

Emission of Extremely High Concentrations of PM_{2.5} and Ultrafine Particles during Firewood Combustion

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Abstract

Firewood still remains a primary cooking and space heating fuel in many rural homes in Bhutan, the combustion of which presents substantial indoor air quality problems for the occupants. Between 2012 and 2013 a team of experts from Bhutan and Australia undertook a major air quality study in rural areas in Bhutan. During the study we observed emission of extremely high concentrations of particles when stoves were operated for cooking/heating. While larger findings of the study have been published in several journals, so far, we have not reported the emission of episodic peak concentrations when fuelwood was burned. This is important since the short-term peak particle concentrations contribute substantially to the total exposure and the associated health risks. Therefore, the aim of this paper is to report extremely high concentrations of fine (PM_{2.5} <2.5 µm) and ultrafine particles (UFP <0.1 µm) observed during cooking/heating, when firewood was used as a fuel. Two sets of measurements were done (i) indoor fixed measurements during cooking and heating in two village homes and (ii) personal exposure monitoring for 59 village children, using real-time instruments DustTrak for PM_{2.5} and NanoTracer for UFP. The mean UFP and PM_{2.5} concentrations (2.0×10^5 particles/cm³ and 1329 µg/m³) observed during heating were 3 and 4 times higher than the mean concentrations measured before the heating. Likewise, the mean UFP and PM_{2.5} concentrations (6.8×10^5 particles/cm³ and 4429 µg/m³) during cooking were 64 and 69 times the mean concentrations measured before the cooking. For personal exposure monitoring, a mean UFP concentration of 1.1×10^5 particles/cm³ was observed corresponding to time spent by children in the kitchen during cooking. This activity contributed to 64% of the daily exposure, even though children spent only 9% the day in the kitchen while cooking. The results reveal (i) a substantial contribution of biomass fuels to indoor pollution levels, (ii) importance of treating peak concentrations separately when average exposure is computed from extended measurements.

Keywords: *Firewood, Rural, Combustion, Cooking, Heating*

An estimated three billion people world-wide depend on solid fuels for cooking (Legros, Gitonga, & Rijal, 2011). In rural areas of developing countries, solid fuels are mostly burned in inefficient traditional cookstoves, without proper smoke venting system, and often in poorly ventilated kitchens. The resulting household air pollution (HAP) presents significant health risks to rural population (Balakrishnan, Sambandam, Ramaswamy, Metha, & Smith, 2004). The recent World Health Organization (WHO) report has estimated 4.3 million premature deaths in low and middle income countries from exposure to HAP (WHO, 2014).

While number of global households relying on solid fuels for cooking has decreased by some 21% in the last decade, the population at risk from exposure to combustion products has remained more or less same due to growth in population (Bonjour et al., 2013). The Population and Housing Census reports present a similar trend for Bhutan. The proportion of households using firewood for cooking in rural areas has decreased to 36.7% in 2017 (NSB, 2018) from 56.6% reported in 2005 (OCC, 2006). However, in the last 12 years, Bhutan has seen a 16% growth in total population, with 62.2% living in rural areas (NSB, 2018). In the last decade, several studies have characterized HAP in rural areas in some of the least developed countries, for example (Begum, Paul, Dildar Hossain, Biswas, & Hopke, 2009; Devakumar et al., 2014; Dionisio et al., 2012; Kang, Li, Wang, Zhang, & Cong, 2009;

Li et al., 2012; Morawska et al., 2011; Nia et al., 2016; Singh, Tuladhar, Bajracharya, & Pillarisetti, 2012; Tian et al., 2009). However, most studies have relied on instruments that provide time averaged data, the resulting mean concentrations of which do not give information on time-series concentrations, which is crucial for understanding source contribution and emission characteristics. The short-term episodic peak concentrations make substantial contribution to total exposure (Buonanno, Marini, Morawska, & Fuoco, 2012; Mazaheri et al., 2014). Hence, the use of time averaged concentration from extended measurements will result in inaccurate assessment of exposure (Ezzati & Kammen, 2001).

Between 2012 and 2013, a team of academics from Bhutan and Australia (including the first author of this paper) undertook the first ever quantitative air quality study in rural areas in eastern Bhutan. Our research activities involved mobile assessment of on-road air quality for a highway, assessment of children's personal exposure, characterization of air quality in school environment and a real-world characterization of emissions from biomass stoves during cooking and heating. The findings of the study have been reported elsewhere (Wangchuk, 2017a, 2017b; Wangchuk, He, Dudzinska, & Morawska, 2015; Wangchuk, He, Knibbs, Mazaheri, & Morawska, 2017; Wangchuk, Knibbs, He, & Morawska, 2015; Wangchuk, Mazaheri, et al., 2015). To our knowledge, no quantitative air quality study has been reported in the scientific literature from Bhutan prior to our study.

We used real-time portable instruments to measure particle mass and number concentrations. Our measurements during real-world cooking and heating inside kitchens, and personal exposure monitoring for children revealed peak concentrations of UFP ($<0.1 \mu\text{m}$) and $\text{PM}_{2.5}$ ($<2.5 \mu\text{m}$), exceeding the upper detection limit of the instrument. The emission of peak particle concentrations corresponded with the time when stoves were operated for cooking/heating and when children spent time inside kitchens during cooking. Therefore, the aim of this paper is to report emission of extremely high concentrations of particles during firewood combustion. This is important since most of the previous studies have reported time averaged concentrations without any mention of episodic peak concentrations during combustion process. For example, studies in Nepal have reported mean 24 hours indoor $\text{PM}_{2.5}$ concentrations of $2070 \mu\text{g}/\text{m}^3$ (Singh et al., 2012) and $656 \mu\text{g}/\text{m}^3$ (Pokhrel et al., 2015), in houses using traditional biomass cook stoves. If the studies have captured short-term peak concentrations when stoves were operated, the resulting concentrations would have been some order of magnitude higher than the reported mean concentrations. Similarly, a study in nomadic tents in Tibet has reported a mean 24 hours $\text{PM}_{2.5}$ concentration of $1420 \mu\text{g}/\text{m}^3$ (Li et al., 2012). At the same time, this study has reported that average peak concentration when stoves were operated was five times higher than the daily mean concentration. Therefore, since short-term peak particle concentrations make substantial contribution to daily exposure, this must be emphasized when assessing exposure to air pollution.

Methods and Methods

Study area and participants

The study was conducted in the rural villages of Kanglung within the Trashigang district in eastern Bhutan, which is one of the largest and the most densely populated districts in the country. Although 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.

For indoor monitoring, two houses (H1 and H2) located in different villages ~5 km apart were selected to represent the most common stove types and cooking/heating activities. Both the houses were traditional structures built from mud, wood and stone, except for the walls of the H1, which were strengthened with concrete. H1 used LPG and electricity for cooking meals and a wood fed metal chimney stove for space heating (locally called bukhari) which had an enclosed combustion chamber (Figure 1a). H2 used traditional biomass **cook stove** made of mostly mud for all cooking activities as well as for space heating (Figure 1b). The stove had two open potholes and an open combustion chamber. As in all the village homes, both H1 and H2 relied on natural ventilation, doors and windows.

For personal exposure monitoring, 59 village children attending three primary schools in Kanglung participated in the study. The schools were located approximately 4 to 10 km from each other. The typical school hours were from 8 am to 4 pm on weekdays, and until mid-day on Saturdays. All children walked to schools from their homes in the villages. Children's participation was based on their willingness and consent from the parents, and in consultation with their teachers.



Figure 1: (a) stove used for heating in H1 and (b) stove used for cooking in H2, and (c) personal exposure monitoring of children

Instrumentation

The instruments used in the study were shipped to Bhutan from the International Laboratory for Air Quality and Health (ILAQH), Queensland University of Technology, Brisbane, Australia. Prior to shipment, all of the instruments were tested and calibrated at ILAQH.

PM_{2.5} was measured using a DustTrak aerosol photometer (TSI Model 8520, TSI Inc., St. Paul, MN, USA), that operates on a light scattering technique, where the amount of scattered light is proportional to the mass concentration of the aerosol. DustTrak was tested and calibrated for ambient urban concentrations against the Tapered Element Oscillating Microbalance (TEOM 1405-DF, Thermo Fisher Scientific Inc.), which is a robust reference instrument for PM_{2.5} measurements and uses gravimetric detection technique. DustTrak was not calibrated for the biomass emission, therefore, the measured PM_{2.5} concentrations represent approximations of the actual values. For simplicity, the DustTrak results discussed in this paper are referred to as PM_{2.5} from now on (omitting the term 'approximation'). Prior to each sampling, the instrument was set to a 10 second averaging interval, zero calibrated, flow rate checked, and time stamps synchronized with the local time.

UFP was measured using Nano Tracer (NT, Philips Aerasense, Netherlands), which works by diffusion charging and measures particle number (PN) concentrations up to 1×10^6 particles/cm³ in the size range of 10-300 nm. The instrument operates in two modes: (i) *Advanced* mode, with 16 second 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 seconds, 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, Voetz, and Kiesling (2010).

The NT's time stamp was synchronized to the local time using the NanoReporter software prior to each measurement. The two NTs 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. A correction factor for each NT was computed by using the following equation as described by Mazaheri et al. (2014):

$$CF = \frac{C_{CPC}}{C_{NT}}$$

Where, C_{CPC} and C_{NT} refer to the concurrent total PN concentrations measured by the CPC and the NT unit, and CF is the correction factor. It should be noted that CPC 3787 has a lower cutoff size of 5 nm in comparison to 10 nm for NT. Particles from 5-10 nm may account for important fraction of total PN concentrations, in which case results of this study could be underestimated.

Air quality monitoring

Indoor fixed monitoring

PM_{2.5} and UFP were measured in two houses, H1 and H2 in January 2013. Monitoring in the two houses were done to measure particle emissions from metal chimney stove during space heating in family's living room in H1 (Figure 1a) and during liquor distillation (cooking activity) from traditional cook stove inside kitchen in H2 (Figure 1b). The measurements were conducted in three stages, with 45 to 60 minutes of background measurement before the activity, followed by the activity (stove operation), and at least an hour after the activity has ceased.

Two measurements each on different dates were done in both the houses for the same activity. The average duration of stove operation was 5 hours for heating and 2 hours for liquor distillation.

Instruments were placed at 1.5 meters above the floor and at least 3 meters away from the stoves, depending on the size of the space. The standardization of instrumentation location with respect to combustion source is important given potential spatial gradient in concentration. However, in real-world measurements (particularly in houses) it has to be determined by convenience of the occupants. Therefore, location of the instrumentation setup was not standardized in the present study.

Personal exposure monitoring

The measurements were done between May and October 2013. Personal UFP exposure was measured by securing the NT to child's waist using a dedicated belt (Figure 1c). 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 (24 hours). 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. All children were trained to maintain their time activity diary for the duration of measurement. A total of six distinct microenvironments/activities have been considered for exposure monitoring: (1) school indoors (2) school outdoors (3) home cooking (4) home sleeping (5) home others and (6) commuting. The consent for the study was obtained from all the children and their parents, and from the school authority.

Data preparation and analysis

The data were downloaded from instruments after each measurement and checked for anomalies immediately. Of the 59 personal monitoring of children only 48 were used for analyses. For the remaining 11 children there was no complete 24 hours data due to children failing to charge the instrument as instructed and due to occasional malfunction of the instrument. The UFP concentrations were multiplied by the corresponding NT correction factors. The corrected data were grouped according to the 'stages' for indoor monitoring and 'micro-environments/activities' for personal monitoring.

Based on real-time NT concentration data, average UFP exposure for different activities was calculated for each child. The personal UFP exposure was defined as the product of UFP concentration and the duration of exposure (Morawska et al., 2013). Personal UFP exposure (particles/cm³) due to specific activity over the total personal monitoring period was derived using Equation 1:

$$\vec{E}_x = \frac{\sum_{i=1}^n \Delta C_{x_i} \times \Delta t_{x_i}}{24 \text{ hours}} \quad (1)$$

where \vec{E}_x is average personal exposure due to the specific activity (x) for each child, ΔC_{x_i} is average UFP concentration (particles/cm³) due to the specific activity, Δt_{x_i} is activity duration and $i = (1- n)$ is the frequency of activity during the day.

For both indoor and personal exposure monitoring, a high UFP concentrations exceeding the maximum NT detection of 1×10^6 particles/cm³ were observed corresponding to

time when stoves were operated and when children spent time inside kitchens during cooking. These values were therefore set to be 1×10^6 particles/cm³ 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).

Results and Discussion

Table 1 presents the mean UFP and PM_{2.5} concentrations for the background, during the activity and after the activity has ceased for indoor measurements. The mean UFP and PM_{2.5} concentrations in H1 during heating with metal chimney smoke were 3 and 4 times higher than the mean concentrations measured before the heating. Likewise, the mean UFP and PM_{2.5} concentrations in H2 during cooking (liquor distillation) with the traditional mud stove were 64 and 69 times, respectively, the mean concentrations measured before the cooking. However, it should be noted that background levels were influenced by the neighborhood emissions. Both H1 and H2 were located in the settlement zone and smoke infiltration from adjacent houses was evident at the time of measurement. Further, it can also be seen that concentrations remained elevated even after the activity has ceased, at least an order of magnitude higher than the background level. This was contributed by smoldering firewood combustion. It was observed that after cooking and heating, the flaming woods were disassembled inside the combustion chamber and this promoted smoldering combustion, thereby extending the source emission time. This presents a major difference between use of biomass fuels and gas for cooking and heating. When gas is used source emission stops immediately after the activity has ceased.

A comparison between the activities revealed that mean concentrations for both UFP and PM_{2.5} during cooking were over three times higher than the mean concentrations observed for heating. This was expected since traditional cookstove used had an open combustion chamber without a chimney. This means all primary particles emitted during the combustion simply diffused inside the kitchen. Moreover, traditional liquor distillation (a customary cooking activity in village homes in Bhutan) is energy and time intensive, lasting at least two hours compared to cooking meals which can be done at a relatively shorter duration (~30 to 45 minutes for average Bhutanese family). For heating, although the fuel used was also firewood, the enclosed combustion chamber and chimney played a part in discharging most of the emissions outdoor.

Morawska, Ristovski, Jayaratne, Keogh, and Ling (2008) reported mean particle number concentrations (particles/cm³) in different ambient environments worldwide ranging from 2.6×10^3 for clean background to 1.1×10^4 for urban, and 4.2×10^4 for street canyon to 1.7×10^5 for tunnel environment, respectively. Surprisingly, the overall mean concentration of 4.4×10^5 particles/cm³ during the activity (when stoves were operated) in this study was nearly three times the concentration reported in the tunnel environment. Likewise, the mean PM_{2.5} concentrations of 4429 µg/m³ in H2 during cooking was comparable with concentrations reported elsewhere for rural homes where open biomass stoves were used (Table 1). For instance, Brauer, Bartlett, Regalado-Pineda, and Perez-Padilla (1995) reported mean cooking time concentration of 5310 µg/m³ when biomass fuel was used in open fire stove. The measurement in their study was also done using time-series instrument. Further, it can be seen in Table 2 that extended measurements reporting lower mean concentrations for studies which used time-series as well as time average instruments. This is because computing mean concentrations from extended measurement levels the peak concentration

(Manigrasso, Stabile, Avino, & Buonanno, 2013), and as indicated earlier this approach will result in inaccurate assessment of exposure.

Table 3 presents the mean UFP concentrations in different microenvironments/activities measured from personal monitoring of children. The bigger results and discussions on this was reported in (Wangchuk, Mazaheri, et al., 2015). The focus of this section is to highlight peak UFP emissions during cooking time and its contribution to personal exposure. The 'home cooking' presented the highest mean UFP concentration of 1.1×10^5 particles/cm³, which was one to two orders of magnitude higher than concentrations observed in other microenvironments/activities. This was during the time when children were indoors (inside kitchens) when cooking was done. Again, this extremely high UFP concentration was contributed by biomass fuels during cooking using traditional open cook stoves without chimney. The resulting exposure computed using equation 1 for this activity was 1.21×10^4 particles/cm³ and accounted for 64% of the daily exposure. This was despite the fact that children spent only 9% of the total daily time inside kitchens during cooking. Therefore, this confirms that children living in the villages received intense short-term exposure to UFP during cooking time. Further, the overall mean UFP concentration for H1 and H2 (see Table 1), when stoves were operated was six times higher than the mean concentration observed during cooking time from personal monitoring of children (see Table 3). Although stoves used in the houses where children lived were similar to H2, indoor fixed measurement captured point concentrations (3 m from the stove), and while personal monitoring of children involved mobile measurement during which children moved around in the kitchen. However, it was not known how close and further away children spent their time from the stove. Currently, there are no similar studies from rural areas in developing countries with which this result can be compared.

Table 1: Summary statistics of UFP (particles/cm³) and PM_{2.5} (µg/m³) for background (prior to activity) during and after the activity.

| House [*] | Parti- cles | Background | | During Activity | | After Activity | |
|--------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|
| | | Mean | SD | Mean | SD | Mean | SD |
| H1 | UFP | 7.7×10^4 | 4.1×10^4 | 2.0×10^5 | 1.5×10^5 | 8.7×10^4 | 1.9×10^5 |
| | PM _{2.5} | 94 | 57 | 1329 | 986 | 121 | 204 |
| H2 | UFP | 1.1×10^4 | 3.0×10^3 | 6.8×10^5 | 3.5×10^5 | 9.8×10^4 | 1.1×10^5 |
| | PM _{2.5} | 64 | 5 | 4429 | 6219 | 195 | 124 |
| Overall Mean | UFP | 4.4×10^4 | 2.2×10^4 | 4.4×10^5 | 2.5×10^5 | 9.3×10^4 | 1.5×10^5 |
| | PM _{2.5} | 79 | 31 | 2879 | 3602 | 158 | 164 |

^{*}House 1: heating/metal chimney stove, House 2: liquor distillation/traditional mud stove.

Figure 2 presents time-series UFP concentrations during heating using metal chimney stove in H1 (a), during cooking using traditional mud stove in H2 (b), while (c) and (d) present time-series concentrations measured during personal monitoring from two children. The peak concentration during heating (Figure 2a) was the result of initial ignition of the stove. A small amount of kerosene and readily flammable wood chips were used for ignition. During this activity the combustion chamber was kept open for the air influx necessary for combustion. As a result, much of the initial particle emissions remained suspended in the

space. Once the combustion has progressed the combustion chamber was closed and immediate drop in concentration was observed. Although ignition of cookstove in H2 also involved similar process, peaks can be observed for the entire duration of combustion. This was due to open fire combustion of firewood during cooking.

The time-series concentrations for personal monitoring of children reveal two sets of distinct peaks. This corresponded with the evening cooking time after children arrived home from the school and morning cooking time next day before children left for the school. No distinct peaks were observed when children were at school since cooking was not done in the school campus. All the children carried their lunch pack from homes. This time-series variation in concentrations was demonstrated in all the children measurements.

The important observation to note in all the figures, however, is the peak concentrations exceeding 1.0×10^6 particles/cm³, which was beyond factory recommended upper detection limit of the NT used for measuring UFP. This was also captured during personal monitoring from most of the children. In such a situation it was difficult to tell what the true peak concentration was, thereby contributing to a certain degree of data uncertainty. This can also potentially lead to malfunctioning of the instrument during extended measurements. Even for PM_{2.5}, maximum peak concentrations of 1.3×10^4 µg/m³ during heating and 3.3×10^4 µg/m³ during cooking were observed. While TSI DustTrak 8520 used for this study has a factory recommended upper detection limit of 1.0×10^5 µg/m³, the concentrations measured in the study were high enough to draw user's attention for data quality and performance of the instrument. Therefore, it is recommended that future studies in similar environments consider using dilution system, so that instruments measure within the recommended concentration range and data quality is not compromised.

Table 2: PM_{2.5} concentrations reported in indoor measurements using biomass fuels and open Cook stoves

| Study | Location | Fuel(s) | AM (µg/m³) | Sampling Duration |
|--|--|-----------------------------------|------------------------------|--------------------------|
| This study | Kanglung, Bhutan | Biomass | 3764 TS | Cooking time |
| Brauer et al. (1995) | San Jose ´de Solis, Mexico | Biomass | 554.7 TA | 9 hr |
| Naeher, Smith, Lederer, Mage, and Grajeda (2000) | Xela, Guatemala | Biomass | 5310 TS | Cooking time |
| (Siddiqui et al., 2009) | Rehri Goth, Pakistan | Wood | 2740 TS | 8 hr |
| Li et al. (2012) | Nam Co & Anduo region, Tibet | Dung | 1420 TS | 24 hr |
| Singh et al. (2012) | Dang, western region; Dolakha, central region; Ilam, eastern region of Nepal | Wood, dung & agricultural residue | 2070 TS | 24 hr |
| Pokhrel et al. (2015) | Rural Bhaktapur, Nepal | Biomass | 811 S | 24 hr inside Kitchens |
| Nia et al. (2016) | Tibetan Plateau | Biomass | 508 TA | 48 hr |

AM: Arithmetic Mean, TS: Time-series concentration, TA: Time average concentration

Table 3: Summary statistics for UFP concentrations (particles/cm³) in different microenvironments/activities during personal monitoring of children. (N = 48 children).

| Activities | Mean | Standard Deviation | Time Spent (%) |
|-----------------|-------------------|--------------------|----------------|
| School outdoors | 4.8×10^3 | 2.5×10^3 | 9 |
| School indoors | 4.1×10^3 | 1.7×10^3 | 23 |
| Home sleeping | 7.8×10^3 | 1.2×10^4 | 38 |
| Home cooking | 1.1×10^5 | 1.5×10^4 | 9 |
| Home others | 1.3×10^4 | 2.3×10^4 | 16 |
| Commuting | 2.1×10^4 | 5.9×10^3 | 5 |

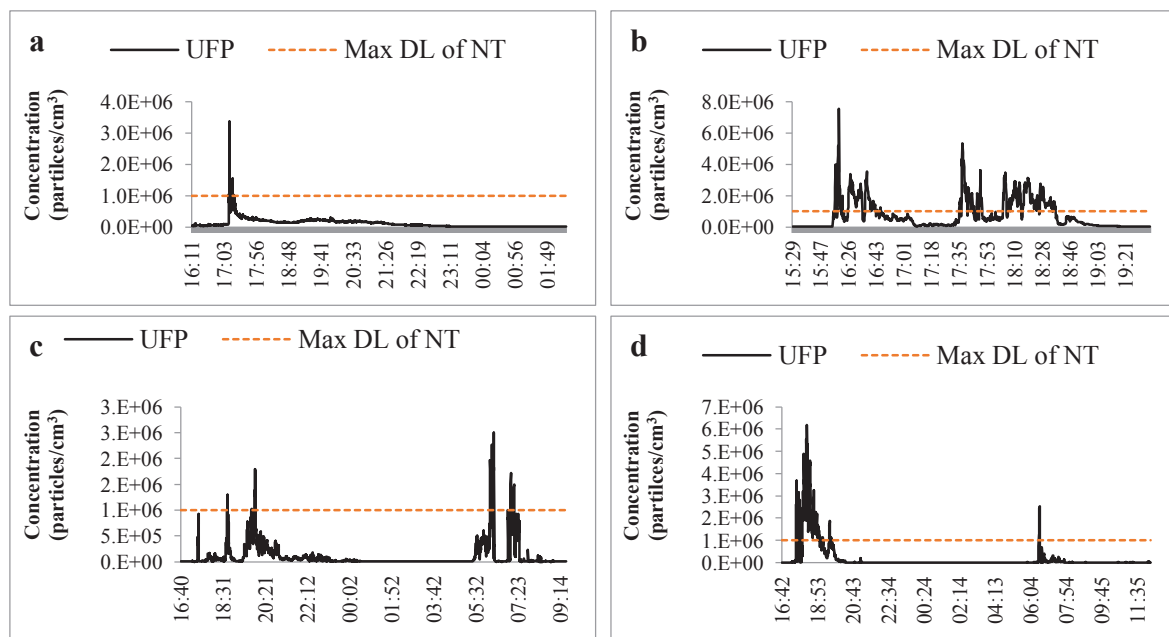


Figure 2: Time-series UFP concentrations (a) during heating using metal chimney stove in H1, (b) during cooking using traditional mud stove in H2, (c) and (d) during personal exposure monitoring of two children. Max DL of NT: Maximum detection limit of NanoTracer. The concentrations in the figures have not been truncated at 1×10^6 particles/cm³ as done for the presentation of results in the text. This was deliberately left to highlight the peak concentrations observed. Further, these figures do not represent simultaneous house and personal exposure measurements

Conclusion

This study reported extremely high concentrations of fine and ultrafine particles observed during combustion of firewood in rural houses in Bhutan. From indoor fixed measurements, the mean UFP concentrations ranging from 1.5×10^5 to 6.8×10^5 particles/cm³ were observed during heating and cooking time. Likewise, the mean PM_{2.5} concentrations ranged from 1329 µg/m³ during heating to 4429 µg/m³ during cooking. Similarly, personal monitoring revealed a very high mean UFP concentrations (1.1×10^5 particles/cm³) during cooking time (when children were present inside kitchens), which was one to two orders of magnitude higher than mean concentrations observed in other microenvironments. Despite children spending only 9% of the time inside kitchens during cooking, this activity contributed to 64% of the daily exposure. These findings clearly highlight (i) the extent to which household fuel combustion contributes to indoor air pollution in rural areas when biomass fuels are used for cooking and space heating (ii) the importance of highlighting peak concentrations when averages are computed from extended measurements. This is crucial during exposure assessments since short-term peak particle concentrations make substantial contribution to health outcomes, and such contributions are not known when only diurnal concentrations are considered. Further, it is recommended that studies focusing on measurements inside kitchens in rural areas use dilution system for data to be more reliable and to prevent instrument malfunction from extreme particle concentrations.

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