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Full length article

Monitoring and apportioning sources of indoor air quality using low-cost particulate matter sensors

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ABSTRACT

Air quality is one of the most important factors in public health. While outdoor air quality is widely studied, the indoor environment has been less scrutinised, even though time spent indoors is typically much greater than outdoors. The emergence of low-cost sensors can help assess indoor air quality. This study provides a new methodology, utilizing low-cost sensors and source apportionment techniques, to understand the relative importance of indoor and outdoor air pollution sources upon indoor air quality. The methodology is tested with three sensors placed in different rooms inside an exemplar house (bedroom, kitchen and office) and one outdoors. When the family was present, the bedroom had the highest average concentrations for PM_{2.5} and PM₁₀ ($3.9 \pm 6.8 \mu\text{g}/\text{m}^3$ and $9.6 \pm 12.7 \mu\text{g}/\text{m}^3$ respectively), due to the activities undertaken there and the presence of softer furniture and carpeting. The kitchen, while presenting the lowest PM concentrations for both size ranges ($2.8 \pm 5.9 \mu\text{g}/\text{m}^3$ and $4.2 \pm 6.9 \mu\text{g}/\text{m}^3$ respectively), presented the highest PM spikes, especially during cooking times. Increased ventilation in the office resulted in the highest PM₁ concentration ($1.6 \pm 1.9 \mu\text{g}/\text{m}^3$), highlighting the strong effect of infiltration of outdoor air for the smallest particles. Source apportionment, via positive matrix factorisation (PMF), showed that up to 95 % of the PM₁ was found to be of outdoor sources in all the rooms. This effect was reduced as particle size increased, with outdoor sources contributing >65 % of the PM_{2.5}, and up to 50 % of the PM₁₀, depending on the room studied. The new approach to elucidate the contributions of different sources to total indoor air pollution exposure, described in this paper, is easily scalable and translatable to different indoor locations.

1. Introduction

Particulate matter (PM) is known to have diverse negative health effects, and is one of the major causes of premature deaths globally (Liu et al., 2019; Pascal et al., 2013; Pope et al., 2009; Rivas et al., 2021; Shiraiwa et al., 2017). It also contributes to the deterioration of the wider environment (Manisalidis et al., 2020; Mannucci & Franchini, 2017). Air pollution has become one of the most important issues concerning public health, causing the premature loss of millions of lives worldwide (Fuller et al., 2022) as well as increasing the hospitalisation cost (Birnbaum et al., 2020; Pimpin et al., 2018; Wei et al., 2019) and ambulance callouts (Sangkharat et al., 2019). It is estimated that about

half a million people have died in 2019 in Europe, due to direct or indirect effects of deteriorated air quality (EEA, 2020). Exposure to PM_{2.5} is considered among the top ten factors leading to life expectancy reduction (Forouzanfar et al., 2015), hence the new stricter guidelines set by the World Health Organisation, further lowering the recommended PM concentration exposure limits (WHO, 2021). The vast majority of the studies on this field have focused on assessing the air quality near pollution hot spots or background sites mainly located outdoors. While such studies provide useful information about the sources of pollution and their contribution to the deterioration of the atmospheric environment, they are mainly focused on assessing the air quality outside people's residences and working environment. This practice fails

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to consider the air quality in the environments where most people spend the greatest part of their day (Duan, et al., 2021; Klepeis et al., 2001). Presently, the number of people that work either indoors or from their homes is increasing. It is estimated that at present about 9.9 million people work from their homes in the U.K., which is more than double compared to pre-COVID19 periods (ONS, 2022a). Working from home is associated with higher levels of job satisfaction due to increased autonomy and life-work balance (Charalampous et al., 2019; Wheatley, 2017, 2021). Similarly, many that are not able to completely accomplish working at home are increasingly utilising hybrid working schedules, combining both working from home as well as from their usual workplace (ONS, 2022b; Eurostat, 2022). This trend has become more prominent especially after the COVID19 pandemic, the lockdowns that were imposed, and the social distancing measures that were implemented in public spaces and in workplaces. Thus, indoor air quality assessment is an increasingly important factor for public health problems related to air pollution (Vardoulakis & Kinney, 2019). Air pollution has been shown to affect not only physical health but also cognitive ability and mental health (Shehab & Pope, 2019; Wen & Burke, 2022; Ke et al., 2022). Hence work productivity whilst working in the home will likely scale with indoor air quality.

Indoor air quality assessment has been studied for many years, though not as intensively as the outdoor environment. This is likely due to the diversity of the indoor environments, the high cost that comes with such studies, as well as the perceived importance of indoor versus outdoor studies and the lack of regulations for indoor environments. Until recently, the instruments used for air quality assessment were solely of regulatory grade, thus having a high acquisition, deployment, and maintenance cost, as well as being bulky and heavy. Within the last decade, the emerge of low-cost sensors (LCS), which have a considerably lower cost and a smaller footprint, has made air pollution measurements easier and more diverse (Lewis et al., 2018; Penza, 2019). These sensors in most cases lack the accuracy of the regulatory grade instruments (Austin et al., 2015; Sousan et al., 2016). Additionally, they often are affected by atmospheric conditions which reduce their ability to provide useful outputs, as well as having a constant need of calibration (Hagan & Kroll, 2020; Wang et al., 2021). Regardless, in recent years LCS have been used in many studies which were not possible with traditional instruments, due to cost or difficulties in instrument deployment (Jovasević-Stojanović et al., 2015; Pope et al., 2018). Successful applications of LCS have shown that regardless of the shortcomings that come with their use, they can fulfil several tasks for which absolute accuracy is not the main consideration. Thus, applications in schools or working areas, multi-site studies and mobile measurements were successfully carried out (Ionascu et al., 2018; Miskell et al., 2018; Weyers et al., 2018), showing their capability in either working alongside and expanding the current monitoring infrastructure (Weissert et al., 2020), or potential with further development to replace the existing instruments. More extensive deployment of LCS will create a denser monitoring network, which will help in better assessing and improving the air quality in more areas, even at neighbourhood level (Feinberg et al., 2019; Prakash et al., 2021; Shindler, 2021).

Indoor air quality levels have been studied in working areas, transportation, and residences (Isa et al., 2022; Wu et al., 2023), with the indoor air quality in most cases being similar and sometimes worse than the respective outdoor environment (Jones et al., 1999; Du et al., 2022). Patel et al., (2020) quantified the effect of both cooking and post cooking decay periods on indoor air quality, identifying a great increase in ultrafine particle concentrations associated with these periods. Additionally, outdoor air infiltrating indoor environments introduces ambient particles (El Orch et al., 2014; Johnson et al., 2017). Matson, (2005) and Zhu et al., (2015), highlight the importance of outdoor sources on the PM₁ and PM_{2.5} concentrations in several indoor environments. Furthermore, Morawska et al., (2017), Vardoulakis et al., (2020) and Kalimeri et al., (2019) discussed the differences found on the I/O ratios of PM and particulate number depending on the use of the

indoor environment. For all the indoor environments studied, the effect of the outdoor sources was crucial for the indoor air quality. While most of the indoor studies were conducted using regulatory grade instruments, the use of LCS expands the range of studies possible, such as the one performed by Ferro et al., (2022) in which the levels of PM_{2.5} were measured simultaneously in 50 houses in Monroe County, USA. Similarly, Krebs et al., (2021) explored the temporal and spatial variability of PM using 1000 LCS placed indoors and outdoors in California, USA. Such studies are becoming more common, increasing the spatial coverage of air quality measurements and improving the output and usability of the LCS (Bi et al., 2022; Weyers et al., 2018).

In the present study, the air quality of a typical family house in the county of Worcestershire, UK has been studied. Three LCS measuring particle number concentrations were deployed for a period of about 50 days in different rooms within the house, as well as an additional sensor measuring the outdoor conditions next to the house. These measurements were compared to measurements from two regulatory grade instruments for quality checking and calibration. The air quality within the house is studied and the possible effect of the indoor particle sources on the occupants' health is considered. Additionally, the effect of the I/O interactions is studied, and with the use of Positive Matrix Factorisation (PMF), indoor and outdoor sources of particles are separated, and their effect is quantified. A number of studies have successfully applied PMF, Non-Negative Matrix Factorisation and other methodologies on LCS data for source identification and apportionment, however, these were only applied on outdoor datasets (Bousiotis et al., 2021, Bousiotis et al. (2022a,2022b; Mills et al., 2023; Hagan et al., 2019). While there are many previous studies that assess the air quality of residential environments (Tryner et al., 2021; Zamora et al., 2020; Manibusan & Mainelis, 2020), to the authors' knowledge this is the first study to quantify the effect of the indoor and outdoor particle sources using source apportionment methods. Furthermore, the effect of specific activities in the house as well as pollution hot spots are considered and studied, and the overall health effect of the indoor air quality is discussed. Finally, while the specific house is located in a rather clean semi-rural area, the effect of the outdoor environment is discussed, and the possible effect of a more polluted outdoor environment is considered. The methodology is easily translatable and scalable to different indoor environments.

2. Methodology

2.1. Study site and materials

The study site is a typical family house in a semi-rural area in the county of Worcestershire, that is approximately 15 km southwest of Birmingham, UK, the UK's second largest city. The house (Fig. 1) accommodates a family of four (two parents and two children - all non-smokers). The measurement period covered 1.5 months between 16/12/2021 to 2/2/2022, a period which included both the Christmas break in which the whole family spent most of their time at home, as well as normal activity periods. The house has central gas heating and a fully electric cooker, incorporating hob; oven and grill, used for the majority of the meals. In rare cases a toaster was used for quick meals. During cooking periods, the extractor fan is typically used.

Four Alphasense OPC-N3 sensors were deployed at the house collecting particle number size distribution data for the study period at a 10 s resolution. The Alphasense OPC-N3 is a low-cost (costing approximately GBP250) optical particle counter measuring particles in the range between 0.35 and 40 μm . The lowest PM concentration reported by the sensor was 0.025 $\mu\text{g m}^{-3}$ (0.01 $\mu\text{g m}^{-3}$ is the manufacturer's stated lower measurement limit). Three of the sensors were placed in different rooms inside the house (a bedroom (volume = 40 m^3) on the upper floor, and the kitchen (58 m^3) and the office (46 m^3) on the ground floor), while the fourth one was placed outside the house (about 2 m away from the back door and 1 m above the ground). In each room there were

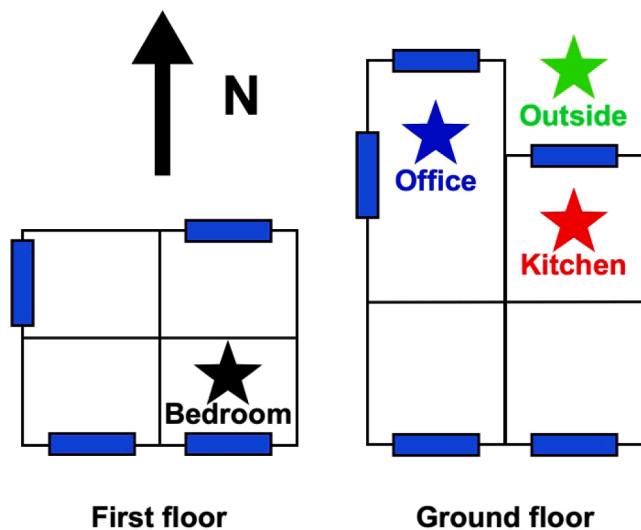


Fig. 1. Approximate plan of the house. Blue rectangles represent windows which contain trickle vents. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

trickle vents that allowed the entrance of ambient air. A trickle vent is a device fitted to the top of a window that allows fresh air to circulate naturally through a room. The ratio of room surface area to the number of trickle vents is 16 for bedroom, 10.7 for the kitchen and 5.3 for the office (greater ratio indicates fewer trickle vents per surface area). Each room also had windows, but these were rarely opened due to the study being in the cold season. The kitchen also has an external door which provides significant ventilation when opened. The data availability for the measurement campaign period was 100 %, and the measurements were averaged up to a resolution of 10 min for the analysis in this study.

Calibration is crucial for the integrity of the measurements of LCS. Specifically, Alphasense OPCs are known to overestimate PM mass concentrations (especially PM_{10}) when ambient levels of relative humidity (RH) are high due to PM hygroscopicity effects (Crilley et al., 2018, 2020). PM hygroscopicity effects tend to become important at $RH > 70$ %. Within the study household, RH was typically below this threshold, but the outdoor sensor was often exposed to higher RH.

The LCS were calibrated using two TSI-3330 Optical Particle Sizers. The TSI-3330 is a calibrated research grade instrument measuring particle size distributions in the range between 0.3 and 10 μm . While these TSI instruments are considered more capable compared to the low-cost sensors, they have reported limitations. Non-existent peaks and zero concentrations in very clean conditions were reported in previous studies with similar instruments (Rivas et al., 2017). Due to this, measurements from these instruments should be handled with care. In the present study the TSIs were collocated with the sensor located in the office and the sensor located outside for four periods during the campaign. The total measurement time for the TSI instruments was about 14 days for each. The sampling time was 1 min per sample, and data were also averaged in a 10-minute resolution for consistency. During the experiment, all the LCS were collocated together for two periods in the same room to test the consistency between the four LCS used. Consistency between the LCS was good and there was no need to inter-calibrate the LCS.

Temperature and relative humidity data (RH) data were collected using BME-280 sensors attached on the sensors. Synoptic meteorological data (wind direction, speed and visibility) were considered from the nearby meteorological station at the Birmingham International Airport (about 15 km NE from the house). Finally, no changes in their everyday routine were asked from the members of the family for the period of the campaign, apart from keeping a detailed diary of events for reference. The diary included information about activities that could affect the

particle composition within the house, such as cooking times, meal types, cleaning sessions, extreme outdoor conditions, collocation periods and several notable events in the house.

2.2. Positive matrix Factorisation and estimation of the PM concentrations

Positive Matrix Factorisation (PMF) is a commonly used multivariate data analysis method proposed and developed by Paatero & Tapper, (1993; 1994). It has been applied successfully for apportioning sources of pollution in multiple air quality studies (Beddows et al., 2015; Cesari et al., 2016; Harrison et al., 2011; Rivas et al., 2020). In atmospheric studies the method describes the relations between variables' measurements (concentrations of chemical species, particle concentrations etc.) using a least-squares technique (Reff et al., 2007). It considers the measured observed data and the experimental uncertainty and outputs a matrix of factors (F) which best represent the average particle concentrations (in this case) accounted for the different sources of pollution and a matrix of contributions (G) which represent the relative contribution of each factor in a given timestep. Being a descriptive model, PMF does not have an objective criterion for the optimal number of factors, thus the best solution is the one that best describes the conditions for the specific case analysed (Paatero et al., 2002). For the specific case study, the datasets from the rooms of the house were combined and the factors formed present particle profiles that are commonly found between the rooms, which allows for direct comparison of the conditions between the rooms.

For the estimation of the PM concentrations, the elements of the F matrix representing the average mass particle concentration for each size bin were multiplied with the elements of the G matrix. As the elements of G are normalised (having an average value of 1), the fluctuation of G provides a metric of the effect of each of the variables of the F matrix, also considering the non-explained variance. This method provides an estimation for the PM concentrations at a given time for each factor, though it may be subject to inaccuracies, as the PM concentrations are calculated according to an average PM concentration for all rooms. Nevertheless, the method provides an estimate of the contribution of each factor on the PM concentrations and can be reliably used for the description of the conditions in the house, as well as for the comparisons done in the present study.

The PMF software developed by Paatero (2004a, b) was used. Additional analysis of the data was done using the Openair package for R developed by Carslaw & Ropkins, (2012).

2.3. Time shifting

The datasets of the different rooms were combined and then analysed using PMF. In some cases, time shifting of the datasets was applied to elucidate possible interactions between the rooms, as well as with the outside. Time shifting involves the omission of some data points (time periods) at the beginning of one room's dataset and recalculation of the Pearson correlations (r) between the factors or the PM concentrations between the rooms. An increase on the r between the PM concentrations in two rooms when e.g. 10 min of data from one room is removed means that the variation of the specific PM or factor contributions coincides better between the rooms with 10 min' delay. This can be an estimation for the time needed for changes in one room to affect the other i.e. travel time of particles between rooms. In the present study multiple time shifts were attempted for factor contributions and PM concentrations and the ones that presented the highest r are reported.

3. Results

3.1. LCS calibration

To calibrate the LCS mass concentrations in the PM_{10} , $PM_{2.5}$ and PM_{10}

size fractions, the collocated TSI 3330 measurements in the same size fractions were used as the reference values. The correlation between the indoor LCS and TSI instrument was good, with $r > 0.9$ for all PM size fractions, but there was an offset of approximately a factor of 2. The outdoor LCS measurements were clearly affected by high RH in the outdoor location, because of its increasing effect upon particle water content as determined by particle hygroscopicity. Hence, a RH dependent calibration was needed for the LCS to obtain the regulatory relevant dry mass concentrations that the TSI instruments measure. The relationship between the ratio of the measured OPC and TSI mass concentrations against RH was well approximated using an exponential curve for the combined indoor and outdoor data. This relationship is expected

since the particle hygroscopicity increases exponentially with RH. This RH dependent relationship was used to calibrate all LCS data regardless of whether it was taken indoors or outdoors.

After the RH dependent calibration, the correlation between the indoors LCS and TSI was good and very similar to that without calibration. The performance of the outdoor LCS was not as good, as extreme and nonsensical outdoor PM measurements were sometimes measured, which were not captured by the collocated TSI instrument. These time periods coincided with low visibility (less than 1000 m), see figure S2, suggesting that the extreme values were due to mist measurement rather than PM measurement. Mist events, as diagnosed by visibility being less than 1000 m, were removed from the dataset. The removed data

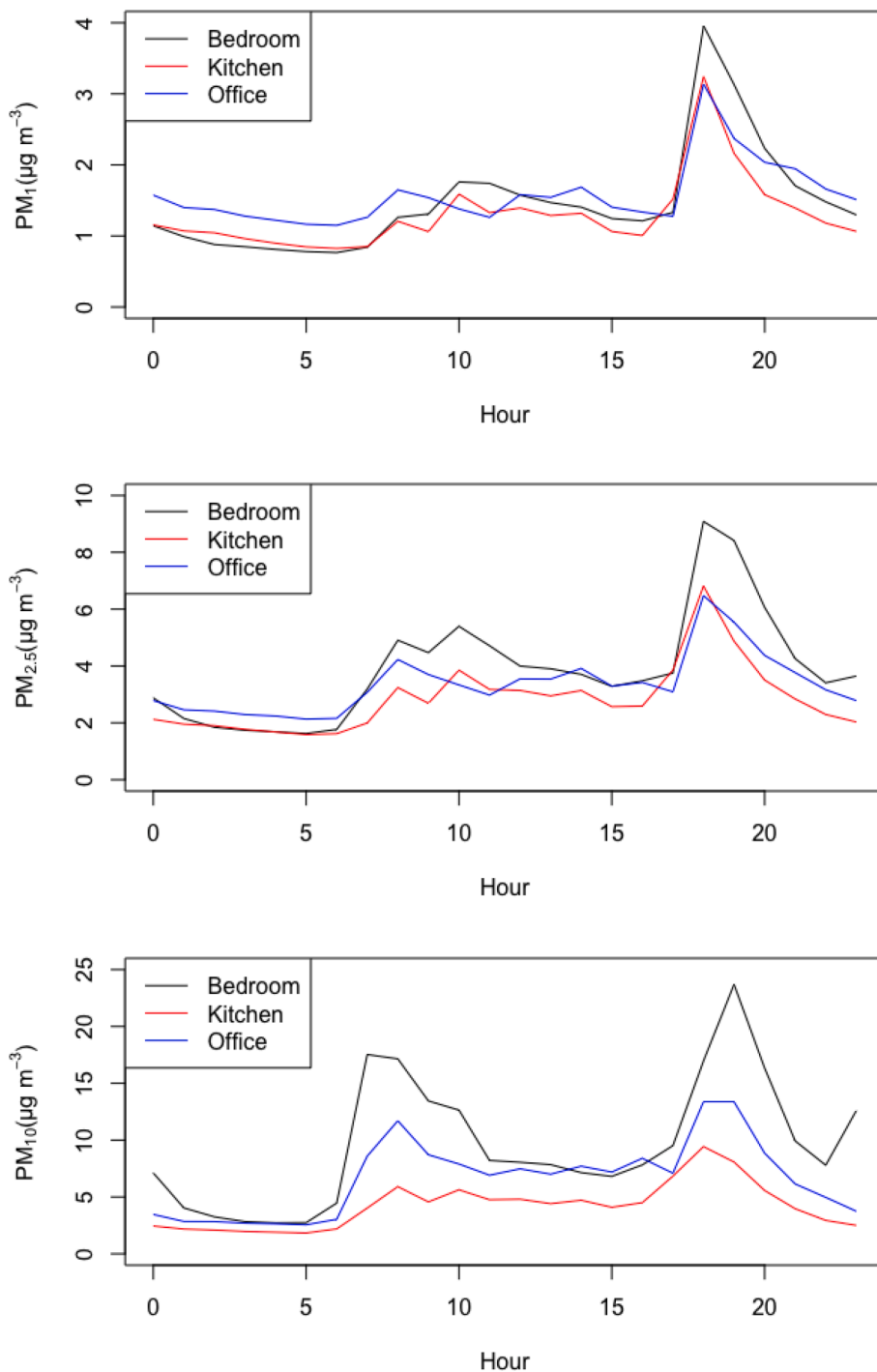


Fig. 2. Diurnal variation of indoor PM during the full measurement period for the PM₁, PM_{2.5} and PM₁₀ size fractions.

accounted for less than 5 % of the outdoors OPC dataset. Once the mist events were removed, the RH dependent calibration greatly increased the correlation between the LCS and TSI instrument, especially for the PM_{10} readings, where before calibration $r = 0.19$, and after calibration $r > 0.70$. During the calibration periods when the LCS and TSI instruments were collocated. The greatest discrepancy between the outdoors LCS and TSI was found during the first period ($r = 0.35$ for PM_{10}), when there were long intervals with high RH and mist. For the other 3 periods the meteorological conditions were more favourable improving the performance of the OPC ($r > 0.75$). Finally, using the curve formed by the calibration against the research grade instruments as well as the two collocation periods (total of 30 h when all the sensors were put together in the same room (figure S1), the additional indoor OPCs were also calibrated.

3.2. Averages, comparison between the rooms, outdoor conditions, and diurnal variability

The diurnal variation of the PM, in the PM_1 , $PM_{2.5}$ and PM_{10} size fractions, for each room is presented in Fig. 2. PM_1 values were found to be similar across all rooms without any notable difference in their diurnal variation. The greatest increase occurs about 18:00, which coincides with the typical time of dinner preparation. While this increase is simultaneous among all the rooms for the PM_1 , it is also observed for the larger PM in the kitchen. The other two rooms, while having a significant increase on the PM concentrations at the same time, presented higher peaks an hour later for $PM_{2.5}$ and PM_{10} , probably associated with other activities after dinner time. The differences become more significant with the greater PM sizes, pointing to the more limited spread of the larger particles formed in one room to the rest of the house rather than the effect of external sources. The concentrations of $PM_{2.5}$ and especially PM_{10} were higher in the office and more than double in the bedroom (for PM_{10}) compared to the kitchen, which appears to have similar PM ratios with outdoors. The increased values found in the office are attributed to the high number of trickle vents in the office, continuously allowing outdoor particles to enter the room, as well as the soft furnishings found there (more cloth, pillows etc.). The high ratio of trickle vents to room volume also likely explains the increased PM_1 and $PM_{2.5}$ concentrations in the office compared to the other rooms during the night. The soft furnishings in the bedroom are accompanied by carpeting on the floor, not present in the other rooms, which captures more larger particles, thus increasing the PM resuspension effect in the room. Furthermore, peaks early in the morning (at 06:00) and late at night (23:00) observed mainly for the PM_{10} and to a lesser extend for $PM_{2.5}$, especially in the bedroom, were associated with the family waking up or the adults going to bed. For these activities no changes are observed for the smaller PM_1 .

The outdoor average PM concentrations for the measurement period are also presented in the Table S1, and consistent with many other studies, they were higher than indoor concentrations (Liu & Zhang, 2019). Being in a semi-rural area, the PM concentrations are relatively low, with the average of both $PM_{2.5}$ and PM_{10} being lower than the guidelines (annual means of $5 \mu\text{g m}^{-3}$ for $PM_{2.5}$ and $15 \mu\text{g m}^{-3}$ for PM_{10}) set by the World Health Organization (WHO, 2021). Higher PM concentrations are observed mainly with winds from the eastern sector where the major town of Redditch is located (though the possibility of regional pollution cannot be excluded), peaking at both evening and night hours, affecting all particles sizes, with a more significant effect on smaller ones (figure S3). The average indoor/outdoor (I/O) ratios found were 0.40, 0.44 and 0.56, for PM_1 , $PM_{2.5}$ and PM_{10} respectively. These ratios are at the lower end of those found in previous indoor studies (Niu et al., 2015; Sangiorgi et al., 2013; Zhao et al., 2015), which is probably due to the lesser ventilation expected in the colder season.

While the concentration of the PM provides a general picture of the conditions in each room an estimation of the nature of the sources that affect them can also be considered by the spread of these values (Fig. 3). The standard deviation between the rooms as well as the outside is

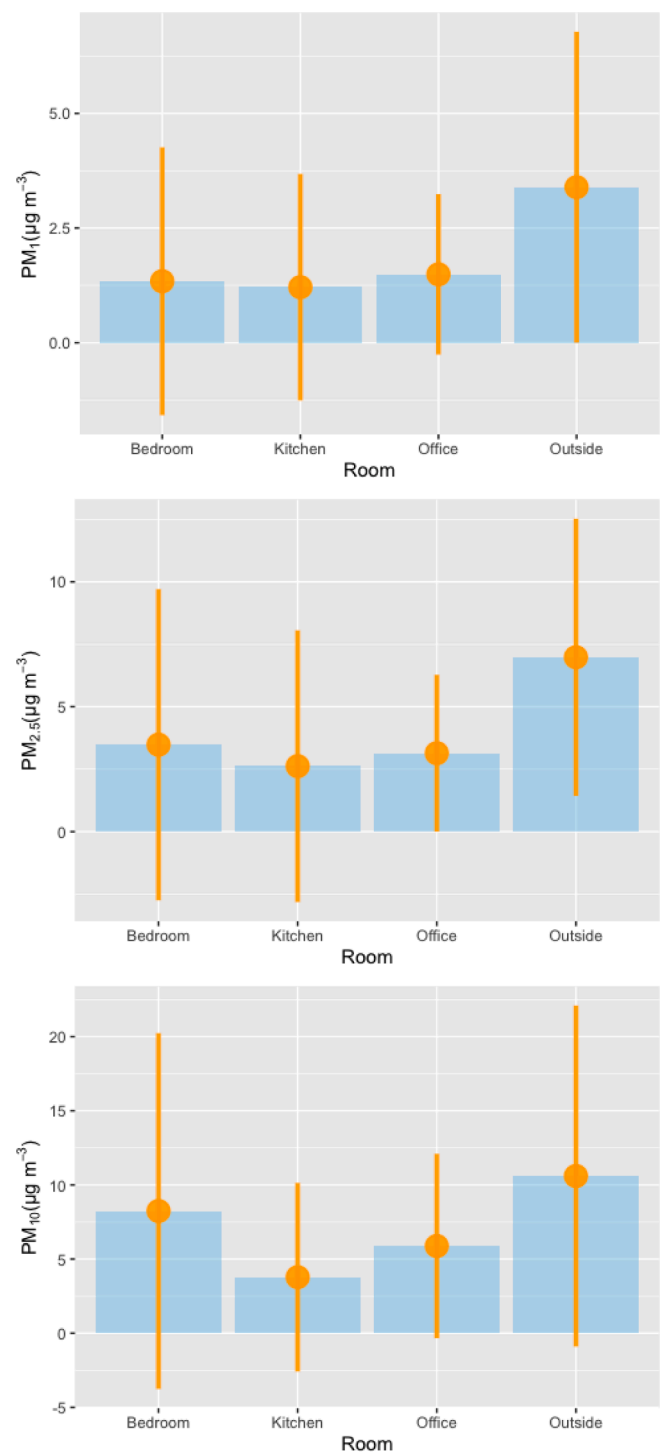


Fig. 3. PM_1 , $PM_{2.5}$ and PM_{10} averages (light blue bar and orange dot height) and standard deviations (orange vertical line) per room. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

decreasing with the range of the expected effect from outdoor sources, as regional sources which are more common at a background site tend to be more stable and fluctuate less compared to local sources, such as the ones from the indoor environment. Thus, the bedroom and kitchen which are the rooms with the greater expected effect from indoor sources, present a greater spread of their values for all the PM sizes, as displayed by the higher standard deviation. The office showed the lowest spread of values due to both the increased ventilation from the

trickle vents as well as the limited indoor factors (furnishing and activities) compared to the other rooms. The spread in the outside data is further limited as, being a background site, it is mainly affected by regional sources. This agrees with the findings of Frederickson et al., (2022), who noted that regional sources of pollution tend to have a more constant presence and lower fluctuations on their effect compared to more local ones.

3.3. Absent periods and I/O interactions

During the campaign there were two periods when house was unoccupied. These were between the 17/12/2021 – 21/12/2021 (ABS1) and 29/12/2021 – 02/1/2022 (ABS2). During these periods there was no activity in the house and the observed variation of the particles was mainly due to the I/O interactions. PM concentrations for all size ranges were similar for all the rooms, especially for the smaller PM sizes (Table 1). They were consistently higher in the office though, due to the greater presence of trickle vents in that room, which was the main entrance point for outdoor particles. While there was a variation in the ratio of PM concentrations between the outside and office values, office PM₁ values occasionally exceeded 50 % of the outside values (Fig. 4 & S4). This shows the range of the effect of the I/O interactions, although in both cases the office windows were not downwind of the average wind direction (figure S5). This I/O effect reduced as particle size increased, though remains significant throughout the absent period. In general, the I/O ratios for both absent periods were 0.41, 0.29 and 0.09 for PM₁, PM_{2.5} and PM₁₀, respectively, in the office and were reduced moving to the other rooms in the house, though this varied depending on the outdoor conditions. Additionally, the Pearson correlation between the office and outside PM concentrations is 0.57, 0.70 and 0.38 for PM₁, PM_{2.5} and PM₁₀, respectively, with an increasing trend for the PM₁ and the PM_{2.5} when time shifting them (for a 20-minute time shifting the correlations were 0.71, 0.76 and 0.38 respectively). The correlations are a lot higher between the rooms. Using the time shifting method the travelling time between the office and the kitchen for all PM sizes was found to be less than 10 min. This travelling time increased to 30 min between the office and the bedroom for the PM₁ and the PM_{2.5}. This is consistent with the travel time of the kitchen-bedroom couple being 20 min, though the maximum r between office and bedroom for PM₁₀ is found 10 min earlier, probably due to the faster settling of the larger PM₁₀.

The greater ventilation in the office is also confirmed by the PM variation during the absent periods, in which any significant change in their concentration is detected first from the outside sensor, followed by the office sensor (with a 10–20-minute delay) and finally by the other two sensors in the house (once again with about 10-minute delay). While this spread was a common observation throughout the absent period, the PM increase was not the same between the rooms, being in some cases

Table 1

Mean concentrations of PM₁, PM_{2.5}, PM₁₀ for each room during the Present and Absent periods and their ratios.

Present	PM ₁ (µg m ⁻³)	PM _{2.5} (µg m ⁻³)	PM ₁₀ (µg m ⁻³)	PM ₁ /PM ₁₀
Bedroom	1.47 ± 3.19	3.89 ± 6.76	9.62 ± 12.7	0.15
Kitchen	1.29 ± 2.69	2.84 ± 5.93	4.22 ± 6.88	0.31
Office	1.57 ± 1.88	3.36 ± 3.34	6.64 ± 6.53	0.23
Absent	PM ₁ (µg m ⁻³)	PM _{2.5} (µg m ⁻³)	PM ₁₀ (µg m ⁻³)	PM ₁ /PM ₁₀
Bedroom	0.72±0.45	1.45±0.94	1.52±1.55	0.47
Kitchen	0.78±0.51	1.54±0.99	1.58±1.06	0.49
Office	1.07±0.80	2.05±1.45	2.21±1.64	0.48
Ratio (present/absent)	PM ₁ (µg m ⁻³)	PM _{2.5} (µg m ⁻³)	PM ₁₀ (µg m ⁻³)	
Bedroom	2.04	2.68	6.33	
Kitchen	1.65	1.84	2.67	
Office	1.47	1.64	3.00	
Average	1.72	2.05	4.00	

reduced by up to 50 % from the office to the other rooms, though a direct effect from the limited number of trickle vents in these rooms should also be considered. This trend was found throughout the measuring period even for sources within the house, as discussed later. Particles, mainly in the PM₁ range were spread throughout the house in about 10 min, regardless of the room these were generated in. This led to a rather homogenised PM₁ profile throughout the house. Larger particles had a greater variation and were not similarly spread.

The absent period measurements can be used to calculate the extent of the effect of human activity, which is strongly dependent on the activities in each room. An increase of about 150 % on average was found during the present period for PM₁ (Table 1). This increase is more significant as the particle size increases, surpassing 600 % in the bedroom for PM₁₀. I/O interactions are greater for the smaller sized particles, whereas indoor activities are more important for larger particles. It should be noted that the lower particle size measurement limit may bias these results, as particles below 350 nm are not measured.

3.4. PMF analysis

3.4.1. Model performance

A combined dataset from all the rooms in the house was analysed using PMF, and after testing solutions with more and less factors, a 5-factor solution was chosen as the best solution, since all factors present unique variations with low covariance. While a per-room analysis was also attempted, which could provide better accuracy on the PM concentrations, the combined dataset method was chosen as it provides information on common household sources.

The PMF performed very well in portraying the PM concentrations in the house (Fig. 5). A very strong R² was found between the measured and modelled values for all PM sizes varying from 0.861 up to 0.996 (table S2). The model accuracy is comparatively lower for PM₁₀, because the very sharp peaks in the measurements in most cases were underestimated by the model. This is likely due to the variety and combination of PM₁₀ sources within the house which cannot be fully explained by the 5 factor model. Nevertheless, even though this methodology considers the fluctuation of a single average PM concentration per factor, for all rooms, contrary to an individual room analysis. The model successfully quantifies the effect of the different factors inside the house and provides an accurate estimate of the effect of each one of them based on the different contributions found in each room. As a result, due to the combination of the datasets the values of the PM were slightly underestimated for the rooms with higher concentrations (bedroom and office) while they were similarly slightly overestimated for the room with the lower concentrations (kitchen).

The adequate separation of the factors can be seen by looking at the difference of the factors affecting the particle composition within the house between the absent and present periods, as well as within the diurnal profiles. This will be further discussed later in the paper.

3.4.2. PMF results

The average PNSD, the explained variance for the PM, and diurnal variation of the contribution G for each factor are presented in figures S6 and S7. Out of the five factors, two were found to have the greatest similarity among the rooms (the first and the fourth factors) and are associated with outdoor particle sources, while the other three factors (the second, third and fifth) were associated with activities within the house. While the indoor factors were clearly associated with indoor activities in general, it was not possible to pinpoint the exact activity or combination of activities each one was associated with. Nevertheless, looking at the variation during the absent periods, the two outdoor factors are dominant across all rooms during both periods. This can be clearly observed in the results of the 2/1/2022 when the family returned to the house after ABS2 (figure S8). There is an almost complete absence of the other three factors until about 14:30 when the family returned. After that time there is a shift to the other three factors. Additionally, in

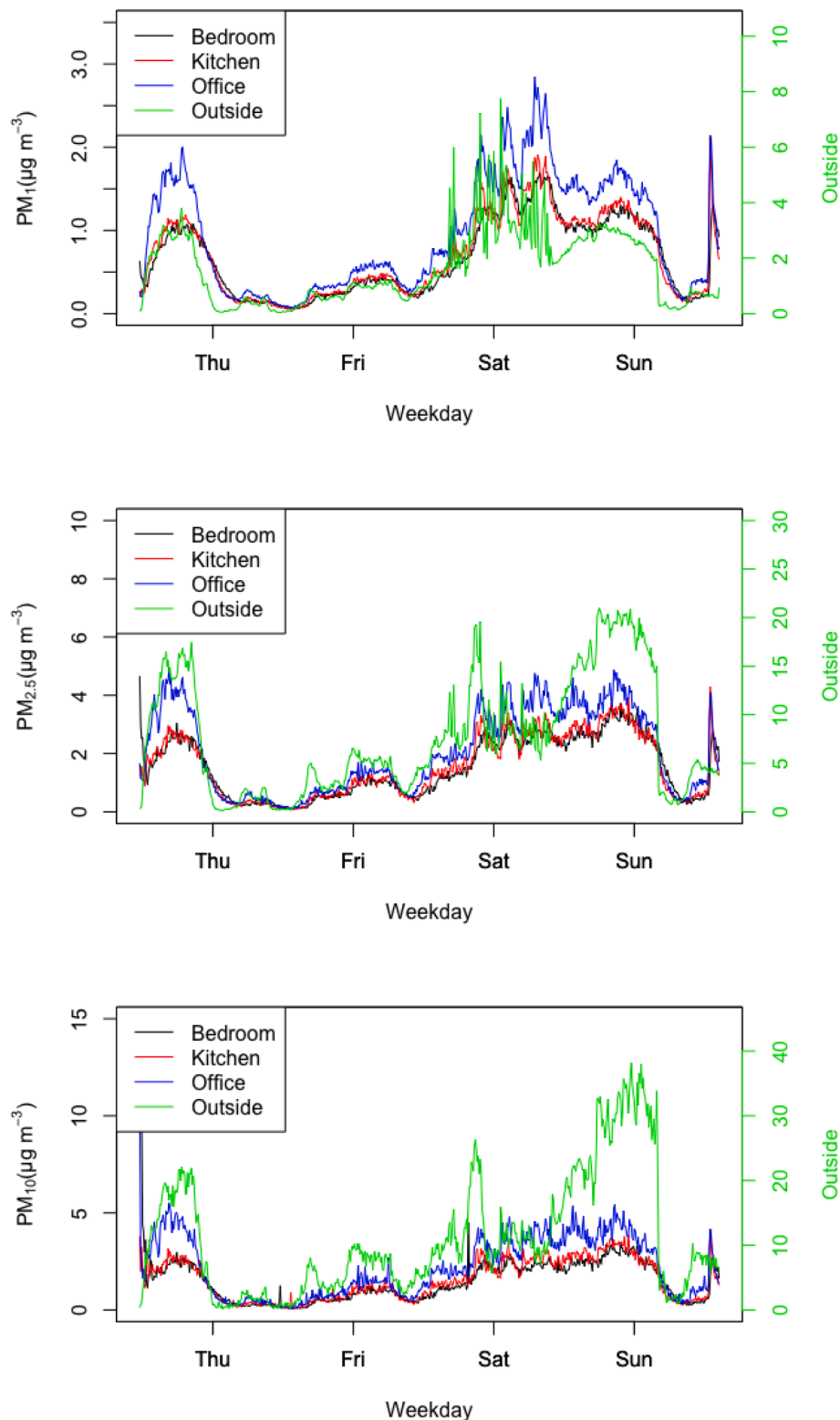


Fig. 4. PM concentrations per room and outside during ABS2. Note that the outside range is $3 \times$ greater than the rooms' range.

the evening the sharp increase of the PM₁ coincides with the cooking time and it is attributed to outdoor factors. This is expected as it is more likely that the OPC can measure particles from effects associated with cooking (e.g. open windows for ventilation or rubbish disposal), rather than those deriving from the cooking process itself. The use of the extractor fan whilst cooking will also reduce any particles associated with cooking. While these factors appear in both ABS1 and ABS2 there is

a great difference in the factors' contribution between them, with the first period being mostly affected by the first factor and the second period by the fourth. As there was no activity within the house for these periods, and no obvious indoor sources, these factors are the effect of outdoor particles entering the house. These external factors present different characteristics (table S3), with the first one having an almost exclusive presence of small particles (PM₁), while the larger particles are

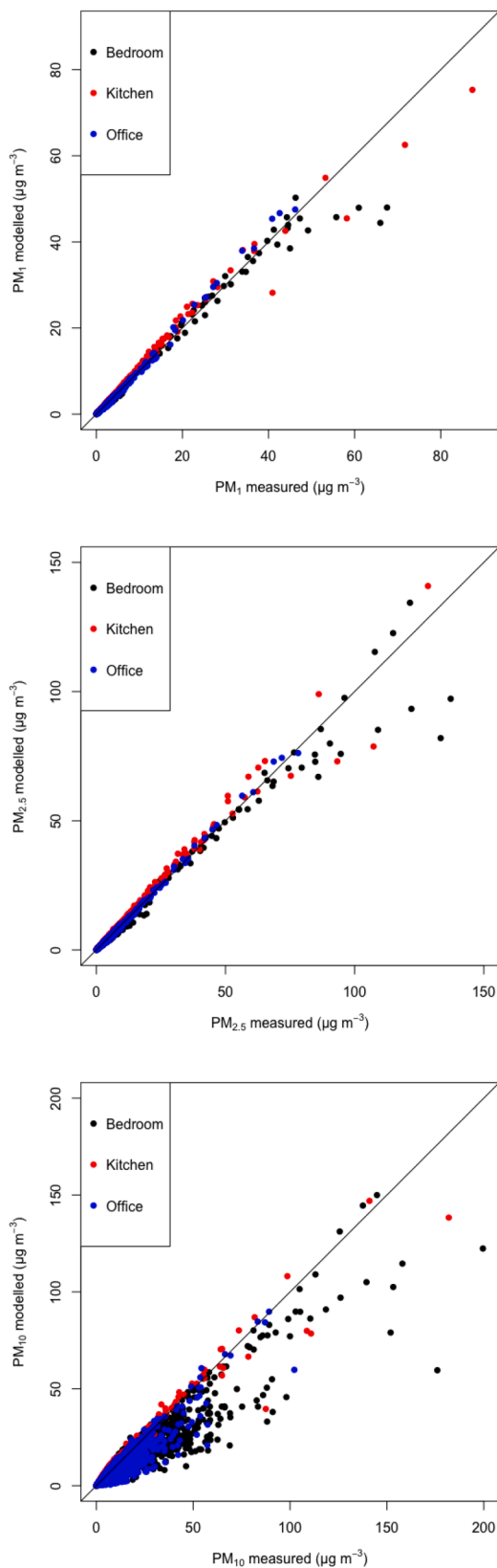


Fig. 5. Correlation plots for PM_1 , $PM_{2.5}$ and PM_{10} of the PMF modelled PM against the measured PM against concentrations for all rooms. The black line represents the 1:1 ratio.

almost non-existent (this results in erroneous ratios higher than 1 in the PM concentration estimation in some cases – table S4). The fourth factor also has relatively high PM_1 concentrations, though there is a significant presence of larger particles as well, mainly up to the size range of $PM_{2.5}$. This factor appears to have an entrance point from the kitchen, as it is the only factor of the two for which the correlations increase when the contributions of the fourth factor in the other rooms are time shifted ten minutes later (0.74 with the B4 and 0.45 with O4). This does not exclude the possibility of them being associated with specific meteorological conditions (mainly wind direction) which enhances their presence specifically from the kitchen as a point of entrance. Similarly, looking at the diurnal contributions of each factor (for the indoor sensors), apart from them being associated mainly with SE wind directions, a difference is also observed in their peak times, with the first one presenting a peak during night and early morning hours, while the fourth one peaking mainly during afternoon and evening times, associating them with different outdoor activities and/or sources (figure S9). The variation of the factors between the two periods is also observed in the measured PM values as well, with ABS1 presenting higher PM_1/PM_{10} ratios compared to ABS2. This PM ratio variability is similar to that found for the outside measurements as well, showing the direct effect of the outdoor environment on indoors. The difference between the two periods is probably due to the different meteorological conditions found during them, mainly the wind conditions (figure S2), which point to different sources of particles affecting the measuring site.

The other 3 factors are all associated with the activities of the occupants in the house. They present distinct particle profiles with a peak on the smaller size measured for one of them and peaks on coarser sizes for all (figure S6). Great similarity is found in the diurnal variation of these factors between the rooms as well, with a notable peak on the indoor ones one hour earlier in the morning at the bedroom, showing the effect of the family getting out of bed before doing activities in the other rooms studied (figure S7). The measurement limit of the sensors, can be a limiting factor in fully capturing the effect of the activities associated with combustion sources from cooking (frying, grilling etc.), which typically result in ultrafine particles. Thus, on every occasion when cooking is mentioned in the diary, almost all these factors present an increase in their values pointing in the presence of people in the specific room and the activities that increase mainly the formation and resuspension of larger particles rather than the activity itself, as no notable differences were observed with different types of cooking. While this may be surprising, as previous studies have detected the effect of cooking even at these size ranges (Farmer et al., 2019), in the present study the focus was the general conditions in the house rather than the effect of each specific activity. Thus, the sensor in the kitchen was put approximately 2 m away from the cooking appliances and an extractor (or an open window) was used in every cooking session, which are probably the reasons for this discrepancy. Similarly, the indoor factors in the bedroom all present an increase during bedtime and waking hours. Within the same rooms these indoor factors are in most cases well associated with each other ($r > 0.60$), which suggests they are associated with similar activities that take place in each room (table S5). These correlations are reduced when compared between rooms, indicating PM moving between rooms, though without a clear picture of a specific source room.

3.4.3. PM contribution of the factors

Using the PM concentrations estimated by the factors, their nature can be further explained and their effect can be quantified between the rooms. Specifically, we can see that the first and fourth factors have the greatest contribution to the smaller PM, being almost the sole contributors for PM_1 . A small contribution of the third factor is also observed throughout the house, though it is only present in the bedroom during the night-time (Fig. 6). As the size range increases the effect of the indoor factors increase compared to outdoor factors. It should be noted that due to a reduced effect from ventilation and increased effect from

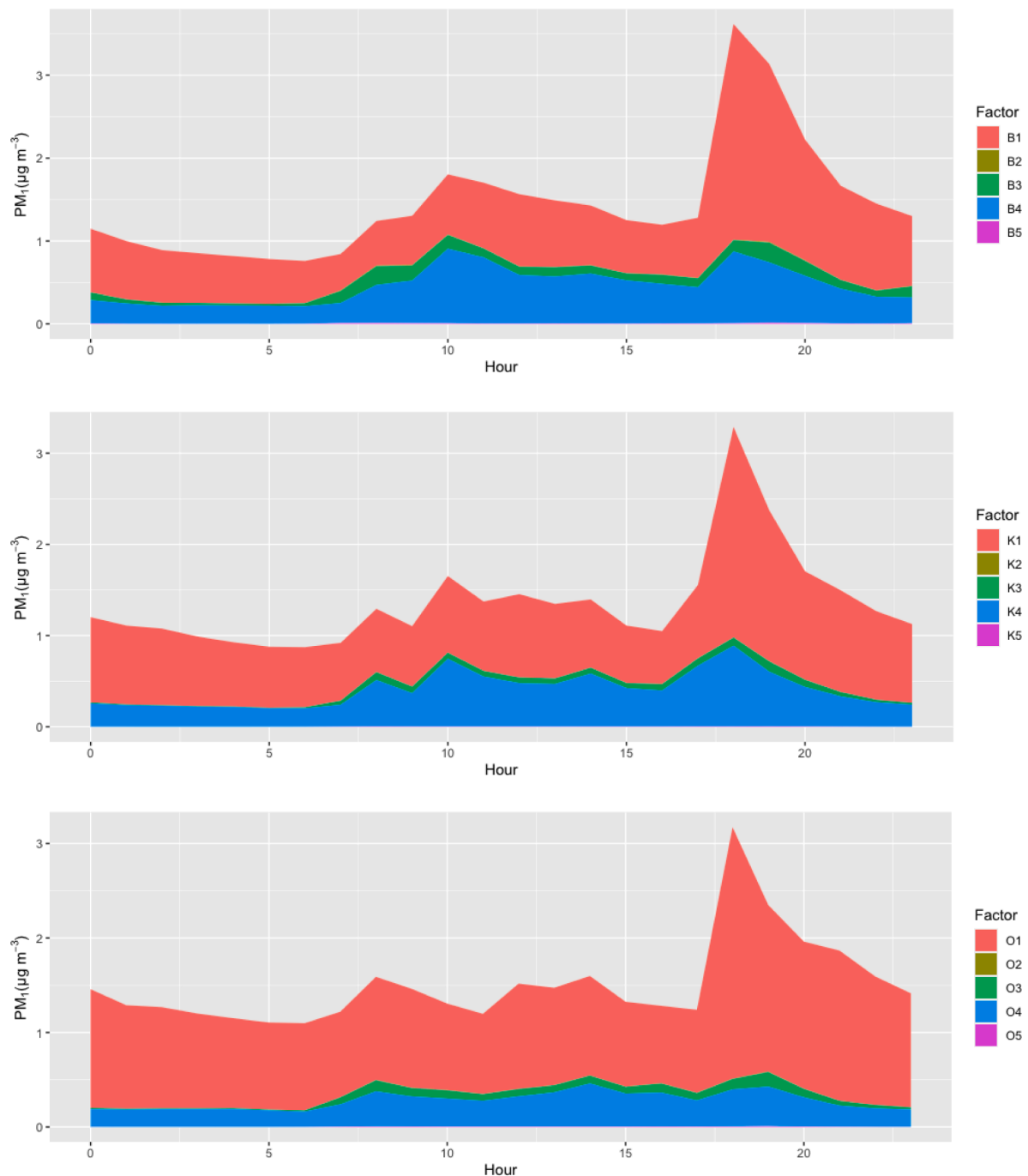


Fig. 6. Average diurnal variation of the factors affecting the PM₁, PM_{2.5} and PM₁₀ at the rooms of the study during the periods when the family was present in the house. The values derive from the common factors between the rooms and their separate contribution for each room (B1-B5 for the bedroom, K1-K5 for the kitchen and O1-O5 for the office).

the activity in the bedroom, the outdoor factors have a smaller relative contributions than in the other rooms. Furthermore, the third and fifth factors present an increase in their contribution approaching bedtime, especially for the larger PM sizes, which probably indicates their greater association with activities specifically in this room. A peak on the fifth factor is also observed in all rooms during the waking up hours, pointing to particles associated either with activities during that time throughout the house or with particles formed in the bedroom and then spread throughout the house by the occupants' movement. The association of the fifth factor with bed-related emissions was also confirmed on the 29/1/2022, when the children in the house were asked to bounce on the bed

for about 5 min at about 15:00 (figure S10). While all indoor factors are increased from this activity, a sharper increase of the fifth factor is observed from the bedroom sensor along with an increase on PM_{2.5} and PM₁₀. A smaller increase is also observed about 10–20 min later in the other rooms as well, though this is greatly reduced compared to that in the bedroom. A sharp increase especially of the PM₁ later in the same day was probably correctly attributed to outdoor factors as it should be associated with the effect of the storm "Malik", since no other significant activity was done at that point according to the diary.

The effect of the indoor and outdoor factors while the occupants of the house were present is found in Table 2. During that period the effect

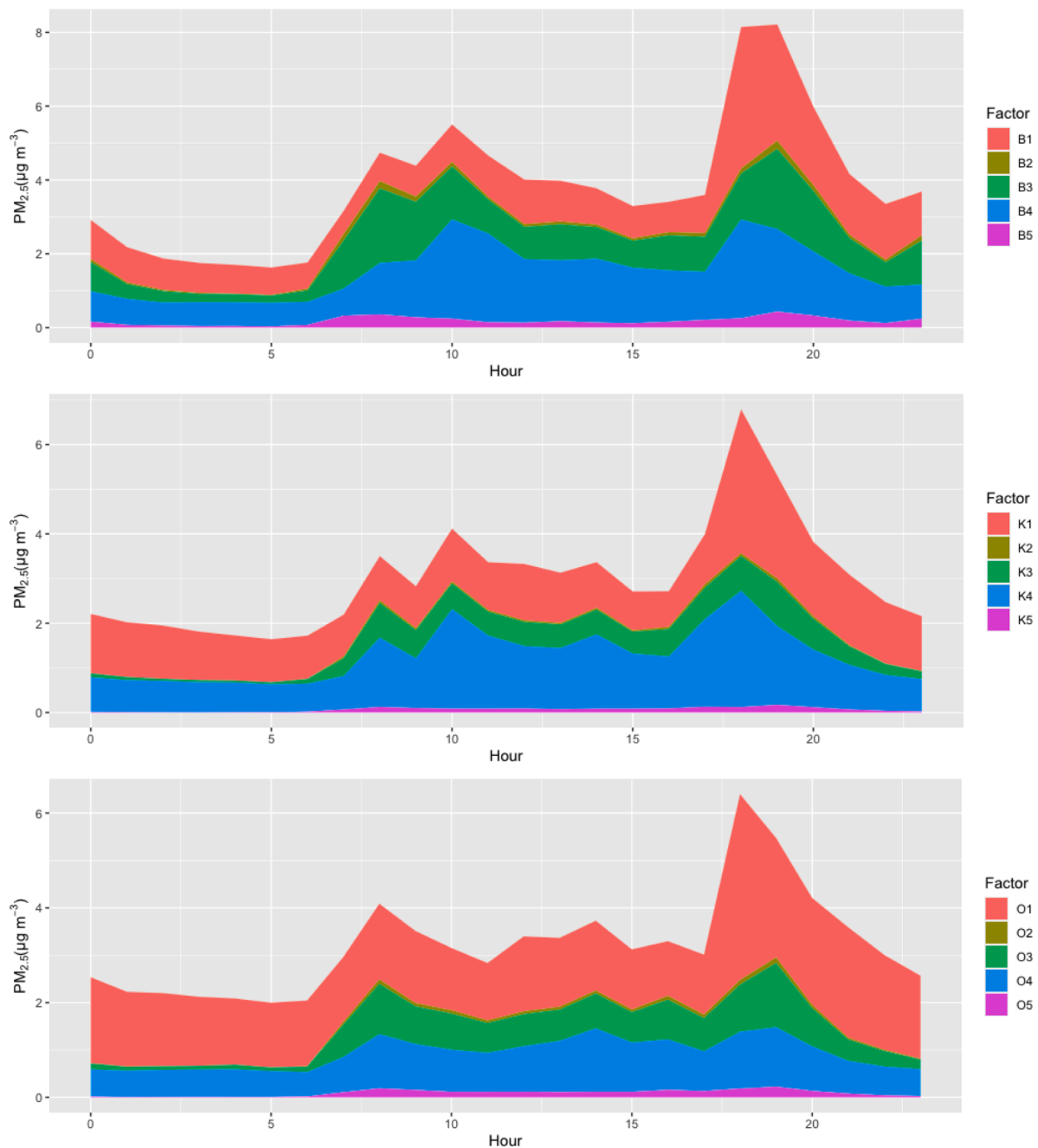


Fig. 6. (continued).

of the indoor factors was minimal on the PM_{10} concentrations in all rooms, being lower than 5 % for all rooms (apart from the bedroom being at about 8 %). As the size increases this effect is also increased being about 20 % for the $PM_{2.5}$ (up to 33 % in the bedroom) and >50 % for the PM_{10} in all the rooms, despite the presence of a single trickle vent in that room as well. The nature of the furniture and activities that take place in the bedroom are the most probable reasons for the greatest effect of the indoor factors in the bedroom, reaching up to 69 % of the PM_{10} in that room. Contrarily, in the kitchen this effect is much lower, which is due to the reduced effect of the indoor activities in that room, as the effect of the outdoor factors on the PM concentrations is almost similar as that found in the other rooms.

A notable feature successfully captured by the model is the different settling periods for the different size ranges. As expected, smaller PM presents milder fluctuations due their slower settling times. This can be

an interesting consideration on health effect studies as regardless of their sources, PM_{10} (even though still not regulated) tend to stay airborne for longer periods, thus affecting the air quality for longer periods especially in indoor environments.

3.5. Average day exposure, peaks, and health effect

As there is an increasing tendency of people working from home, especially after the COVID lockdowns, indoor air quality and exposure becomes a more important factor on the overall health effects from PM exposure. The exposure of an average work-at-home day is estimated here, with a schedule of 7 h of working, 4 h of preparing and having meals and 13 h of sleeping and personal time, which is assumed to be mostly spent in the bedroom. The daily schedule considered here is presented in the table S6. The average daily exposure for each PM size

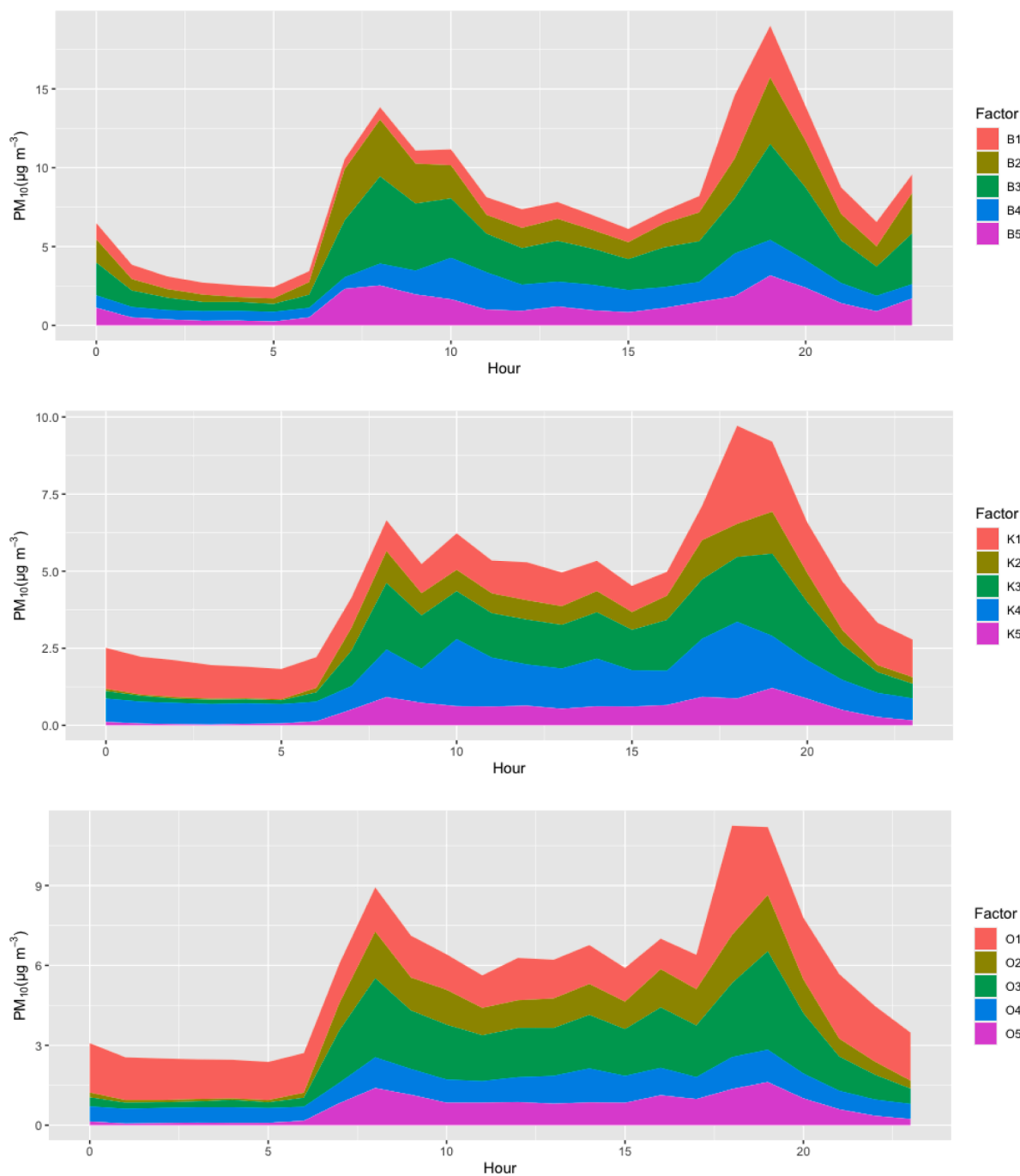


Fig. 6. (continued).

Table 2

Estimated percentage of the effect of internal and external factors on PM (Internal are F2, F3 and F5 and external are F1 and F4) for the periods when the occupants were present.

	PM ₁	PM _{2.5}	PM ₁₀
Bedroom Internal	8.1 %	32.1 %	68.5 %
Bedroom External	91.9 %	67.9 %	31.5 %
Kitchen Internal	3.8 %	17.4 %	47.2 %
Kitchen External	96.2 %	82.6 %	52.8 %
Office Internal	4.4 %	21.3 %	54.4 %
Office External	95.6 %	78.7 %	45.6 %

range is presented in the Table 3, along with the relative contribution of the internal and external sources as calculated by the PMF analysis for the period when the family was present. While the average PM concentrations appear to be relatively low, it should be considered that these are daily averages, in which 7 h of sleeping are also included. Furthermore, while the kitchen was the room with the lowest average PM₁₀ values, during the campaign, peaks well above 50 µg m⁻³ were observed (though these were rather rare) in some cooking sessions. These were associated mainly with outdoor sources, probably resulting from a combination of increased outdoor PM₁₀ concentrations and room ventilation (either by the open windows or the cooker extraction hood), further showing the detrimental effect of the outdoor environment. These were also complemented by PM from the activities of the occupants which further increased the PM concentrations.

Table 3
Work-at-home average exposure for PM1, PM2.5 and PM10 and estimated internal and external contribution.

PM ₁			PM _{2.5}			PM ₁₀		
Internal	External	Average (µg m ⁻³)	Internal	External	Average (µg m ⁻³)	Internal	External	Average (µg m ⁻³)
6.3 %	93.7 %	1.4 ± 0.6	27.5 %	72.5 %	3.3 ± 1.3	63.6 %	36.4 %	6.5 ± 2.8

Looking at the PM diurnal variation, we can see that for the smaller particle size ranges the biggest contributor remain the outdoor sources (in most cases the fourth factor), though their effect is rather significant for the PM₁₀ as well (Fig. 7). As the period studied is in the winter, the effect of the outdoor sources is expected to be smaller compared to the warmer months, as open windows and increased ventilation should be

more common. However, ambient PM concentrations are expected to be higher during the winter in the UK (Harrison et al., 2012).

The outdoor conditions are clean and the house is equipped with newer and high quality furniture, carpeting and cushions which would result in a limited formation of particles due to abrasion. Considering these along with heating, and cooking appliances and the season of the

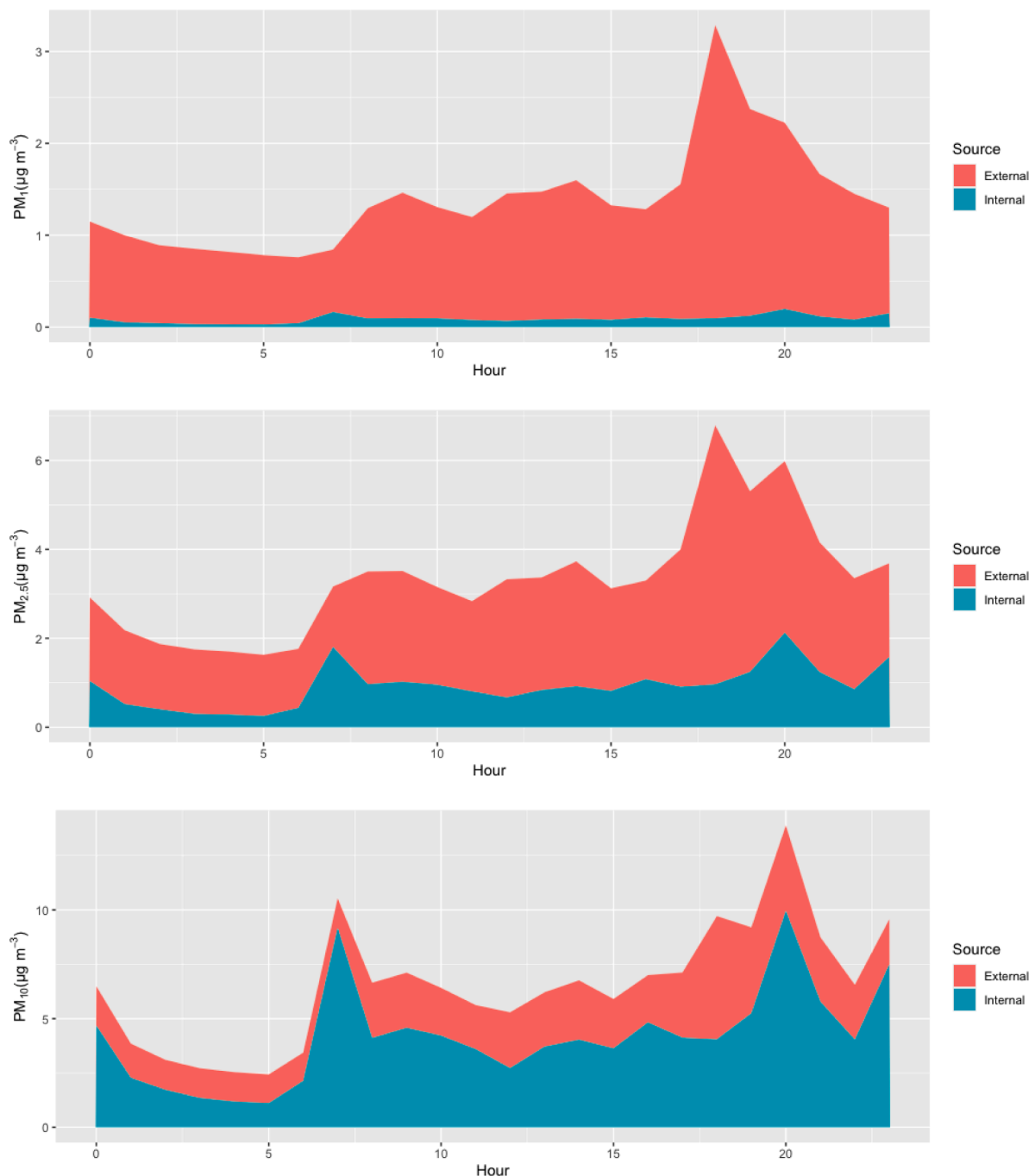


Fig. 7. Average diurnal variation of the sources of the PM on a work-at-home day.

study in the present application, it is expected that the effect of all the factors found would be more significant in other case studies (different seasons, sites etc.). With all these favourable conditions, the indoor $PM_{2.5}$ average concentration observed is more than half the annual mean suggested by the new WHO guidelines and more than a third of those for PM_{10} . A significant proportion of the $PM_{2.5}$ and PM_{10} , and almost all PM_1 , originate from outdoor sources, which makes such studies also important for working places or schools, which are usually located in more polluted areas, making the potential effect of the outdoor sources even more significant. If a similar setup is considered (house and activities) located in an area with an average $PM_{2.5}$ concentration higher than about $12 \mu\text{g m}^{-3}$ and PM_{10} of about $50 \mu\text{g m}^{-3}$, PM concentrations would exceed the new annual guidelines set by the WHO. These are concentrations commonly reached in many urban environments. Similarly, the effect of the indoor particle sources would be increased with more soft furnishings, pollution-generating or less modern appliances, or an increase in activities that affect the particle concentrations. Different heat generation via combustion of solid fuels (wood or coal burner) would also likely introduce a new and significant internal factor.

4. Discussion

There is a rapidly increasing literature on indoor air quality measurements. This study adds to this literature by simultaneously measuring PM in different rooms. Furthermore, and uniquely, it provides a new methodology for estimating the contribution of different sources to the PM concentrations in the different rooms. The indoor air quality literature shows that indoor PM concentrations vary significantly from study to study, due to different indoor conditions and outdoor conditions that impact upon the indoor environment. Hence, it is difficult to use individual studies to make generalizations. For example, Kaliszewski et al., (2020) and Zamora et al., (2020) reported higher indoor PM concentrations in Warsaw, Poland and Baltimore, USA respectively, however, in both cases the houses were within urban areas. Hegde et al., (2020) reported similar concentrations to this study (about $3 \mu\text{g}/\text{m}^3$ in the kitchen and $6 \mu\text{g}/\text{m}^3$ in the bedroom) in a summer campaign in Utah, USA, with higher concentrations observed in the winter period that tracked outdoor concentrations. Similarly, Shen et al. (2021) reported slightly higher $PM_{2.5}$ average concentrations ($5.7 \mu\text{g}/\text{m}^3$) in Beijing during the COVID-19 lockdown period, even though the outdoor concentrations were double the ones found in the present study ($14 \mu\text{g}/\text{m}^3$).

Study limitations include LCS constraints, and lack of information on activities within the household. The lower size limit of the LCS is 350 nm, hence, smaller particles often associated with combustion sources (heating and cooking) are not fully captured. However, the inclusion of smaller particles would not change the results significantly, as smaller particles contribute less to the PM mass concentrations, as well as having less indoor emissions compared to outdoors. The LCS are well equipped to capture the regulatory important $PM_{2.5}$ and PM_{10} size ranges (WHO, 2022). There is an increasing interest in particle number concentrations in relation to public health (Brown et al., 2001; Phalen et al., 2010; Wittmaack, 2007), and the LCS used in this study do not characterise this metric particularly well. The absence of a detailed household activity diary limits the ability to connect indoor factors with specific actions and rooms. Additional sensors measuring temperature and space activity and a diary of the opening of outdoor doors and windows would have been useful in the analysis.

Regardless of the highlighted limitations, this study shows that LCS coupled with relatively little effort by the occupants, can provide detailed information on air quality concentrations and sources. The study was able to measure air quality levels, subset by room, and was further able to separate and quantify the indoor and outdoor sources. It successfully identified the major pollution hot spots within the indoor environment, providing a better understanding of the processes

affecting air quality as well as their magnitude. Such analyses would also be extremely useful in public indoor environments, such as schools, offices, and hospitals. These buildings are often located in more polluted (urban) environments and their air quality impacts significantly on the health of the occupants.

Measurements conducted during the absent period, when the household was empty, showed that household PM concentrations were very homogenous and dependent only upon outdoor sources. During non-absent periods, different indoor sources were identified. Smaller particles tended to spread evenly throughout the house, regardless of the source room, within time periods of 10–30 min. Larger particles tended to be much more room specific with less homogenous mixing throughout the house. Between the rooms, the greatest concentrations for the larger PM were found in the bedroom. This is ascribed to the great number of soft furnishings in this room, including bed covers and carpeting, and the activities that take place in there, showing the importance of the nature and activities that take place in the room studied. For the smaller PM_1 size fraction, the greatest concentrations were found in the office, which has the highest level of ventilation, showing the greater influence of outdoor source in this room. Regardless of the low average concentrations found in the kitchen, it was the room with the highest peaks for all PM sizes, especially during cooking times, making it an important pollution hot spot for short periods of time. As found by the PMF analysis, these PM_1 peaks were mainly accounted for by outdoor particles. Somewhat unexpectedly the actual use of the cooking appliances caused little effect on the PM_1 concentrations, and the variation found associated with cooking activities for the indoor sources may depend on the type of meals cooked.

This study was conducted in a household within a semi-rural environment with low levels of outdoor pollution. The new methodology of source apportionment using LCS data provided allowed for estimation of the PM levels, and was able to separate the effects of outdoor and indoor sources. Even though the measured outdoor pollution was low, it was still possible to clearly identify the effect of the outdoor pollution sources upon the indoor PM concentrations. It was observed that the influence of outdoor PM reduced as the PM size increased. For households situated within more polluted outdoor environments the measured level of ingress would deteriorate indoor air quality more significantly.

5. Conclusions

In this study, four LCS were put in three different rooms and the garden of a typical family house in a semi-rural area to the southwest of the city of Birmingham, UK. In general, the performance of the LCS sensors was good, with the only significant shortcoming being during periods of outdoor mist formation. After calibrating the LCS against two research grade instruments, the LCS performed exceptionally well, especially indoors, showing their potential when properly deployed. The average PM concentrations inside the house were lower than the ones found outside the house by a factor of about 2 depending on the room studied. For the periods when the family was not in the house, these concentrations were significantly lower (being about 10 % of the outdoor guidelines set by the WHO) and the outdoor environment played a crucial role in the indoor environment. Furthermore, the effect of the I/O interactions was found to be decreasing with PM size, dependent on the room studied. The effect of the outdoor sources varied from as high as 95 % for PM_1 down to about 35 % for PM_{10} . The indoor sources are also significant, as when the family was present average $PM_{2.5}$ and PM_{10} concentrations surpassed 50 % and 40 % of the guidelines set by the WHO respectively. Another interesting finding of the study is the variable airborne lifetime of the different sized PM . Smaller particles stay airborne for longer periods and spread among the rooms, making them a greater threat for the health of the occupants, compared to the larger particles which had faster settling times. Finally, the exposure of a work-at-home day was considered. It was found that even in favourable conditions measured, the average $PM_{2.5}$ concentrations were >60 % of

the WHO guideline values. Even a small increase on the outdoor PM_{2.5} and PM₁₀ concentrations would lead to an exceedance of the WHO guidelines.

As technological advances have made it possible for more people to work from home or other indoor working places, the need for assessment of the air quality in such environments becomes crucial. The sensors used in this study, come at a relatively small cost of less than GBP1500, which is considerably less than the cost of the house and similar to other household appliances. This paper clearly highlights the power of source apportionment using LCS data. The approach allows for an understanding of the different indoor and outdoor PM sources that contribute to indoor air pollution concentrations, on a room by room basis. This information will lead to a better understanding of indoor health effects of PM, thereby allowing for informed measures to be taken against processes that increase air pollution within living and working environments. The new approach to elucidate the contributions of different sources to total indoor air pollution exposure, described in this paper, is easily scalable and translatable to different indoor locations.

6. Data accessibility

Data supporting this publication are openly available from the UBIRA eData repository at <https://doi.org/10.25500/edata.bham.00000893>.

Author contributions

The study was conceived and planned by FDP who also contributed to the final manuscript and collected data, and DB who also carried out the analysis and prepared the first draft of the manuscript. LNSA assisted in data collection. RMH and DCSB provided feedback on the analysis. All authors reviewed and edited the final paper.

CRediT authorship contribution statement

Dimitrios Bousiotis: Methodology, Data curation, Formal analysis, Visualization, Writing – original draft. **Leah-Nani S. Alconcel:** Writing – review & editing. **David C.S. Beddows:** Formal analysis, Writing – review & editing. **Roy M. Harrison:** Conceptualization, Writing – review & editing. **Francis D. Pope:** Conceptualization, Funding acquisition, Supervision, Writing – review & editing.

Declaration of Competing Interest

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

Data availability

Data supporting this publication are openly available from the UBIRA eData repository at <https://doi.org/10.25500/edata.bham.00000893>.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2023.107907>.

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