Indoor air quality in a home improvement store: gaseous pollutants, bioburden and particle-bound chemical constituents

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Abstract

Ensuring good air quality is essential for safeguarding human health, requiring monitoring to comprehend air composition and formulate effective interventions. This study focused on indoor and outdoor air quality monitoring within a home improvement and gardening franchise store in northeastern Portugal. Real-time optical monitors recorded levels of particulate matter below 10 µm (PM_{10}), while PM_{10} gravimetric sampling was performed to analyse carbonaceous constituents and metal(loid)s. Continuous monitoring also included CO_2 and comfort parameters. Volatile organic compounds (VOCs), bacteria, and fungi were passively sampled. During labour hours, indoor PM_{10} concentrations exceeded the national protection threshold, reaching 45.4 ± 15.2 µg/m^3, compared to 27.1 ± 9.96 µg/m^3 outdoors. The presence of elements from tyre and brake wear and road dust resuspension suggested that outdoor particles were mainly from non-exhaust traffic emissions. Indoors, the abundance of soil-related elements points to the resuspension of mineral dust as an important source of PM_{10}. Also, anthropogenic constituents associated with activities (e.g., wood sawing) and products sold in the store contributed to indoor particle levels. Dominant indoor VOCs were α-pinene, limonene, and hexanal. The most abundant elements in PM_{10} indoors were Ca, Fe and Zn, with values of 658 ± 297, 273 ± 141, 172 ± 67.4 ng/m^3, respectively. Common fungi included *Trichoderma* sp. and *Penicillium* sp., while colony-forming bacterial units were most prevalent in the gardening and heating sections. This comprehensive study highlights the need to implement indoor air quality monitoring strategies in commercial spaces, particularly with regard to particulate matter and associated pollutants.

Keywords: Retail store, IAQ, PM_{10}, VOCs, bioburden

1 INTRODUCTION

As its name suggests, indoor air quality (IAQ) refers to the quality of air inside buildings and structures [1]. To better define the term, Fanger [2] argued that IAQ is characterised by the possible adverse effects on humans, whether on health, well-being, productivity or learning. These effects are caused by a series of air pollutants, making the monitoring of their levels essential for identifying sources and creating action plans to ensure good air quality. The most common indoor air quality determinants are carbon dioxide (CO_2), carbon monoxide (CO), particulate matter lower than 10 and 2.5 µm (PM_{10} and $PM_{2.5}$), volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs), nitrogen dioxide (NO_2), radon, microorganisms and pollen grains [3]. The World Health Organisation (WHO) has developed guidelines for these pollutants, based on scientific evidence on their health effects [4]. There is no specific reference directive on IAQ in European legislation, although some countries have started to adopt specific legislation. Portugal, in particular, promulgated the Decree-Law No. 101-D/2020, in which it is stated that commercial, service and other type of buildings must comply with air quality and ventilation standards in order to safeguard public health and enhance overall quality of life. Among the pollutants subject to guidelines by the WHO or limit values by national legislation, PM is considered the most worrying given that it was classified as carcinogenic to humans (IARC Group 1). Scientific evidence has shown that PM is associated with respiratory diseases, including asthma, reduced lung function, bronchitis, and cancer [5–7]. Exposure to PM has also been linked to Alzheimer’s disease [8], preterm birth and low birth weight [9,10], myocardial infarction [11], and other cardiovascular diseases [12]. Additionally, knowing that PM is a complex mixture of solid and liquid particles, its toxicity can vary greatly due to its composition, and therefore, a detailed characterisation is essential to determine possible health effects [13].

When exploring IAQ, it is important to note that urban populations spend up to 90% of their time indoors [14]. Urban centres have a wide variety of stores that are central to people's daily lives, whether for work, leisure or shopping. Therefore, optimal indoor environmental conditions are essential to ensure a good working environment and a more attractive and healthier place for customers. Retail stores are among the numerous indoor environments that are part of everyday life in urban centres. These refer to stores that sell products and provide services to the final consumer, covering a wide variety of establishments, including supermarkets, bars, clothing stores, workshops, and many others, which have diverse indoor characteristics, with different sources of air pollutants.

Many studies have been focused on stores where smoke is present, such as bars, restaurants, and waterpipe cafes, in which PM mainly originates from smoking and cooking activities [15,16]. However, the IAQ of several other types of highly frequented stores was not or was very little characterised. A review by Zaatari et al. [17] on IAQ in retail stores pointed out seven pollutants that generally exceed the regulated or recommended limit values: PM_{10}, $PM_{2.5}$, acrolein, formaldehyde, acetaldehyde, trichloroethylene, and benzene. The sources of these pollutants range from building materials and furnishings to cleaning products and electronic equipment. Moreover, human activities such walking, vacuuming, sweeping, and smoking contribute to the indoor pollutant loads. The ventilation system's effectiveness in retail spaces can greatly influence the concentration of these air pollutants. Proper ventilation, alongside regular monitoring of air quality, represents a crucial step towards a better indoor environment.

The objective of this work was to carry out a detailed characterisation of the indoor and outdoor air quality of a type of increasingly frequented commercial establishment that until now has not been the target of this type of study: a home improvement and gardening store. In addition to traditional pollutants covered in legislation and other IAQ studies, the monitoring campaign included the speciation of VOCs, the detection of bacteria and fungi in different culture media and a detailed analysis of PM_{10}-bound chemical constituents. This study is part of a larger initiative that aims to analyse the air quality of different types of large globalised commercial establishments. Despite their representativeness, these indoor spaces have not been subject to detailed investigation, as demonstrated by a search of the Scopus and Web of Science databases. Monitoring and apportioning sources of indoor air pollution are crucial for more effective management and taking measures to promote the health and well-being of both workers and customers of these establishments.

2 METHODOLOGY

2.1 Sampling site

This work was conducted in a home improvement store from December 15th, 2021, to January 4th, 2022, involving indoor and outdoor measurements. Given that the measurements were carried out in winter, a period in which ventilation conditions are more hermetic and there is an intensification of outdoor emissions due to residential biomass combustion, this study must be seen as a “worst case scenario”. The store is located in the northern region of Portugal (Lat. 41.8072, Long. -6.75919), in an urban industrial area composed of various commercial activities, including building materials stores, car dealers, supermarkets, furniture shops, electrical and lighting stores, and auto repair centres. The main area of activity of the store is the home improvement sector. It is part of a franchise present throughout the national territory and several countries worldwide. Home improvement stores, also known as hardware stores or DIY (do-it-yourself) stores, sell products for repair, maintenance, home improvement, construction, and gardening. Their products include paints and adhesives, hand and electrical tools, heating and ventilation systems, gardening tools, bathroom furniture, electrical accessories, wooden boards, plumbing parts and fittings, and decorative items. The opening hours were from Monday to Saturday, 9:00 AM to 8:00 PM, and Sunday, 9:00 AM to 5:00 PM. Figure 1 provides an overview of the hardware store and the sampling locations.

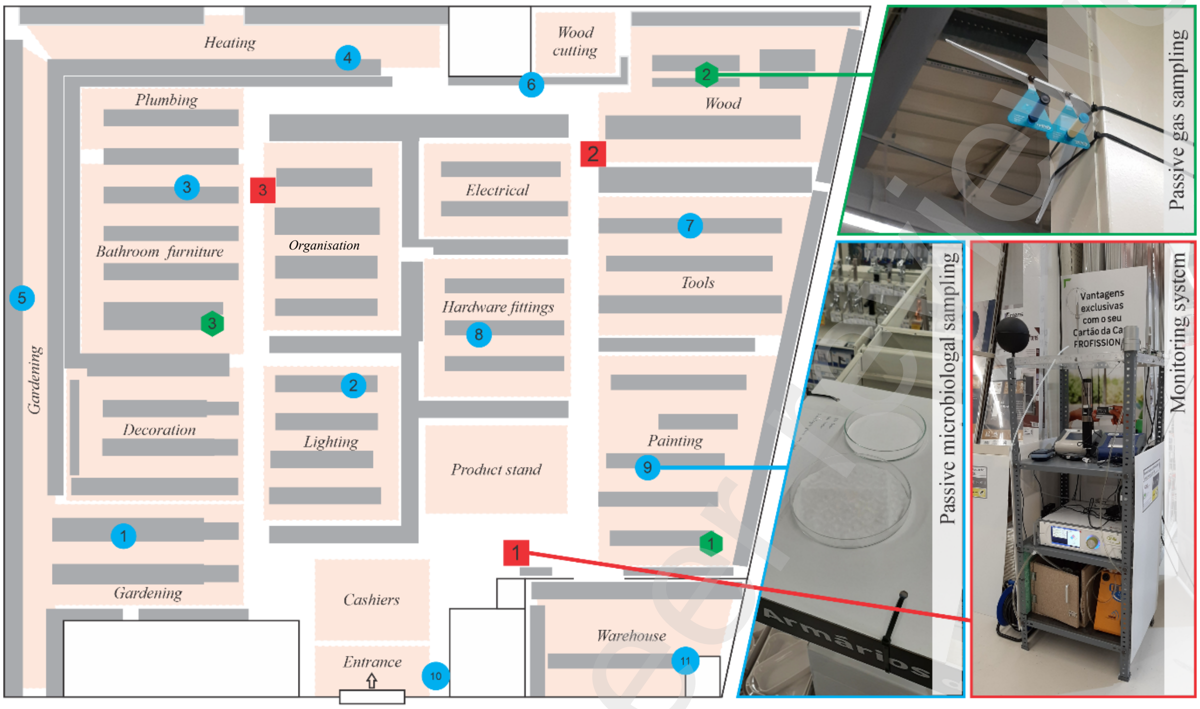


Figure 1 – Home improvement store overview. The monitoring locations are shown as follows: in red, continuous monitoring of comfort parameters, gaseous compounds and particulate matter, and PM_{10} sampling on filters; in blue, passive sampling of microorganisms and settleable dust; in green, passive sampling of volatile organic compounds.

2.2 Sampling and instrumentation

Sampling followed the principles defined in the ISO 16000-1:2004 norm (Indoor air - Part 1: General aspects of sampling strategy), using two systems, one indoors and one outdoors. The indoor system was composed of a low volume sampler (FAI Instruments, SILENT Sequential Air Sampler) for collection of PM_{10} on 47 mm quartz fibre filters, an optical particulate matter monitor (OPS 3330 from TSI), an air quality probe (GrayWolf, WolfSense IQ-610) for CO_2, a multi-gas analyser (Gasera, Gasera ONE) for measuring CH_4, CO_2, NH_3, an air temperature and relative humidity monitoring and a thermal comfort and microclimate station (DeltaOhm, HD32. 3). The outdoor system included a low volume sampler (Tecora, Echo PM) for PM_{10} sampling and an optical particulate matter monitor (DustTrak DRX 8533 from TSI). Data acquisition was performed every minute. A total of 17 filter pairs were sampled, 14 pairs during opening hours (daytime) and 3 filter pairs during closing hours (nighttime), collected simultaneously indoors and outdoors. In addition, data from nitrogen oxide (HORIBA, APNA-370) and ozone

(HORIBA, APOA-370) gas analysers and a weather station near the store were used to characterise the outdoor atmosphere. The indoor system was positioned in three locations to obtain a more significant representativeness of the home improvement store, while the outdoor system remained fixed during the entire sampling campaign. As seen in Figure 1, the first location was between the cashiers, the exhibition area, and the painting section. The second location was near the laminate flooring shelf in the wood section. Lastly, the monitoring system was positioned between the bathroom furniture and organisation sections. It should be noted that all the sampling points were close to the circulation areas, which, as agreed with the store, would have the least impact on its normal operation.

Furthermore, diffusion tubes (Radiello) for passive sampling of VOCs and carbonyls were installed in three indoor and one outdoor location. Indoors, the diffusion tubes were installed in the paint and adhesives, wood, and bathroom furniture sections for seven days. Outdoors, the diffusive tubes were placed in front of the store, near an urban road for fourteen days. Eleven electrostatic dust collectors (EDC) - polypropylene electrostatic cloths with a diameter of 140 mm – and 110 mm quartz fibre filters dust collectors (QFDC), were also placed indoors for microbiological sampling and the determination of settleable dust [18,19]. Each pair (EDC and QFDC) was distributed across the following locations: gardening, lighting, bathroom furniture, heating and ventilation, fencing, wood cutting, hardware fittings section, painting, entrance, and warehouse. Settleable dust was collected for approximately 50 days.

2.3 Analytical procedures

The quartz fibre filters of the PM_{10} samplers, as well as those used for collection of settleable dust, were weighed using a microbalance (RADWAG, MYA 5/2Y/F) with an accuracy of 1 µg. The mass of the filters was obtained from the average of six weightings performed after a stabilisation period of two days (20 ºC, 50% relative humidity). PM_{10} concentrations obtained by the gravimetric method were used to correct the DustTrak data (Figures S1 and S2 of the supplementary material).

The determination of the carbonaceous fractions in the PM_{10} samples was made by thermo-optical analysis of small portions of the filters. In this method, organic carbon (OC) is determined by heating filter punches in an inert atmosphere of 100% nitrogen, to vaporise the organic fraction, followed by the oxidation in an atmosphere composed of 4% oxygen and 96% nitrogen for the determination of elemental carbon (EC). The CO_2 released during these phases is continuously monitored by a FTIR gas analyser allowing the determination of the carbon concentration. Detailed information on the method and instrumentation can be found elsewhere [20,21]. However, in this study, due to the small mass of particulate material accumulated on the filters, it was difficult to separate OC and EC, so only total carbon (TC) is reported.

In addition, the quartz fibre filters were analysed by X-ray fluorescence (XRF) to determine the concentration of S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Br, Pb and Sr, using an ARL Quant'X EDXRF spectrometer (Thermo Scientific Inc). The complete procedure is available in Chiari et al. [22]. The analyses of diffusive samplers were carried out at Fondazione Salvatore Maugeri (Padova, Italy). Briefly, carbonyls were determined by desorption with acetonitrile and analysis by high performance liquid chromatography (HPLC). VOCs were quantified by thermal desorption with subsequent analysis by gas chromatography – mass spectrometry (GC/MS).

For microbiological analyses, EDCs were extracted with a NaCl saline solution. Later, each sample was inoculated in four different culture media: malt extract agar (MEA) and dichloran-glycerol agar (DG18) for the determination of fungi, and tryptic soy agar (TSA) and violet red bile agar (VRBA) for bacteria. After incubation for one week, the fungal and bacterial density was calculated. Detailed information can be found in Viegas et al. [23,24].

2.4 Data analyses

Data from continuous instruments were used to create temporal profiles of the air pollutants and weather variables. On December 25th, 2021, and January 1st, 2022, the store was closed, thus these days were not used in the calculation of the temporal profile, as they did not correspond to the normal operation. The weekly profile of pollutants and meteorological variables was created by grouping the data from the same day of the week, same hour of the day and averaging the values. In addition, the decomposition of the time series for indoor CO_2 and PM_{10} was also made using the additive model to check the trend, cycle, and randomness of the series.

To analyse the normality of the data, the Kolmogorov-Smirnov normality test (K-S test) was used due to the large volume of data. For all variables the p-value was below the 5% confidence level, indicating that all variables did not have a normal distribution. Following the normality test, Spearman's correlation was applied to determine the level of correlation between the variables. Spearman's correlation is considered a nonparametric test. It has shown better results when applied to large samples when compared to Pearson, as highlighted by Bishara et al. [25]. The Spearman's correlation analysis was performed applying the two-sided test with a significance level of 0.05, 0.01 and 0.001.

Considering the number of samples, which limited the use of robust techniques for identifying the sources of the elements, such as factor analysis with varimax rotation [26] and positive matrix factorisation [27], elemental composition data were analysed by means of I/O ratios, enrichment factors (EF) and correlations between the elements. The elemental concentrations were used to calculate the enrichment factors of each element in relation to its concentration in the earth's crust [28]:

$$EF = \frac{(X/Fe)_{air}}{(X/Fe)_{crust}} \quad (1)$$

In this work, Fe was used as reference to calculate the enrichment factors for all elements except Fe, for which Ca was used. Thus, in Equation (1), $(X/Fe)_{air}$ corresponds to the ratio of element X to the concentration of Fe in the same filter, whereas $(X/Fe)_{crust}$ corresponds to the ratio of the element X to the concentration of Fe in the crust. The reference concentration of crustal elements was obtained from Wedepohl [29]. The different pollution classes proposed by Sutherland [30] were adopted: EF < 2 Depletion to minimal enrichment, suggestive of no or minimal pollution; EF 2 – 5 Moderate enrichment, suggestive of moderate pollution; ER 5 – 20 Significant enrichment, suggestive of a significant pollution signal; EF 20 – 40 Very highly enriched, indicating a very strong pollution signal; EF > 40 Extremely enriched, indicating an extreme pollution signal. Furthermore, for ER>10, a more significant contribution from anthropogenic sources is assumed, as observed in the study of Chiarenzelli et al. [31]. To determine the ventilation rate of the building, the CO_2 decay method was used [32]:

$$AER = \frac{\ln((C_1 - C_R)/(C_0 - C_R))}{t} \quad (2)$$

where AER is the air exchange rate in 1/h; C_0-C_R is the difference between the concentration at time 0 and the reference concentration, C_1-C_R is the difference between the concentration at time 1 and the reference concentration, and t is the measurement time in hours between time 0 and 1. To convert the ventilation values to L/s.m^2, equation 3 (modified from [33]) was applied for comparability with ventilation values.

$$Ventilation\ rate\ (\frac{L}{s\ .\ m^2}) = \frac{\mathrm{AER}\ .\ height\ (m)}{3.6} \quad (3)$$

During the sampling period, a Saharan dust intrusion was registered. This phenomenon was also detected in other study conducted in the region [34]. To confirm this source, backward trajectories were calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory Model (HYSPLIT) from NOAA [35]. The days with the highest residuals of the time series analysis of PM were chosen to run the model, with 5-day back trajectories at 100.

3 RESULTS AND DISCUSSION

3.1 Particulate matter temporal profiles and correlations

The indoor PM_{10} concentrations were much higher than those measured outdoors, averaging 45.4 ± 15.2 μg/m^3 during the opening hours (diurnal period) and 27.1 ± 9.96 μg/m^3 outside the building. The higher indoor concentrations of PM_{10} are probably the result of the building's airtightness to improve thermal efficiency and reduce space heating costs, as well as human-induced resuspension. It was also found that the dust on the shelves was cleaned very infrequently. Therefore, the handling of products by employees and customers may also have contributed to the increase in PM_{10} levels indoors. When the store was closed (night period), the indoor and outdoor averages were 39.5 ± 8.43 and 21.3 ± 1.49 μg/m^3, respectively. In the absence of activities, during the night, coarser particles settle down quickly, contributing to reducing the difference between indoor and outdoor concentrations. However, even at night, high indoor concentrations of PM_{10} indoors were observed. This is probably indicative that a significant mass fraction of PM_{10} is made up of fine particles that take longer to sediment by gravity. On the other hand, at night in winter, there is an intensification of particulate emissions from residential biomass combustion, which, due to their essentially ultrafine nature, have a great capacity for infiltration into indoor environments [36].

The indoor concentrations were well above those recommended by the World Health Organisation [37], which specifies a 24-h average of 45 μg/m^3, and also above the protection threshold of 50 μg/m^3 for PM_{10} stipulated by the Portuguese legislation on indoor air quality [38]. At the first sampling point, near the cashiers, in the wood section, and in the bathroom furniture and organisation sections, the diurnal PM_{10} concentrations were 41.5 ± 7.27 μg/m^3, 46.9 ± 18.6 μg/m^3 and 49.2 ± 17.2 μg/m^3, respectively. In the supplementary material, Fig. S3 depicts weekly profiles of pollutant concentrations, temperature, and relative humidity in the indoor environment. PM_{10} registered two concentration peaks, one between 10:00 AM and 12:00 PM and another between 4:00 PM and 7:00 PM, likely related to resuspension caused by the movement of occupants. During lunchtime, from 12:00 PM to 2:00 PM, the PM_{10} concentrations tend to decrease due to the lower number of customers and fewer work activities. Using time series decomposition (Figure 2), in the cycle component, it is possible to see these two peaks with an amplitude of 70 μg/m^3.

The residuals, which correspond to the values remaining after removing the trend and cycle from the time series, provide useful information about the occurrence of singular events contributing to the decrease and increase of PM_{10} concentrations.

The weekly profile for outdoor pollutants and the meteorological variables for the monitoring campaign can be found in Fig. S4 of the supplementary material. Outdoors, on Monday of the weekly profile, a PM_{10} peak of 250 µg/m^3 was recorded. This peak may be related to a Saharan dust outbreak, as demonstrated by the Hysplit model (Fig. S5 of the supplementary material). Although the Hysplit indicates air masses coming from North Africa, only the filter from January 3rd showed signs of this origin, due to the high concentrations of Fe, Ca and Ti. It should be emphasised that there are no industries with high emission rates of particulate matter in the industrial area. The store's proximity to the street is an indication that PM_{10} concentrations are related to vehicle emissions and resuspension. The outdoor PM_{10} profiles reveal two daily maxima, one between 8:00 AM and 10:00 AM, and another from 5:00 PM to 7:00 PM, which match the NOx peaks, coinciding with periods of higher traffic intensity. Higher PM_{10} concentrations during the night on Thursday and Friday of the weekly profile are attributed to residential biomass burning, a common practice in the city.

Due to the central heating system, temperature and relative humidity indoors (Fig. S3 of the supplementary material) presented a smaller amplitude than outdoors, with mean values of 18.3 ºC and 41.7%, respectively. The standards EN 16798-1 [39] and ISO 17772-1 [40] specify, for department stores, a temperature range for existing buildings between 15 and 23 ºC in the winter season and from 22 to 26 ºC in the summer season. For offices and spaces with predominantly sedentary activities, a temperature between 19 and 25 ºC and from 22 to 27 ºC is recommended in the winter and summer season, respectively. In relation to relative humidity, it is recommended to maintain the values between 20 and 70%. As the study was conducted in winter, the values of temperature and relative humidity indoors are within the recommended ranges for this season.

CO_2 concentrations (Fig. S3 of the supplementary material), followed the occupancy patterns, increasing with the opening of the shop, slightly decreasing during lunchtime, rising again after 2:00 PM and declining after 6:00 PM. The Portuguese Law [41], aiming to improve energy performance of buildings, applies a lower limit, stipulating a protection threshold of 1250 ppm. The highest CO_2 values were close to 850 ppm, and the maximum 8-h average was 680 ppm, well below the national threshold. Outdoor NO_X concentrations followed the traffic pattern. The maximum NO_X level registered was 61.5 ppb. Although outdoor O_3 had a weak correlation with solar radiation (r^2=0.13, p<=0.01), its concentration followed the intensity of sunlight, raising at 09:00 AM and reaching its maximum between 1:00 PM and 4:00 PM. On Sunday, although the NO_X concentration remained very low and presented a negative moderate correlation with O_3 (r^2=-0.57, p<=0.001), the ozone profile was similar to that of the other days of the week, indicating its formation in the region is not only influenced by NO_X emissions. Portugal has stipulated a maximum daily 8 hour mean for O_3 of 120 µg/m^3 (60 ppb) [42]. During the monitoring period, the ozone concentrations remained below this threshold with the maximum concentration being 42.8 ppb.

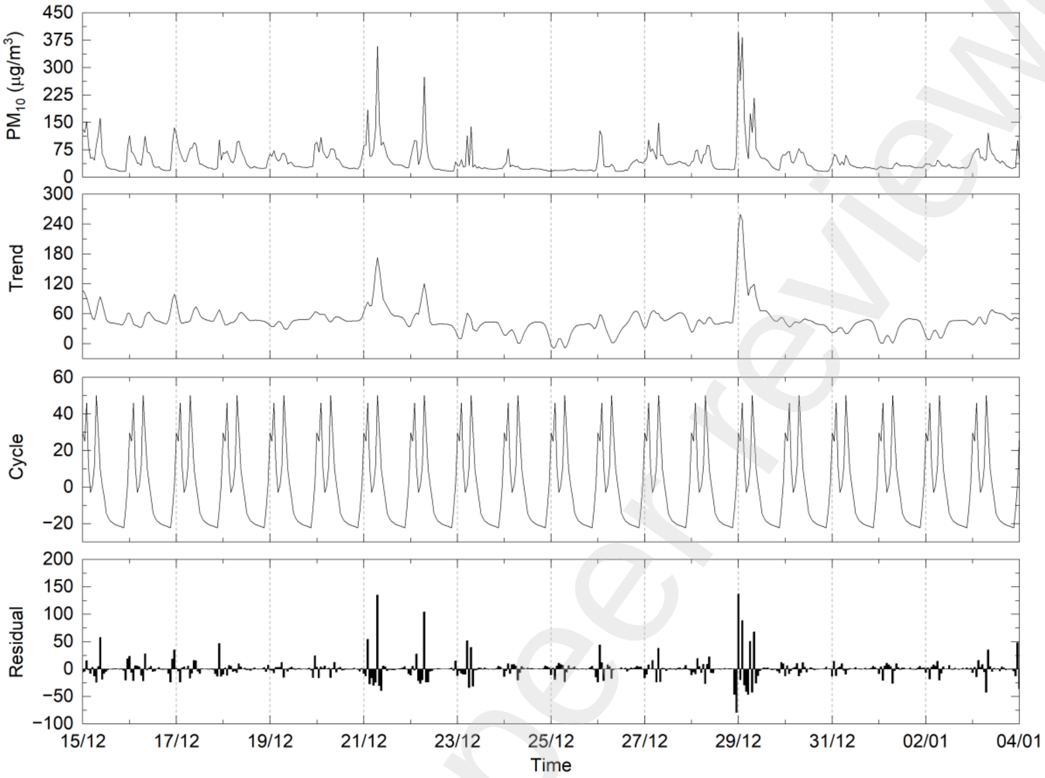
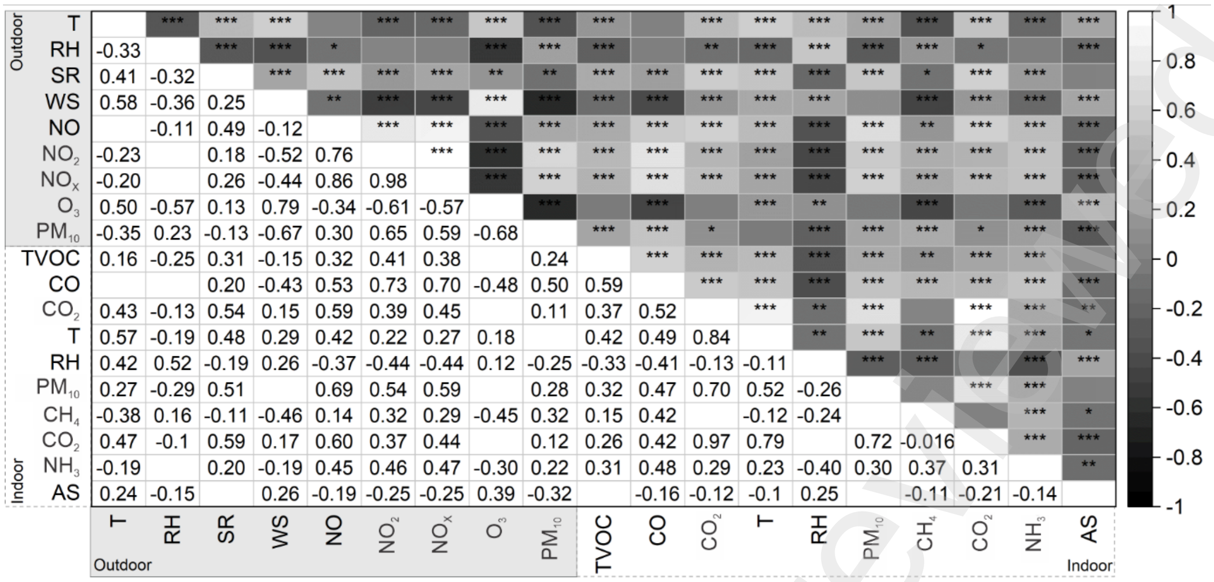


Figure 2 – Time series decomposition for indoor PM$_{10}$. The first graph corresponds to the hourly data between December 15th, 2021, and January 4th, 2022. The second graph corresponds to the time series trend. The third graph indicates the cycle component, showing the estimated variation of PM$_{10}$ throughout the day. The fourth graph indicates the residuals, the values subtracting the trend and cycle from the time series.

Air change rates were estimated to be between 0.54 AER/h and 0.96 AER/h. Considering the average height of the store building as 5 m, the ventilation rate ranged from 0.75 L/s.m^2 to 1.33 L/s.m^2. The ASHRAE standard 62.1, Ventilation and Acceptable Indoor Air Quality [43], sets a minimum ventilation rate for sales buildings of 0.6 L/s.m^2. This category is the closest to the store studied, which is characterised by a moderate level of occupant activity, and by having products with a potentially high impact on IAQ.



		Outdoor									Indoor									
		T	RH	SR	WS	NO	NO_2	NO_x	O_3	PM_{10}	TVOC	CO	CO_2	T	RH	PM_{10}	CH_4	CO_2	NH_3	AS
Outdoor	T		***	***	***		***	***	***	***	***		***	***	***	***	***	***	***	***
	RH	-0.33		***	***	*			***	***	***		**	***	***	***	***	*		***
	SR	0.41	-0.32		***	***	***	***	**	**	***	***	***	***	***	***	*	***	***	
	WS	0.58	-0.36	0.25		**	***	***	***	***	***	***	***	***	***		***	***	***	***
	NO		-0.11	0.49	-0.12		***	***	***	***	***	***	***	***	***	***	**	***	***	***
	NO_2	-0.23		0.18	-0.52	0.76		***	***	***	***	***	***	***	***	***	***	***	***	***
	NO_x	-0.20		0.26	-0.44	0.86	0.98		***	***	***	***	***	***	***	***	***	***	***	***
	O_3	0.50	-0.57	0.13	0.79	-0.34	-0.61	-0.57		***		***		***	**		***		***	***
	PM_{10}	-0.35	0.23	-0.13	-0.67	0.30	0.65	0.59	-0.68		***	***	*		***	***	***	*	***	***
Indoor	TVOC	0.16	-0.25	0.31	-0.15	0.32	0.41	0.38		0.24		***	***	***	***	***	**	***	***	
	CO			0.20	-0.43	0.53	0.73	0.70	-0.48	0.50	0.59		***	***	***	***	***	***	***	***
	CO_2	0.43	-0.13	0.54	0.15	0.59	0.39	0.45		0.11	0.37	0.52		***	**	***		***	***	**
	T	0.57	-0.19	0.48	0.29	0.42	0.22	0.27	0.18		0.42	0.49	0.84		**	***	**	***	***	*
	RH	0.42	0.52	-0.19	0.26	-0.37	-0.44	-0.44	0.12	-0.25	-0.33	-0.41	-0.13	-0.11		***	***		***	***
	PM_{10}	0.27	-0.29	0.51		0.69	0.54	0.59		0.28	0.32	0.47	0.70	0.52	-0.26			***	***	
	CH_4	-0.38	0.16	-0.11	-0.46	0.14	0.32	0.29	-0.45	0.32	0.15	0.42		-0.12	-0.24				***	*
	CO_2	0.47	-0.1	0.59	0.17	0.60	0.37	0.44		0.12	0.26	0.42	0.97	0.79		0.72	-0.016		***	***
	NH_3	-0.19		0.20	-0.19	0.45	0.46	0.47	-0.30	0.22	0.31	0.48	0.29	0.23	-0.40	0.30	0.37	0.31		**
	AS	0.24	-0.15		0.26	-0.19	-0.25	-0.25	0.39	-0.32		-0.16	-0.12	-0.1	0.25		-0.11	-0.21	-0.14	

* p<=0.05 ** p<=0.01 *** p<=0.001

Figure 3 - Spearman correlation matrix for indoor and outdoor variables. The correlations with no significant level were excluded. T- temperature, RH – relative humidity, SR – solar radiation, WS – wind speed, AS – air speed.

PM_{10} and CO_2 indoors showed a strong correlation (r^2=0.72 $p \leq 0.001$, Figure 3), indicating that human activities significantly influence the concentrations of particulate matter, since the main source of CO_2 in the store was human respiration. In retail stores, the variety of products and services provided may have a great impact on PM levels. Furthermore, indoor PM_{10} showed a moderate correlation with outdoor NO_X (r^2=0.59, $p \leq 0.001$), indicating the infiltration of urban traffic emissions. It is also observed that solar radiation has a moderate positive correlation with indoor PM_{10} (0.51, $p \leq 0.001$), but a weak negative correlation with outdoor PM_{10} levels (r^2=-0.13, 59 p<=0.01). CH_4, N_2O and NH_3 in the store showed little to no variation during the monitoring period with averages of 2.58 ± 0.33, 0.46 ± 0.04 and 0.68 ± 0.26 ppm, respectively.

3.2 Carbonaceous content of PM_{10}

Indoors, the total carbon (TC) concentration for the diurnal period was 14.1 ± 6.53 μg/m^3, while for the night period it was 5.13 ± 0.82 μg/m^3. On average, TC accounted for 31% and 13% of the PM_{10} mass for the diurnal and nocturnal period, respectively. Using Kruskal-Wallis ANOVA at 5% significance level, followed by Dunn's test, to compare indoor day with outdoor day, and indoor night with outdoor night, the only significant difference was observed between indoor day and outdoor day with a p=0.03. Thus, it is possible to infer that indoor sources contribute to the increase of TC concentrations during the opening period.

The paint and wood sections showed the most remarkable differences between indoor and outdoor concentrations (Figure 4). As explained earlier, these areas have greater movement of workers and customers, so there may be a more significant contribution of dust resuspension to carbon levels. Product handling also contributes to semivolatile compounds that are part of OC. Furthermore, considering that the study was carried out in winter, when people tend to wear more garments, the contribution of clothing fibres to carbon levels cannot be ruled out [44].

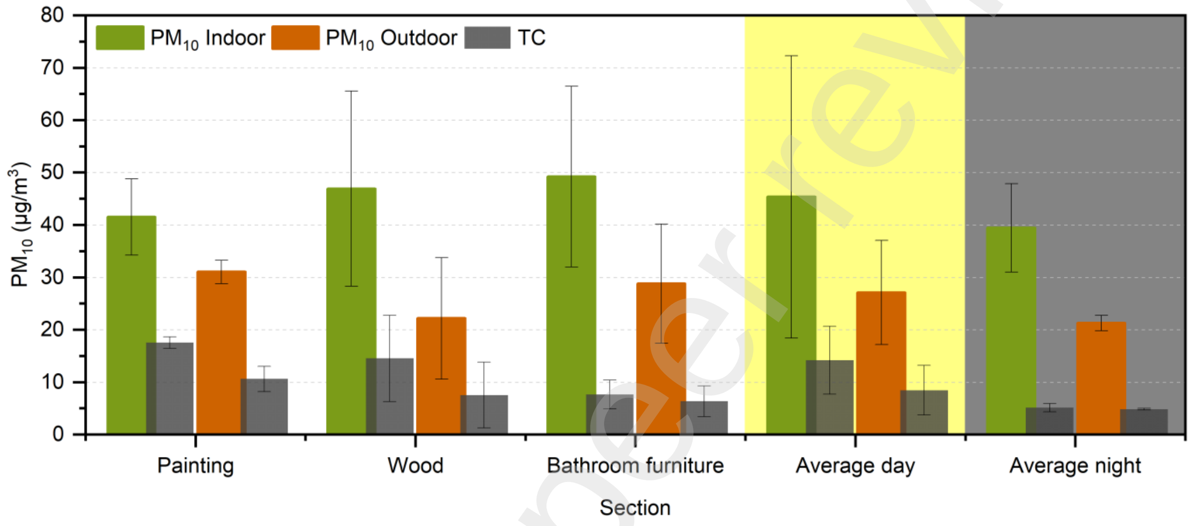


Figure 4 – Carbonaceous content of indoor and outdoor particles. The indoor results for each sampling area are presented along with the corresponding outdoor concentration for that period, as the outdoor system were positioned at the same spot. Additionally, the averages of all the samples taken during the day and night, both indoors and outdoors, are also provided.

3.3 PM_{10}-bound elements

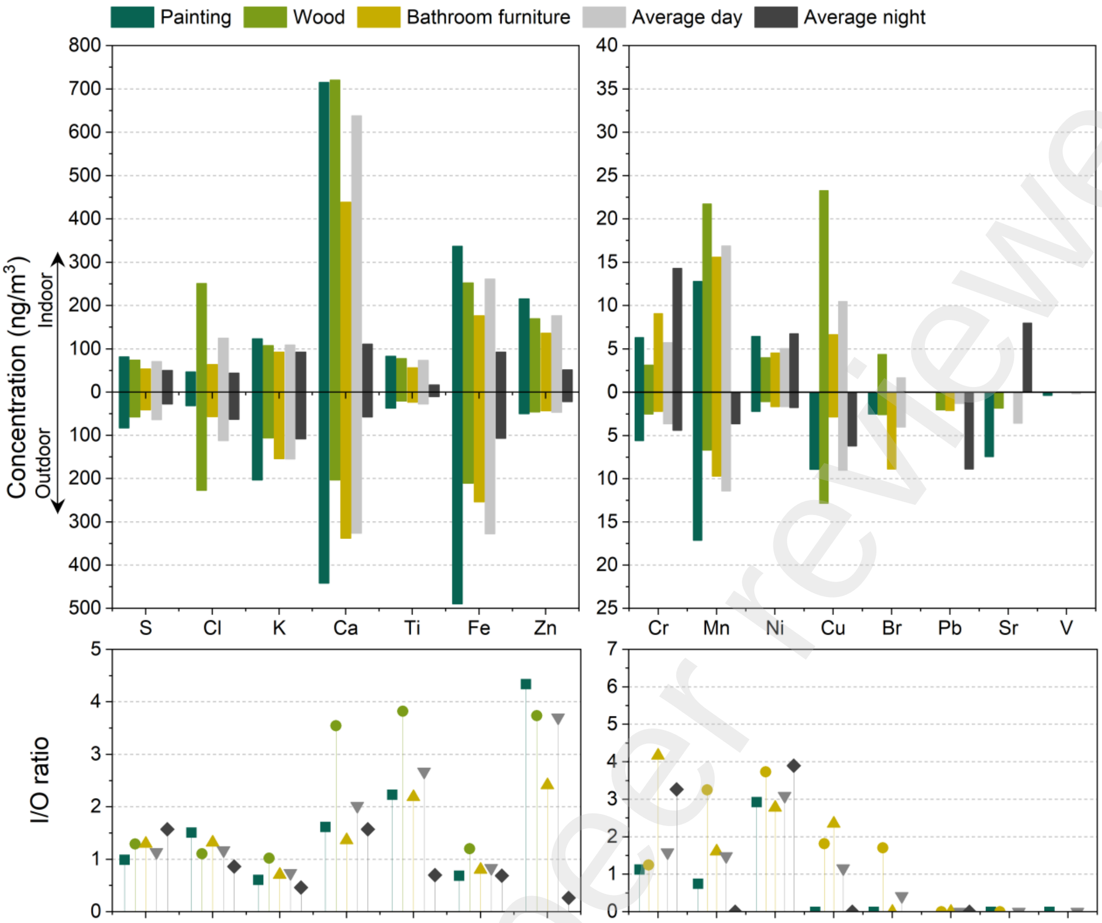


Figure 5 – Top: Concentrations of PM_{10}-bound elements in different store sections and mean value for the day and night periods. Bottom: Indoor / Outdoor ratio.

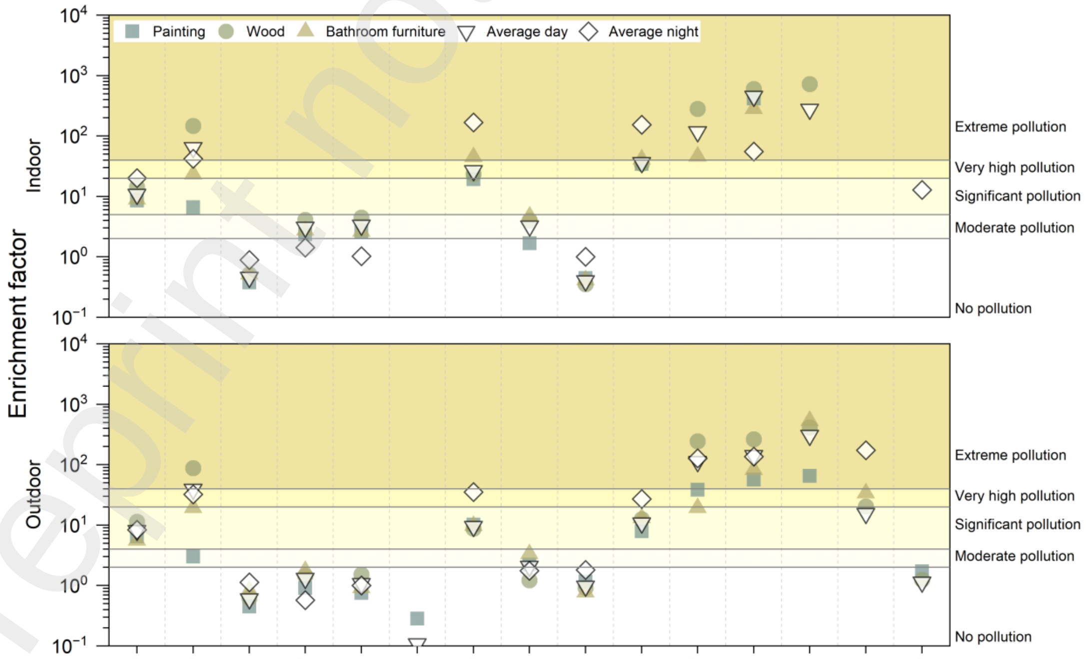


Figure 6 – Element enrichment factors for different store sections and mean values for the day and night periods.

In general, indoor elemental concentrations were higher than the outdoor levels, even for elements commonly related to vehicular emissions and mineral dust. The most predominant elements were Ca, Fe, Zn, K, Cl, S and Ti with average indoor concentrations for the opening period of 658 ± 297, 273 ±.140, 172 ± 67.4, 114 ± 79.6, 131 ± 172, 72.3 ± 32.8, and 73.5 ± 21.9 ng/m^3, respectively. Cl was the element with greatest amplitude between the sections within the store. However, its diurnal I/O ratio was close to 1, showing a high correlation coefficient with outdoor Cl, a strong indication of external sources. Additionally, the EFs placed the average daytime concentrations of Cl at the extremely polluted range, suggesting an origin in anthropogenic activities. One of the possible causes is biomass burning, as stated in the study of Pio et al. [27], and strengthened by the work of Cipoli et al. [34], which pointed out wood combustion for residential heating as a common practice in the region. Also, the high concentrations of K, a tracer of biomass burning, reinforces this evidence [45].

The major elements Ca, Ti and Zn presented the highest I/O ratios, especially in the wood sector. Indoor Ca highly correlated with indoor concentrations of S, Ti, Fe and Zn. Ca and Fe are major components of the region's soil [46]. Thus, their relationships suggest that resuspension of soil dust is a contributor to these PM_{10}-bound elements. The higher concentrations of some elements in the sawmill section may be related to the composition of the wood itself. Some studies have shown that pellets and wood from the genus *Pinus* are enriched in Ca and K, and also contain Ti, although at lower levels [47,48].

Mean daytime I/O ratios of 2.01, 2.67 and 3.70 were obtained for Ca, Ti and Zn, respectively, but at nighttime, the ratios decreased considerably to 1.57, 0.70 and 0.26, indicating that these constituents originate mostly from indoor activities during the occupancy period. Both the indoor and outdoor Ti EFs were below 10, suggesting a crustal origin often associated with resuspension [49]. Zn showed the highest I/O ratio and indoor and outdoor EFs well above 10, pointing to anthropogenic sources. Besides infiltration of vehicular emissions, the contribution of a diverse range of galvanized steel products, including profiles, squares, wires, screens, plates, and ventilation tubes that use Zn as a protective coat, cannot be ruled out [50]. Zinc is also an important constituent of rubbers and paints. In addition, in the form of zinc chloride, it is an ingredient of deodorants. Therefore, the use of these personal care products may explain the higher concentrations of both Cl and Zn inside the store. The minor elements Cr, Mn, Ni, Cu and Br showed high I/O ratios. Regarding indoor Cr, a significant pollution level was observed with EFs above 10, and an I/O ratio of 3.8 in the bathroom furniture section. The literature shows that stainless steel products are a source of Cr and Ni [51]. Several stainless-steel items can be found in the bathroom section, such as shower hoses, shower heads, faucets, support bars and towel racks. It is very likely that these items are the sources of these elements.

Among the elements analysed, the one with the greatest potential to harm human health is Cr. Its hexavalent form Cr(VI) is classified as category 1 (carcinogenic to humans) by the International Agency for Research on Cancer (IARC). However, in this study, total Cr concentrations are reported. Sheehan et al. [52] found that Cr(VI) represented 21% of total airborne Cr in indoors samples. Considering that percentage, the highest Cr(IV) concentrations would be in the bathroom furniture section (0.006 μg/m^3). In its Integrated Risk Information System (IRIS), the U.S. Environmental Protection Agency stipulates a reference concentration (R_fC) for chromium (VI) in the particulate matter of 0.1 μg/m^3.

Most metal(oids) are typically found in the air in the form of oxides [53]. Monsé et al. [54] showed that the "No Effect Exposure Level" (NOEL) for ZnO should be between 500 and 1000 μg/m^3. Hadrup et al [55] reviewing scientific studies for setting a health-based occupational exposure limit for ZnO, suggested an occupational exposure limit of 40 μg/m^3. In this study, after converting Zn to ZnO, the highest concentrations were found in the paint section (0.32 μg/m^3) and in the wood section (0.29 μg/m^3), values far below the exposure limits. The highest copper concentration was registered in the wood section, with a value of 0.04 μg/m^3. The Scientific Committee on Occupational Exposure Limits (SCOEL), in its publication "Recommendation from the Scientific Committee on Occupational Exposure Limits for Copper and its inorganic compounds" [56], suggested a time weighted average (TWA) of 10 μg/m^3 for Cu in any 8-hour work shift for respirable dust. The same committee recommended a TWA for Mn of 200 μg/m^3 for the inhalable fraction and 50 μg/m^3 for the respirable fraction [57]. In this study, the highest concentrations of Mn were 0.04 μg/m^3 in the wood section and 0.03 μg/m^3 and bathroom furniture sections. For Ni, the SCOEL [58] recommend an 8-hour TWA of 5 μg/m^3. In this study, the highest concentration was 0.009 μg/m^3. Thus, metal(loid) concentrations found in this study are far below the occupational exposure limits set by international organisations.

Br, Pb, Sr and V were mainly detected outdoors. High EFs, above 100, were observed for outdoor concentrations of Cu, Zn, Br and Pb, including these elements at the extreme pollution class, and indicating anthropogenic sources. Outdoors, Cu showed a high correlation with Mn and Zn, suggesting the contribution of non-exhaust vehicle emissions, such as brake and tyre wear [59]. Additionally, Cu, Pb and Zn are road dust marker elements [60].

3.4 Volatile organic compounds

A total of 21 volatile organic compounds were analysed (Table 1). As observed in other retail stores [17], the most prevalent VOCs in the indoor air were α-pinene, ethylbenzene, formaldehyde, hexanal, limonene, m,p-xylene, o-xylene, and toluene. All VOCs presented I/O ratios higher than 1, indicating the contribution of indoor sources to their concentrations.

α-Pinene and limonene are non-carcinogenic monoterpenes mainly released by plants. Their presence in indoor environments is generally related to wooden emissions and the use of air fresheners [61]. Most of the woods offered by the hardware store are from angiosperm trees of the genera *Abies, Betula, Fagus, Pinus* and *Quercus*. Risholm-Sundman et al. [62], in a study of VOC emissions from different types of wood, found that most hardwoods emit hexanal and a small amount of pentanal. Additionally, the study showed that α-pinene is predominantly emitted by conifers. The higher concentrations of hexanal and formaldehyde in the wood section, may also be associated with wood particleboard emissions [63]. Also, formaldehyde and acetaldehyde are common products from the oxidation of many VOCs [64–66]. Limonene and α-pinene showed the highest mean I/O ratios. Their concentrations were 415 and 365 times higher than those recorded outdoors, respectively. If only the wood section is considered, the I/O ratios reached values 634 and 612 times higher.

Benzene, toluene, ethylbenzene, and xylene isomers (BTEX) are emitted into the indoor environment from a large variety of sources, including industrial paints, adhesives, degreasing agents, and outdoor vehicle emissions ([67] and references therein). They are pollutants of great interest because of their adverse effects on human health, as potential carcinogens. Benzene showed similar concentrations in all sections of the shop (~1.4 $\mu g/m^3$). Toluene exhibited a higher concentration in the paint and adhesives section (53 $\mu g/m^3$), where a variety of potentially emitting products are used, such as solvents, paints, varnishes, adhesives, and sealants [68,69]. Martins et al. [69] also documented higher concentrations of toluene in painting and varnishing workshops, showing that the solvents used in those workshops did not contribute significantly to benzene concentrations. Similarly, the concentrations of ethylbenzene and xylene isomers were higher in the paint and wood sections, probably because they are part of paint and varnish formulations.

Outdoor concentrations of formaldehyde, acetaldehyde, toluene, and benzene, commonly related to car emissions [70,71] were low. The mean I/O ratio of these pollutants were 5.5, 7.5, 9.2 and 2.8, respectively. This indicates that local traffic does not have a strong influence in the indoor concentrations, and that these VOCs are mostly released by products and activities inside the store, as the indoor concentrations were higher than the outdoor levels.

Table 1 – VOC concentrations in different sections of the hardware store

VOC (µg/m³)	Indoor			Outdoor	Average I/O ratio
	Paint section	Wood section	Furniture section	Urban Road	
1,4-Dichlorobenzene	< 0.045	< 0.045	< 0.045	< 0.024	1.9*
Acetaldehyde	5.6	6.6	5.8	0.8	7.5
Acrolein	< 0.44	< 0.44	< 0.44	< 0.22	2*
α-Pinene	39	98	38	0.16	365
Benzaldehyde	1.3	1.5	1.5	0.24	6
Benzene	1.5	1.4	1.4	0.51	2.8
Butanal	< 1.5	< 1.5	< 1.5	< 0.8	1.9*
Ethylbenzene	9.5	5	3	0.36	16
Formaldehyde	7.7	12	8.5	1.7	5.5
Hexanal	16	36	17	< 0.5	46*
Isopentanal	1.1	1.4	1.1	0.1	12
Limonene	13	26	12	< 0.041	415*
m,p-Xylene	25	15	9.4	0.96	17.2
Naphthalene	0.5	0.5	0.45	0.13	3.7
o-Xylene	12	6.2	3.9	0.51	14
Pentanal	2.7	6	2.9	< 0.4	9.7*
Propanal	1.7	3	1.7	0.2	11
Styrene	2.5	2	2	0.077	28
Tetrachloroethylene	0.63	0.51	0.51	0.041	13
Toluene	53	21	14	3.2	9.2
Trichloroethylene	< 0.037	< 0.037	< 0.037	< 0.019	1.9*

* I/O ratio considering the detection limit

3.5 Microorganisms in settleable dust

To date, only a few countries have limits on the deposition of particulate matter. In Australia, the state of Queensland adopted the Environmental Protection Act 1994, which sets a limit of 120 mg/m^2/day to avoid nuisance [72]. The United Kingdom suggested a deposition limit of 200 mg/m^2/day in its guidelines TGN M17 [73]. All the values registered in the present study were below these thresholds (Figure 7). However, it should be noted that the values established by these guidelines are commonly used for ambient air. The highest dust deposition rate was observed in the wood section (15.5 mg/m^2/day), probably due to particle emissions during wood cutting and resuspension of wood dust.

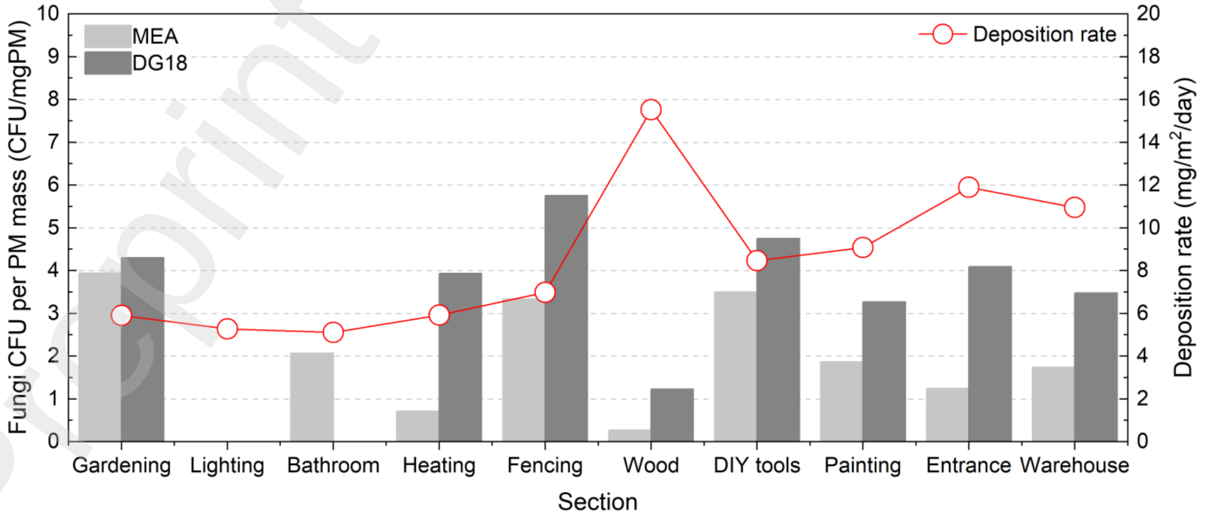


Figure 7 – Particulate mass fraction of colony forming units and dust deposition rates. MEA: malt extract agar, DG18: dichloran glycerol.

The emission of particles during the cutting of wood and the resuspension of fine sawdust fragments certainly contribute to higher values of settleable dust in this area of the store. It was also observed that the areas between the entrance and the wood section - right side of the store, see Figure 1 – also presented higher dust deposition rates. In contrast, the sections on the left showed the lowest depositions. This observation indicates a preferential air path on the right side of the store and a higher circulation of people and machines since the wood section and warehouse have a more intense traffic of hand pallet trucks.

The dust deposition rate had the following decreasing order: Wood section > Entrance > Warehouse > Painting > DIY tools > Fencing > Heating > Gardening > Bathroom furniture section. Despite presenting the highest deposition rate, the wood section registered the lowest concentration of fungal colonies with a value of 2.61 CFU/m^2/day in MEA and 14.4 CFU/m^2/day in DG18. The lighting section showed no fungal growth. This finding may be explained by the effects of light on fungal growth and germination. Some works [74,75], studying the effects of blue light on *Penicillium digitatum* and *Penicillium italicum* in citrus, found that high light intensities cause inhibition of spore germination, while low light intensities have a fungicidal effect. The largest number of colonies on MEA were identified in the tool section, with a total of 18.31 CFU/m^2/day; on DG18 the highest fungal concentration was at the entrance, with 30.01 CFU/m^2/day.

On MEA, *Trichoderma* sp. were the most common fungal species, corresponding to 41% of the species in this media. However, it should be considered that fast growing fungi, such as *Trichoderma* sp. can inhibit the growth of other fungi with higher clinical and toxicological relevance [76]. On DG18, the most prevalent fungal species were the ones belonging to *Penicillium* genus, accounting for 44% of the colonies found in this media. Special focus is given to the DIY tools section, which exhibited in all culture media the highest concentrations of *Penicillium* sp., 17% of the total species on MEA and 11% of the total species on DG18. DG18 also showed a greater variety of species, explained by the fact that dichloran restricts and limits the development of fast-growing species, such as the ones belonging to Mucorales (*Mucor, Lichtheimia* and *Rhizopus* genera) order and *Trichoderma* sp., decreasing competition [76,77].

In 2022, WHO listed *Aspergillus* section *Fumigati* as of critical priority due to this section clinical relevance and link with antifungal resistance, posing a menace for the treatment of invasive aspergillosis [78]. The fact that it was identified in different areas from the assessed indoor environments should be highlighted (Table 3). However, the list of WHO neglected the toxigenic potential from fungi and several of the identified fungi present toxigenic potential, such as the species belonging to *Penicillium* and *Aspergillus* genera. In fact, *Aspergillus* section *Flavi* was observed in DIY tools area, and it is the main producer of Aflatoxin B1, classified as Group 1A carcinogen

[79,80]. Thus, the fungal burden obtained must be considered as a threat to customers and workers since it presents clinical and toxicological potential.

Table *2* – Bacterial deposition rates in the different sections of the hardware store

Sampling Section	TSA		VRBA	
	CFU/m²/day	%	CFU/m²/day	%
Gardening	14.8	18.9	0	0
Lighting	12.7	16.2	0	0
Bathroom furniture	6.34	8.11	2.11	0.14
Heating	6.34	8.11	1535	99.7
Fencing	6.34	8.11	0	0
Wood	6.34	8.11	0	0
DIY tools	6.34	8.11	0	0
Painting	6.35	8.11	0	0
Entrance	6.34	8.11	2.11	0.14
Warehouse	6.35	8.11	0	0
Total	78.2	100	1539	100

TSA: tryptic soy agar VRBA: violet red bile agar

Table *3* – Deposition rates of fungi grown in different media for the different sections of the hardware store

Sampling Section	Fungi	MEA		DG18	
		CFU/m²/day	%	CFU/m²/day	%
Gardening					
	Aspergillus section *Aspergilli*			7.00	4.18
	Cladosporium sp.	3.91	4.34	6.52	3.89
	Aspergillus section *Nidulantes*			1.30	0.78
	Penicillium sp.	7.82	8.68	5.22	3.12
	Trichoderma sp.	2.61	2.90		
Bathroom furniture					
	Cladosporium sp.	1.30	1.44		
	Penicillium sp.	2.61	2.90		
	Trichoderma sp.	2.61	2.90		
Heating					
	Cladosporium sp.			2.61	1.56
	Aspergillus section *Fumigati*			3.91	2.33
	Penicillium sp.			7.83	4.67
	Trichoderma sp.	2.61	2.90		
Fencing					
	Cladosporium sp.	9.13	10.14	15.7	9.37
	Aspergillus section *Fumigati*			1.30	0.78
	Lichtheimia sp.			5.22	3.12
	Penicillium sp.			2.61	1.56
	Trichoderma sp.	5.22	5.79		
Wood					
	Aspergillus section *Aspergilli*			2.61	1.56
	Penicillium sp.			9.13	5.45
	Trichoderma sp.	2.61	2.90		
DIY tools					
	Cladosporium sp.			3.92	2.34
	Aspergillus section *Flavi*			2.61	1.56
	Penicillium sp.	15.7	17.43	18.3	10.93
	Trichoderma sp.	2.61	2.90		
Painting					
	Aspergillus section *Aspergilli*			9.14	5.46
	Chrysonilia sitophila			2.61	1.56
	Aspergillus section *Restricti*			2.61	1.56
	Penicillium sp.	6.53	7.25	3.92	2.34
	Trichoderma sp.	3.92	4.35		
Entrance					
	Cladosporium sp.			10.4	6.21
	C. sitophila			2.61	1.56
	Aspergillus section *Nidulantes*			1.30	0.78
	Penicillium sp.			15.7	9.37
	Rhizopus sp.				
	Trichoderma sp.	9.13	10.14		
Warehouse					
	Aspergillus section *Aspergilli*			1.31	0.78

	Cladosporium sp.	3.92	4.35	10.4	6.21
	Aspergillus section *Fumigati*	1.31	1.45		
	Mucor sp.	2.61	2.90		
	Penicillium sp.			11.7	6.99
	Trichoderma sp.	3.92	4.35		
Total		90.1	100	167	100

MEA: malt extract agar DG18: dichloran glycerol

As regards bacteria, for TSA, the highest numbers of colonies were observed in samples from the gardening and lighting sections, representing 18.9% and 16.2% of the total. A hypothesis raised is the proximity of plants and organic matter from compost bags or other substrates for application to the soil.

4. CONCLUSIONS

Indoor air quality was monitored in a home improvement and gardening store in northern Portugal during winter. As far as it is known, this is the first time that a comprehensive monitoring was carried out in this type of commercial establishment, despite its globalisation. A special focus was given to PM_{10}, which was analysed for elemental composition and carbonaceous content. The PM_{10} concentration was higher than the WHO guideline of 45 $\mu g/m^3$ and the CO_2 levels were below the protection threshold of 1250 ppm stipulated by Portuguese legislation. Using correlations, enrichment factors and I/O ratios, it was possible to identify the major sources of PM_{10}. Outdoors, exhaust and non-exhaust emissions from traffic, and biomass burning represented the most important contributors. Additionally, on one day, long range transport of dust from the Saharan desert was demonstrated by the high contents of crustal elements. Dust resuspension and several products were identified as major PM_{10} sources indoors. PM_{10}-bound Cr and Zn were likely linked to stainless steel and galvanized steel, respectively. VOCs were mainly associated with paints and wood. *Trichoderma sp.* and *Penicillium sp.* were the most common fungal species, mostly identified in the gardening, fencing and DIY tools section. Fungal species with clinical relevance (*Aspergillus* section *Fumigati*) and toxigenic potential (*Penicillium* and *Aspergillus* genera) were widespread in the different assessed areas. Concerning bacteria, there was not a great difference in the TSA media between sections. However, in VRBA, the heating section presented the highest number of colonies, above 1500 CFU/m^2/day.

High concentrations of particulate matter reveal that the health of customers, and especially workers who are subject to chronic exposure for at least 40 hours a week, may be at risk. The detection of toxigenic fungi and concentrations of particulate matter that exceed WHO guideline values and indoor air quality standards, especially in some sections of the commercial establishment, alerts those responsible for the company and occupational health authorities to the need to carry out more frequent disinfection and dust cleaning, among other possible measures.

Given that the concentrations of atmospheric pollutants may present seasonal variations, it is recommended to repeat the monitoring campaign in the summer period. In future studies, to allow more robust statistical treatments and the application of Positive Matrix Factorisation (PMF) or other source apportionment model, monitoring should be longer to allow obtaining a higher number of samples. Since polycyclic aromatic hydrocarbons, brominated flame retardants, plasticisers, and other organic compounds are important risk factors for humans, the speciation of these PM-bound constituents should also be addressed.

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