Seasonal variations of outdoor air pollution and factors driving them in the school environment in rural Bhutan

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Abstract

A quantitative understanding of outdoor air quality in school environments is crucial given that air pollution levels inside classrooms are significantly influenced by outdoor pollution sources. To date, only a handful of studies have been conducted on this important topic in developing countries. The aim of this study was to quantify pollutant levels in the outdoor environment of a school in Bhutan and assess the factors driving them. Measurements were conducted for 16 weeks, spanning the wet and dry seasons, in a rural school in Bhutan. PM_{10}, $PM_{2.5}$, particle number (PN) and CO were measured daily using real-time instruments, while weekly samples for volatile organic compounds (VOCs), carbonyls and NO_2 were collected using a passive sampling method. Overall mean PM_{10} and $PM_{2.5}$ concentrations ($\mu g/m^3$) were 27 and 13 for the wet, and 36 and 29 for the dry season, respectively. Only wet season data were available for PN concentrations, with a mean of 2.56×10^3 particles/cm^3. Mean CO concentrations were below the detection limit of the instrumentation for the entire measurement period. Only low levels of eight VOCs were detected in both the wet and dry seasons, which presented different seasonal patterns in terms of the concentration of different compounds. The notable carbonyls were formaldehyde and

hexaldehyde, with mean concentrations (μg/m^3) of 2.37 and 2.41 for the wet, and 6.22 and 0.34 for the dry season, respectively. Mean NO_2 cocentration for the dry season was 1.7 μg/m^3, while it was below the detection limit of the instrumentation for the wet season. The pollutant concentrations were associated with a number of factors, such as cleaning and combustion activities in and around the school. A comparison with other school studies showed comparable results with a few of the studies, but in general, we found lower pollutant concentrations in the present study.
Keywords: school, rural, pollutants, outdoor, season, Bhutan

1. Introduction

Air pollution is a major public health issue, and both short and long-term health effects of exposure to a range of air pollutants have been documented by many epidemiological studies (Hussein et al., 2005, Pope III and Dockery, 2006, Kattan et al., 2007, Fullerton et al., 2008, Kim et al., 2011, Buonanno et al., 2013b). While air pollution affects people of all ages, children represent the most vulnerable sub-population. This is due to their developing organs and the fact that they breathe more air relative to their body size compared to adults (Buonanno et al., 2012b, Zhang and Zhu, 2012, Demirel et al., 2014). For school children, the adverse health outcomes from exposure to air pollution lead to absenteeism from schools, and consequently, poor academic achievement (Gilliland et al., 2001, Mendell and Heath, 2005).

Schools have the biggest congregation of children compared to any other environment and children spend a significant amount of their daily time in the school microenvironment (Buonanno et al., 2012a, Mazaheri et al., 2013). The recent literature reviews have established that a substantial portion of children's daily exposure to particles occur in school environments (Mejía et al., 2011, Morawska et al., 2013). Therefore, characterization of air pollution in schools is an important undertaking, in order to enable a reduction in human health risks (Raysoni et al., 2013).

Data from nearby air quality monitoring stations have been frequently used to characterize the air quality in schools (Gilliland et al., 2001, Mejía et al., 2011). However, given the spatial and temporal heterogeneity of pollution levels, in-situ measurements provide a much better assessment of local air quality (Rivas et al.,

2014). In recent years, several air quality studies have been published based on measurements in schools around the world (Lee and Chang, 2000, Mullen et al., 2011, Chithra and Shiva Nagendra, 2012, Buonanno et al., 2013a, Polednik, 2013, Raysoni et al., 2013, Amato et al., 2014, Fonseca et al., 2014, Rivas et al., 2014). However, reports of air quality studies in schools in developing countries are limited. Of the above cited studies, one was conducted in India (Chithra and Shiva Nagendra, 2012) and another in Hong Kong (Lee and Chang, 2000). The study in India was done in a school located near a busy traffic junction, while the study in Hong Kong investigated five schools from residential, industrial and rural areas. However, several deficiencies were identified in these studies, including relatively short measurement periods and often only a single parameter being assessed. Overall, extended measurement periods capturing seasonal variations and multi-parameter assessments were generally lacking, even in school studies conducted in developed countries. Furthermore, only a few studies have investigated UPF levels in school environments (Morawska et al., 2013).

Many studies have reported that pollution levels inside classrooms are largely influenced by outdoor sources (Lee and Chang, 2000, Mullen et al., 2011, Chithra and Shiva Nagendra, 2012, Amato et al., 2014, Fonseca et al., 2014, Rivas et al., 2014). This is because outdoor pollutants can penetrate into the classrooms through ventilation intakes, and open doors and windows (Chithra and Shiva Nagendra, 2012, Rivas et al., 2014). A study in six elementary schools in California found higher average particle number (PN) concentrations inside classrooms when ventilation rates were high, corresponding to higher outdoor concentrations (Mullen et al., 2011). In 39 schools in Barcelona, 53% of the measured $PM_{2.5}$ concentrations inside classrooms were explained by penetration from outdoors (Amato et al., 2014). This study also found significantly higher $PM_{2.5}$ concentrations inside classrooms with windows oriented towards the main street than those away from it. Likewise, a study in Hong Kong has linked higher PM_{10} concentrations inside the classrooms (with natural ventilation) to infiltration from outdoors (Lee and Chang, 2000). In three Portuguese preschools, mean indoor-outdoor (I/O) ratios ranging from 0.54 – 0.93 were reported for UFP, indicating a significant contribution from outdoor sources to indoor concentrations (Fonseca et al., 2014). The mean I/O ratios for PN in three schools in Italy were 0.63 – 0.74 (Buonanno et al., 2013a), while the same ratio for CO in a

103 school in India was 0.51 (Chithra and Shiva Nagendra, 2012). Therefore, it is very
104 crucial for air quality investigations in schools to include outdoor measurements in
105 their experimental designs (Morawska et al., 2013).

106 The aim of the present study was to quantify and characterise the outdoor air quality
107 of a rural school in eastern Bhutan. The primary objectives were: (i) to quantity
108 particle mass (PM_{10}, $PM_{2.5}$), PN, CO, volatile organic compounds (VOCs), carbonyls
109 and NO_2 in the school outdoor environment, (ii) to characterise the sources of
110 pollution and factors driving it, and (iii) to compare the results of this study with those
111 derived from other studies.

112 # 2. Methods

113 ## 2.1 Study Site

114 Bhutan is a small eastern Himalayan country bordered by India and China. Nearly
115 70% of Bhutan's population live in rural areas and are subsistence farmers (RGoB,
116 2006). In general, the environmental conditions, as well as social characteristics are
117 largely comparable with the rest of the Himalayan region. As of 2013, Bhutan had
118 348 primary schools with a gross enrolment of 47,511 children (MOE, 2013). Most of
119 them were day schools and children spent nearly eight hours each day at these
120 schools, for nearly eight months a year, making it one of the most important
121 microenvironments for exposure after homes. Currently, no air quality data are
122 available for schools in Bhutan.

123 This study was conducted in a rural primary school (hereafter called school) located
124 in Kanglung within the Trashigang district in eastern Bhutan (Supporting Information
125 (SI Figure S.1). The school (altitude 1900 m) was centrally located within the block
126 and had the highest enrolment (around 500 children at the time of this study) among
127 the five primary schools in the same block. It was established in 1974 and functioned
128 as a day school, with school activities taking place between 8 am and 4 pm on
129 weekdays and 8 am to 12 pm on Saturdays. All school buildings were two storey
130 traditional structures, constructed in the last five to eight years, replacing the
131 structures built in 1970s. The school did not use any heating or cooling systems and
132 relied on natural ventilation at the time of this study. Further, the main road (East-

West highway) connecting the eastern districts to the districts in the west runs along the school boundary. There are around 20 small shops on either side of the school (along the East-West highway, spread over a kilometre) and village settlements from some 500 meters radius of the school. While the school may not be representative of the whole of Bhutan, the characteristics such as school infrastructures, children's activities, including surrounding and traffic volume are very typical of the schools in rural areas across the country.

2.2 Instrumentation and Quality Assurance

Particle mass (PM_{10} and $PM_{2.5}$) were measured using two DustTraks (TSI Model 8520 aerosol monitor, TSI Incorporated, St. Paul, MN, USA). The DustTrak operates based on a light scattering technique where the amount of scattered light is proportional to the volume concentration of the aerosol. The approximations of PM_{10} and $PM_{2.5}$ values obtained using this instrument were not actual gravimetric values, as the instrument was not calibrated for each specific aerosol studied. However, for simplification, all of the DustTrak results discussed in this paper are referred to as PM_{10} and $PM_{2.5}$, omitting the term 'approximation'.

PN was measured using a Philips Aerasense NanoTracer (NT). The NT measures PN concentrations up to 1×10^6 particles/cm^3 in the size range 10-300 nm and also provides an indication of mean particle diameter. In brief, the instrument operates in two modes: (i) *Advanced* mode, with 16 seconds sampling intervals, allowing for measurement of both PN and mean particle diameter; and (ii) *Fast* mode, which allows for adjustment of sampling intervals down to 3 seconds, but measures only PN. Advanced mode was used in this study. The details of the design and operational procedures for NT can be found elsewhere (Buonanno et al., 2012a, Mazaheri et al., 2013).

Indoor Air Quality Meter (Q-Trak) (IAQ-CALC Model 7545, TSI Incorporated, USA) was used to measure CO, temperature and relative humidity. The Q-Trak uses sensors to monitor different parameters, namely an 'electro-chemical' sensor for CO, 'thermistor' for temperature, and 'thin-film capacitive' for relative humidity. Prior to their shipment to Bhutan, all of the instruments were calibrated and tested at the International Laboratory for Air Quality and Health, Queensland University of Technology, Brisbane, Australia.

2.3 Sampling Protocols

The outdoor air quality measurements were conducted for 16 weeks in two campaigns. The first campaign (8 weeks) was conducted during the wet season, between 27/05/2013 and 04/08/2013, and the second campaign (8 weeks) during the dry season, between 02/10/2013 and 28/11/2013. The wet season (May to August) in the study area is characterised by a warm and wet weather, while the dry season (October to December) is characterised by cool and windy weather. All of the sampling was conducted from the window of the first floor of the school administrative building, located in the centre of the school complex.

2.4 Outdoor particle mass, PN and CO measurements

The DustTraks and Q-Trak were set at a 10 second averaging interval and the NT was set at 16 seconds. The flow rate for the DustTraks was maintained at 1.7 L/min and they were zero calibrated prior to each measurement. The Q-Trak probe assembly was extended outside the window, in the shade, in order to protect the sensors from extreme weather conditions. Sampling was done 24 hours a day and data from the instruments were downloaded every two to three days.

2.5 Outdoor VOCs, Carbonyls and NO_2 Measurements

VOCs, carbonyls and NO_2 were sampled passively using Radiello dosimeters, RAD 130, RAD 165 and RAD 166, respectively. The dosimeters were exposed to outdoor air, under the appropriate shed, for seven days. A total of 16 samples for the wet and dry seasons (8 each) for VOCs and NO_2, and 12 samples for carbonyls (8 for the wet and 4 for the dry season) were collected. Due to budgetary constraints, only a limited number of carbonyl dosimeters could be procured. Diffusive bodies were reused for the subsequent measurements, after washing them as per the manufacturer's protocol. Thereafter, the GC/FID analysis (Trace Ultra, Thermo Scientific) for VOCs, HPLC analysis (Water Action Analyzer) for carbonyls and UV spectrometry (U-1500, Hitachi) for NO_2 were performed. 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. The target carbonyls were formaldehyde, acetaldehyde, acrolein, acetone, propionaldehyde, butyraldehyde and hexaldehyde. Detection limits

for VOCs ranged from 0.01 to 0.05 μg/m^3 depending on the compound, as well as 0.1 to 0.9 μg/m^3 for carbonyl compounds and 0.9 μg/m^3 for NO_2.

We quantified the above gaseous pollutants because of their associated adverse health effects. Most VOCs have the potential to cause sensory irritation and impairment of the central nervous system (Zhang and Smith, 2003) while benzene, toluene, ethylbenzene and xylenes (BTEX) are known carcinogens (Pegas et al., 2011, Demirel et al., 2014). Therefore, the World Health Organization (WHO) has not proposed any threshold level for carcinogens like benzene (WHO, 2010). Likewise, carbonyl compounds are toxic and present carcinogenic effects, and formaldehyde levels exceeding 1 μg/m^3 are considered a concern (Pegas et al., 2011). Among others, the reported health effects of NO_2 include wheezing and exacerbation of asthma (Kattan et al., 2007, Kim et al., 2011). While the WHO guidelines for NO_2 are 200 μg/m^3 for 1 hour and 40 μg/m^3 for annual average (WHO, 2010), significant health effects from NO_2 exposure at much lower levels than the WHO guideline have been reported (Jantunen et al., 1999).

2.6 Other Data

The rainfall, wind direction and wind speed data were collected from the nearest weather station, located a kilometre from the monitoring site, owned by the Department of the Hydromet Services, Bhutan. Only daily average data were available for these meteorological parameters.

2.7 Data Processing and Analysis

NT concentrations were multiplied by the correction factor that was computed at the International Laboratory for Air Quality and Health, Queensland University of Technology, Brisbane, Australia before the instruments were shipped to Bhutan. The correction factor was derived by running the NT side by side with a TSI Model 3787 condensation particle counter (CPC) as follows (Mazaheri et al., 2013):

$$CF = C_{CPC}/C_{NT}$$

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.

Erroneous data occurred due to the malfunction of the instrument and tube obstruction by insects during the wet season. On a few occasions it was found that insects had made their way into the sampling tubes and impeded the airflow. The NT failed completely in the middle of the wet season measurements, therefore providing data for the first quarter of the measurement period only. Data was also lost due to power outages over the course of the measurement periods. In total, 106 days of PM_{10}, 97 days of $PM_{2.5}$, 28 days of PN and 114 days of CO data were available for analysis.

Further, during the dry season campaign, the school organised two major non-academic events. On 18-19 October (campaign week 11), the school organised a sports competition for the children, while on 15-16 November (campaign week 15), a religious ceremony was conducted at the school. On those four days, the meals for all of the students and teachers at the school were cooked out in the open on the school grounds using open wood fire. Significantly higher pollution levels were observed during these event days and therefore, the data for those four days were treated separately from the overall analysis.

2.8 Statistical Analysis

Statistical analyses were performed using SPSS version 21 (SPSS Inc.). A 5% level of significance was used for all analyses ($p < 0.05$). The Mann-Whitney U test (a non-parametric equivalent to student's t-test) was used to test the mean differences between two independent variables, while the Spearman's rho correlation was used to analyze the correlation between daily mean pollutant concentrations and meteorological parameters. The pollution rose diagrams were plotted using the 'openair' R package (Carslaw, 2012).

3. Results

3.1 Meteorological Parameters

The mean and standard deviation for temperature, relative humidity and wind speed for the wet season were 19.8 ± 0.9 °C, 89.7 ± 5.1% and 0.58 ± 0.32 m/s, respectively. The same statistics for the dry season were 13.7 ± 2.8 °C, 77.6 ± 13.8% and 1.14 ± 0.64 m/s, respectively. There was only a marginal difference in temperature and relative humidity between the wet and dry season campaigns. However, these

parameters can get much lower in January and February. Total rainfall during the measurement periods were 459 and 122 mm for the wet and dry seasons, respectively.

3.2 Particle Mass, PN and CO Concentrations

Mean hourly time-series concentrations for the entire measurement period presented distinct peaks for all of the particle fractions, generally between 8-8:30 am (Figure 1). The mean hourly CO concentrations were less than the detection limit of the instrument (0.01 ppm), except on the four days when there was open wood fire cooking in the school grounds (the results are presented separately in the subsequent section). The mean 24 hour concentrations for the wet and dry season days ranged from 10 to 64 $\mu g/m^3$ and 11 to 158 $\mu g/m^3$ for PM_{10}, and 3 to 49 $\mu g/m^3$ and 5 to 147 $\mu g/m^3$ for $PM_{2.5}$, respectively (Figure 2). The daily mean PN concentrations for the wet season campaign ranged from 1.28×10^3 to 4.35×10^3 particles/cm^3 (Figure 3). There were no PN data for the dry season due to instrument malfunction. Unlike PM_{10} and $PM_{2.5}$, there are currently no established guidelines or permissible standards for PN.

The seasonally segregated summary statistics for PM_{10}, $PM_{2.5}$ and PN concentrations are presented in SI Table S.1. The overall mean PM_{10} and $PM_{2.5}$ concentrations were higher for the dry season than the wet season (Figure 4). However, only mean $PM_{2.5}$ concentrations were significantly different between the seasons. The reason for this is explained in the discussion section. The overall mean PN concentration for the wet season was $2.35 \times 10^3 \pm 7.25 \times 10^2$ particles/cm^3.

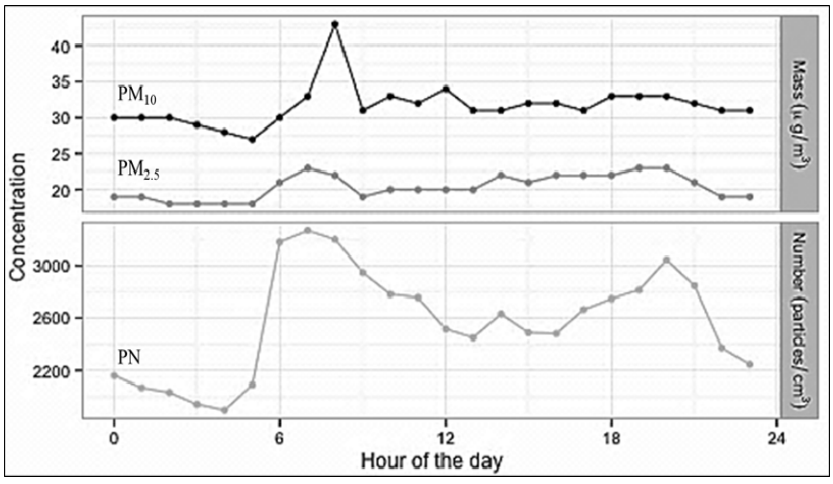

Figure 1. Mean hourly time-series concentrations of particle mass and PN for the entire measurement period.

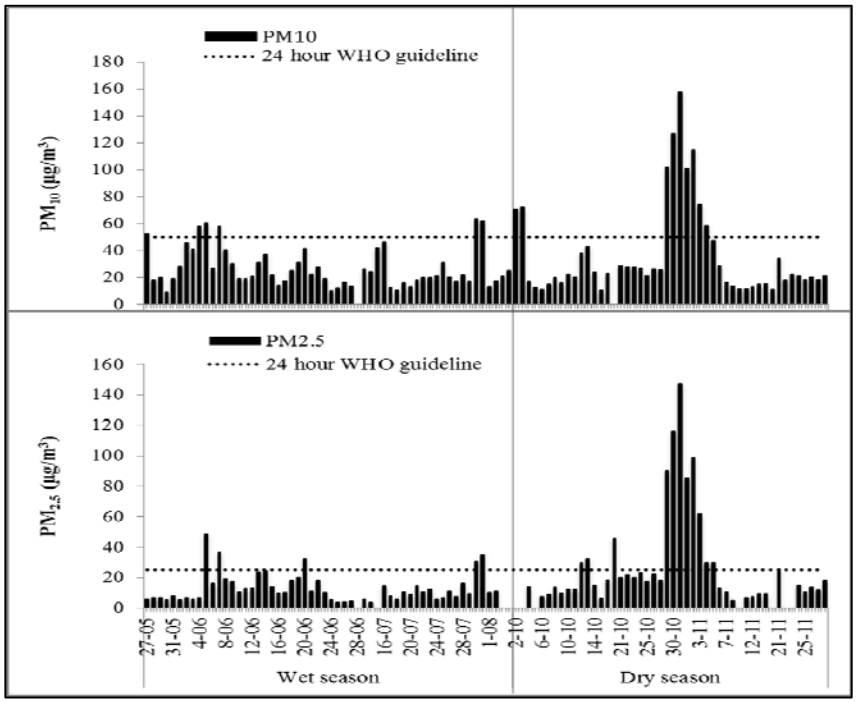

Figure 2. Variations of daily mean PM_{10} and $PM_{2.5}$ concentrations for the wet and dry seasons.

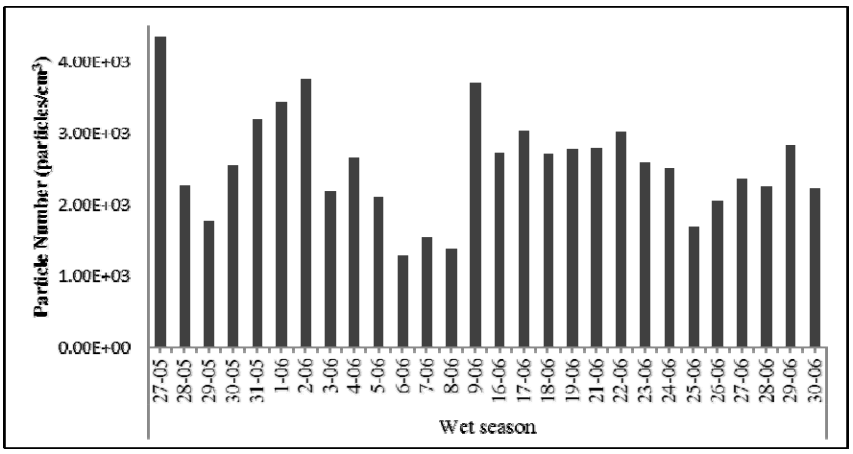

Figure 3. Variations of daily mean PN concentrations for the wet season.

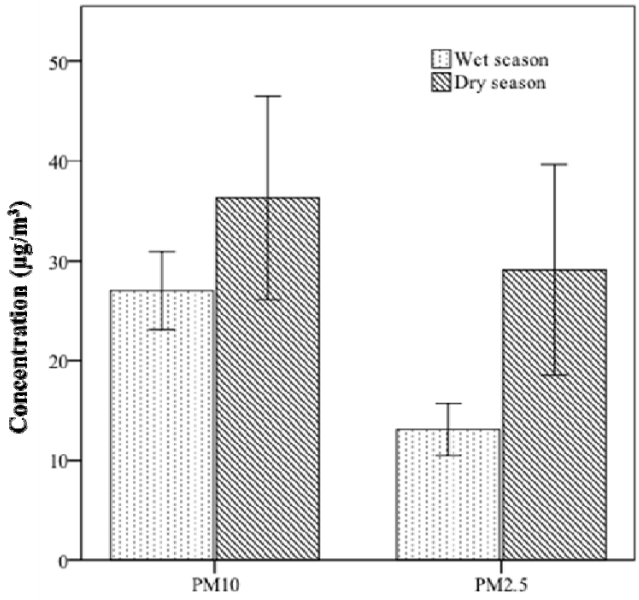

Figure 4. Overall mean concentrations for PM_{10} and $PM_{2.5}$ for the wet and dry seasons. Error bars show 95% confidence interval.

Figure 5 and SI Table S.1 show the summary statistics for particle mass and PN concentrations on school and non-school days. As explained in the previous section, children also attended school on Saturdays until mid-day. Therefore, only data from Sundays were used to compute the statistics for non-school days. No distinct variation was observed (for both mean and median concentrations) for particle mass between school and non-school days, although maximum concentrations were higher on school days. In contrast, PN presented higher mean and median concentrations on non-school days than school days. However, it should be mentioned that there were limited data for non-school days compared to school days.

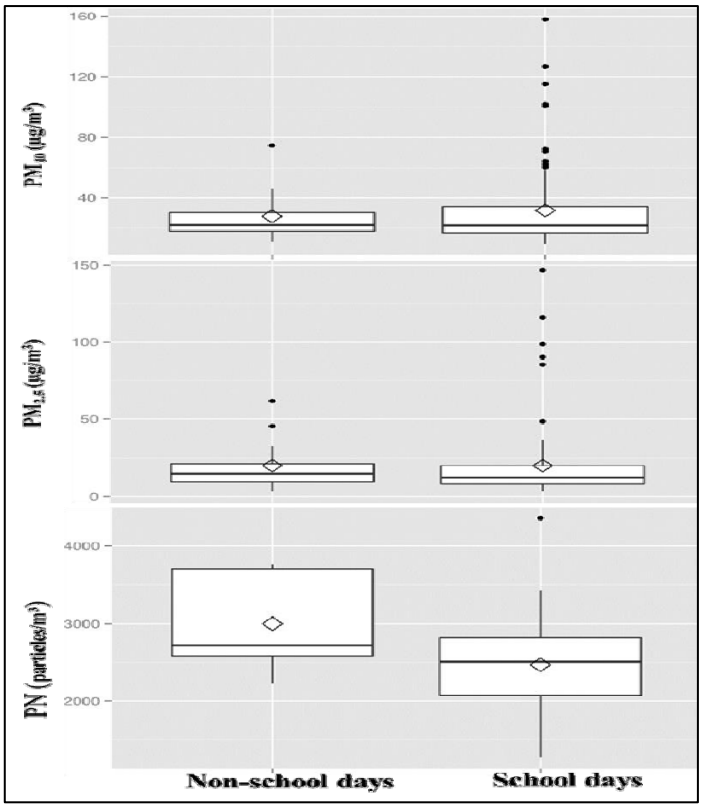

Figure 5. Box plot for PM_{10}, $PM_{2.5}$ and PN presenting the maximum, minimum, median (middle dark line), mean (square box), first and third quartile values for school and non-school days.

3.3 VOCs, Carbonyls and NO_2 Concentrations

Figure 6 and SI Table S.2 present the seasonally segregated summary statistics for VOCs and carbonyls. Only low levels of eight VOCs were detected for both the wet and dry seasons. Ethylbenzene was detected only in the dry season, while 1,2-dichloropropane, trichloroethylene, chlorobenzene and styrene were not detected on neither of the seasons. The mean concentrations of benzene and 1,4-dichlorobenzene were higher for the dry season, while toluene, α-pinene, limonene and 1,2,4-trimethylbenzene were higher for the wet season. However, mean differences were not statistically significant for any of the detected VOCs.

Among the carbonyls, acrolein and butyraldehyde were not detected for the dry season. The mean formaldehyde concentrations were similar for both the wet and dry seasons, while acetaldehyde, acetone and propionaldehyde were marginally higher for the dry season. The mean hexaldehyde concentration was significantly higher for the

wet than the dry season. Mean NO_2 cocentration for the dry season was 1.7 µg/m^3, while it was below the detection limit for the wet season.

It should be noted that the results for VOCs, carbonyls and NO_2 were based on passive measurements, quantifying weekly average concentrations. Therefore, the data do not show possible peak concentrations, and also cannot be apportioned to examine differences on school and non-school days.

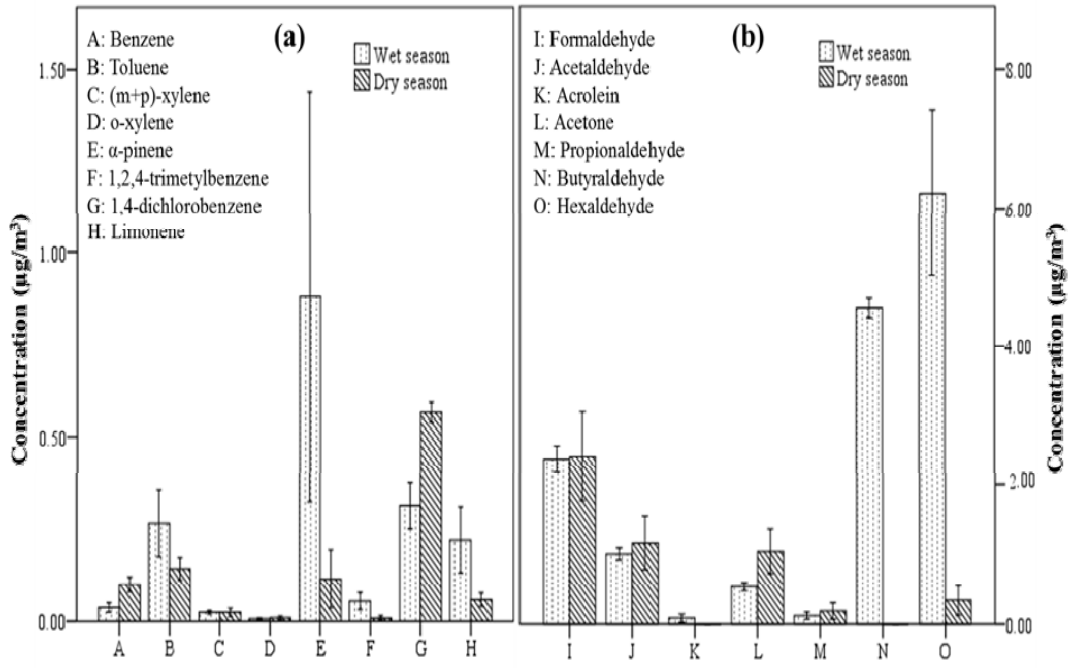

Figure 6. Average VOCs (a) and carbonyl (b) concentrations for the wet and dry seasons. Error bars present 95% confidence interval.

3.4 Influence of Cooking on School Outdoor Pollution Levels

The highest pollution levels were detected on the four days during which open wood fire cooking took place in the school grounds, on weeks 11 and 15. The mean concentrations ranged from 116 to 434 µg/m^3 for $PM_{10,}$ 99 to 327 µg/m^3 for $PM_{2.5,}$ and 0.36 to 1.56 ppm for CO, respectively. This was the only time when measurable concentrations of CO were detected, whereas for other measurement days, mean CO concentrations were below the detection limit. Among the VOCs, ethylbenzene, (m+p)-xylene, and o-xylene concentrations were the highest on weeks 11 and 15. Likewise, NO_2 concentrations were also the highest on those two weeks, being 3.18 µg/m^3 for week 11 and 5.17 µg/m^3 for week 15, respectively. Among the carbonyls, formaldehyde, acetaldehyde and acetone concentrations were highest on week 15, being 4.85, 2.97 and 2.54 µg/m^3, respectively. A similar finding, with these three

carbonyls being the dominant compounds during wood (cooking fuel) combustion, was reported by Zhang and Smith (1999). Carbonyl measurements were not conducted on week 11, due to the limited number of dosimeters available.

3.5 Correlation between Pollutant Concentrations and Meteorological Parameters

Spearman's correlation analyses were conducted between pollutant concentrations (PM_{10}, $PM_{2.5}$, PN, dominant carbonyls and NO_2) and meteorological parameters (SI Table S.3). VOCs were not considered, since their concentrations were very low. The particle concentrations (PM_{10}, $PM_{2.5}$ and PN) and rainfall showed a significant negative correlation for the wet season. Among the carbonyls, formaldehyde showed a significant negative correlation with the rainfall for the wet season and acetaldehyde for the dry season. Only PM_{10} and $PM_{2.5}$ showed a significant negative correlation with wind speed for the dry season. It should be noted that there were less observations for carbonyls and NO_2 compared with PM_{10} and $PM_{2.5,}$ and this could have biased the statistical correlation results.

SI Figure S.2 shows the pollution rose diagrams for the wet and dry season for PM_{10} and $PM_{2.5}$, and wet season only for PN. South-westerly winds were associated with the maximum percentage of particles on both the wet and dry seasons. The highest PM_{10} and $PM_{2.5}$ concentrations for the dry season were associated with westerly winds. For the wet season, the highest PM_{10} concentrations were associated with south-westerly and southerly winds, while the highest $PM_{2.5}$ concentration was associated with south-westerly winds only. The highest PN concentrations were also associated with southerly winds. The likely sources are discussed in the next section.

4. Discussion

The time-series concentrations presented distinct peaks, broadly coinciding with the morning cleaning time at the school. All the children were engaged in cleaning both inside and outside school areas daily, and some trash was occasionally burnt. A more pronounced peak of PM_{10} than $PM_{2.5}$ or PN concentrations indicated the higher impact of resuspension of coarse particles from the school grounds during cleaning activities, rather than contributions from burning the trash (Figure 1). As expected, particle mass concentrations were higher for the dry season than the wet season. The

daily mean PM_{10} and $PM_{2.5}$ concentrations in this study exceeded the WHO guidelines (PM_{10} = 50 μg/m^3, $PM_{2.5}$ = 25 μg/m^3) (WHO, 2006) on 12% and 18% of the measurement days, the majority of which occurred during the dry season days (Figure 2). Likewise, seasonally segregated overall means of both PM_{10} and $PM_{2.5}$ were above the WHO annual guidelines (PM_{10} = 20 μg/m^3, $PM_{2.5}$ = 10 μg/m^3) (WHO, 2006) for both the wet and dry seasons. This was surprising since the air quality in a rural Himalayan location like Bhutan is expected to be pristine, yet explainable high dust levels were observed. Although the results reported in this study were based on four months of outdoor monitoring, a comparison with the annual WHO guidelines was made based on the assumption that significant variations in particle mass concentrations were not expected for the months not covered by the monitoring.

A number of factors may have contributed to particle concentrations and their trends at the study site. At the time of this study, the school had a dirt playground and an unpaved assembly ground in the middle of the school complex (SI Figure S.1). All children and teachers gathered in the assembly ground for prayer before the lessons started each day. During the lunch break and after school hours, children were found to use the ground for different outdoor activities, such as playing and walking. Therefore, it is expected that particle mass concentrations were a result of the dust resuspended from the bare ground. Children played on the school ground even on Sundays and the fact that school and non-school days presented comparable mean and median concentrations (Figure 5) indicates that the particle resuspension rate from the school ground remained similar throughout the week. While we were not able to conduct any indoor measurements at the school due to limited instrumentation, resuspension of coarse particles from classroom floors, due to cleaning and movement of children can be the dominant indoor source.

The predominant wind directions during the measurement period were from the west and southwest, and much of the particle mass and PN were associated with these winds (SI Figure S.2). There were several potential sources upwind of the predominant wind directions. The shops and settlements located immediately to the west of the school were expected to have contributed to particle concentrations through activities such as burning trash and crop residues, and outdoor incense burning (a Buddhist ritual carried out by some people each morning, during which

478 leaves and branches of certain plants are burnt). A statistically significant higher mean
479 $PM_{2.5}$ concentration for the dry season than the wet season (but not for PM_{10}) (Figure
480 4) indicated that the contribution of combustion sources to fine particle concentrations
481 was more pronounced for the dry season. It should be noted that while burning trash
482 and incense are regular activities, burning crop residues, which was by far the
483 dominant source, is usually done during the dry season. The higher mean and median
484 concentrations of PN on non-school days (Figure 5) could be due to trash burning in
485 the vicinity of the school, as people have more time for cleaning on Sundays.

486 As reported in the literature, the major outdoor sources of VOCs, carbonyls and NO_2
487 are industrial processes, biomass and fossil fuel combustion (Zhang and Smith, 1999,
488 Linaker et al., 2000, Demirel et al., 2014). Carbonyls are also generated through
489 secondary atmospheric oxidation of VOCs and hydrocarbons (Pang and Mu, 2006).
490 Therefore, higher carbonyl concentrations during the summer season can result from
491 photo-oxidation of VOCs, while combustion is likely to be the major source during
492 the winter months (Pang and Mu, 2006). There were no industrial sources in the
493 vicinity of the school, and so the low concentrations of gaseous pollutants in this
494 study can be explained as being negligible contributions from other combustion
495 sources, such as biomass burning and traffic emissions. Among the VOCs, α-pinene
496 and limonene are terpenes, a class of organic compounds commonly emitted by trees,
497 particularly conifers. The school surrounding had good natural forest cover, including
498 planted coniferous trees within the school compound. Therefore, higher α-pinene and
499 limonene concentrations during the wet season were likely to be biogenic emissions
500 from trees induced by the hot temperature during the day. It should be noted that
501 daytime temperatures could be high at the study site during the wet season when it is
502 not raining.

503 On four days when there was open wood fire cooking on the school grounds, children
504 were actively engaged in different outdoor activities (no classroom lessons). The
505 inhalation rate for children is generally highest during outdoor playing and sports
506 (Buonanno et al., 2011). Therefore, open wood fire source in the school when
507 children were engaged in outdoor activities was likely to subject them to a higher risk,
508 by increasing their inhaled pollutant doses. Further, these events were noted to occur
509 each year as annual school programs. In future, making alterative cooking plans or

using cleaner fuels can minimise the health risks that the children are currently subjected to.

The results for particle mass and number concentrations obtained in this study were compared with other school studies which reported outdoor concentrations (SI Table S.4). It should be noted that sampling durations varied between the studies, which could have influenced the mean concentrations. The overall mean PM_{10} concentration for the present study was two times higher than the mean outdoor concentrations reported in USA (Raysoni et al., 2013), but two and six times lower than in Poland (Zwoździak et al., 2013) and Hong Kong (Lee and Chang, 2000), respectively. While the overall mean $PM_{2.5}$ concentration was comparable with the concentration reported in Spain (Rivas et al., 2014), it was two times higher in Sweden (Wichmann et al., 2010) and USA (Raysoni et al., 2013), and two times lower in Belgium (Stranger et al., 2008) and Poland (Zwoździak et al., 2013).

A meta study of particle number concentrations (particles/cm^3) in different ambient environments found values of 2.61×10^3 for clean background to 1.68×10^5 for tunnel environment (Morawska et al., 2008). Therefore, overall mean PN concentration (2.35×10^3 particles/cm^3) in this study was comparable with the worldwide ambient clean background. The mean outdoor PN concentrations reported in other schools, in Italy (Buonanno et al., 2013a), Canada (Weichenthal et al., 2008), USA (Mullen et al., 2011), Spain (Rivas et al., 2014), and Australia (Guo et al., 2008) were four to eleven times higher than the present study (SI Table S.4). In most of these studies, traffic emissions influenced PN concentrations in the school environment. The Italian study, in particular, reported higher PN concentrations for schools located within urban areas (higher traffic density) and lower concentrations for a school located in a rural area (lower traffic density) (Buonanno et al., 2013a). For the present study, while no traffic data were collected, it was observed that only a few tens of cars per day travelled along the stretch of the East-West highway where the school was located. This is because of remote location of the study site, as well as low traffic volume in this part of the country, which explains the low PN concentrations for the present study. However, as discussed earlier, the PN concentrations reported were for the wet season only, during which the study site

541 received more rainfall than the dry season. Further studies quantifying PN levels in
542 the dry season are needed.

543 In SI Table S.5, the mean VOCs, formaldehyde and NO_2 concentrations of the current
544 study are compared with other schools studies. Other carbonyls were not included,
545 since we have not come across any such investigations in school outdoor
546 environments. The mean VOCs concentration in this study were broadly comparable
547 with a study in USA (Godwin and Batterman, 2007). However, school studies
548 elsewhere have reported VOCs concentration several orders magnitude higher than
549 the present study, for example in Turkey (Demirel et al., 2014), USA (Raysoni et al.,
550 2013), Italy (Gennaro et al., 2013) and Belgium (Stranger et al., 2008). Although
551 formaldehyde was one of the dominant carbonyls in the present study, its
552 concentration was two times lower than in Turkey (Sofuoglu et al., 2011) and Serbia
553 (Jovanović et al., 2014). The mean NO_2 concentration of the present study was 13 to
554 46 times lower compared to studies in Turkey (Demirel et al., 2014), Sweden
555 (Wichmann et al., 2010), Spain (Rivas et al., 2014) and Belgium (Stranger et al.,
556 2008).

557 # 5. Conclusions

558 Since air quality inside school classrooms is significantly influenced by outdoor
559 sources, it is crucial to characterize and quantify outdoor air quality in school
560 environment. In this study, for the first time, we quantified multiple outdoor
561 pollutants for 16 weeks (with measurements spanning wet and dry seasons) in a rural
562 primary school in Bhutan. The results showed seasonal variations in pollutant
563 concentrations, with higher PM_{10}, $PM_{2.5}$ and NO_2 during the dry season. However, the
564 overall mean concentrations of PM_{10} and $PM_{2.5}$ during both the wet and dry seasons
565 were over the WHO annual guidelines. The detected VOCs and carbonyls presented
566 different seasonal patterns in terms of concentrations of different compounds. Since
567 this school is considered as representative of schools in rural areas, we expect
568 comparable pollution levels in the outdoor school environments in rural Bhutan.

569 Overall, a comparison with the studies conducted in developed countries showed
570 comparable outdoor pollutant concentrations with a few of the studies, but in general,
571 the pollutant levels were lower at the Bhutanese school environment. The major

572 contributors were non-traffic sources such as dust resuspension (from bare ground),
573 and biomass and trash combustion, as opposed to traffic emissions in developed
574 countries. Even though the school is located next to the main road, traffic contribution
575 was expected to be negligible because of the very low traffic volume. The
576 Government of Bhutan is planning to develop the area around the school into an
577 urban centre. In future, while there is likely to be a suppression of current dust sources
578 with the installation of pavement and roads, the traffic volume is expected to increase,
579 thereby leading to increased pollution.

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SUPPORTING INFORMATION (SI)

Seasonal variations of outdoor air pollution and factors driving them in the school environment in rural Bhutan

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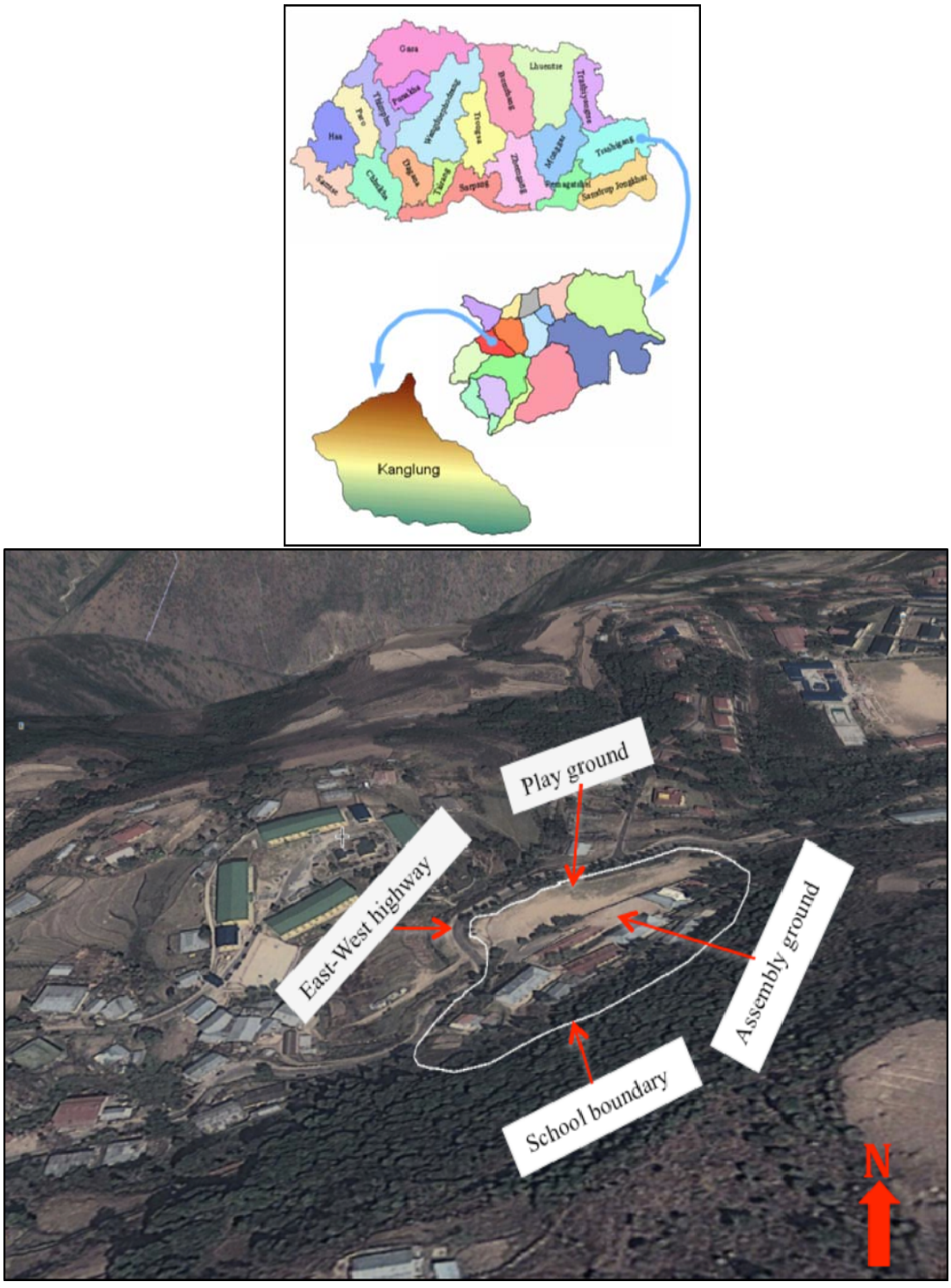

Figure S.1: Map of Bhutan showing the study site.

784 Table S.1: Summary statistics for PM_{10}, $PM_{2.5}$ ($\mu g/m^3$) and PN* ($particles/cm^3$) for the
785 wet and dry seasons, and on school and non-school days.

	Particles	Minimum	Maximum	Median	Mean	SD
Wet season	PM_{10}	10	64	21	27	15
	$PM_{2.5}$	3	49	10	13	9
	PN	1.28×10^3	4.35×10^3	2.57×10^3	2.56×10^3	7.25×10^2
Dry season	PM_{10}	11	158	22	36	34
	$PM_{2.5}$	5	147	17	29	33
School days	PM_{10}	10	158	21	32	27
	$PM_{2.5}$	4	147	12	20	25
	PN	1.28×10^3	4.35×10^3	2.51×10^3	2.47×10^3	7.11×10^2
Non-school days	PM_{10}	11	75	22	28	18
	$PM_{2.5}$	3	62	15	20	17
	PN	2.22×10^3	3.78×10^3	2.72×10^3	3.00×10^3	3.36×10^2

786 *No PN data for the dry season due to instrument malfunction. SD: standard deviation
787

788 Table S.2: Summary statistics for VOCs, carbonyls and NO_2 ($\mu g/m^3$) for the wet and
789 dry seasons.

Compounds	Wet season				Dry season			
	Min	Max	Mean	SD	Min	Max	Mean	SD
Benzene	0.00	0.14	0.04	0.05	0.03	0.22	0.10	0.08
1,2-dichloropropane	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Trichloroethylene	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00
Toluene	0.00	1.06	0.27	0.35	0.05	0.36	0.14	0.12
Chlorobenzene	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Ethylobenzene	0.00	0.00	0.00	0.00	0.00	0.04	0.01	0.02
(m+p)-xylene	0.00	0.05	0.02	0.02	0.00	0.12	0.02	0.04
Styrene	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
o-xylene	0.00	0.02	0.01	0.01	0.00	0.05	0.01	0.02
α-pinene	0.00	6.35	0.88	2.21	0.00	0.89	0.12	0.31
1,2,4-trimetylobenzene	0.00	0.22	0.05	0.09	0.00	0.07	0.01	0.02
1,4-dichlorobenzene	0.06	0.69	0.31	0.24	0.48	0.78	0.57	0.11
Limonene	0.00	0.91	0.22	0.35	0.00	0.18	0.06	0.07
Formaldehyde	1.62	4.08	2.37	0.81	0.85	5.40	2.41	2.03
Acetaldehyde	0.61	1.65	1.01	0.37	0.47	2.97	1.17	1.20
Acrolein	0.00	0.68	0.09	0.24	0.00	0.00	0.00	0.00
Acetone	0.31	1.06	0.54	0.26	0.32	2.54	1.05	1.00
Propionaldehyde	0.00	0.49	0.12	0.22	0.00	0.75	0.19	0.37
Butyraldehyde	3.80	5.41	4.55	0.60	0.00	0.00	0.00	0.00
Hexaldehyde	2.27	18.12	6.22	5.12	0.00	1.35	0.34	0.68
NO_2	0.00	0.20	0.07	0.07	0.76	5.17	1.73	1.61

790 Min: Minimum, Max: Maximum, SD: standard deviation

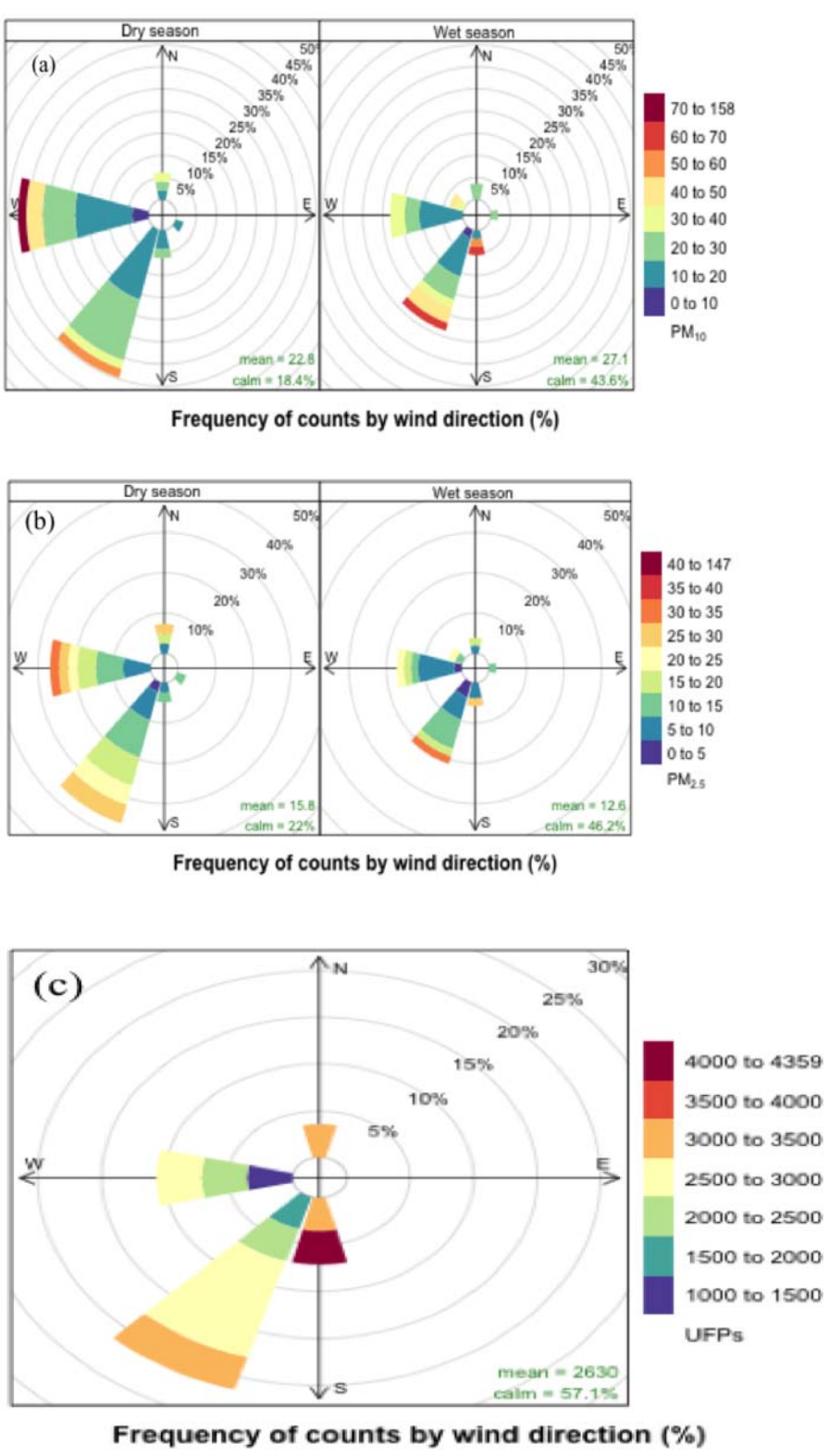

Figure S.2: Pollution rose diagrams as function of wind direction (a) for PM_{10}, (b) for $PM_{2.5}$ and (c) for PN.

833 Table S.3: Correlation matrix for pollutant concentrations and rainfall/wind speed.

Pollutants	Wet season		Dry season	
	Rainfall	Wind speed	Rainfall	Wind speed
PM_{10}	-0.43**	0.04	0.15	-0.62**
$PM_{2.5}$	-0.30*	0.10	0.23	-0.62**
PN	-0.49**	0.11	NM	NM
Formaldehyde	-0.83**	0.00	-0.40	0.20
Acetaldehyde	-0.52	0.26	-0.20	0.40
Acetone	-0.55	-0.32	-1.00**	0.80
Butyraldehyde	-0.50	0.36	ND	ND
Hexaldehyde	-0.12	-0.05	0.78	-0.26
NO_2	-0.02	-0.14	-0.17	0.17

834
835 NM: No measurement, ND: Not detected
836 ** $p < 0.01$, * $p < 0.05$
837

838 Table S.4: Comparison of mean PM_{10}, $PM_{2.5}$ and PN concentrations.

Study	PM_{10} ($\mu g/m^3$)	$PM_{2.5}$ ($\mu g/m^3$)	PN (particles/cm^3)
This study	32	21	2.561×10^3
Lee and Chang (2000)	200		
Stranger et al. (2008)		52	
Guo et al. (2008)			2.90×10^4
Weichenthal et al. (2008)			1.40×10^4
Wichmann et al. (2010)		10	
Mullen et al. (2011)			1.80×10^4
Raysoni et al. (2013)	28	12	
Zwoździak et al. (2013)	59	49	
Buonanno et al. (2013)			1.04×10^4
Rivas et al. (2014)		29	2.34×10^4

839
840 This Study: Bhutan, mean for 1 rural school, weekly sampling. Lee and Chang
841 (2000): Hong Kong, 1 rural school with light industrial area, 24 hours sampling.
842 Stranger et al. (2008): Antwerp, Belgium, mean for 27 schools (15 urban and 12
843 suburban), 24 hours sampling. Wichmann et al. (2010): Stockholm, Sweden, mean for
844 6 urban schools, 14 days sampling. Raysoni et al. (2013): El Paso, USA, mean for 4
845 urban schools, 48 hours sampling. Zwoździak et al. (2013): Warclaw, Poland, 1
846 school, 24 hours sampling. Rivas et al. (2014): Barcelona, Span, mean for 39 urban
847 schools, weekly sampling. Weichenthal et al. (2008): Ontario, Canada, 1 rural school,
848 7 hours sampling. Guo et al. (2008): Australia, 1 school located in a rural area with
849 low level of local traffic, monitoring from 4/9/2006 to 29/9/2006. Mullen et al.
850 (2011): California, USA, 6 elementary schools, 9 hours average. Buonanno et al.
851 (2013): Cassino, Italy, 3 schools (2 urban and 1 rural), 8 hours average.

852

Table S.5: Comparison of mean VOCs, formaldehyde and NO_2 concentrations ($\mu g/m^3$).

Study	1	2	3	4	5	6	7	8	9	10	11	12
This study*	0.07	0.02	0.01	0.02	ND	0.01	0.50	0.03	0.44	0.14	2.38	1.39
Godwin and Batterman (2007)	0.06	0.52	<0.01	0.00	<0.01	<0.01	0.11	0.01		0.29		
Gennaro et al. (2013)*	0.52 to 1.51	1.16 to 4.25	0.19 to 1.70	0.41 to 2.38	0.12 to 1.20		0.03 to 0.08		0.01 to 0.07	0.36 to 32.15		
Raysoni et al. (2013)	0.52 to 1.51	1.16 to 4.25	0.27 to 0.86	0.68 to 2.26		0.28 to 0.88						
Demirel et al. (2014)	0.75 to 1.30	0.39 to 27.86	0.01 to 0.22	0.14 to 0.56		0.28 to 0.88						18.28
Stranger et al. (2008)*	1.12	3.00	0.52	1.51		0.47						64
Rivas et al. (2014)												47
Sofuoglu et al. (2011)											5.59	
Jovanović et al. (2014)											5.07	
Wichmann et al. (2010)												20.50

1: Benzene, 2: Toluene, 3: Ethylbenzene, 4: (m+p)-xylene, 5: Styrene, 6: o-xylene, 7: α-pinene, 8: 1,2,4-trimethylbenzene, 9: 1,4 dichlorobenzene, 10: Limonene, 11: Formaldehyde, 12: NO_2; *used radiello samplers, other studies used different samplers.

This Study: Bhutan, mean for 1 rural school, weekly sampling. Godwin and Batterman (2007): Michigan, USA, mean for 9 suburban schools, 4.5 days sampling. Gennaro et al. (2013): Bari, Italy, mean range for 8 urban schools, weekly sampling. Raysoni et al. (2013): El Paso, USA, mean for 4 urban schools, 48 hours sampling. Demirel et al. (2014): Eskisehir, Turkey, mean for 2 schools (1 urban and 1 suburban), 24 hours sampling. Stranger et al. (2008): Antwerp, Belgium, mean for 27 schools (15 urban and 12 suburban), 24 hours sampling. Rivas et al. (2014): Barcelona, Span, mean for 39 urban schools, weekly sampling. Sofuoglu et al. (2011): Izmir, Turkey, mean for 3 schools (2 urban and 1 suburban), 5 hours sampling. Jovanović et al. (2014): Zajecar, Serbia mean for 1 school in residential area, 10 days sampling. Wichmann et al. (2010): Stockholm, Sweden, mean for 6 urban schools, 14 days sampling.

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