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Utility of outdoor central site monitoring in assessing exposure of school children to ultrafine particles



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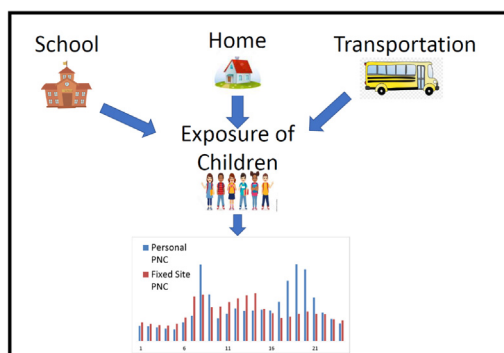
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HIGHLIGHTS

- Particle number concentrations were monitored in three widely separated cities.
- Data from personal sampling and fixed station measurements were compared.
- Agreement was good over long averaging periods such as 24 h.
- Agreement was poor over short periods such as hourly.
- Reasons for the differences are presented and discussed.

GRAPHICAL ABSTRACT



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ABSTRACT

Epidemiological studies investigating the association between daily particle exposure and health effects are frequently based on a single monitoring site located in an urban background. Using a central site in epidemiological time-series studies has been established based on the premises of low spatial variability of particles within the areas of interest and hence the adequacy of the central sites to monitor the exposure. This is true to a large extent in relation to larger particles ($PM_{2.5}$, PM_{10}) that are typically monitored and regulated. However, the distribution of ultrafine particles (UFP), which in cities predominantly originate from traffic, is heterogeneous. With increasing pressure to improve the epidemiology of UFP, an important question to ask is, whether central site monitoring is representative of community exposure to this size fraction of particulate matter; addressing this question is the aim of this paper. To achieve this aim, we measured personal exposure to UFP, expressed as particle number concentration (PNC), using Philips Aerasense Nanotracers (NT) carried by the participants of the study, and condensation particle counters (CPC) or scanning mobility particle sizers (SMPS) at central fixed-site monitoring stations. The measurements were conducted at three locations in Brisbane (Australia), Cassino (Italy) and Accra (Ghana). We then used paired *t*-tests to compare the average personal and average fixed-site PNC measured over the same 24-h, and hourly, periods. We found that,

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at all three locations, the 24-h average fixed-site PNC was no different to the personal PNC, when averaged over the study period and all the participants. However, the corresponding hourly averages were significantly different at certain times of the day. These were generally times spent commuting and during cooking and eating at home. Our analysis of the data obtained in Brisbane, showed that maximum personal exposure occurred in the home microenvironment during morning breakfast and evening dinner time. The main source of PNC for personal exposure was from the home-microenvironment. We conclude that the 24-h average PNC from the central-site can be used to estimate the 24-h average personal exposure for a community. However, the hourly average PNC from the central site cannot consistently be used to estimate hourly average personal exposure, mainly because they are affected by very different sources.

1. Introduction

There is a large body of epidemiologic evidence on the health impacts of ambient particulate matter characterised in terms of mass concentration. Exposure to PM_{2.5} (typically measured as particles smaller than 2.5 µm) is among the top 10 global health risks (Gakidou et al., 2017). Time-series analyses have consistently shown the impact of daily PM_{2.5} concentrations on mortality around the world (Liu et al., 2019).

While it is imperative to control PM_{2.5} concentrations to reduce health impacts, this alone will not eliminate all impacts of ambient particulate matter on health; it will have only a limited impact on the smallest particles, in the ultrafine size range (UFP), below 0.1 µm (de Jesus et al., 2019). UFP are so small that their mass contributes little to PM_{2.5} and they are typically measured in terms of number, rather than mass concentration.

Most UFP originate from sources other than those generating large particles. Combustion and new particle formation processes are the key contributors to ambient UFP levels, while mechanical processes are the main source of larger particles (de Jesus et al., 2019; Morawska et al., 2008). In contrast with large particles, the spatial distribution of UFP is inhomogeneous across cities. Additionally, UFP are much more difficult to measure and characterize than large particles. For these reasons, there is a poor quantitative understanding of the urban physiochemistry of these particles and the health effects they cause. The epidemiology of UFP is much less advanced than that of PM_{2.5}, with contradictory conclusions derived from various studies. This has been noted in the White Paper on “Ambient ultrafine particle evidence for policymakers” (Morawska et al., 2019).

Numerous open questions hinder the advancement of UFP epidemiology, such as the use of central site monitoring versus personal monitoring. Typically, PM_{2.5} concentrations are sufficiently spatially homogeneous within urban scales of interest (Morawska et al., 2008). Most time-series epidemiological studies (short-term studies) on acute effects of PM_{2.5} assume that central site monitoring is representative of the concentrations to which the communities are exposed. Puustinen et al. (2007) showed that using a central site to characterize exposure in epidemiological time series studies does not result in substantial more measurement error for PNC than for PM_{2.5}. Cyrus et al. (2008) observed high correlation of PNC between four urban background sites in Augsburg, implying that temporal variations in PNC are similar for the whole city area and that it could be reflected by one central monitoring site. It suggests that using one carefully chosen monitoring site might be a proper approach to characterize adequately temporal variation of exposure to UFP in short-term epidemiological studies (for example to characterize the day to day variation in the entire city, which is mostly driven by varying emissions levels and meteorological conditions), which is in line with conclusions of (Puustinen et al., 2007).

As far as long-term studies are concerned, it is not the temporal variation but the spatial variation that is important. While the temporal variation of UFP is quite strongly correlated between different monitoring sites, UFP concentrations vary significantly in space, often by as much as one or two orders of magnitude within distances of just tens of meters (Johansson et al., 2007; Kumar et al., 2014; Morawska et al., 2008; Weber, 2009). The larger spatial variation of the absolute concentration level of PNC compared to PM_{2.5} across a city suggests that it is virtually impossible to characterize the average spatial distribution of PNC using one monitoring

site. This implies that epidemiological long-term studies assessing health effects related to long-term average exposure between cities or within one city, should not rely on a single central site. Cyrus et al. (2008) showed that the PNC measured at different background locations in Augsburg, Germany, were not homogeneously distributed over the study area. This indicated that one fixed measurement site was not a good approximation for spatial distribution of PNC over a wide urban area. The authors concluded that for long-term epidemiological studies other approaches should be considered to properly estimate of the UFP exposure in space. For example, one might include spatial variability modeling of PNC and/or increase the number of monitoring sites.

Personal exposure monitoring is always more demanding than central site monitoring, requiring a substantial number of volunteer participants and, in the case of UFP, much more expensive and complex instruments than those required for PM_{2.5} personal monitoring. This raises the question: is it necessary to conduct assessment of personal exposure to UFP as the only means to obtain information on community exposure to UFP, or is central site monitoring adequate? (Deffner et al., 2016). Several recent studies have investigated personal exposure monitoring and its outcomes for school-aged children. The studies were conducted in Brisbane, Australia (Mazaheri et al., 2014), Cassino, Italy (Buonanno et al., 2012) and Accra, Ghana (Nyarku et al., 2019). In all cases, the research teams also simultaneously measured UFP concentrations and in some locations also size distribution at central monitoring sites, usually for other purposes related to the studies. Comparison of UFP concentration data from central sites and personal exposure monitoring have also been carried out by Gu et al. (2015) and Peters et al. (2015). The results of these studies provide the opportunity to address the question: are the central site monitoring data representative of personal exposure, and thus community exposure to UFP? The specific objective of the present study was to investigate this relationship in more detail by 1) comparing 24-h, and 1-h, UFP exposure in children based on personal exposure and central site monitoring data; and 2) analysing the factors driving the variation of the outcomes between these two methods of assessing exposure.

2. Materials and methods

2.1. Study design

Two types of UFP data were used for this study: obtained based on personal exposure measurements and on measurements at a fixed monitoring site. The data were collected from three different cities, Brisbane (Australia), Cassino (Italy) and Accra (Ghana), as shown in Fig. 1.

Personal exposure data in Brisbane were collected during the project ‘Ultrafine Particles from Traffic Emissions and Children’s Health – UPTECH’ (Mazaheri et al., 2014). The study was carried out between November 2010 and August 2012, and data were collected from 25 schools in the Brisbane Metropolitan school district, over a period of two weeks at each school. Students enrolled in these schools typically lived within the school catchment area, which was defined by equidistant trafficable routes between one school and its neighbouring schools. A total of 137 school children with ages ranging from 8 to 11 years participated in the study. Information about the characteristics of the schools and emission sources in their vicinity is provided by Clifford et al. (2018), Crilley et al. (2016), Laiman et al. (2014), Mazaheri et al. (2014), and Mazaheri et al. (2016).

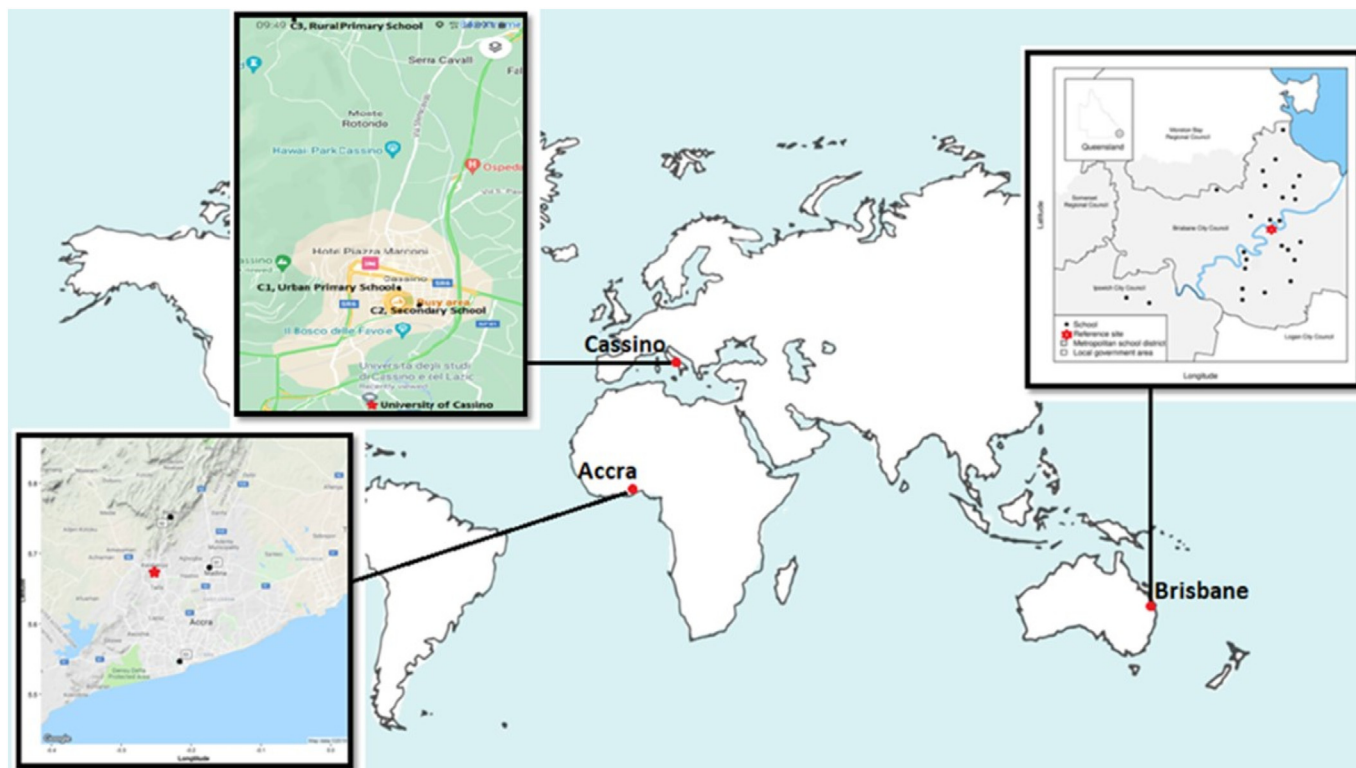


Fig. 1. Map showing the location of the three cities. The black dots indicate the locations of the schools and the red stars show the locations of the fixed monitoring stations.

The measurements in Cassino were carried out between October 2011 and March 2012 (Buonanno et al., 2012). A total of 103 children aged between 8 and 11 years from three schools (C₁, C₂ and C₃) participated in the study. C₁ was a primary school located near an urban street with traffic consisting mostly of light diesel vehicles. The peak traffic hours were between 8.30 am and 1.30 pm. C₂ was a secondary school located near the intersection of an urban street with moderate to heavy traffic and peak traffic hours similar to C₁. C₃ was a primary school located in a rural area with very low traffic density.

In Accra, personal exposure measurement data were collected during weekdays over ten weeks between October and December 2017 (Nyarku et al., 2019). A total of 61 school children with ages ranging from 11 to 16 years participated in the study. They were drawn from three junior high schools (A₁, A₂ and A₃) located in suburbs in and around Accra. A₁ was located 17 km away from Accra in Madina near a heavily trafficked Accra-Aburi highway, A₂, 23 km away from Accra in Berekuso, which is a rural community and A₃, in Ayalolo, in the heart of the city of Accra.

The personal exposure data collected in Brisbane were categorized into the same types of microenvironments for analysis as discussed by Mazaheri et al. (2014). A microenvironment analysis was previously conducted for the children in Cassino (Buonanno et al., 2012) for their study on individual dose and exposure of Italian children to ultrafine particles. The personal exposure of the children is the amount of time spent and the weighted average of concentrations they were present in each microenvironment and is drawn upon but not repeated here. For Accra, it was not possible to conduct a microenvironment analysis because the entries in the activity diaries were incomplete (Nyarku et al., 2019).

2.2. Personal monitoring

In each of the cities, the personal exposure of the children to PNC was measured for 24 h using a portable Philips Aerasense Nanotracer (NT), which measures PNC up to $1 \times 10^6 \text{ cm}^{-3}$ in the particle size range of 10 to 300 nm. Before the start of the experimental campaign, the NT was calibrated and data quality assurance was done by comparing the data

against a Condensation Particle Counter (CPC); TSI Model 3787 for Brisbane, 3775 for Cassino, and 3025A for Accra. Owing to the nature of the project in widely separated locations, all three CPCs could not be calibrated at the same location nor compared against each other. However, the Brisbane CPC (3787) and the Accra CPC (3025A) were compared against each other in Brisbane and found to be consistent. The Cassino CPC (3775) was calibrated in the standard manner using a TSI 3068B aerosol electrometer, so may be assumed to be accurate. Further details of calibration of NTs have been provided by Mazaheri et al. (2014) for Brisbane, Buonanno et al. (2012) for Cassino, and Nyarku et al. (2019) for Accra. In all three studies, the NTs were operated in the 'advanced mode' at a sampling frequency of 16 s. The participating school children in all the cities were also provided with an activity diary in which they recorded their daily activities and the times spent in different microenvironments.

2.3. Fixed-site monitoring

The fixed-site monitoring in Brisbane was carried out through the window of a sixth-floor room, approximately 18 m above the ground, at the Gardens Point Campus of the Queensland University of Technology (QUT). The site is located at the south-western end of the central business district (CBD) approximately 100 m east of a busy freeway. PNC was monitored and recorded continuously at intervals of 30 s using a TSI 3787 water-based CPC. The data was later averaged to one hour for the purpose of comparison between personal and stationary monitoring.

The fixed-site monitoring in Cassino was carried out near the Faculty of Engineering of the University of Cassino, located in the central park of the town and away from roads with high traffic volumes. The PNC was continuously measured at a frequency of 30 s using a CPC TSI 3775, calibrated against a TSI 3068B aerosol electrometer in the European Accredited Laboratory at the University of Cassino.

In Accra, the stationary site was located on the grounds of the Ghana Atomic Energy Commission (GAEC) in Kwabenya. The site is surrounded by the following communities: Ashongman Estate in the north, Atomic Down in the west, Dome in the southwest, Dome-Pillar-2 in the south,

Haatso in the east, and Narma in the northeast. Residents in these communities engage in environmental practices such as trash burning and the use of biomass fuels for cooking along the street by food vendors. UFP concentrations were measured using an NT in the advanced mode, so particle number concentration was measured, with a sampling frequency of 16 s. Prior to using the NT, it was calibrated against a CPC using both laboratory-generated aerosols and urban outdoor air.

3. Data analysis

The analysis presented in this paper is based on the data from the participating children for whom at least 23 h of data were available: 84 (out of 137) children in Brisbane, 20 (out of 103) children in Cassino, and 13 (out of 61) children in Accra. Some children did not meet this requirement due to, for example, misplacing their activity diary or they did not charge the NT as per the study protocol.

3.1. Analysis of 24-hour average PNC

To compare personal and stationary PNC, the personal PNC values for each child were matched with the stationary PNC values corresponding to the same 24-h period and city. The 24-h average PNC value was determined using Eq. (1) for personal measurements and Eq. (2) for stationary measurements.

$$PNC_{\text{personal}} = \frac{1}{n_p} \sum_{i=1:n_p} PNC_i \quad (1)$$

$$PNC_{\text{stationary}} = \frac{1}{n_s} \sum_{j=1:n} PNC_j \quad (2)$$

PNC_i corresponds to the i -th personal PNC measurement and n_p is the number of personal measurements, over the 24 h. PNC_j and n_s correspond to the stationary PNC measurements. The number of measurements over the 24 h is $n_p = 5400$ and $n_s = 2880$ for data recorded every 16 and 30 s, respectively.

The distributions of 24-h average PNC values for personal and stationary measurements were visually summarised and compared using interval plots, which depict the means and the spreads of the data; the mean values and standard deviations were also calculated and compared. Additionally, the personal and stationary PNC values were compared by calculating the percentage difference between the overall average personal PNC and stationary PNC, using Eq. (3):

$$\frac{\text{Average}(PNC_{\text{personal}}) - \text{Average}(PNC_{\text{stationary}})}{\text{Average}(PNC_{\text{stationary}})} \times 100\%. \quad (3)$$

A paired t -test was used to assess if the 24-h average personal PNC and 24-h average stationary PNC differ, at a 5 % significance level. Scatter plots of the 24-h average personal PNCs and 24-h average stationary PNCs for each city were also produced to inspect how well these two values correlate.

3.2. Analysis of hourly average PNC

Eq. (1) and Eq. (2) were also used to calculate hourly average personal and stationary PNC values, respectively. The number of measurements in the hour is $n_p = 225$ and $n_s = 120$ for data recorded every 16 and 30 s, respectively.

For each hour of the day, the hourly average personal PNC values and the hourly average stationary PNC values were compared using a paired t -test to determine whether the differences were statistically significant, at a 5 % significance level. For Brisbane, the daily activities of the children were categorized into different microenvironments based on the information recorded in the activity diary by the children. Based on (Mazaheri et al., 2014), the microenvironments were commuting, home-eating-cooking, home-indoors, home-sleeping, other-indoors, other-outdoors,

school-indoors and school-outdoors. For those hours of the day where the differences between personal and stationary PNC values were statistically significant, further analysis of the data was performed to identify the possible drivers. This was done by investigating the microenvironments that contributed the most to the differences in PNC values.

4. Results

4.1. Comparison of 24-hour average PNC

Fig. 2 shows, for each city, the interval plots of the 24-h average stationary and personal PNC values, with the values themselves presented in Table 1S (Supplementary Information). We see that, essentially, Brisbane is a much ‘cleaner’ city than either Accra or Cassino. The ambient PNC concentration in Brisbane is in the range 5000–10,000 cm^{-3} , whereas in the other two cities, it is in the range 40,000–50,000 cm^{-3} . Furthermore, the indoor exposure of children in Australia is relatively lower because cooking is not the main activity for the family due to the reliance on take-away and ready-made foods which are readily available in the supermarkets. Smoking and burning of mosquito coils in the home-indoor microenvironment also contributes to the total PNC and level of exposure of the children. These are activities that are not very prevalent in Australia. The commuting times of the students were also relatively short as most of the students study in schools close to where they live. Based on visual observation (Fig. 2), for all cities, the means of the 24-h average personal PNC values are marginally higher than the means of the 24-h average stationary PNC values, but these differences are not large enough to be of any practical importance. Further, the statistical findings from the paired t -tests concluded that the mean differences between 24-h average stationary and personal PNC were not statistically significant for Brisbane ($p = 0.11$), Cassino ($p = 0.65$) and Accra ($p = 0.74$).

Scatter plots of 24-h average stationary PNC values versus 24-h average personal PNC of all children in Brisbane, Cassino and Accra are presented, respectively, in Fig. 3S (a)-(c) (Supplementary Information). All plots exhibit a moderate to weak correlation between personal and stationary 24-h average PNC ($r = 0.29, 0.29, \text{ and } -0.46$, for Brisbane, Cassino and Accra respectively).

4.2. Comparison of hourly average PNC

Fig. 3(a) presents the differences between the average hourly personal PNC and the average hourly stationary PNC for children in Brisbane.

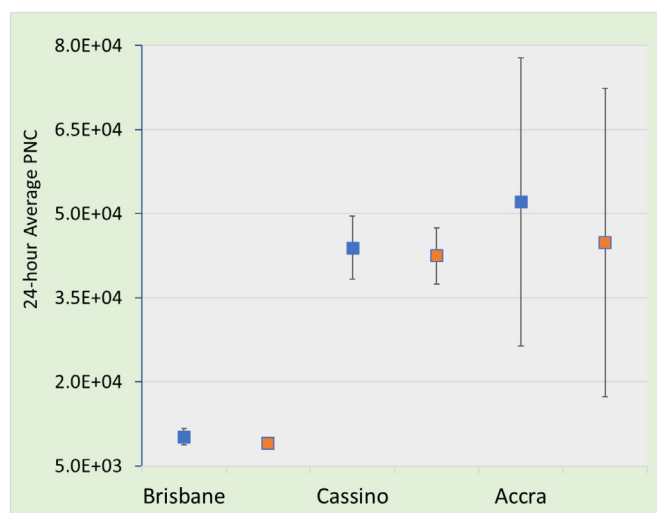


Fig. 2. 24-h average personal and stationary PNC for children in Brisbane, Cassino and Accra. Blue squares are the 24-h average personal PNC, averaged over all the children in the relevant city. Orange squares are the overall 24-h average stationary PNC. The error bars represent 95 % confidence intervals of the mean, i.e., two standard errors either side of the mean.

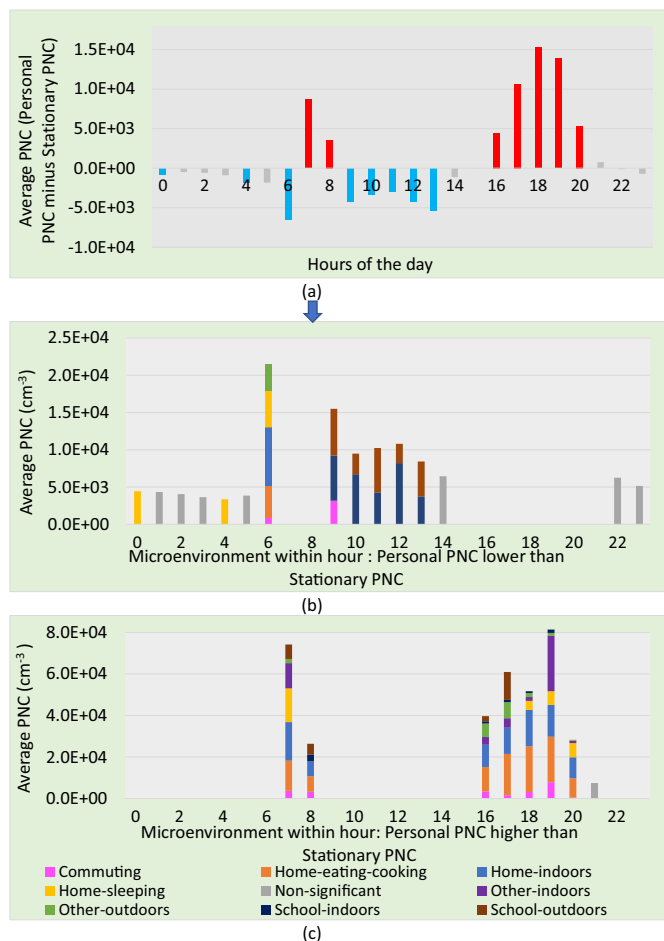


Fig. 3. Comparison of hourly average PNC for children in Brisbane. Fig. 3(a) shows the differences between hourly average personal PNC and hourly average stationary PNC for each hour of the day. Blue bars correspond to the hours in the day where the average personal PNC was significantly lower than the average stationary PNC. Red bars are the hours where the average personal PNC was significantly higher than the average stationary PNC. Grey bars are those hours of the day where the difference between the average personal PNC and the stationary PNC are not statistically significant. Fig. 3(b) and (c) shows the hourly contribution of each microenvironment for hours where personal PNC was significantly (b) lower and (c) higher than stationary PNC.

Fig. 3(b) shows the contribution of each microenvironment to the hourly personal PNC, for the hours where personal PNC was lower than stationary PNC, while Fig. 3(c), for the hours when personal PNC was higher than stationary PNC. Each stacked bar in the charts indicates the average exposure of all the children for the actual time spent in each of the microenvironments within that hour. Fig. 4 presents the differences between the average hourly personal PNC and the average hourly stationary PNC, for children in Cassino. The mean and standard deviations of hourly average personal and stationary PNC are provided in Tables 2S (a) and (b). The absolute difference (personal minus stationary), the percentage difference using Eq. (3), and the significance of the difference based on the paired *t*-tests, are provided in Table 2S(a) for Brisbane and Table 2S(b) for Cassino. Table 3S and 4S provide the values for the microenvironments corresponding to the hours when personal PNC was lower and higher than stationary PNC, respectively.

In Brisbane in the hours commencing 12 pm, 4 am, 6 am, and 9 am–1 pm, the average personal PNC was lower than the average stationary PNC; in the hours commencing 7 am–8 am, and 4 pm–8 pm, personal PNC was higher than stationary PNC. In Cassino in the hours commencing 6 am–10 am (except 7 am), and 10 pm–11 pm, the average personal PNC was significantly lower than the average stationary PNC; in the hours

commencing 2 am, and 1 pm–2 pm, personal PNC was higher than stationary PNC. The hourly average data for children in Accra could not be obtained due to some inconsistencies in reporting of the measurement times of the children as discussed in Section 2.1 Study design.

For all three locations, there were no statistically significant differences between the 24-h average personal and fixed-site PNCs. Next, a pairwise comparison between hourly average personal PNC and hourly average stationary PNC for each hour of the day was carried out for Brisbane and Cassino to identify possible drivers for the differences between these two types of assessment. There were significant differences between hourly average personal and central-site PNCs at different times of the day. During some hours of the day, personal PNC hourly averages were significantly higher than the central-site PNC values, while at other hours they were significantly lower. For Cassino, the hours where personal PNC hourly averages was higher for the children occurred at the times when children were in transportation and when they were exposed to emissions from cooking/eating activities (Buonanno et al., 2013a, b). For the children in Brisbane, higher personal exposure occurred mainly in the home-eating-cooking microenvironment. The comparison of hourly average PNC for children in Brisbane between different microenvironments demonstrates that the highest average was in the home-indoors microenvironment with the mean value of $1.06 \times 10^4 \text{ cm}^{-3}$ and the lowest in home-sleeping with the mean value of $3.39 \times 10^3 \text{ cm}^{-3}$. In the case of Cassino, personal PNC was significantly higher in the hours commencing at 1 pm and 2 pm mainly due to the generation of indoor UFP from cooking activities during lunchtime (Buonanno et al., 2012). The cooking activity increases the UFP level and exposure of the children during the eating time and the time spent at home-microenvironment. The level of exposure was generally higher in the indoor microenvironment as opposed to outdoor as children spent a longer period of time indoors both at home and school. Students in Brisbane typically spend around 6 h in the school (9.00 am – 3.00 pm) and their exposure at school can be attributed to the emission from traffic as there were no indoor sources of particle emission.

5. Discussion and conclusion

The majority of epidemiological time-series (short term) studies assess the acute health effects of ambient pollutants in which the measurement of ambient PM at a fixed monitoring site is the method of choice. A total personal exposure to PM (T) has an ambient exposure component (A) resulting from exposure to ambient PM while outdoors, and to ambient PM that has infiltrated indoors while the person is indoors, plus a non-ambient exposure component (N) resulting from exposure to indoor generated PM while in various indoor environments, so that $T = A + N$. If we are interested in the effects of ambient PM, the ambient concentrations measured at the central monitoring site (C) could be a better indicator for the ambient exposure component (A) than the total personal exposures (T). The correlation that interests many epidemiologists is not that between total personal exposure and outdoor concentrations, but the correlation between that component of personal exposure due to outdoor particles and the outdoor concentrations (Wallace and Williams, 2005). Ebelt et al. (2005) as well Wilson and Brauer (2006) demonstrated the usefulness of separating total exposure into its ambient and nonambient components in epidemiological studies of acute health effects (short term studies).

Epidemiological time-series studies and risk assessment of the impacts of ambient air pollution (PM_{2.5} and PM₁₀) on health use data from the closest central-site monitoring stations as representative of exposure to the community. Since spatial distribution of PM_{2.5} and PM₁₀ in typical urban environments is reasonably homogenous, this approach is justifiable. There are still potential differences between personal exposures compared to those assessed based on data from central ambient monitoring stations due to indoor sources of PM_{2.5} and PM₁₀, which can lead to elevated concentrations of these particle fractions. For schools it was shown that dust brought inside by children contributed to elevated PM₁₀ and PM_{2.5} (Amato et al., 2014; Fromme et al., 2008; Stranger et al., 2008), while for homes, indoor combustion sources contributed to PNC (Morawska et al., 2017).

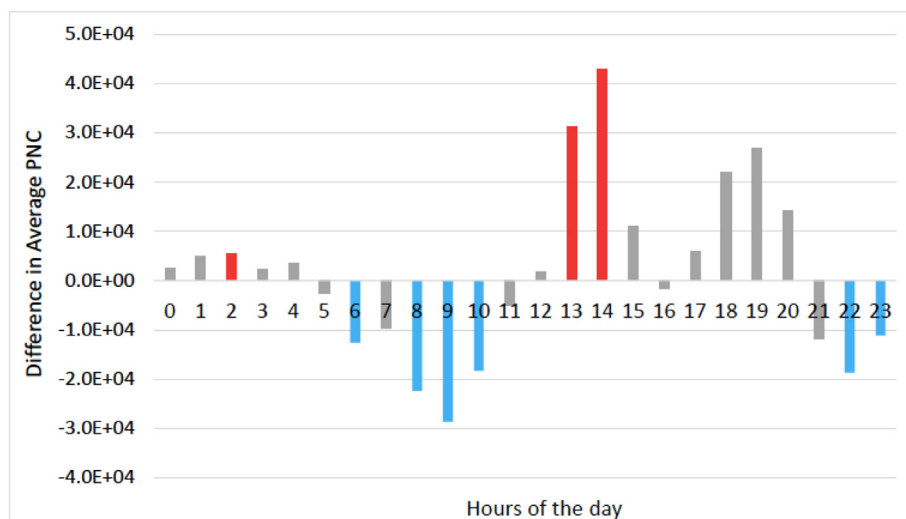


Fig. 4. Difference between hourly average personal and stationary PNC for each hour of the day for children in Cassino. The colours of the bars are the same as in Fig. 3(a).

Assessment of exposure to UFP is significantly more complicated, because concentrations of these particles display large spatial variations within urban environment (Buonanno et al., 2013a, b; Buonanno et al., 2011; de Jesus et al., 2019; Hofman et al., 2016; Kumar et al., 2014; Morawska et al., 2009a, b; Morawska et al., 2008; Saha et al., 2019), therefore, even in the absence of contribution from indoor sources it is possible that central monitoring stations do not provide data representative of community exposure. For this reason, it could be questioned if central site UFP monitoring data can be used for exposure assessment and in turn for epidemiologic studies. Contribution from indoor sources, particularly any type of indoor combustion further contributes to the variation between personal and central site ambient air monitoring station. UFP concentrations during cooking for example, could be orders of magnitude higher than the indoor background (Bhangar et al., 2011; Buonanno et al., 2012; Buonanno et al., 2009; Jeong et al., 2019; Morawska et al., 2003; Mullen et al., 2011; Wallace and Ott, 2011).

The obtained values are comparable to the results from a handful of studies investigating the relationship between measured personal exposure to $PM_{2.5}$, and central site monitoring (Borgini et al., 2011; Kim et al., 2006). Other studies reported varying relationships depending on the type of the environment (Nerriere et al., 2005) with low correlations between personal exposure and background concentrations (Kaur et al., 2007). However, the study carried out by Braniš and Kolomazníková (2010) found a strong relationship between the 24-h average $PM_{2.5}$ at a central site and the personal exposure of one person monitored for a year. In the present study, the variation in the average 24-h personal exposure of the participants was due to proximity to local sources and the variation in ambient background concentrations. For example, the high personal exposure values above $20,000 \text{ cm}^{-3}$ as seen in Fig. 3S(a) for Brisbane), and values above $80,000 \text{ cm}^{-3}$ in Fig. 2S (c) for Accra, can be attributed to the exposure of the children during home-eating-cooking and commuting microenvironments.

In previously reported studies, the personal exposure of school children to ultrafine particles varied according to the type of microenvironment (Ryan et al., 2015) and the time spent in different microenvironments. For example, compared to the home environment, riding in a car and walking demonstrated 1.36 and 2.51 times higher UFP concentrations, respectively, at a CI of 95 %. Many studies of personal exposure to PNC in the home microenvironment, have shown that the highest exposures were generally due to emissions from cooking (Bordado et al., 2012; Broich et al., 2012; Buonanno et al., 2012; Dennekamp et al., 2002). (Morawska et al., 2003) have shown that the PNC level in a kitchen can be 100 times higher than the background level. Previous studies have also shown that the maximum PNC in a kitchen during cooking was $2.3 \times 10^5 \text{ particles cm}^{-3}$ (Buonanno et al., 2012) $8.17 \times 10^5 \text{ particles cm}^{-3}$ (Morawska et al., 2003).

High concentrations are not only associated with frying or grilling of food, but the type of stove or oven used. Studies carried out by Benka-Coker et al. (2020) and de la Sota et al. (2018) found out that PNC was lower among the households with improved cookstoves than with traditional cookstoves. The increase in indoor personal exposure was also associated with indoor activities like burning of candles (Deffner et al., 2016), mosquito coils (Liu et al., 2003; Mazaheri et al., 2019; Nyarku et al., 2019; Shu-Chen et al., 2008) and incense (Chen et al., 2017; He et al., 2018; Tse et al., 2011). In summary, what we have learnt from all these studies in relation to the research question of this study is that, in general, indoor exposure is greater than outdoor exposure with the main indoor source being cooking emissions.

Previous studies carried out by Morawska et al. (2009a, b) observed an increase in indoor particle concentration during children's painting activities. It was shown that this was due to the formation of secondary organic aerosols. Laiman et al. (2014) identified a number of school activities such as painting and printing in the classroom, grilling in the school tuckshop, which led to a significant elevation in personal exposure. A study carried out in 10 Portuguese primary schools by Rufo et al. (2015), suggested that a range of parameters such as emissions from wood-based furniture and laminated blinds, physical activities and behaviour of occupants, may affect the indoor UFP concentration. Wangchuk et al. (2015) demonstrated that children in Bhutan were exposed to UFP due to the use of biomass burning for cooking, preparation of cattle feed, brewing of local liquor and heating. A study in China by Bai et al. (2018) concluded that mosquito repellent incense was associated with large concentrations of particulate matter and gaseous pollutants which are hazardous to human health, with the greatest exposure observed within the juvenile age group. Children in Accra were exposed mainly to emissions from traffic and cooking. Also, the use of a mosquito coils in homes contributed greatly to increased levels of pollution, especially in poorly ventilated houses (Nyarku et al., 2019). It was found that the type of microenvironment, activities, average time spent, and mode of transport used by children, can result in different level of exposure even though they might be attending the same school. Mazaheri et al. (2019) highlighted children's indoor exposure in Heshan, China was associated with the use of mosquito repellent incense and smoking at home. Morawska et al. (2009a, b) showed that children in urban areas have a higher exposure level than that of rural areas due to the presence of trafficked roads close to urban schools. Previous studies have shown that ambient outdoor PNC closely followed the temporal variation in traffic density with the highest levels observed on weekdays during rush hours (Buonanno et al., 2010; Hussein et al., 2004; Morawska et al., 2008). High exposure levels may also be experienced during commuting times. However, in the present study, this was not a major contributor due to the short commuting time of the participants.

Within the constraints of the project, it was not possible to carry out a seasonal investigation.

Owing to the nature of the study only short-term personal monitoring data campaigns were carried out. Hence the relationship between central monitors and personal exposure could not be assessed for different seasons. However, we suggest that seasonal differences are secondary to the much more obvious differences between personal and stationary sampling.

In summary from this study, no significant differences were seen when the 24-h average personal PNC, averaged over all the students, and the 24-h average stationary PNC, computed over the same time periods, were compared (Fig. 2). The weak correlation in Fig. 1S indicates that the 24-h average personal PNC values were significantly different to the corresponding 24-h average stationary values. As such, we conclude that the 24-h average stationary values cannot be used as a proxy for the 24-h personal PNC which is the parameter of interest with respect to health issues.

However, when the hourly average personal PNC and stationary PNC were compared, significant differences were seen at certain hours of the day. The question is whether the differences are significant in terms of health effects or not. Over the 24 h, whether the effect is cumulative or short exposure to high concentration has a different effect than the cumulative exposure, which is unknown and masked in the 24-h analysis. This potentially has an impact on human health and would require more in-depth epidemiological studies in terms of health or physiological effects. Previous studies have recommended that in the assessment of personal exposure in epidemiological studies, activities in different microenvironment and average time spent should be taken into consideration. The results of this study suggest that epidemiological studies should be based on a much shorter time scale rather than the 24-h period which is yet to be implemented by WHO.

CRedit authorship contribution statement

Basant Pradhan: Methodology, Data curation, Writing- Original draft preparation; **Rohan Jayaratne:** Supervision, Writing- Reviewing and Editing; **Helen Thompson:** Supervision, Formal analysis, Writing- Reviewing and Editing; **Giorgio Buonanno:** Writing- Reviewing and Editing; **Mandana Mazaheri:** Writing- Reviewing and Editing; **Mawutorli Nyarku:** Writing- Reviewing and Editing; **Weiwei Lin:** Writing- Reviewing and Editing; **Marcelo Luiz Pereira:** Writing- Reviewing and Editing; **Josef Cyrus:** Conceptualization, Writing- Reviewing and Editing; **Annette Peters:** Conceptualization, Writing- Reviewing and Editing; **Lidia Morawska:** Supervision, Conceptualization, Project administration, Writing- Reviewing and Editing.

Data availability

Data will be made available on request.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2022.160162>.

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