contributions to the project management.

The authors gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model (<https://www.ready.noaa.gov>) used in this publication.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.atmosenv.2022.119260>.

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