



Chemical speciation and oxidative potential of PM₁₀ in different residential microenvironments: Bedroom, living room and kitchen

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ABSTRACT

Exposure to particulate matter (PM) and its chemical constituents in residential microenvironments has become a major health concern worldwide. The oxidative potential (OP) has been proposed as a metric for estimating the PM capacity to induce oxidative stress and, consequently, health effects. In the present study, PM₁₀ was daily monitored simultaneously in the bedroom, living room and kitchen of three dwellings for one week in a small town of Portugal, to perform a detailed characterisation of its organic and inorganic constituents and the determination of the OP. Bedrooms (B) were found to be a hotspot of PM₁₀ concentrations (B1 = 22.7 μg m⁻³, B2 = 19.5 μg m⁻³; B3 = 68.1 μg m⁻³). PM₁₀-bound elements varied significantly between microenvironments in all dwellings. Lower molecular weight polycyclic aromatic hydrocarbons (PAHs) were found to be between 14 and 72 times higher than high molecular weight PAHs in bedrooms. The mean volume-normalised OP determined by the dithiothreitol and ascorbic acid assays varied within the 0.01–0.38 nmol min⁻¹ m⁻³ and 0.03–0.53 nmol min⁻¹ m⁻³ ranges, respectively. Quinones, oxy-aromatic, aromatic and alkyl-aromatic compounds stood out in bedrooms. Strong and significantly positive relationship between OP and black carbon, Cu and Br were observed, indicating common redox active species mainly associated with traffic emissions. Sr, Fe, Zn and Zr presented higher concentrations in dwelling 3, exhibiting excellent positive correlation with OP, indicating that the Sahara dust intrusion recorded in that house may have contributed to the formation of more redox active species thought to drive antioxidant depletion responses.

1. Introduction

Over the last recent decades, increased technology and urbanisation have remarkably upgraded people's lifestyle in developing countries. However, a considerable attention has been given to the notably harmful effects of atmospheric pollutants on human health over the years. Recent reports have indicated that exposure to air pollution causes about 7 million premature deaths each year [1]. Air pollution ranked 5th among 87 risk factors assessed by the Global Burden of Diseases (GBD) study [2]. This study pointed out that household air pollution accounted for

210 million disability-adjusted life-years and 6.45 million deaths, highlighting the significant impact on public health. Among the different air pollutants, particulate matter (PM) is particularly concerning, as it not only contributes to global climate deterioration, but also to adverse health outcomes [3]. Thus, PM concentrations are commonly used as an indicator of air pollution [4–7]. Air quality management policies with more stringent control regulations have been gradually implemented in most countries [8]. However, ambient concentration levels assessed by national air quality networks in fixed monitoring stations may mislead people's real exposure. Since people

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spend about 90 % of their time indoors [9–11], exposure in these environments has a more significant impact on human health than outdoor PM concentrations [12–14]. Several studies have reported two to five times higher concentrations of pollutants indoors and twice as toxic when compared to levels recorded outdoors [15,16]. Thus, attention should be paid to indoor air quality, which is only regulated in some countries (e.g., Portugal, Finland, Slovenia, and Lithuania).

Indoor air quality has been assessed for many years, though not as intensively as outdoors, mainly in schools and offices (e.g., Refs. [17–20]). However, only a few studies have focused on specific housing microenvironments [21–26]. This is likely due to the high costs of such studies and the difficulty of implementing experimental setups in environments where people rest and socialise (e.g., bedroom and living room), which can be disrupted by the noise of equipment. Furthermore, the estimation of the health effects due to exposure to indoor particles is one of the main challenging tasks, because the harmful effects of these particles depend on the chemical composition, duration of exposure, emission sources, and specific characteristics of the target population [13]. So far, most work in residential environments has used low-cost and/or photometric devices, which generate less discomfort for occupants [25,27]. On the other hand, studies that assessed the chemical composition, toxicity and oxidative potential of PM were mainly conducted in only one specific microenvironment (e.g., living rooms or kitchens) [15,28–31]. Either the mass concentrations of PM or its chemical composition in a specific microenvironment may not be representative of the exposure in the entire home. Given the lack of information for specific residential microenvironments, the present study focuses on the simultaneous daily monitoring of PM₁₀ in three different microenvironments (bedroom, living room and kitchen) of several houses in a small town of Portugal to assess the chemical composition and oxidative potential. Given that the representativeness of the exposure may vary with the microenvironment, this study can provide important information to direct future research on indoor air quality and human health in residential settings.

2. Material and methods

2.1. Monitored microenvironments

The indoor sampling campaign took place in dwellings located in Bragança, an inland city with approximately 35,000 inhabitants, 1/3 of whom are students [32], located in the northeast of Portugal in a mountainous region (altitude 690 m). Characterised as a temperate climate (Csa) according to the Köppen–Geiger classification [33], the region is characterised by high thermal amplitudes, with frequent rains during the winter and dry and hot summers [34]. Three naturally ventilated dwellings were selected for this study (Fig. 1): one located on the ground floor in a residential neighbourhood with low traffic density (Dwelling 1), a second one situated on the first floor facing a main street with intense traffic (Dwelling 2), and a third one positioned on the third floor away from the central area in a building complex (Dwelling 3). In each one, the monitoring campaign covered three specific microenvironments: bedroom (B), living room (LR) and kitchen (K). All dwellings had the same number of rooms: 3 bedrooms, 1 living room and 1 kitchen. Given the technical difficulties in operationalising the monitoring campaign, mainly due to the annoying noise of the equipment (especially in the bedrooms), the target homes and specific microenvironments were selected among the residents who volunteered to participate in the study under these conditions.

All dwellings were composed of non-smokers students aged between 25 and 30 years. A common characteristic between dwellings was that occupants spent more time in the bedrooms than in other microenvironments, even outside of sleeping hours. General tasks, such as eating, studying and physical activities, were generally carried out in the bedroom, followed, sometimes, by the use of the living room. The different configurations and characteristics of the microenvironments are described in Table 1.

The occupants were instructed to carry out their usual activities and to record their departure and arrival times, opening/closing of windows and doors, use of the kitchen, cleaning, among other general activities in a logbook (Table S1). Ventilation conditions refer to the prevailing settings, as reported by occupants in the logbook. In general, all microenvironments were individual rooms, divided by walls and

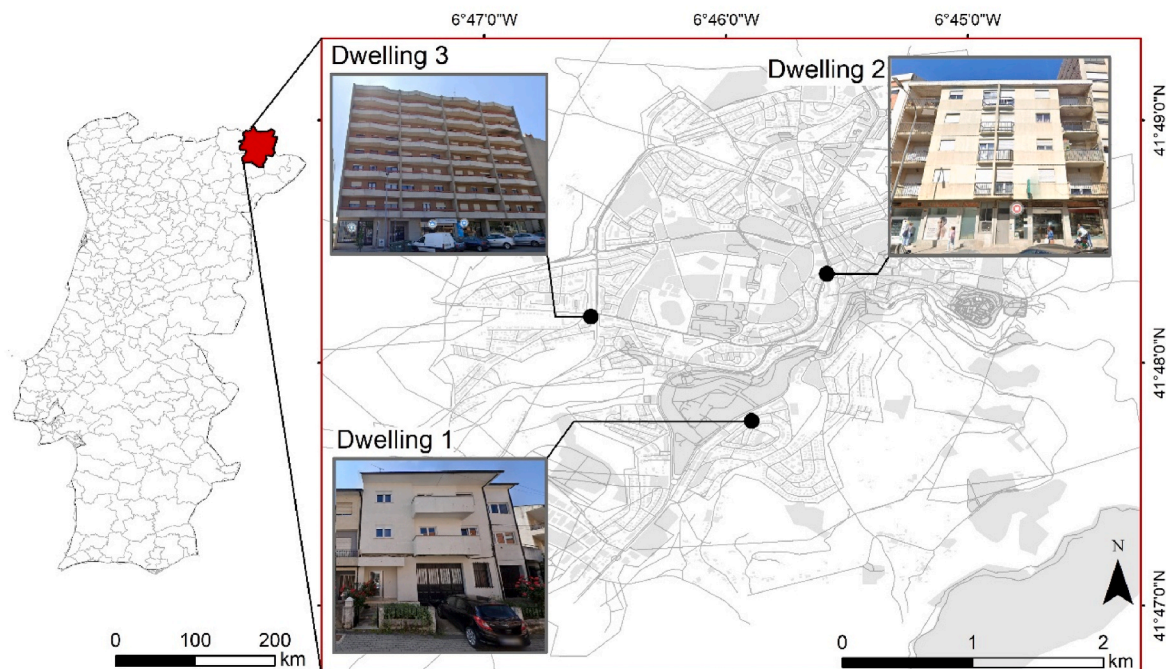


Fig. 1. Municipality of Bragança, map of the city and location of the residences targeted in the study.

Table 1
Characteristics of each microenvironment in the 3 dwellings.

	Dwelling 1			Dwelling 2			Dwelling 3		
	B	LR	K	B	LR	K	B	LR	K
Windows orientation	Yard	Street	Street	Street	Yard	Yard	Parking	Parking	Street
Area (m ²)	16	30	30	12	20	10	10	40	20
Ventilation settings ^a	CD/SW	OD/SW	OD/OW	CD/SW	CD/CW	CD/CW	CD/SW	OD/OW	OD/CW
Permanent occupants	4			3			4		
Daily occupancy (h)	20			14			24		
Source of energy for cooking	Gas			Gas			Gas		
Pets	2 cats			–			1 dog		

^a OD/CD - open doors/closed doors; OW/SW/CW—open windows/semi-open windows/closed windows.

communicating with each other through hallways. The average distance between microenvironments was 7 m.

2.2. PM₁₀ measurements

Gravimetric and continuous PM₁₀ concentrations were consecutively monitored in 3 residential indoor microenvironments, with simultaneous measurements in bedroom, living room and kitchen. Sample collection took place over 3 weeks in the 2022 spring (end of May to mid-June), covering one week in each dwelling. For each microenvironment, the setup consisted of one photometric and gravimetric device. Continuous monitoring of PM₁₀ in the kitchen and living room was performed with a light-scattering laser photometer (DustTrak DRX 8533, TSI inc., Shoreview, MN, USA), whereas in the bedroom an Optical Particle Sizer (OPS 3330, TSI inc., Shoreview, MN, USA) was used. All equipment was calibrated at the manufacturer before the monitoring campaign. The monitors were operated with a 1-min time resolution, and before moving to the next dwelling, zero check, inlet cleaning and impactors greasing were performed. Simultaneously, the gravimetric PM₁₀ collection took place for 24 h and was performed by low-volume samplers (Echo Tecora, Italy – bedroom and kitchen, and Leckel LVS6, Germany – living room) at a flow rate of 2.3 m³ h⁻¹. The gravimetric devices were equipped with 47 mm pre-weighed PTFE filters (Pall) and were positioned at a minimum distance of 1 m from the walls. Their inlets were at 1.5 m high from the ground, corresponding approximately to the breathing zone. All filters were stored in petri slides, conditioned at -20 °C in a freezer between collection and analyses to avoid, as much as possible, degradation.

2.3. Analytical details

2.3.1. Black carbon measurements

A multi-wavelength absorption black carbon instrument (MABI, Ansto) was employed to measure the BC content in all PM₁₀ samples. The light transmission was measured through unexposed filters (I₀) and sampled filters (I) at seven wavelengths (405 nm, 465 nm, 525 nm, 639 nm, 870 nm, 940 nm, and 1050 nm). In this study the reported BC mass concentrations were determined for the central wavelength of 639 nm. This corresponds to the total BC and includes both fossil fuel and biomass burning emissions. The BC was determined by Equation (1):

$$BC (\mu\text{g m}^{-3}) = \frac{10^2 \cdot A}{\varepsilon(\lambda_{639}) \cdot V} \cdot \ln \left[\frac{I_0}{I} \right] \quad (1)$$

where $\varepsilon(\lambda_{639})$ is the mass absorption coefficient with a value of $\varepsilon = 6.036 \text{ m}^2 \text{ g}^{-1}$, which was recommended for 47 mm PTFE filters [35,36]. A is the sampled filter area (cm²) and V is the sampled air volume (m³).

2.3.2. Elemental analysis

Elements with $Z > 10$ (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Br, Rb, Sr, Y, Zr, Mo, Ba, Pb) were directly measured in PTFE filters by particle-induced X-ray emissions (PIXE). This technique had been widely tested and intercomparisons were made with ion

chromatography on a vast array of data to ensure high quality standards, as described in detail by Lucarelli et al. [37]. Each sample was submitted to irradiation with a 3.2 MeV proton beam (~2 mm² spot, 5–50 nA intensity) for ~5 min. After filter scanning to analyse the deposited area, the GUPIX code was employed to fit the PIXE spectra. Quality control of PIXE measurements was carried out in accordance with best practices: at least one sample per sample holder was re-measured, and a reference material (NIST SRM2783 - Air Particulate on Filter Media) was routinely analysed and checked for agreement with the certified values. Elemental concentrations were calculated from a calibration curve from a set of thin standards (Micromatter Inc.) of known areal density (uncertainty of 5 %) A set of thin standards of known areal density (Micromatter Inc.) was used for calibration. The method detection limit (3 σ) ranged from 0.5 ng m⁻³ for Se to 11.6 ng m⁻³ for Ba.

2.3.3. Organic speciation

Half of each filter was extracted 3 times by ultrasonication for 10 min each with three aliquots (20 mL each) of hexane-dichloromethane (DCM) (3:7 v/v) with a 5-min stop between them. The organic extracts were concentrated using a Turbo Vap® II evaporation system (Biotage), dried under a gentle nitrogen stream and analysed in a gas chromatographer-mass spectrometer (GC-MS) with a single quadrupole (models 6890 and 5973, respectively), from Agilent Technologies (USA) equipped with a TRB-5MS 60 m × 0.25 mm × 0.25 μm column. Data were acquired in electron impact (EI) mode at 70 eV using helium as carrier gas at 1.2 mL min⁻¹. The oven temperature program was as follows: 60 °C (1 min); 60–150 °C (10 °C min⁻¹), 150–290 °C (5 °C min⁻¹), 290 °C (35 min). The quantitative analysis was performed by single ion monitoring (SIM) for PAHs and in full scan for other organics. Blank filters were subjected to the same analytical procedures, and their concentrations were subtracted from those of real samples. Calibration was performed with authentic PAH standards (Sigma-Aldrich) at five different concentration levels. Tetracosane-d50 and surrogate standards from Supelco (acenaphthene-d10, benzo(a)pyrene-d12, chrysene-d12, naphthalene-d8, perylene-d12 and phenanthrene-d10), diluted in GC grade solvents (Sigma Aldrich), were used as internal standards. The precision for naphthalene, acenaphthylene, acenaphthene and phenanthrene was 16.9 ± 2.5 %, while for other PAHs it was 4.86 ± 1.78 %. The limit of detection (LOD) for PAHs was 0.26 ± 0.16 ng mL⁻¹. In a previous work, a set tests was carried out to evaluate the efficiency of the extraction process Recoveries ranged from 53.0 ± 10.6 % for phenanthrene to 108 ± 10.6 % for benzo[k]fluoranthene [38].

2.3.4. Oxidative potential assessment

The oxidative potential was assessed with both the dithiothreitol (OP_{DTT} response) and ascorbic acid (OP_{AA} response) acellular methods. For both assays, the OP response was measured as the antioxidant depletion rate of known quantities of DTT and AA, following the experimental procedure described in Pietrogrande et al. [39]. Briefly, the assays were performed on 3 mL of the aqueous extract of a portion of the sampled filter. Filters were previously extracted for 15 min in an ultrasonic bath using 10 mL of 0.1 M buffer at pH 7.4, filtered on a

regenerate cellulose syringe filter (13 mm, 0.22 μm , Kinesis) to remove the suspended solid particles and then introduced into an amber vial at a constant temperature of 37 °C using a dry bath. DTT and AA depletion rates (nmol min^{-1}) were measured by spectrometry by computing the slope of the straight line obtained by linear fitting of 5 points of known reagents concentrations of both reagents at different times (5, 10, 15, 25, and 40 min) [40].

2.4. Enrichment factor

The enrichment factor (EF) was used to assess possible contributions from anthropogenic sources to PM_{10} -bound elements. EF was calculated following Equation (2):

$$EF = \frac{\frac{C_s}{C_{ref}} \rightarrow \text{PM}_{10} \text{ sampled}}{\frac{C_s}{C_{ref}} \rightarrow \text{Crustal concentration}} \quad (2)$$

The EF relates the concentration of a specific element (C_s) with their crustal concentration (C_{ref}), using a reference element to normalise the calculations. The calculation was based on the natural abundance (clarkes) in the upper continental crust [41]. Aluminium (Al) was used as the reference element due to its crustal abundance and generalised use to quantify the EF in environmental samples [42,43]. EFs were calculated for Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Rb, Sr, Zr, Ba, and Pb. Values > 10 suggest that anthropic sources contribute to their excess [44].

2.5. Tracing long range transport with backward trajectories

Saharan dust and wildfire smoke can contribute to the increased levels of ambient PM concentrations in the Iberian Peninsula [45,46]. To confirm the regional transport of PM_{10} during the sampling period, backward trajectories were calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT 4.0) [47] from the National Oceanic and Atmospheric Administration (NOAA). Fire foci locations (<http://effis.jrc.ec.europa.eu>) were checked and combined with the backward trajectories to assess the possible transport of smoke plumes from wildfires in Spain during the last week of monitoring. The 5 day-backward trajectories were calculated using the Global Forecast System Reanalysis (0.25°, global) for the days with PM_{10} concentrations higher than the 90th percentile, arriving at 500, 1000 and 1500 m above the ground level.

2.6. Data quality assurance

For data quality assurance, procedures including field blanks and regular operation checks were followed. Blank concentrations were subtracted from those of real samples. Furthermore, it is widely reported that PM concentrations obtained by photometric devices can provide valuable information with high spatiotemporal resolution [48–50], but can suffer several deviations related to different responses to aerosol properties [51]. In general, DustTrak devices tend to overestimate PM_{10} concentrations [52,53], while OPS underestimates them [54]. Thus, the calibration process is essential to ensure accurate and comparable measurements under the prevailing ambient conditions. The calibration was made using the Tecora and Leckel samplers as reference instruments for PM_{10} measurements, both of which comply with the European norm EN12341:2014. Comparisons of daily filters collected at each microenvironment versus PM_{10} mean concentrations for the sampling time by photometric devices provided a good reproducibility, with r^2 varying from 0.82 to 0.99, slopes from 0.69 to 2.6 and offset from -28 to 1.8 (Fig. S1). Subsequently, the PM_{10} concentrations measured by photometric devices were rectified using the equations for each specific microenvironment.

2.7. Statistical analyses

The descriptive statistics of PM_{10} concentrations and its constituents were performed using the SPSS software (IBM Statistics software v. 24) and MATLAB 2018a (The MathWorks Inc., Natick, MA, USA). The normality of PM_{10} and BC concentrations were tested by the Shapiro-Wilk test. Nonparametric methods were used to evaluate the differences between microenvironments (Mann-Whitney). Spearman correlations between PM_{10} -bound elements (Spearman) were calculated to obtain statistically significant differences at a confidence level of 95 % ($p < 0.05$). Pearson's correlations were used to establish linear relationships between gravimetric and light-scattering measurements.

3. Results and discussion

3.1. PM_{10} mass concentrations

PM_{10} concentrations from gravimetric equipment ranged from 6.53 to 68.1 $\mu\text{g m}^{-3}$. Dwelling 3 showed the highest concentrations for all microenvironments, with daily PM_{10} exceeding the 24-h guideline of 50 $\mu\text{g m}^{-3}$ imposed by the European legislation (Directive 2008/50/EC) on 50 % of measurement days. On the other hand, no exceedances were observed for dwellings 1 and 2. However, the real-time data measured by the photometric monitors showed strong variations over the days (Fig. 2). Inspection of daily concentrations and notes in the logbooks showed that the highest values for all dwellings were recorded in the bedrooms between 8 and 10 a.m. and from 6 to 7 p.m., when the occupants were getting up and returning in the late afternoon. During this time, occupants generally left the doors closed and windows half open when leaving the bedroom. The highest PM_{10} levels in bedrooms of dwellings 1, 2 and 3 were 156 $\mu\text{g m}^{-3}$, 256 $\mu\text{g m}^{-3}$ and 1705 $\mu\text{g m}^{-3}$, respectively. In fact, specific activities such as making beds and using aerosol sprays and incense markedly increased PM_{10} concentrations. Details of the main activities can be seen in Table S2. In addition to indoor sources, outdoor contributions to PM levels were notable in house 3 for all microenvironments. During the sampling period, contributions from long-range transport of smoke from wildfires in Spain and dust from the Sahara Desert in north Africa affected the PM_{10} concentrations (Fig. S2). PM_{10} levels for this dwelling did not show statistically significant differences between the monitored microenvironments. However, the higher concentrations in the bedroom may be associated with the opposite orientation of the kitchen and living room windows, and the activities that favour dust resuspension and the release of clothing fibres in that microenvironment. Although outdoor measurements were not carried out, the influence of infiltration on PM_{10} concentrations in the bedroom of dwelling 2 is notable. The bedroom of dwelling 2 faces an avenue with intense traffic, and even if the windows were ajar, exhaust and non-exhaust emissions may have contributed to the increment in PM_{10} levels. Similar results were reported by Abdel-Salam [21] who found strong correlations between indoor and outdoor PM_{10} concentrations during periods of open windows and argued that infiltration can contribute to the accumulation of particles in rooms. Furthermore, house 2 is the only one in which statistically significant differences ($p < 0.05$) in PM_{10} concentrations between microenvironments were registered.

Overall, PM_{10} levels in this work are comparable to those reported in previous studies. Abdel-Salam [21] measured PM_{10} concentrations in 28 dwellings of older adults in Egypt, reporting mean concentrations of $87.1 \pm 21.2 \mu\text{g m}^{-3}$, $129.8 \pm 23.0 \mu\text{g m}^{-3}$ and $169.5 \pm 58.5 \mu\text{g m}^{-3}$ for bedrooms, living rooms and kitchens, respectively. Similar findings were documented by Madureira et al. [25] for 65 Portuguese households, with PM_{10} concentrations in bedrooms ($49 \mu\text{g m}^{-3}$) 1.2 times lower than in living rooms. Almeida-Silva et al. [55], who measured PM_{10} levels in bedrooms and living rooms of Portuguese elderly care centres, registered mean concentrations of $20.3 \mu\text{g m}^{-3}$ and $28.3 \mu\text{g m}^{-3}$, respectively. Results from the various studies indicate that, in general,

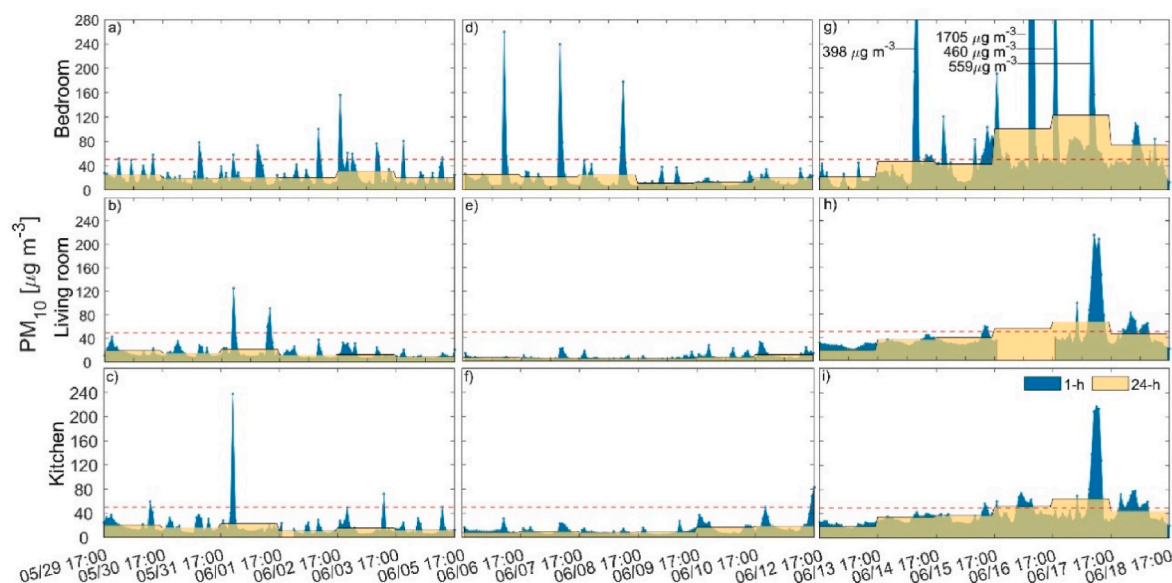


Fig. 2. PM₁₀ 24-h mean concentrations (yellow area) by gravimetric equipment and hourly concentrations measured with photometric devices (blue area) in dwelling 1 (a, b and c), dwelling 2 (d, e, and f) and dwelling 3 (g, h, and i). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

PM₁₀ levels are higher in living rooms, probably due to more intense dust resuspension. The results of the present study are not in line with those reported in the literature, since the PM₁₀ ratios between living room and bedroom were 0.67, 0.32 and 0.63 for dwellings 1, 2 and 3, respectively. This fact indicates the importance of occupant profiles, types of activities, duration of occupancy in each microenvironments and outdoor influences on PM₁₀ concentrations. It is, however, necessary to highlight that these studies may report overestimated concentrations of PM, considering the use of DustTraks and the lack of reference equipment to validate the data, which may represent multiplicative biases of 2–2.5 [51,56]. In this study, the multiplicative factor

varied from 0.69 to 2.6, depending on the specific microenvironment.

3.2. PM₁₀ chemical composition

In addition to BC, major PM₁₀ constituents in all dwellings were Si, Al, Mg, Ca, S, Na, K, Cl and Fe (Table 2). BC is a good indicator of traffic-related PM pollution because it is a side product of incomplete combustion [57–59]. However, residential activities such as cooking, burning candles and incense sticks, smoking and fireplaces can contribute to high indoor concentrations [60–63]. In this study, the mean contributions of BC to the PM₁₀ mass were 13 %, 18 % and 10 %

Table 2
Mean concentrations (±SD) of PM₁₀, BC and elements for each microenvironment in the 3 dwellings.

	Dwelling 1			Dwelling 2			Dwelling 3		
	B	K	LR	B	K	LR	B	K	LR
[µg m ⁻³]									
PM ₁₀	22.7 ± 4.56	16.0 ± 4.89	15.2 ± 5.01	19.5 ± 6.33	11.7 ± 4.71	6.53 ± 2.52	68.1 ± 38.3	42.1 ± 15.7	43.3 ± 16.6
BC	3.63 ± 4.39	1.78 ± 0.56	1.83 ± 0.23	2.93 ± 0.74	1.82 ± 0.95	1.48 ± 0.83	5.12 ± 1.95	4.35 ± 2.01	5.18 ± 2.03
[ng m ⁻³]									
Na	379 ± 121	399 ± 131	390 ± 129	345 ± 169	160 ± 30.4	111 ± 49.7	688 ± 444	397 ± 108	319 ± 94.0
Mg	741 ± 457	201 ± 74.1	197 ± 58.2	92.4 ± 19.8	88.9 ± 25.1	47.8 ± 11.3	526 ± 192	532 ± 160	571 ± 184
Al	642 ± 225	333 ± 87.4	348 ± 74.3	1386 ± 863	438 ± 138	261 ± 81.9	2017 ± 812	2643 ± 1066	3048 ± 1294
Si	1767 ± 876	658 ± 176	669 ± 123	1995 ± 1370	721 ± 312	397 ± 148	4900 ± 1999	5133 ± 1952	5709 ± 2320
P	28.1 ± 8.02	14.9 ± 3.88	16.8 ± 3.04	16.5 ± 14.1	50.9 ± 69.9	11.5 ± 7.58	30.2 ± 11.0	33.8 ± 10.2	39.6 ± 13.6
S	387 ± 214	389 ± 221	399 ± 221	188 ± 56.8	189 ± 53.3	175 ± 60.4	506 ± 34.5	460 ± 28.3	506 ± 37.2
Cl	231 ± 105	229 ± 80.7	210 ± 91.6	357 ± 260	206 ± 219	47.9 ± 16.4	1015 ± 1096	292 ± 213	132 ± 75.1
K	277 ± 69.5	179 ± 40.8	193 ± 35.7	144 ± 75.8	353 ± 396	92.5 ± 56.5	649 ± 273	673 ± 215	753 ± 266
Ca	424 ± 159	204 ± 56.4	215 ± 46.9	294 ± 97.1	260 ± 83.0	101.5 ± 62.1	2857 ± 2621	1449 ± 651	1333 ± 462
Ti	28.3 ± 8.18	15.7 ± 5.63	16.5 ± 3.76	8.49 ± 3.22	12.1 ± 6.47	5.59 ± 4.79	86.5 ± 38.3	115 ± 47.4	133 ± 58.4
V	–	–	–	–	–	–	3.76 ± 0.98	3.55 ± 0.90	4.20 ± 1.01
Cr	3.68 ± 1.09	2.64 ± 0.4	3.03 ± 0.74	2.49 ± 0.64	2.42 ± 0.87	2.05 ± 1.23	2.72 ± 1.14	4.04 ± 1.58	4.38 ± 2.04
Mn	6.49 ± 1.51	4.24 ± 1.08	4.25 ± 0.81	2.36 ± 0.64	2.61 ± 1.05	1.88 ± 0.66	16.1 ± 6.04	22.3 ± 7.99	26.9 ± 11.3
Fe	225 ± 49.2	172.8 ± 38.4	170 ± 41.6	109 ± 15.4	101.7 ± 37.7	59.8 ± 18.9	806 ± 324	1046 ± 392	1210 ± 484
Ni	1.07 ± 0.32	1.02 ± 0.43	0.99 ± 0.39	0.73 ± 0.28	0.89 ± 0.09	0.46 ± 0.15	2.04 ± 0.44	2.29 ± 0.71	2.33 ± 0.64
Cu	7.82 ± 8.67	4.76 ± 4.27	3.58 ± 1.67	19.9 ± 25.7	8.89 ± 12.0	5.84 ± 7.24	6.55 ± 1.81	6.96 ± 2.98	6.97 ± 3.14
Zn	16.7 ± 2.61	10.6 ± 2.16	11.3 ± 2.67	31.7 ± 28.6	16.9 ± 16.6	9.4 ± 6.01	21.4 ± 9.55	15.9 ± 3.66	16.7 ± 3.94
Br	2.38 ± 1.05	2.12 ± 0.68	2.16 ± 0.57	0.73 ± 0.34	0.83 ± 0.41	0.71 ± 0.33	5.28 ± 3.26	4.23 ± 1.20	4.10 ± 1.75
Rb	–	–	–	–	–	–	4.64 ± 1.88	5.87 ± 2.99	5.62 ± 2.83
Sr	2.15 ± 1.65	–	–	–	–	–	5.91 ± 3.63	4.78 ± 1.51	5.34 ± 2.20
Zr	–	–	–	2.85 ± 0.78	1.60 ± 0.08	–	3.54 ± 1.07	3.80 ± 1.51	5.17 ± 1.37
Ba	–	–	12.1 ± 1.87	–	–	10.2 ± 1.91	38.4 ± 14.5	46.3 ± 17.1	51.9 ± 19.1
Pb	4.50 ± 0.86	2.37 ± 0.83	2.99 ± 1.03	1.26 ± 0.27	1.01 ± 0.42	–	4.71 ± 0.96	3.66 ± 1.54	4.01 ± 1.33

for dwellings 1, 2 and 3, respectively.

BC concentrations in dwelling 1 were influenced by cooking and burning incense activities, while in dwelling 2 its main source was traffic-related, as the occupants did not cook often and never lit candles or incense. Dwelling 3, as mentioned previously, was affected by long-range transport of smoke, which was mixed with some indoor emissions from cooking. For all microenvironments, statistically significant differences were observed for BC, indicating that ventilation conditions can favour accumulation or help dispersion.

To perform a PM₁₀ mass balance, concentrations of major and trace elements were converted into the equivalent oxide species (CaO, Al₂O₃, Fe₂O₃, SiO₂, etc.) and summed. Element oxides accounted for PM₁₀ mass fractions of 26 %, 29 % and 32 % for dwellings 1, 2 and 3, respectively. Similar results were reported for a university cafeteria [64], where the dominant elements coincided with those of the present study. Some of these elements (e.g., Ca, Mg, K, Si) are common constituents of mineral matter and can be related to soil dust that is brought in from outdoors on shoes or that infiltrate from outdoors through doors or windows. Furthermore, certain floor materials, humidity, cleaning activities, and construction wear (e.g., walls) can contribute to their accumulation and amounts [65]. The intrusion of Saharan dust in dwelling 3 contributed to a strong increment in concentrations of Si, Ca, Al, Cl, Fe, Na, K, Mg and S. Their values were 1.5 to up 14 times higher than those in other dwellings. The only exception was Mg in the bedroom of dwelling 1, which may have been affected by the presence of a litter cat box. Magnesium is excreted in urine and faeces. Some elements may have resulted from non-exhaust emissions, abrasion of metallic components of building materials or cooking activities. Zn and Cu, for example, have been used as tracers of traffic emissions (exhaust and non-exhaust), especially from tyre and brake wear [66]. In a source apportionment study conducted in Bragança, these elements were dominant in the traffic source profile [67]. Bedroom of dwelling 2 showed EFs from 1.3 to 8 times higher than other dwellings for these elements, with EFs >10 (Fig. 3), reinforcing a traffic-related origin.

Pb and Zn in dwelling 1 displayed higher enrichments than in other dwellings. These elements can accumulate in house dust from common building materials and products, especially in paint pigments that are dispersed into the indoor environment from the cracking and flaking processes caused by oxidation and physical wear [68,69]. Br was highly enriched in all microenvironments. Brominated compounds are used as flame retardants and disperse dyes, being detected in clothing lint. Their presence in indoor dust has been attributed to fibrous components [70]. EFs below 10 were found for Na, Mg, Al, Si, K, Ti, V, Mn, Rb, Sr, Zr, Fe, Ca, and Ba, suggesting that these elements have a crustal origin.

The particulate matter organic extracts encompassed, among others, several polycyclic aromatic hydrocarbons (PAHs), alcohols, acids, phenolic compounds, and other oxygenated compounds (Table 3). PAH concentrations were obtained from calibration solutions containing the

16 EPA priority PAHs (US EPA 610 mix). However, only naphthalene, phenanthrene, fluoranthene, pyrene and chrysene were detected in the samples. The absence of HMW PAHs (e.g., benzo(a)pyrene) may be due to the extraction of only half of each filter and the small amount of accumulated mass. Longer sampling times would probably be necessary. On the other hand, it cannot be ruled out that some of the undetected compounds are present in samples from the winter period, when concentrations are generally higher due to the intensification of residential biomass combustion. The total concentrations of PAHs detected in the bedrooms were higher than those recorded in kitchens or living rooms. The most abundant PAH in PM₁₀ was naphthalene, ranging from 1.11 ± 1.56 to 2.87 ± 6.42 ng m⁻³. This compound was only detected in the bedrooms, with the exception of the kitchen of dwelling 1. This PAH is known as a component of aromatic naphta, naphtalin, cleaning and health care products. PAHs are divided into two groups, depending on the number of benzene rings: LMW (low molecular weight) and HMW (high molecular weight). LMW PAHs include naphthalene and phenanthrene, while HMW PAHs encompass fluoranthene, pyrene and chrysene. Previous studies have associated LMW with greater acute toxicity, while HMW PAHs show a higher potential carcinogenic risk [71]. LMW/HMW ratios <1 for indoor environments have been generally reported in the literature [72–74], indicating a predominance of pyrogenic sources. Contrasting LMW/HMW ratios were obtained in the present study: dwelling 1 (B: 72.3, LV: 0.67, K: 2.43), dwelling 2 (B:14.2, LV: 0.85, K: 0.83) and dwelling 3 (B: 19.2, LV: 0.45, K: 0.54). According to other studies focused on cooking emissions, the proportion of naphthalene is usually higher while cooking [75]. However, in this study, the concentrations of naphthalene during cooking activities were not consistent with these findings. Disparities between results may be related to cooking frequency, ventilation conditions, and differences in sampling methods and analytical techniques.

Some quinone and oxy-aromatic compounds were found in the PM₁₀ samples. Concentrations of 2,6-di-tert-butyl-1,4-benzoquinone (DTBB) and 7,9-di-tert-butyl-1-oxaspiro[4.5]deca-6,9-diene-2,8-dione (DTBO) stood out in dwellings 1 and 3. DTBB is widely used as a chemical intermediate, polymerisation inhibitor and rubber antioxidant. This quinone has been identified in diverse plastic food packaging's [76] and industrial and commercial products to retard oxidative reactions [77]. It has been reported that DTBB cause cleavage of supercoiled DNA at very low concentrations in *in vitro* experiments [78] and can be more toxic than their corresponding parent chemicals [77]. This quinone was previously observed in outdoor PM_{2.5} samples collected near a chemical complex at an average concentration of 115 ng m⁻³ [79]. Lower levels (0.44 ng m⁻³) were reported for indoor campaigns, including a university cafeteria [64] and in a living room (1.34 ng m⁻³) where different vacuum cleaners were used [80]. Given that the indoor concentrations reported in previous studies were significantly lower than those in dwellings 1 and 3, it is assumed that other emission sources or processes

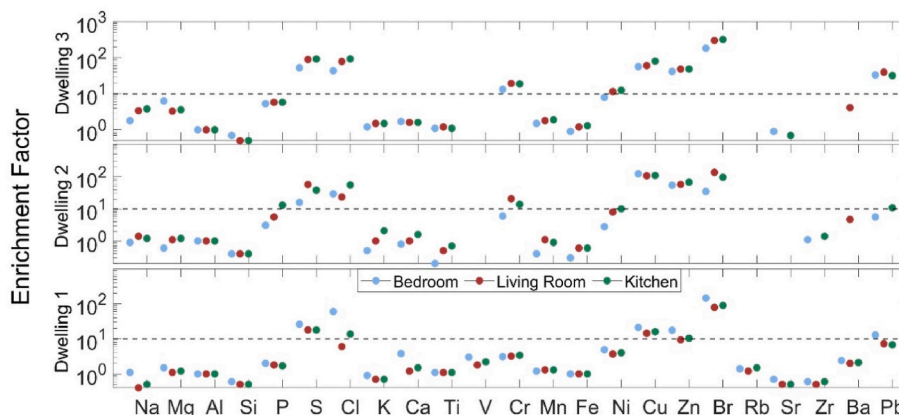


Fig. 3. Element enrichment factors using Al as a reference.

Table 3
Concentrations (ng m^{-3}) of some organic compounds detected in PM_{10} for each monitored microenvironment.

[ng m^{-3}]	Dwelling 1			Dwelling 2			Dwelling 3		
	B	K	LV	B	K	LV	B	K	LV
Aromatic and alkyl-aromatic compounds									
Naphthalene	2.87 ± 6.42	0.11 ± 0.25	<LOD ^a	1.11 ± 1.56	<LOD	<LOD	1.48 ± 1.66	<LOD	<LOD
Phenanthrene	0.02 ± 0.01	0.06 ± 0.04	0.04 ± 0.02	0.03 ± 0.04	0.05 ± 0.03	0.06 ± 0.04	0.06 ± 0.03	0.06 ± 0.03	0.05 ± 0.04
Fluoranthene	0.01 ± 0.01	0.02 ± 0.02	0.02 ± 0.01	0.02 ± 0.01	0.01 ± 0.01	0.02 ± 0.02	0.03 ± 0.01	0.03 ± 0.01	0.03 ± 0.02
Pyrene	0.02 ± 0.02	0.04 ± 0.03	0.03 ± 0.02	0.04 ± 0.03	0.04 ± 0.02	0.04 ± 0.03	0.04 ± 0.02	0.05 ± 0.02	0.05 ± 0.02
Chrysene	0.01 ± 0.01	0.01 ± 0.01	0.01 ± 0.01	0.02 ± 0.01	0.01 ± 0.01	0.01 ± 0.01	0.01 ± 0.01	0.03 ± 0.01	0.03 ± 0.01
1,3,5-Trimethylbenzene	0.87 ± 0.23	0.28 ± 0.18	0.24 ± 0.09	1.16 ± 1.87	0.08 ± 0.07	0.46 ± 0.02	0.34 ± 0.07	0.27 ± 0.05	0.43 ± 0.08
Unidentified aromatic compounds	62.2 ± 77.8	34.6 ± 53.4	55.8 ± 24.5	4.58 ± 8.09	11.6 ± 22.7	15.1 ± 28.8	11.8 ± 8.86	56.6 ± 18.2	78.3 ± 35.4
Quinones and oxy-aromatic compounds									
2,6-Ditert-butyl-1,4-benzoquinone	61.9 ± 80.4	14.7 ± 14.8	6.14 ± 8.15	1.68 ± 3.77	0.24 ± 0.54	1.97 ± 4.39	4.27 ± 6.18	2.03 ± 3.1	4.48 ± 6.78
7,9-di-tert-butyl-1-oxaspiro[4.5]deca-6,9-diene-2,8-dione	132 ± 215	84.8 ± 183	21.6 ± 48.4	10.1 ± 22.5	<LOD	5.06 ± 11.3	47.5 ± 38.6	108 ± 61.1	137 ± 89.2
Benzyl alcohol	1.25 ± 1.38	1.15 ± 0.50	0.59 ± 0.36	0.42 ± 0.15	0.34 ± 0.12	0.28 ± 0.18	0.78 ± 0.31	1.01 ± 0.45	0.77 ± 0.28
BHT-aldehyde (3,5-Di-tert-butyl-4-hydroxybenzaldehyde)	2.04 ± 4.09	0.95 ± 1.11	0.02 ± 0.03	0.16 ± 0.36	0.09 ± 0.21	0.25 ± 0.57	0.56 ± 0.29	0.58 ± 0.42	0.63 ± 0.47
Phenolic compounds									
4-tert-Butylphenol	2.28 ± 1.77	0.65 ± 0.14	0.62 ± 0.26	0.19 ± 0.15	0.57 ± 0.16	0.46 ± 0.13	<LOD	0.36 ± 0.13	<LOD
2,4-Di-tert-butylphenol	4.43 ± 3.06	2.62 ± 0.55	1.69 ± 0.52	0.73 ± 0.28	1.28 ± 0.18	1.29 ± 0.41	0.59 ± 0.25	1.49 ± 0.31	2.49 ± 2.93
Alkyl esters of acids									
Hexadecanoic acid methyl ester	1.32 ± 1.23	1.20 ± 1.91	1.33 ± 0.83	0.59 ± 0.27	0.64 ± 0.35	0.35 ± 0.34	0.52 ± 0.38	2.28 ± 0.57	1.10 ± 0.59
Methyl ester of 3-(3,5-di-tert-butyl-4-hydroxyphenyl)-propionic acid	0.16 ± 0.22	0.96 ± 0.60	0.18 ± 0.27	0.57 ± 0.63	0.18 ± 0.40	0.46 ± 0.77	2.65 ± 3.91	1.02 ± 0.67	1.04 ± 0.41
9,12-Octadecadienoic acid, methyl ester	0.98 ± 1.58	0.67 ± 0.24	0.30 ± 0.26	1.09 ± 0.32	1.47 ± 0.46	1.30 ± 0.20	0.92 ± 0.22	1.04 ± 0.16	0.73 ± 0.22
9-Octadecenoic acid (Z)-, methyl ester (Oleic acid methyl ester)	3.01 ± 1.87	1.47 ± 2.10	1.79 ± 0.53	0.90 ± 0.42	0.83 ± 0.61	0.84 ± 0.51	0.44 ± 0.35	1.94 ± 0.43	0.92 ± 0.31
Octadecanoic acid, methyl ester	1.04 ± 1.26	0.60 ± 1.25	1.28 ± 0.84	0.31 ± 0.15	0.40 ± 0.12	0.41 ± 0.28	0.25 ± 0.21	1.28 ± 0.13	0.61 ± 0.14
Decanedioic acid, bis(2-ethylhexyl) ester	0.64 ± 1.42	<LOD	<LOD	3.66 ± 3.83	<LOD	<LOD	8.52 ± 7.97	<LOD	<LOD
Other oxygenated compounds									
Triethylenediamine	<LOD	5.58 ± 10.6	<LOD	<LOD	<LOD	<LOD	<LOD	8.12 ± 3.4	<LOD
3,5-di-tert-Butyl-4-hydroxyacetophenone	1.08 ± 1.75	0.28 ± 0.26	<LOD	0.11 ± 0.14	0.04 ± 0.06	0.1 ± 0.14	0.14 ± 0.09	0.12 ± 0.13	0.25 ± 0.15
Nonanal	2.97 ± 1.93	11.5 ± 10.8	4.63 ± 7.11	0.59 ± 0.41	1.17 ± 0.85	0.49 ± 0.60	4.31 ± 2.77	1.96 ± 1.05	1.05 ± 0.38
2-Propanol, 1-chloro-, phosphate	1.40 ± 0.82	1.02 ± 0.75	0.39 ± 0.34	0.27 ± 0.14	0.31 ± 0.13	0.28 ± 0.11	0.18 ± 0.18	0.42 ± 0.14	0.31 ± 0.16
Nicotine	0.58 ± 0.65	1.93 ± 0.93	5.13 ± 3.78	<LOD	0.14 ± 0.17	0.04 ± 0.09	<LOD	0.13 ± 0.14	0.27 ± 0.28
Dihydro methyl jasmonate	0.58 ± 0.30	0.99 ± 1.29	0.24 ± 0.08	0.10 ± 0.07	0.10 ± 0.05	0.07 ± 0.05	0.31 ± 0.15	0.37 ± 0.07	0.28 ± 0.13
Tributyl acetylacrylate	2.60 ± 3.74	1.10 ± 0.96	0.88 ± 1.36	0.21 ± 0.45	<LOD	<LOD	3.78 ± 3.73	0.38 ± 0.85	0.33 ± 0.52
Cannabinol	0.49 ± 0.72	2.17 ± 1.62	4.34 ± 3.91	<LOD	<LOD	<LOD	<LOD	<LOD	6.85 ± 15.3
Irgafos 168	13.6 ± 25.2	5.49 ± 10.6	31.2 ± 31.5	81.5 ± 179	18.5 ± 19.8	10.5 ± 9.24	<LOD	<LOD	<LOD

^a LOD – Limit of detection.

took place in these indoor environments. In fact, in these dwellings, DTBO presented higher concentrations and a statistically significant correlation ($p < 0.05$) with DTBB, suggesting plastic materials as a common source. The mean values of DTBO, varying from 5.06 ± 11.3 to $137 \pm 89.2 \text{ ng m}^{-3}$, were higher than that (18.5 ng m^{-3}) obtained in industrial areas of Estarreja, Portugal [79] and India (5.2 ng m^{-3}) [81].

Cannabinol was detected in dwellings 1 and 3. Cannabinol is an abundant substance in cannabis smoke [82,83]. Although only non-smoking homes were selected and occupants were asked not to smoke, the rule may have been broken during the monitoring campaign. Oxidised irgafos 168 (tris(2,4-ditert-butylphenyl) phosphate) was detected in the bedroom of dwelling 2 at relatively high concentrations

($81.5 \pm 179 \text{ ng m}^{-3}$). It is an antioxidant commonly used in industrial formulations of plastic polymers to protect them from aging and oxidation. This organophosphorus compound is also present in polymeric materials commonly used in auto parts, such as rubbers, elastomers and plastic component parts [84]. Furthermore, oxidised irgafos 168 is employed as a stabiliser in brake fluids to enhance their useful life and durability. It was previously detected as a component of brake wear particles [85]. High concentrations in bedroom of dwelling 2 are most likely related to non-exhaust traffic emissions due to the orientation of the facade towards a very busy street. Moreover, BC was correlated with pyrene and chrysene in this bedroom, also reported as traffic-related PAHs [86–88].

Excellent correlations were noticed for dwellings 2 and 3 between PM_{10} and organic acids and alkyl esters ($r = 0.80$ and $r = 0.68$ with $p < 0.05$, respectively), while for dwelling 1 the PM_{10} concentrations were correlated with quinone and oxy-aromatic compounds ($r = 0.72$). In fact, benzyl alcohol and BHT-aldehyde (3,5-Di-tert-butyl-4-hydroxybenzaldehyde) showed the most abundant concentrations in dwelling 1.

3.3. Oxidative potential

The DTT- and AA-OP responses were measured for 37 p.m.₁₀ samples (10 for dwelling 1, 9 for dwelling 2 and 18 for dwelling 3), since it was not possible to assess the OP response of all filters. In some cases, due to the low masses, it was necessary to combine two filters to carry out the extractions. The OP determined by the DTT and AA assays varied within the 0.32–3.93 and 0.45–6.21 nmol min^{-1} ranges, respectively. The OP depletion rate was normalised by the sampled volume (OPv) as an exposure metrics accounting for inhaled air (Table 4). Both assays provided OPv values within similar ranges: 0.01–0.38 $\text{nmol min}^{-1} \text{m}^{-3}$ for DTT and 0.03–0.53 $\text{nmol min}^{-1} \text{m}^{-3}$ for AA. These values are in good agreement with the data reported by Pietrogrande et al. [39] for a suburban area away from large sources of local pollution, where OPvDTT ranged from 0.02 to 0.53 $\text{nmol min}^{-1} \text{m}^{-3}$ and OPvAA from 0.03 to 0.69 $\text{nmol min}^{-1} \text{m}^{-3}$. Zeng et al. [89] reported OPvDTT for $\text{PM}_{2.5}$ in an unoccupied apartment in Chicago ranging from 0.02 to 0.179 $\text{nmol min}^{-1} \text{m}^{-3}$. The present study showed values at the lower end of the typical ranges observed for ambient particles, with OPs for the DTT and AA assays spanning from ~ 0.2 to 2 $\text{nmol min}^{-1} \text{m}^{-3}$ and ~ 0.3 –4 $\text{nmol min}^{-1} \text{m}^{-3}$, respectively [90–93].

Janssen et al. [90] assessed OP of $\text{PM}_{2.5}$ and PM_{10} samples impacted by different sources and found that the responses of DTT and AA assays varied significantly across sampling sites due to changes in particle properties, especially absorbance and mass concentrations. In the present study, it is remarkable that, even at similar PM_{10} concentrations, different responses of OPvAA and OPvDTT were found, mainly related to the PM_{10} -bound elements. The highest OP values for both assays were found in dwelling 3, where the highest concentrations of PM_{10} and elements were observed. In relation to the microenvironments of

Table 4
Oxidative potential values measured with DTT (OPvDTT) and AA assays (OPvAA) normalised by the sampled volume.

Sampling site		OPvAA ($\text{nmol min}^{-1} \text{m}^{-3}$)			OPvDTT ($\text{nmol min}^{-1} \text{m}^{-3}$)		
		min	max	mean \pm SD	min	max	mean \pm SD
Dwelling 1	K	0.05	0.13	0.08 ± 0.03	0.04	0.12	0.08 ± 0.03
	LV	0.05	0.08	0.06 ± 0.01	0.06	0.10	0.08 ± 0.02
	B	0.07	0.31	0.16 ± 0.1	0.07	0.12	0.09 ± 0.02
Dwelling 2	K	0.03	0.22	0.13 ± 0.08	0.01	0.06	0.04 ± 0.02
	LV	0.03	0.08	0.06 ± 0.02	0.03	0.07	0.05 ± 0.02
	B	0.07	0.28	0.14 ± 0.09	0.05	0.17	0.10 ± 0.05
Dwelling 3	K	0.18	0.37	0.25 ± 0.06	0.13	0.38	0.24 ± 0.09
	LV	0.19	0.53	0.33 ± 0.11	0.17	0.25	0.21 ± 0.03
	B	0.10	0.47	0.21 ± 0.12	0.13	0.33	0.22 ± 0.07

dwellings 1 and 2, as observed for PM_{10} concentrations, OPvAA and OPvDTT assays showed higher values in bedrooms, while for dwelling 3 OPvAA and OPvDTT were higher for the living room and kitchen, respectively.

Correlations between the different OPv assays are shown in Table 5. To increase statistical representativeness, the data were grouped by household. Table 5 shows only the PM_{10} -bound elements for which significant Pearson correlations ($p < 0.05$) with OPv were obtained. For dwellings 1 and 2, where lower concentrations were recorded, more species were correlated with OPvAA than with OPvDTT. The inverse was observed for dwelling 3, for which 8 species were correlated with OPvAA and 14 with OPvDTT. On the other hand, PM_{10} concentrations presented no significant correlations with OP assays in dwellings 1 and 2, but showed strong correlations in dwelling 3, suggesting that higher PM_{10} levels can induce higher depletion in the OP assays.

In general, no significant correlation was found between OPvDTT and OPvAA ($r = -0.15$ for dwelling 1, $r = -0.10$ for dwelling 2 and $r = -0.17$ for dwelling 3), suggesting that DTT and AA assays may have different associations with PM_{10} -bound elements. This is in agreement with findings from some previous studies [40,94,95], but in contrast with others reporting significant sensitivity of the two assays to the same chemical species [39,96,97]. In fact, OPvDTT for dwellings 1 and 2 showed significant positive correlation with BC, while OPvAA was correlated with Mg, Al, Si, Cu and Zn in dwelling 1 and with P, S, Cl, K, Ca, Mn, Cu and Zn in dwelling 2. BC, Cu and Br always showed positive correlations for all residential sites, indicating that they act as common redox active species. However, Cu and Br instigated OPvDTT in dwelling 3, while in dwellings 1 and 2 these species induced OPvAA. These results highlight the importance of evaluating the oxidative potential using different assays, since concentration, particle properties and emission sources can affect the sensitivity of the tests.

Table 5

Correlations between OPvDTT and OPvAA values and PM_{10} , BC and PM_{10} -bound chemical constituents. Spearman correlation coefficients (r) computed for the whole monitoring period in each dwelling.

Chemical Species	Dwelling 1		Dwelling 2		Dwelling 3	
	OPvAA	OPvDTT	OPvAA	OPvDTT	OPvAA	OPvDTT
PM_{10}	ns	ns	ns	ns	ns	0.80^a
BC	ns	0.70^a	ns	0.78^a	ns	0.74^a
Aromatic & alkyl-aromatic compounds	ns	ns	ns	ns	0.54^a	ns
Quinone & oxy-aromatic compounds	ns	0.70^a	ns	ns	ns	ns
Na	ns	ns	ns	ns	ns	0.54^a
Mg	0.72^a	ns	ns	ns	ns	0.47^a
Al	0.64^a	ns	ns	ns	0.63^a	ns
Si	0.70^a	ns	ns	ns	ns	0.48^a
P	ns	ns	0.91^a	ns	ns	0.71^a
S	ns	ns	0.68^a	ns	ns	ns
Cl	ns	ns	0.70^a	ns	ns	0.71^a
K	ns	ns	0.77^a	ns	ns	0.89^a
Ca	ns	ns	0.70^a	ns	ns	0.88^a
Ti	ns	ns	ns	ns	0.59^a	ns
Cr	ns	ns	ns	ns	0.50^a	ns
Mn	ns	ns	0.82^a	ns	ns	0.61^a
Fe	ns	ns	ns	ns	0.55^a	ns
Cu	0.65^a	ns	0.84^a	ns	ns	0.74^a
Zn	ns	ns	ns	ns	ns	0.76^a
Br	0.63^a	ns	0.73^a	ns	ns	0.78^a
Sr	ns	ns	ns	ns	ns	0.71^a
Zr	ns	ns	ns	ns	ns	0.61^a
Ba	ns	ns	ns	ns	0.51^a	ns

ns – non-significant spearman correlation.

^a Statistical significance at a level of $p < 0.05$.

It should be noted that some elements, even when present in lower concentrations, may show stronger correlations with OP than other constituents detected at higher levels. For example, taking dwelling 2 as a reference, the mean concentrations of S, K and Ca were 2.2, 1.4 and 1.3 times lower than those found in dwelling 1, but significant correlations for these elements were only observed in that house.

Sr, Fe, Zn and Zr displayed excellent correlations with OP assays in dwelling 3. The occurrence of these elements was associated with the Saharan dust outbreak, highlighting the contribution of this source to redox active transition metals thought to drive antioxidant depletion responses. Mineral elements such as Al, Mg, Si, Fe, and Ca exhibited significant correlations with OP assays, especially during dust events. Similar results were reported previously [98,99], reinforcing that dust periods can contribute to harmful effects on human health.

Aromatic and alkyl-aromatic compounds were correlated with the AA assay values (dwelling 3), while quinone and oxy-aromatic compounds were found to induce the OP by the DTT assay (dwelling 1).

The sum of the concentrations of quinone and oxy-aromatic compounds, as well as the sum of aromatic and alkyl-aromatic constituents were significantly correlated with OPvDTT (dwelling 1) and OPvAA (dwelling 3), respectively, suggesting that these organic compounds are involved in inducing PM oxidative properties. Although PAHs were not significantly correlated with OP responses, some polyaromatics may be oxidised to redox active compounds such as quinones [100]. Caution should be exercised when analysing the OP correlation results, as the causal relationship may be a likely consequence of covariance between species, such as metal(oids) and organic compounds. Moreover, the results can be potentially affected by other PM-bound elements not identified in this study that may induce ROS production. It must be underlined that, in the present study, organic and elemental carbon were not assessed, frequently identified as drivers of OP [4,101].

3.4. Study limitations

This study presents some methodological limitations that may condition or limit the generalisation of the conclusions (1) lack of measurements of CO₂ concentrations as a proxy of air exchange rates; (2) lack of outdoor measurements to estimate particle infiltration and the contribution of external sources to the chemical constituents analysed and, consequently, to the OP; (3) the limited number of samples and houses restricts the application of a source apportionment model; (4) dwellings with different heights in relation to street level, which may introduce some variability in the infiltrated PM component resulting from traffic; (5) due to the difficulties of using pairs of gravimetric samplers, one equipped with quartz filters and the other with Teflon filters, it was not possible to analyse organic and elemental carbon and water-soluble ions, which prevents the calculation of mass balances and the completion of chemical profiles for a better interpretation of both sources and toxicity; (6) measurements were carried out in spring and do not represent seasonal variations that may be observed in indoor air quality. Therefore, future studies should cover a larger number of households in different seasons and simultaneous monitoring indoors and outdoors. In addition, other analyses should be considered to provide more detailed information on PM₁₀-bound chemical constituents.

4. Conclusions

PM concentrations in dwellings are usually represented by measurements in a single microenvironment. In this study, which covered different microenvironments in student homes, statistically significant differences in PM₁₀ and PM₁₀-bound elements were found between monitored rooms, demonstrating that exposure based on measurements in just one space can be uncertain. As the volunteers spent and carried out many activities most of their time in the bedrooms, this microenvironment was identified as a hotspot for exposure to PM₁₀ compounds. The organic compounds found in dwellings were mainly originated from

plastic materials, paint wear and cooking activities. Improving natural ventilation, by opening doors and windows, may increase air change rates but, simultaneously, enhance the infiltration from outdoors. Since PM concentrations in indoor microenvironments can be affected by the penetration from outdoor air, interpretations of the results would have benefited from simultaneous outdoor measurements. Further research on indoor microenvironments should be conducted with different ventilation conditions, whether natural or mechanical, and incorporating outdoor measurements.

Cu, Br and BC were correlated with the oxidative potential at all monitoring sites. A study with a larger number of samples that allows the application of source assignment tools is necessary to confirm more rigorously that, even in indoor microenvironments, exhaust and non-exhaust emissions from traffic contribute to the antioxidant depletion rate and harmful effects on human health. New efforts should be made to elucidate contributions of PM₁₀-bound elements to oxidative potential in different seasons and to better source control and mitigation. Interventions are also needed in residential environments, combining cleaning technologies and improvements in air change rates to reduce the large burden of disease attributable to air pollution exposure.

CRediT authorship contribution statement

Yago Alonso Cipoli: Writing – review & editing, Writing – original draft, Investigation, Formal analysis, Conceptualization. **Estela D. Vicente:** Writing – review & editing, Methodology. **Margarita Evtyugina:** Methodology. **Daniela Figueiredo:** Methodology. **Maria C. Pietrogrande:** Methodology. **Franco Lucarelli:** Methodology. **Manuel Feliciano:** Writing – review & editing, Supervision. **Jiří Ryšavý:** Writing – review & editing, Methodology, Funding acquisition. **Célia Alves:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Funding acquisition, Conceptualization.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.buildenv.2024.112181>.

Data availability

Data will be made available on request.

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