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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 NO₂ 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 NO₂ exposure (μg 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.

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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 NO₂ (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 micro-environment 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 NO₂ 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 distilling 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 mid-day 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 $1 \times 10^6 \text{ cm}^{-3}$ 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

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