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Children's personal exposure to air pollution in rural villages in Bhutan

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ABSTRACT

Exposure assessment studies conducted in developing countries have been based on fixed-site monitoring to date. This is a major deficiency, leading to errors in estimating the actual exposures, which are a function of time spent and pollutant concentrations in different microenvironments. This study quantified school children's daily personal exposure to ultrafine particles (UFP) using real-time monitoring, as well as volatile organic compounds (VOCs) and NO_2 using passive sampling in rural Bhutan in order to determine the factors driving the exposures. An activity diary was used to track children's time activity patterns, and difference in mean exposure levels across sex and indoor/outdoor were investigated with ANOVA. 82 children, attending three primary schools participated in this study; S1 and S2 during the wet season and S3 during the dry season. Mean daily UFP exposure (cm^{-3}) was 1.08×10^4 for children attending S1, 9.81×10^3 for S2, and 4.19×10^4 for S3. The mean daily NO_2 exposure ($\mu g\ m^{-3}$) was 4.27 for S1, 3.33 for S2 and 5.38 for S3 children. Likewise, children attending S3 also experienced higher daily exposure to a majority of the VOCs than those attending S1 and S2. Time-series of UFP personal exposures provided detailed information on identifying sources of these particles and quantifying their contributions to the total daily exposures for each microenvironment. The highest UFP exposure resulted from cooking/eating, contributing to 64% of the daily exposure, due to firewood combustion in houses using traditional mud cookstoves. The lowest UFP exposures were during the hours that children spent outdoors at school. The outcomes of this study highlight the significant contributions of lifestyle and socio-economic factors in personal exposures and have applications in environmental risk assessment and household air pollution mitigation in Bhutan.

1. Introduction

Globally, household air pollution alone accounted for 4.3 million deaths in 2012, mostly in low and middle income countries (WHO, 2014). The health impacts of air pollution are driven by personal exposure to pollutants in different locations where people spend their time (Ashmore and Dimitroulopoulou, 2009). A number of methods have been used for monitoring personal exposure, from indirect methods such as fixed outdoor stations and simultaneous indoor–outdoor area measurements to direct monitoring, using personal monitors (Morawska et al., 2013). Indirect exposure monitoring does not generally provide accurate and representative exposure data (Saarela et al., 2003, Kaur et al., 2007, Dionisio et al., 2012, Buonanno et al., 2013), since some pollutants show a high degree of concentration inhomogeneity with respect to the source, both in space and time (Sarnat et al., 2005, Buonanno et al., 2012a, Mazaheri et al., 2013, Hinwood et al., 2014). This is particularly pronounced for ultrafine particles (UFP), which have been shown to vary by orders of magnitude between different indoor and outdoor environments (Buonanno et al., 2011a). Real time personal monitoring, coupled with individual's time activity data provide a more realistic assessment of the exposure risk on cohorts or population groups, as well as the level and frequency of exposure, and importantly, the microenvironments where high exposure occurs (Chow et al., 2002).

Exposure to emissions from the combustion of fuels used for cooking and heating presents significant health risks in developing

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countries (Balakrishnan et al., 2004). In Bhutan, where this study was conducted, there are no studies on personal exposure to air pollution. As in many developing countries, the use of biomass fuels for cooking in the villages of Bhutan is very common. Exposure studies done in neighbouring and other developing countries where biomass fuels is used have reported particle mass levels exceeding the international guideline limits by several orders of magnitude (Ezzati et al., 2000, Parikh et al., 2001, Balakrishnan et al., 2002, Balakrishnan et al., 2004, Baumgartner et al., 2011, Dionisio et al., 2012, Devakumar et al., 2014).

Children are more sensitive to the effects of air pollution than adults, due to their developing immune system and because they breathe more air relative to their body size (Kulkarni and Grigg, 2008, Buonanno et al., 2012b, Zhang and Zhu, 2012, Buonanno et al., 2013, Demirel et al., 2014). In a typical day, children are exposed to air pollutants in a range of environments, such as at school, home and during transportation (Weichenthal et al., 2008, Ashmore and Dimitroulopoulou, 2009). To date, children's exposure studies have mainly focused on particle mass (Balakrishnan et al., 2004, Almeida et al., 2011, Baumgartner et al., 2011, Dionisio et al., 2012, Devakumar et al., 2014, Hinwood et al., 2014, Rivas et al., 2014), however, toxicological studies have reported significant adverse health effects from exposure to UFP (Oberdörster et al., 2004, WHO, 2005, Weichenthal, 2012, Buonanno et al., 2013). A few studies have quantified children's exposure to UFP around the world, mostly using fixed monitors in school environments (Buonanno et al., 2011b, Mullen et al., 2011, Buonanno et al., 2012b, Zhang and Zhu, 2012, Polednik, 2013, Fonseca et al., 2014). Only two recent studies have quantified children's personal exposure to UFP in different microenvironments in Italy and Australia, using real-time personal particle monitors and time activity diaries (Buonanno et al., 2012a, Mazaheri et al., 2013). To our knowledge, there have been no studies conducted on children's personal exposure to UFP in developing countries. Also, only a handful of studies have assessed children's exposure to volatile organic compounds (VOCs) and NO_2 (Linaker et al., 2000, Rijnders et al., 2001, Kattan et al., 2007, Scheepers et al., 2010, Demirel et al., 2014). Therefore, there is a big gap in knowledge characterising children's personal exposure to air pollution in different microenvironments, as well as the factors driving it. This can only be addressed by conducting studies focused on the personal sampling of pollutant concentrations in every microenvironment that children spend their time.

The aim of the present study was to quantify school children's personal exposure to air pollutants and to determine the factors driving it. The specific objectives of this study were to: (i) quantify children's personal exposure to UFP, VOCs and NO_2 during a typical school day in eastern Bhutan, (ii) apportion sources of the children's daily exposures according to their time spent in different microenvironments, and (iii) assess how children's personal exposure in this study compares with those derived from other studies.

2. Methods

2.1. Study area and participants

This study was approved by the Trashigang District Administration, Royal Government of Bhutan, through letter DAT/DES/[NEC]-22/2012/4637, under whose jurisdiction the three schools function. Verbal consent was obtained from the children and their parents prior to their participation in the study.

Bhutan is administratively divided into 20 districts, and each district into several sub-divisional administrative units called blocks, consisting of clusters of villages. This study was conducted in the Kanglung block within the Trashigang district in eastern Bhutan (Supplementary Information, SI Fig. S.1), which is one of the largest and the most densely populated districts in the country. People in these villages are mostly subsistence farmers, depending mainly on farming and livestock. There are no obvious differences in housing types and lifestyle among the villages. People in Kanglung live in traditional houses made of wood, stone and mud. Although most villages have access to electricity, the use of firewood in traditional stoves is very common for cooking, as well as indoor heating. This is mainly due to intensive cooking activities, such as cattle feed preparation and distiling local liquor, which cannot be done using standard electric or gas stoves due to the size of the pots needed for such activities.

The children attending three rural primary schools (S1–S3) and living in the village settlements around the schools were selected for the study (SI Fig. S.1). All three were day schools, and typical school hours were from 8 am to 4 pm on weekdays, and until midday on Saturdays. S1 was located at an altitude of 1600 m above the sea level, S2 at 1900 m, and S3 at 1400 m. Further, S1 and S2 were located a few metres from the main road connecting the eastern districts to the districts in the west (East–West national highway), while an unpaved farm road connected S3 to the same highway. There were no other roads in close proximity to the schools. S2 was located close to Kanglung, a small town consisting of about 20 shops, while the other two schools were surrounded by farmhouses. The schools were approximately 4–10 km from each other. All school buildings had traditional structures, relying on natural ventilation, and without any heating or cooling systems. Although ventilation was not measured, it was expected to be relatively high because of gaps in the walls and ceilings, which are often associated with traditional building structures.

Children's participation was based on their willingness and consent from the parents, and in consultation with their teachers. A training session on how to handle and charge the instruments was organised for the participating children. Following this, each child was asked to demonstrate the process to the rest of the children and those thought to be competent enough to successfully complete the task were selected.

2.2. Instrumentation and quality assurance

Two Philips Aerasense NanoTracers (NTs) were used to measure personal exposure to UFP. In brief, NT measures particle number (PN) concentrations up to $1 \times 10^6\ cm^{-3}$ in the size range of 10–300 nm and it also provides an indication of mean particle diameter. The instrument operates in two modes: (i) *Advanced* mode, with 16 s sampling intervals allowing for measurement of both PN and mean particle diameter; and (ii) *Fast* mode, which allows for the adjustment of sampling intervals down to 3 s, but only measures PN. The Advanced mode was used in the present study. Details of design and operational procedures for the NT are available in Marra et al. (2010).

The NT's time stamp was synchronised to the local time using the NanoReporter software prior to each measurement. The NTs were tested at the International Laboratory for Air Quality and Health, Queensland University of Technology, Brisbane, Australia prior to their shipment to Bhutan. The two NTs ($n=1,2$) used in this study were run side by side with a TSI model 3787 condensation particle counter (CPC) in order to calibrate the instruments the same way, and ensure the readings from each NT were directly comparable. This approach was employed despite the fact that CPC and NT have different size and concentration specifications in detecting particles (Buonanno et al., 2012a, Mazaheri et al., 2013, Buonanno et al., 2014), and there are differences between the ambient PN concentration profiles in Brisbane (urban area, a developed country) and the study area (rural villages in Bhutan, a developing country). However, since the aim was intercomparison between NTs, rather than between NTs and CPC, and all the three instruments were measuring the same aerosols (ambient air in Brisbane) and there was no other reference instrument available in Bhutan, this approach was identified as the most practical in ensuring that the readings are consistent between the NTs. The correction factors were derived using the following equation:

$$CF_n = \frac{C_{CPC}}{C_{NTn}}$$

where, C_{CPC} and C_{NTn} refer to the concurrent total particle number concentrations in the ambient air, as measured by the CPC and the NTs.

2.3. Measurements

Personal UFP exposure was measured by securing the NT to each child's waist using a dedicated belt. The sample tube was extended close to child's breathing zone. Measurements commenced when children left school for home and concluded the next day at approximately the same time. The children were instructed to keep the instrument charging overnight and while in the classrooms, and to carry it throughout the day except during sleeping, playing and washing. While in the classrooms, the instrument was to be placed in the child's close proximity.

VOCs and NO_2 were sampled using passive sampling on Radiello dosimeters (Fondazione Salvatore Maugeri-IRCCS, Italy), RAD 130 and RAD 166, respectively, for 24 h and concurrently with the UFP personal monitoring. Radiello dosimeters have been widely used for both indoor and outdoor environmental monitoring of VOCs and NO_2, for example (Simon et al., 2004, Charpin-Kadouch et al., 2007, Stranger et al., 2008, Esplugues et al., 2011, Gennaro et al., 2013, Derbez et al., 2014). Passive sampling gives the average results for the duration of measurements and therefore cannot be used to apportion the contributions from different microenvironments. Active sampling enables measurement for shorter time intervals, such as during the peak emission time, but requires a pump, which is noisy and heavier than the NT. Therefore, it could not be used for the present study. The advantages of using passive sampling were highlighted by several review studies (Kot-Wasik et al., 2007, Seethapathy et al., 2008, Barro et al., 2009, Król et al., 2010). There are currently no real time techniques for personal monitoring of VOCs and NO_2.

Diffusive bodies were reused for subsequent measurements after washing them as per the manufacturer's protocol. Because diffusive bodies are quite susceptible to damage, particularly when children carry them for an extended period and also to prevent children from touching with their fingers, a well perforated plastic container was used to house the samplers. This adaption was made based on the similar protective casing designed and recommended for use by the manufacturer for outdoor monitoring, to protect dosimeters from extreme weather conditions. This sampler assembly was attached to child's clothing, close to the breathing zone.

The adsorbed VOCs and NO_2 were analysed by the GC/FID (Trace Ultra, Thermo Scientific) and the UV spectrometry (U-1500, Hitachi), respectively, at the Environmental Engineering Laboratory, Lublin University of Technology, Lublin, Poland. According to the manufacturer prescription, the dosimeters are stable for 4 months after exposure for NO_2 and 6 months for VOCs, if they are properly stored. The recommended low temperature storage was undertaken during the measurement, as well as during the shipment to Poland.

Based on their health significance, thirteen VOCs, namely benzene, 1,2-dichloropropane, trichloroethylene, toluene, chlorobenzene, ethylbenzene, (m+p)-xylene, styrene, o-xylene, α-pinene, 1,2,4-trimethylbenzene, 1,4-dichlorobenzene and limonene were quantified. Detection limits for VOCs ranged from 0.01 - 0.05 $\mu g\, m^{-3}$ depending on the compound, and was 0.9 $\mu g\, m^{-3}$ for NO_2.

Further information on VOCs and NO_2 sampling, analytical procedures and quality assurance are presented in the SI file.

In addition, each child completed a short questionnaire about their houses' characteristics and maintained a time activity diary (SI Appendix 1), keeping records of their main indoor and outdoor activities. Unlike studies in developed countries, where parents assisted in maintaining children's time activity diaries, the parent's role was not possible in this study due to their low literacy levels. Therefore, children were trained to provide this information prior to commencement of the measurements. Further, no traffic data were collected in this study, however, it was observed that only a few tens of cars travelled along the stretch of East–West highway per day (where S1 and S2 were located), as the area is in the remote part of the country.

2.4. Data preparation and analysis

Data from the NTs were downloaded after each measurement and multiplied by the corresponding NT correction factors. The corrected data were grouped according to their location and time period. Based on real-time NT concentration data, average UFP exposure for different activities was calculated for each child. In this paper, personal UFP exposure was defined as the product of UFP concentration and the duration of exposure (Morawska et al., 2013). Personal UFP exposure (cm^{-3}) due to specific activity over the total personal monitoring period was derived using Eq. (1):

$$\vec{E}_x = \frac{\sum_{i=1}^{n} \Delta C_{xi} \times \Delta t_{xi}}{24\ \text{h}} \tag{1}$$

where $\vec{E}_x$ is average personal exposure due to the specific activity (x) for each child, $\Delta C_{xi}$is average UFP concentration (cm^{-3}) due to the specific activity, $\Delta t_{xi}$is activity duration and i=(1–n) is the frequency of activity during the day.

To determine which microenvironments were associated with the highest exposures, the exposure relative intensity (Mazaheri et al., 2013) for each child in each microenvironment was obtained by dividing the fraction of exposure that child received in that microenvironment by the fraction of the day that the child spent in that microenvironment (Eq. (2))

$$\text{Exposure relative intensity (ERI)} = (\text{Fraction of exposure})/(\text{Fraction of the day}) \tag{2}$$

2.5. Statistical techniques

All statistical analyses were performed with SPSS version 21 (SPSS Inc.), with a 5% level of significance ($p < 0.05$). Descriptive statistics were used to characterise UFP exposure in the different microenvironments occupied by the children. The Kolmogorov–Smirnov goodness of fit test and histogram plots were used to check the normality of the distribution of data. All statistical tests were done on the log-transformed data, which showed better normality.

3. Results and discussion

82 children (28 from S1, 34 from S2 and 20 from S3), ranging from 9 to 13 years in age and studying in grades 5 and 6 participated in the study. Measurements for children attending S1 and S2

Table 1
General demographic characteristics of the study population.

Characteristics	S1 (n=28) (%)	S2 (n=34) (%)	S3 (n=20) (%)
Gender			
a. Female	54	38	50
b. Male	46	62	50
Types of house in which children lived			
a. Concrete buildings	4	0	0
b. Traditional buildings	96	100	100
Kitchen types			
a. Indoor	82	79	70
b. Outdoor	18	21	30
Traditional mud cookstove (Yes)	82	77	90
Use of firewood (Yes)	79	85	90
Mode of travel (Walk)	100	100	100

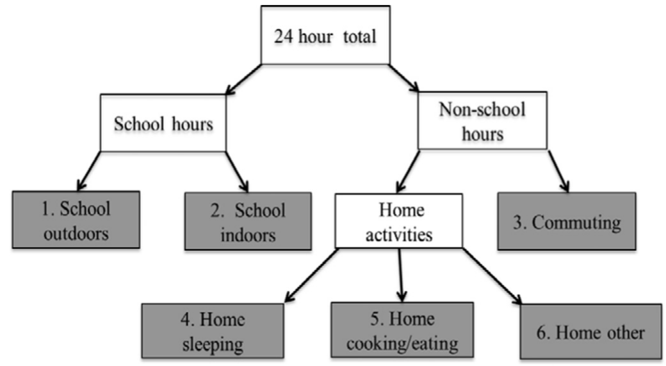

Fig. 1. Classification of children's time activities. The six distinct time activity categories are shaded grey.

were conducted between May and August 2013, which is the wet season in Bhutan, while measurements for S3 children were conducted during the dry season, between October and November 2013. Out of the 82 participating children, personal UFP, VOCs and NO_2 exposure data are available for 59 children. Personal UFP exposure data is not available for the remaining 23 children due to the NT malfunction.

The UFP personal exposure analysis is based on the NT measurements from 48 children, for whom 24 h of data was available. This comprised of 28 children from S1, 7 from S2 and 13 from S3. Missing data was due to children failing to charge the instrument as instructed and due to instrument malfunction. Furthermore, high UFP concentrations were observed for a total of 18 children from S1 and S3 during home cooking/eating, exceeding the maximum NT detection of 1×10^6 cm^{-3}. These values were therefore set to be 1×10^6 cm^{-3} and any analysis involving this data represents a lower bound on the quantity of interest (given the concentration must have been higher than the maximum detection). Of the total 82 VOCs and NO_2 dosimeters, two VOCs and four NO_2 dosimeters from S1 were excluded due to damage sustained by the diffusive bodies.

3.1. General demography of the study population

Table 1 presents the general demographic characteristics of children according to the collected study questionnaires. Almost all children lived in traditional houses. Walking was the only mode of transport for children attending these schools. The number of indoor kitchens was significantly higher than the outdoor enclosed kitchens, while all kitchens were naturally ventilated. Almost all houses had traditional mud cookstoves, without a chimney, and used firewood. It should be noted that some houses used a metal stove with a chimney (called a *bukhari*), both for cooking and heating. Therefore, even without traditional mud cookstoves, firewood was still used.

3.2. Time activity diaries

Based on the information gathered through time activity diaries, a total of six distinct time activity categories were formulated (Fig. 1). School hours were classified as indoors and outdoors, and non-school hours as commuting and home activities. Home activities were further classified as sleeping, cooking/eating and other. Although children have been instructed to place the instrument near them when playing, we do not exactly know where were the instruments placed (except that this was a safe location nearby where they played), and for what time duration since this has not been captured by the time activity diary provided by the children.

SI Table S.1 presents average time spent by children for different activities. Overall, children spent 63% of their daily time at home, with more time spent sleeping than cooking/eating and home other. The time spent at schools was 32%, with considerably more time spent in the indoor than outdoor environment. Commuting accounted for only 5% of the daily activities. The total time spent during school and non-school activities in this study were comparable to similar studies in Australia (Mazaheri et al., 2013) and Italy (Buonanno et al., 2012a). Both the Australian and Italian studies found that, on average, school children spent 28% of their typical day at schools and 65% at home.

The children attending S3 (where measurements were done during the dry season) spent almost 50% more time on cooking/eating than children attending S1 and S2 (measurements done during the wet season). The dry season (October–November) is cool, windy and it gets dark earlier than during the wet season (May–August). Therefore, it is not surprising that children had a different pattern of activities during the cooler months and spent more time inside the kitchen with the fire burning, for both during cooking and heating. Also, the commuting time for children from S3 was half that of the children from S1 and S2, thereby giving them more time at home. This is because all of the S3 children came from just one village, which is closer to their school than children attending S1 and S2, who lived in more spread out villages.

3.3. UFP concentrations

Fig. 2 shows time-series of the mean hourly UFP concentrations for all the children monitored. The peaks during morning and evening hours corresponded to cooking periods when children were at home. This was due to high concentrations from the firewood combustion (the main cooking fuel in the villages of Bhutan) in traditional mud cookstoves, without presence of a chimney. A few of the children's time-series of UFP concentrations also showed sharp peaks during sleeping hours. This may have been due to the use of oil lamps for the religious ceremony in the altar rooms. It should be noted that majority of the Bhutanese population are Buddhist and usually each house has an altar room, which is used for worship and religious ceremonies. A space inside the main house is usually devoted to an altar, where food and water offerings are made daily and small oil lamps fuelled by cooking oil or butter is lit on occasions. Because of its sacredness, normal household activities are usually not carried out inside the altar rooms, however, it was found that some children were using them as bedrooms.

The highest daily (mean $\pm$ standard error) UFP concentration was observed for children attending S3 ($4.19 \times 10^4 \pm 1.04 \times 10^4$), followed by children attending S1 ($1.08 \times 10^4 \pm 1.21 \times 10^3$) and

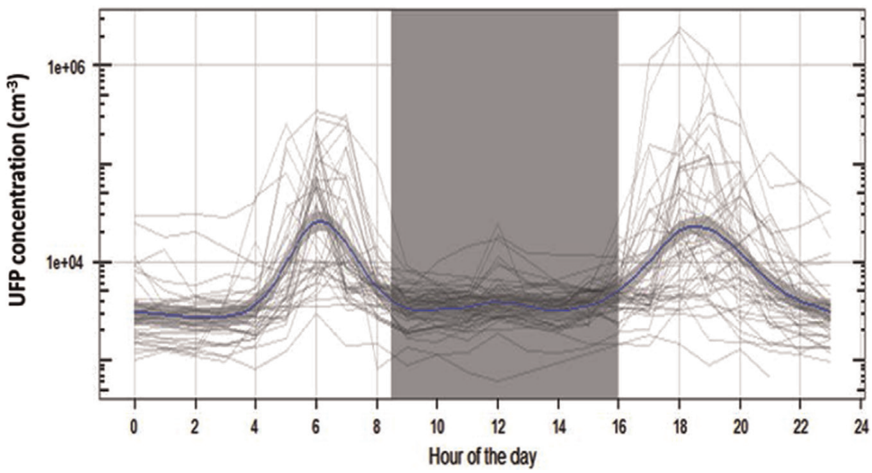

Fig. 2. Mean hourly time-series of UFP concentrations for all the children. The shaded area represents typical school hours.

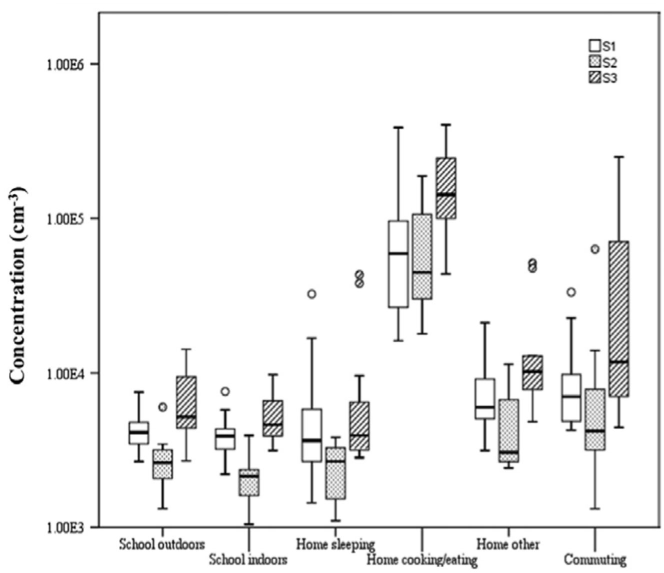

Fig. 3. UFP concentrations for different activities. The boxplot presents minimum, first quartile, median (middle dark line), third quartile and maximum values.

children attending S2 ($9.81 \times 10^3 \pm 3.93 \times 10^3$). Likewise, UFP concentrations for all of the activities were higher for children attending S3 than S1 and S2 (Fig. 3). The cool and windy weather in the dry season meant that children spent more time indoors, where more firewood is used for heating than in the warm, wet season. This explains why the mean concentrations for home activities (and thus daily concentration) were much higher for children from S3.

The daily mean (median) UFP concentration for all of the children monitored was $1.91 \times 10^4 \pm 3.51 \times 10^3$ (1.24×10^4) cm^{-3}. A meta study of particle number concentrations in different environments worldwide reported ambient average PN concentrations (cm^{-3}) of 2.61×10^3 for clean background and 1.08×10^4 for urban ambient to 7.15×10^4 and 1.68×10^5 for on-road and tunnel environments (Morawska et al., 2008). Therefore, the total daily UFP exposure of rural children in this study was comparable to concentrations reported for worldwide ambient urban levels.

The mean PN concentrations during school hours, home and commuting activities were compared with the similar studies in developed countries. Since no similar studies on UFP exposures are available in the literature for the developing world, these rough comparisons highlight the impact of lifestyles and socio-economic factors in the scale of UFP exposure between the study area and the ones conducted in the developed countries. Mean concentration during school hours in this study ($4.54 \times 10^3 \pm 3.02 \times 10^2$ cm^{-3}) was two times lower than the mean concentrations reported for 25 urban schools in Brisbane, Australia (Mazaheri et al., 2013) and three urban secondary schools in Poland (Polednik, 2013), and six times lower than three schools (two urban and one rural) in Italy (Buonanno et al., 2012a). The Polish study used stationary monitors, while the Australian and Italian studies used personal monitors. However, mean home concentration (concentrations during sleeping, cooking/eating and home 'other') was $2.63 \times 10^4 \pm 4.72 \times 10^3$ cm^{-3} three times higher than the Brisbane study. The difference was not as substantial as might have been expected. This is because cooking emissions were the main source of UFP at home in this study and concentrations decreased significantly during sleeping and home 'other'. The mean cooking/eating concentration ($1.07 \times 10^5 \pm 1.47 \times 10^4$ cm^{-3}) was comparable with the mean cooking/eating concentrations reported for the three schools in Italy. Even though the mean commuting concentration in this study ($2.05 \times 10^4 \pm 5.78 \times 10^3$ cm^{-3}) is not associated with vehicular traffic emissions due to the negligible traffic, the magnitude of commuting concentrations were comparable with the Italian study, and nearly two times higher than the Australian study. High commuting concentrations are due to children's commuting time coinciding broadly with cooking periods in the villages and the fact that the routes taken by the children to school and home generally crisscrossed through neighbourhood houses in the village. Therefore, it is hypothesised that children were exposed to neighbourhood cooking smoke during commuting. Particularly, the children from S3 presented the highest commuting concentration among the three schools (Fig. 3), and all of them came from a crowded village (by a local standard) of 79 houses. This was observed even though average time spent for commuting by S3 children was 50% less than S1 and S2. Therefore, commuting concentration for the present study reflects emissions from biomass combustion, unlike the Australian and Italian studies where commuting concentration was largely due to traffic emissions.

3.4. UFP personal exposure and exposure relative intensity

Fig. 4 and SI Table S.2 show the contribution of different activities to children's UFP exposure computed using Eq. (1), while SI Fig. S.2 shows the variation in exposure relative intensity calculated using Eq. (2). The children attending S3 received the highest personal exposure for all the activities compared to children attending S1 and S2. As explained above, this is expected to be due

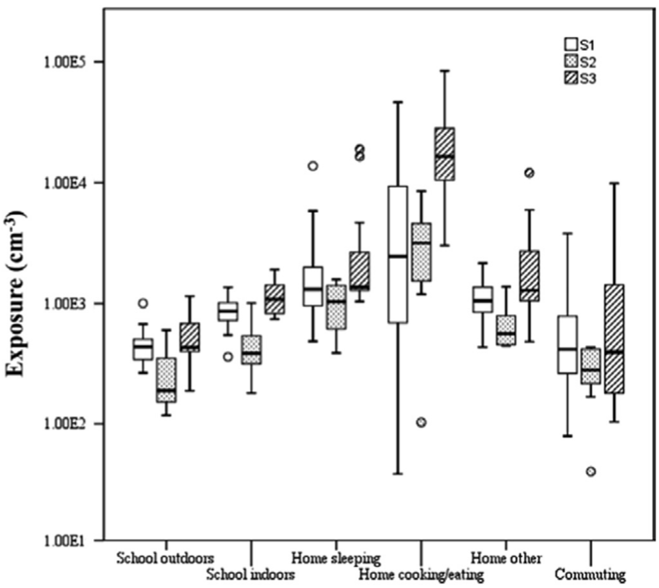

Fig. 4. UFP personal exposure for different activities. The boxplot presents minimum, first quartile, median, (middle dark line), third quartile and maximum values.

to different activity patterns in the wet and dry seasons, resulting in higher exposure to UFP during the dry season. A significant season wise variations of children's (7–9 years old) exposure to particle mass (PM_4) was also reported in a recent study in Nepal (Devakumar et al., 2014).

Personal exposures during school hours were lower than during non-school hours for all the children monitored. This is because, in general, UFP concentrations at schools (both outside and inside) were lower than other microenvironments, particularly at homes. Although school indoor UFP concentrations were lower than the outdoor concentrations (Fig. 3), children's indoor personal exposure was nearly two times higher than the outdoors' in all schools. This is because children spent more time indoors than outdoors at school. However, exposure intensities were approximately the same (a mean of 0.4) for school indoors and outdoors. All children, regardless of school, received the highest exposure from home activities, especially during cooking/eating, characterised by peak UFP concentrations, even though children spent a short time in this microenvironment. Cooking/eating was also associated with the highest exposure intensity (a mean of 8.9), highlighting very high exposure received during the activity per unit time. All children had their breakfast and dinner at home, while they took their homemade cold lunch pack to school. Therefore, children's exposure to particles during cooking was during both breakfast and dinnertime at home. Home sleeping exposure was higher than the home ‘other’ activity. This was due to the much longer time (38% of the day) spent by the children at home for sleeping than for the home ‘other’, which includes activities like attending to household chores, religious ceremonies and feeding cattle outdoors. However, exposure intensity was significantly higher for home ‘other’ activity (a mean of 5.3) than sleeping (a mean of 0.5), due to higher UFP concentrations during the home ‘other’ activity. Even though UFP concentrations during commuting were higher (as explained earlier) than during most other activities (Fig. 3) their contribution to total personal exposure intensity was much lower than from other activities due to the smaller fraction of total time spent commuting compared to other activities.

The exposure intensities in this study were, in general, comparable to the intensities in a similar study in Italy (Buonanno et al., 2013). The differences were observed for cooking/eating (three times higher in this study) and school intensity (two times lower in this study). Similar comparison with the Australian study was not possible, since their reported metrics for children's personal UFP exposure were different from this study and the Italian study (e.g. lung deposited particle surface area) (Mazaheri et al., 2013).

SI Table S.3 shows the relative contributions from different activities to the total daily UFP exposure for all of the children monitored. The home activities contributed to 88% of the total daily exposure, with over 60% contributed by cooking/eating alone, despite only 9% of the daily time being spent on this activity. The lowest contribution of 2% was made by time spent outdoors at school.

The villages where the children lived were fairly homogenous in terms of house characteristics, fuel use and lifestyle. Therefore, overall UFP exposure levels and time activity patterns may be taken as representative for all children in rural villages of Bhutan. The results of this study are consistent with the findings of other studies, in that indoor sources, such as heating and cooking were major contributors to PN concentrations (Buonanno et al., 2009, Mullen et al., 2011, Laiman et al., 2014) and that homes where biomass fuels are used are associated with the highest exposure (Ezzati and Kammen, 2001, Balakrishnan et al., 2002, Balakrishnan et al., 2004, Clark et al., 2010).

3.5. Daily VOCs and NO_2 personal exposure

Benzene was detected in 89% of the dosimeters, toluene in 93%, (m+p)-xylene in 88%, 1,2,4-trimethylbenzene in 85%, 1,4-dichlorobenzene in 99% and limonene in 89%, while the detection percentage for seven other VOCs ranged from 48–70% of the dosimeters.

In general, children attending S3 received higher daily exposures to a majority of the VOCs and NO_2 than those from S1 and S2 (Fig. 5). Only toluene and o-xylene exposures were higher for children attending S2, while chlorobenzene and 1,2,4-trimetylbenzene were higher for children attending S1. The children's exposure to VOCs and NO_2 were lower by several orders of magnitude in this study compared with children exposure reported by

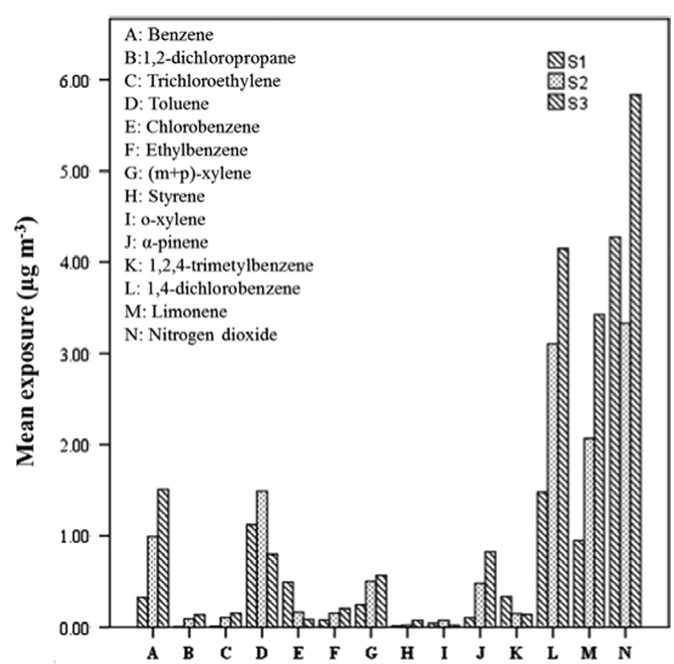

Fig. 5. Daily mean VOCs and NO_2 personal exposure for children attending three schools.

other studies: VOCs (Adgate et al., 2004, Peng and Lin, 2007, Demirel et al., 2014), NO_2 (Alm et al., 1997, Linaker et al., 2000, Rijnders et al., 2001, Demirel et al., 2014). Also see SI Table S.4 for the comparison. Most of these studies have linked children's exposure to VOCs and NO_2 to a range of sources, such as environmental tobacco smoke (ETS), solvent based products, attached garages and traffic emissions. The low VOCs and NO_2 results in this study could be attributed to several factors, such as better ventilation in schools (which would facilitate removal of the compounds emitted by indoor sources) and low traffic emissions in the study areas. Further, since villagers in Bhutan live a fairly traditional lifestyle, modern household products containing VOCs are generally not used. It is also possible that some pores of the diffusive bodies may have been partially blocked by particles (since concentrations were extreme) and therefore, lower amounts may have been sorbed on the cartridge. Further studies on such interactions are required.

Most VOCs are still toxic at low concentrations, and there is no threshold level for carcinogens like benzene (World Health Organization) (WHO, 2010). Similarly, WHO guidelines for NO_2 are 200 $\mu g\,m^{-3}$ for 1 h and 40 $\mu g\,m^{-3}$ for annual average (WHO, 2010), and significant health effects from NO_2 exposure at much lower levels than the WHO guideline have been reported (Jantunen et al., 1999). It should be noted that personal VOCs and NO_2 exposures in this study were based on passive measurements, which do not show possible peak concentrations that may result in high short-term exposure.

3.6. Exposure covariates

In addition to microenvironmental contributions, we investigated whether different exposure covariates, such as gender and house characteristics, influenced the children's exposure levels. SI Fig. S.3 shows the total daily and mean UFP exposures received by male and female children during different activities. Mean commuting exposure was significantly higher for males than females. The differences between exposures during other activities, including total daily UFP exposure, were not significant between male and female children. There was no significant difference in the time spent by male and female children for all the activities, including commuting. This could explain the absence of a statistically significant difference in the mean exposures for activities other than commuting. However, we have no clear explanation why males received higher exposure than females during commuting.

SI Table S.5 shows the variation of mean UFP personal exposure for home activities across house characteristics. During cooking/eating, mean UFP exposure was significantly higher for children living in the houses that used firewood (85% of houses) than those which did not. Also, children living in the houses that had indoor kitchens received significantly higher UFP exposure than those with outdoor kitchens, indicating the presence of cooking emissions inside the homes for long hours after the activity.

SI Fig. S.4 shows the mean daily exposures of NO_2 and the seven dominant VOCs that were detected in the majority of the dosimeters, received by male and female children. There were some variations in the mean exposure between male and female children for different compounds. However, none of the VOCs and NO_2 showed a statistically significant difference between male and female children.

4. Conclusions

We measured children's personal exposure to UFP, VOCs and NO_2 in a rural setting in Bhutan to quantify the magnitude and contribution of different activities conducted in different microenvironments to their daily exposure. 24 h of personal measurements were conducted for 82 children (9–13 years old), attending three primary schools in rural Bhutan (S1–S3). Time-activity data, as recorded in an activity diary, were used to track the daily activity patterns of children. Results showed that children attending S3 experienced higher daily UFP exposure than those attending S1 and S2, and that these were due to differences in activity patterns of children during the dry and wet seasons. For all the children monitored, the highest contribution to the total daily UFP exposure was during times of cooking and eating at homes, due to very high UFP concentrations from firewood combustion in the traditional mud cookstoves used in village homes. The lowest UFP exposures were during the hours that children spent outdoors at school. Likewise, children attending S3 also experienced higher daily exposure to a majority of the VOCs and NO_2 than those attending S1 and S2, but their concentrations were generally very low. Although the measured VOCs and NO_2 exposures using passive 24h sampling do not provide detailed information for source apportionment analysis, but the relatively low VOCs and NO_2 exposures compared to UFP could be attributed to several factors, such as better ventilation in schools, low traffic emissions in the study areas and absence of modern household products containing VOCs in village homes.

As expected, the comparison of UFP personal exposure with similar studies in Australia, Italy and Poland highlighted the significant lifestyle and socio-economic factors affecting these exposures between developing and developed countries. The high UFP exposures at home and during cooking activities using biomass fuels in the present study demonstrate the potential associated health risks.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found in the online version at 10.1016/j.envres.2015.06.006.

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