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QUANTITATIVE ASSESSMENT OF AIR QUALITY IN DIFFERENT INDOOR AND OUTDOOR ENVIRONMENTS IN RURAL BHUTAN

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Abstract

Health effects of air pollution are driven by exposure, which is a function of pollutant concentration and the duration for which a person has been in contact with the pollutant. People are exposed to air pollutants in various microenvironments, importantly at homes, schools and on roads during commuting. Therefore, quantification and characterization of air quality in these environments are crucial for assessing human exposure and evaluating the health risks.

Although studies have been conducted in developing countries focussing on the above mentioned microenvironments, there are several deficiencies with the current literature. In relation to residential environments, the majority of studies have focused on particle mass concentrations. Hardly any study has characterized particle number (PN) concentrations in houses relying on biomass fuels for cooking and heating. Also, only a limited number of studies have reported emission rates from biomass stoves under real-world conditions.

Children are more sensitive to air pollution because of their developing organs and because they breathe more air relative to their body size compared to adults. Outside their homes, children spend the highest time at schools, compared to other environments. Although the indoor environment of schools present a number of pollution sources, many studies have highlighted significant contributions from outdoor sources to indoor levels in schools. Only a handful of studies focussing on conventional pollutants for schools located in major cities and near busy traffic intersections have been conducted in developing countries. Air quality in rural schools, which can be affected by both local sources and long-range transport, has not received any attention.

Human exposures in transportation microenvironments have been less investigated in developing countries. Mobile methods have been successfully used for measuring on-road pollution levels during real-world driving. A few on-road and in-vehicle exposure studies have been done along major urban roads using mobile methods. Apart from traffic emissions,

on-road pollution levels along major roads in rural areas can be affected by local sources such as agricultural and residential wood combustion, and also by long-range transport. This can lead to different exposure patterns and risks compared to primarily vehicle emissions along urban roads. Since road commuting and settlements near roadsides are becoming increasingly common in rural areas, studies characterizing air quality on rural roadways are needed.

Considering the above knowledge gaps, this PhD project undertook a quantitative assessment of air quality in three environments in rural Bhutan (i) in homes, (ii) a school outdoor environment, and (iii) a major roadway, with the following aims.

1. To quantitatively characterize indoor biomass combustion products during cooking and heating in village homes.
2. To characterize and quantify outdoor air quality of a rural school and assess the factors driving pollutant concentrations.
3. To characterize and quantify on-road air quality along the East-West Highway and assess the contributions by different sources.

The first part of the project characterized pollutant concentrations and emission rates from biomass stoves during heating and different cooking activities (rice cooking, fodder preparation and liquor distillation) in four village homes. Results showed that concentrations of $PM_{2.5}$, PN and CO were significantly elevated when stoves were operated, compared to background levels. Extremely high PN concentrations, exceeding 1×10^6 particles/cm^3, which were beyond the upper resolution of the NanoTracer used for PN measurement were observed, when stoves were operated. The $PM_{2.5}$ and CO emission rates were several orders of magnitude higher than the emission rate targets recommended by the recent "World Health Organisation (WHO) guidelines for indoor air quality: household fuel combustion". Further, it was observed that both the pollutant concentrations and emission rates were more dependent on cooking activities than stoves types, with the highest during liquor distillation, which was more time and fuel intensive compared to other cooking activities. Interventions

focussing on dissemination of improved stoves, supported by education on adaptations of customary household activities would lead to significant reduction in exposure to household air pollution (HAP).

The second part of the project characterized air quality in the outdoor environment of a rural school and assessed the factors driving its concentrations. The measurements for PM_{10}, $PM_{2.5}$, PN, CO, VOCs, carbonyls and NO_2 were conducted for 16 weeks, spanning the wet and dry seasons. The overall mean concentrations of both PM_{10} and $PM_{2.5}$ were higher during the dry season than the wet season. However, mean concentration of both the particle mass fractions exceeded the WHO annual guidelines even during the wet season. The PN results were available only for the wet season, which presented a concentration comparable with the worldwide ambient clean background. Likewise, mean NO_2 concentration was higher during the dry season than the wet season. Mean CO concentrations were below the detection limit of the instrument throughout the measurement period. Only low levels of eight VOCs were detected, while dominant carbonyls were formaldehyde and hexaldehyde. The pollutant concentrations were associated with a number of factors, such as cleaning and combustion activities in and around the school.

The third part of the project characterized on-road air quality and dominant sources for the East-West Highway (EWH). Repeated measurements were conducted along the 570 km stretch of the EWH, using a mobile platform method. The entire length of the EWH was divided into five segments (R1-R5), taking the road length between two district towns as a single road segment. The on-road concentrations of PM_{10}, PN and CO were always higher for R5 than other road segments, which is the final segment of the EWH towards the capital, which also presented the highest traffic volume. The major pollution sources, apart from vehicle emissions were roadworks, unpaved roads, and roadside combustion activities. Overall, the highest contribution above the background levels were made by unpaved roads for PM_{10}, and vehicle emissions for PN and CO.

In summary, the findings of this study confirmed significant air quality problems in the three investigated rural environments in Bhutan. The pollutant concentrations in residences during open wood fire cooking were extreme, with concentrations remaining elevated even after the activity had ceased. The mean concentrations of PM_{10} and $PM_{2.5}$ in the outdoor environment of the investigated school were higher than the WHO recommended guidelines. The EWH presented a diversity of both combustion and non-combustion sources, contributing to high pollution levels, especially the PM_{10} and PN. Therefore, people are potentially exposed to high concentrations of multiple pollutants at homes, school, and while commuting along the EWH.

Findings are scientifically novel in that this is the first major scientific investigation of air quality in Bhutan, which will be a significant input for informing policies. Since the entire Himalayan region has similar environmental and social characteristics, the findings of this study will be of significance to other countries in the region in evaluating human exposure in various microenvironments. Further, this is one of the first studies to report emission rates from biomass stoves during cooking and heating under real-world operating conditions, and to validate the robustness of the mobile sampling method for characterization of on-road air quality for a major rural roadway. Finally, the study has provided additional insights about air pollution problems in rural areas.

Keywords

Bhutan, Rural, Emission rate, Firewood, Stoves, School, Seasons, Outdoor, On-road, Mobile platform, PM_{10}, $PM_{2.5}$, Particle Number, Nitrogen dioxide, Carbon monoxide, Volatile Organic Compounds, Carbonyls

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List of Publications

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Wangchuk, T., He, C., Dudzinska, M. R., Morawska, L., 2015. Seasonal variations of outdoor air pollution and factors driving them in the school environment in rural Bhutan. *Atmospheric Environment* 113, 151-158.

Wangchuk, T., Knibbs, L. D., He, C., Morawska, L., 2015. Mobile assessment of on-road air pollution and its sources along the East-West Highway in Bhutan. *Atmospheric Environment* 118, 98-106.

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Acronyms

AER: Air Exchange Rate

BEA: Bhutan Electricity Authority

BTEX: Benzene, Toluene, Ethylbenzene & Xylene

CF: Correction Factor

CNG: Compressed Natural Gas

COHb: Carboxyhaemoglobin

CPC: Condensation Particle Counter

DALYs: Disability-Adjusted Life Years

DoE: Department of Energy

ERT: Emission Rate Target

EWH: East-West Highway

GRADE: Grading of Recommendations Assessment, Development & Evaluation

HAP: Household Air Pollution

IAQ: Indoor Air Quality

ILAQH: International Laboratory for Air Quality & Health

LOAEL: Lowest Observed Adverse Effect Level

LPG: Liquefied Petroleum Gas

MEL: Mobile Emission Laboratory

NEC: National Environment Commission

NO_x: Oxides of Nitrogen

NOAEL: No Observed Adverse Effect Level

NT: NanoTracer

NSB: National Statistical Bureau

PAHs: Polycyclic Aromatic Hydrocarbons

PN: Particle Number

RGoB: Royal Government of Bhutan

RSTA: Road Safety and Transport Authority

SMPS: Scanning Mobility Particle Sizer

TEOM: Tapered Element Oscillating Microbalance

TSP: Total Suspended Particles

UFP: Ultrafine Particles

UNDP: United Nations Development Programme

VOCs: Volatile Organic Compounds

WHO: World Health Organisation

Statement of Original Authorship

The work contained in this thesis has not been previously submitted to meet requirements for any award at any other higher education institution. To the best of my knowledge and belief, the thesis contains no material previously published or written by another person except where due reference is made.

Signature:

(Tenzin Wangchuk)

Date: 26 January 2016

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Chapter 1: Introduction

1.1 Description of scientific problems investigated

Air pollution presents a major public health risks throughout the world. There are numerous epidemiological studies that report both short and long-term health effects of pollutants originating from a diversity of indoor and outdoor sources, for example (Hussein et al., 2005, Pope III and Dockery, 2006, WHO, 2006, Kattan et al., 2007, Fullerton et al., 2008, Kim et al., 2011, Buonanno et al., 2013, WHO, 2014). The health effects of air pollution are driven by the exposure to pollutants in various microenvironments. Important microenvironments where people spend a majority of their time include indoor spaces such as homes, work places, schools and inside vehicles (WHO, 2000), while in rural areas of many developing countries, people spend significant amount of time outdoors such as on farms and construction sites. Quantification and characterization of air quality and human exposure in different indoor and outdoor microenvironments where people spend time, are essential to evaluate the health risks (Morawska et al., 2001).

Air pollution problems are much greater in developing countries than the developed world. A large percentage of the population in rural areas rely on solid fuels for cooking and heating using inefficient traditional stoves, where incomplete combustion of fuels result in severe household air pollution (HAP) (WHO, 2014). In relation to HAP, the studies to date in developing countries have mostly focused on particle mass, with only limited studies characterizing particle number (PN) concentrations. Compared to particle mass, ultrafine particles (UFP, < 100 nm) have higher deposition rate in the alveolar region, enabling easy translocation into the blood stream, and could potentially exhibit higher toxicity than particle mass (WHO, 2005, Buonanno et al., 2012). Also, only very few studies have estimated emission rates from stoves and fuels used for cooking and heating. The recently released World Health Organisation (WHO) guidelines for household fuel combustion, among other measures, recommend emission rate targets for stoves (example, $PM_{2.5}$ and CO) to meet the

WHO guidelines for indoor air quality (WHO, 2014). However, studies are needed with more focus on characterization of PN concentrations and emission rates from stoves under real-world conditions.

In general children are more sensitive to pollutants given their weak immune system and developing organs (Raysoni et al., 2013, Demirel et al., 2014). Apart from homes, children spend a significant portion of their time at schools (Buonanno et al., 2012, Mazaheri et al., 2014), exposed to heterogeneous pollutants originating from both indoor and outdoor facilities. Therefore, school based air quality investigations are important to assess the contribution to children's daily exposure and to reduce health risks. To date, only a handful of studies focussing on air quality in schools have been conducted in developing countries. Further, these studies were conducted in schools located in major cities and near busy traffic intersections, for example (Zhao et al., 2008, Tippayawong et al., 2009, Chithra and Shiva Nagendra, 2012). So far, air quality investigations for schools located in rural areas have not been reported. In addition to local sources, rural areas can be affected by long-range transport of regional and urban pollutants. Hence, there is an urgent need to study the air quality in schools located in rural areas.

Since vehicle emissions contribute significantly to air pollution, transportation microenvironments such as on roads, inside vehicles, and roadside locations experience the highest pollution levels. Because many people commute daily by private cars and public transports, commuter exposure on roadways are crucial to understand the health risks. Measurements along roadways are challenging since it would require a spatially dense network of monitoring sites to capture the spatial and temporal distributions of pollution levels. To address this challenge vehicle-based mobile measurements have been successfully used along urban roadways, including tunnels. In developing countries, human exposure in transportation microenvironments are far less studied, with only a handful of on-road and in-vehicle exposure studies done along urban roads in a few large cities, for example (Wang et

al., 2009, Apte et al., 2011, Colbeck et al., 2011, Both et al., 2013). Pollution levels along major roadways in rural areas, apart from vehicle emissions, can be influenced by local sources such as agricultural and residential wood burning, as well as by long-range transport. Mobile on-road studies are needed for major roadways in rural areas to understand commuter exposure, as well as exposure for roadside residents.

In Bhutan, there is a growing concern about the health and environmental impacts of air pollution due to rapid urbanisation and transport infrastructure development taking place throughout the country. In the villages, where nearly 70% of the population live, a major indoor air quality problem is anticipated due to household use of biomass fuels for cooking and space heating. However, to date no quantitative air quality study has been conducted in Bhutan. Without experimental measurements, actual air quality problems and the resulting health burden cannot be assessed. From the policy perspective and for effective management of the problem, good scientific research findings are essential. Therefore, scientific air quality investigations are needed sooner than later in Bhutan.

In summary, this thesis reports air quality investigations done for homes, a school environment and on the major roadway in Bhutan. This is important since no scientific investigation of air quality has been done in the country so far. Since Bhutan shares similar environmental and social characteristics with that of other Himalayan regions, findings from this study will be of significance for other countries in the region.

1.2 Overall aims of the study

Considering the research problems and gaps in the current literature, which are defined above, the overall aim of this research project was to characterize and quantify pollution levels in different indoor and outdoor microenvironments in rural Bhutan. In particular, the study has the following specific aims:

4. To quantitatively characterize indoor biomass combustion products during cooking and heating in village homes in Bhutan.

5. To characterize and quantify outdoor air quality of a rural school in Bhutan and assess the factors driving the pollutant concentrations.
6. To characterize and quantify on-road air quality along the East-West Highway in Bhutan and assess the contributions by different sources.
7. To establish whether pollutant concentrations in the three dominant microenvironments investigated (homes, school outdoor, and on-road) presented a potential health risks to people.

1.3 Specific objectives

The specific objectives in relation to the above mentioned aims are:

To achieve aim 1:

- To quantify pollutant concentrations ($PM_{2.5}$, PN and CO) during cooking and heating in rural homes.
- To estimate emission rates ($PM_{2.5}$, PN and CO) from biomass stoves used for cooking and heating under real-world operating conditions.

To achieve aim 2:

- To characterize the sources and concentrations of PM_{10}, $PM_{2.5}$, PN, VOCs, carbonyls and NO_2 in the outdoor environment of a rural school.
- To assess the factors driving the sources and concentrations of pollutants in a school outdoor environment.
- To assess the seasonal variations of pollutant concentrations in relation to meteorological parameters, wind speed, wind direction, and rainfall.

To achieve aim 3:

- To identify principal pollution sources and concentrations (PM_{10}, PN, and CO) using a mobile platform method for the East-West Highway.
- To determine the contribution of on-road and other proximate sources to on-road pollution levels.

1.4 Account of scientific progress linking the research papers

In order to address the above outlined aims and objectives, air quality investigations were conducted in homes, at a school and for a major roadway, representing the three important microenvironments where people spend a significant amount of time. Four scientific papers have been developed from the research activity in Bhutan (see the list of publications in the preceding section). The papers have been cohesively integrated to form various chapters of the thesis. Figure 1.1 presents the schematic representation of aims and the methodologies used in the study.

The first paper describes HAP measurements conducted in village homes in Bhutan. In particular, the paper characterized $PM_{2.5}$, PN, CO and CO_2 concentrations, including temperature and relative humidity during cooking and heating using biomass stoves in four homes, capturing common stove types and cooking activities. Measurements were conducted in three stages, with 45 – 60 minutes of background measurements before the heating/cooking activities were started, followed by measurements during the activity, and again at least an hour after the activity has ceased. Air exchange rate for each house was estimated based on CO_2 decay after the activity has ceased. The measured $PM_{2.5}$, PN and CO concentrations were used for estimating emission rates. A complete methodology, results and discussions are presented in the paper, titled "A pilot study of traditional biomass cooking and heating in rural Bhutan: gas and particle concentrations and emission rates". This paper is under review in *Indoor Air* and forms Chapter 3 of the thesis.

The second paper characterized pollution sources and quantified concentrations of PM_{10}, $PM_{2.5}$, PN, VOCs, Carbonyls, and NO_2, including temperature and relative humidity for a school outdoor environment. Measurements were conducted for 16 weeks spanning wet and dry seasons. The factors driving the pollutant concentrations were assessed in relation to activities conducted in the school, as well as outside the school, and correlated with meteorological parameters. A complete methodology, results and discussions are presented in

the paper, titled "Seasonal variations of outdoor air pollution and factors driving them in the school environment in rural Bhutan". This paper has been published in the Journal, *Atmospheric Environment, Vol. 113, pp. 151-158,* and forms Chapter 4 of the thesis.

The third paper characterized the on-road air quality along the 570 km stretch of the East-West Highway using a mobile platform method. A total of six measurements of PM_{10}, PN and CO were conducted for the complete length of the EWH during the real-world journey. The source contributions to on-road pollution levels were estimated based on interception of on-road and proximate sources by the research vehicle and by accounting for the time and location of sources. A complete methodology, results and discussions are presented in the paper, titled "Mobile assessment of on-road air pollution and its sources along the East-West Highway in Bhutan". This paper has been published in the Journal, *Atmospheric Environment, Vol. 118, pp. 98-106,* and forms Chapter 5 of the thesis.

The fourth paper highlights HAP problems in developing countries and the significance of new WHO guidelines for household fuel combustion for tackling the health burden. In particular, this paper has identified specific research areas in developing countries in relation to HAP, using the current situation in Bhutan as an example. A complete paper, titled "Tackling the health burden from household air pollution: Development and implementation of new WHO guidelines" has been published in the journal, *Air Quality and Climate Change* 47, *pp. 32-38,* and forms Chapter 6 of the thesis.

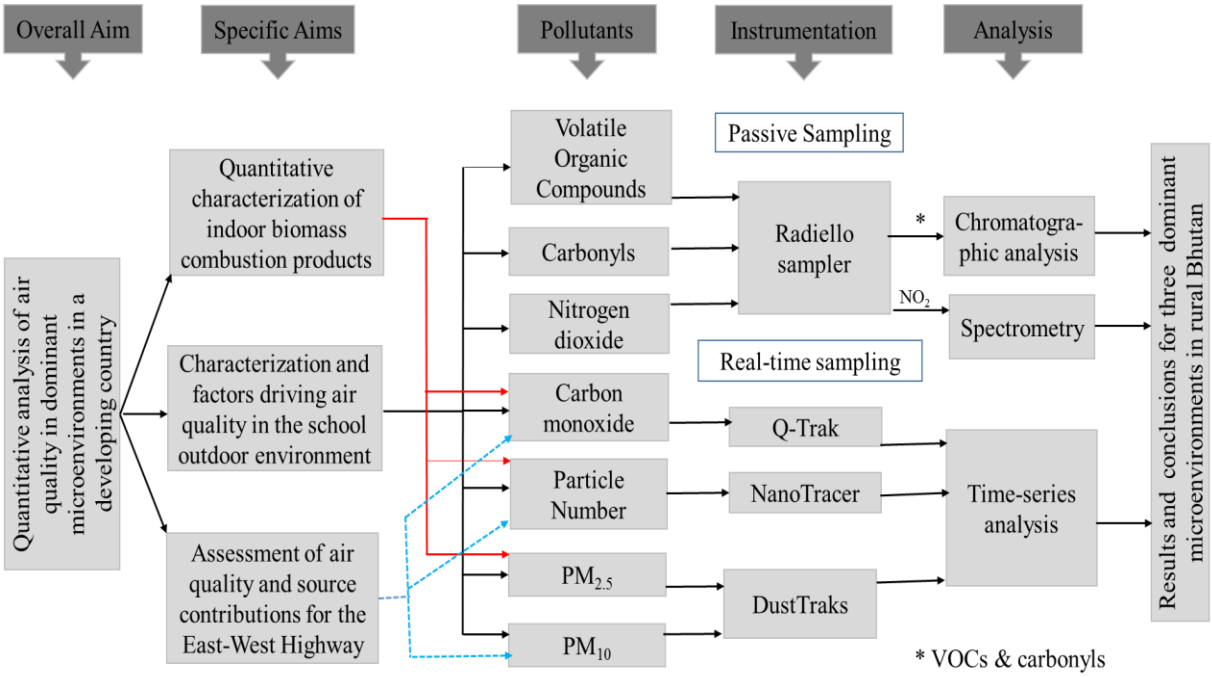

Figure 1.1: Schematic representation of the methodologies used in the study.

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Chapter 2: Literature Review

2.1 Background

Atmospheric pollution is not a contemporary issue. The presence of soot in prehistoric caves (Spengler and Sexton, 1983) and blackened lung tissues observed in preserved ancient human remains, similar to the lung disease suffered by 19th century coal miners (Brimblecombe, 2011), present strong historical evidences of air pollution and its effects on human health. In London, regulations to reduce outdoor air pollution from smoke were introduced as early as the 14th century (Spengler et al., 2001c). Following the industrial revolution, some of the worst pollution disasters in the 20th century were demonstrated by episodes in the Meuse valley in 1932; Donora, Pennsylvania in 1948 and the London smog in 1952, causing thousands of deaths (Spengler et al., 2001c). Burning coal by households was the main cause of Great London Smog, which eventually lead to the banning of household coal fires in London (Zhang and Smith, 2007). Well into the 21st century, air pollution is still the leading cause of the high global burden of disease. Despite conclusive scientific understanding of causes and impacts of air pollution, and abatement strategies in place, it still remains as one the major environmental and public health risks throughout the world.

Emissions from the ever increasing number of vehicles and industries are the primary source of ambient air pollution in urban settings. The use of solid fuels (wood, crop residues, animal dung, charcoal, and coal) for cooking and heating is the leading cause of HAP in rural areas. In fact, the recent WHO report has estimated 4.3 million premature deaths due to HAP and another 3.7 million from ambient air pollution in 2012 (WHO, 2014b). Although the majority of deaths occurred in low and middle-income countries, a significant number of deaths was also reported from high-income countries.

This literature review chapter focuses on air quality problems in three microenvironments: (i) homes, (ii) schools and (iii) transportation microenvironments. Because air quality issues are severe in developing countries, the review centres mostly on

the scientific knowledge and understanding from developing countries. Since this PhD project was conducted in Bhutan, the air quality problems specific to Bhutan have also been discussed. Additionally, cross-cutting concepts in air quality studies, the physical and chemical characteristics of common pollutants, the concept of human exposure and indoor-outdoor relationships have also been discussed.

2.2 Physical and chemical characteristics of pollutants that are of concern in indoor and outdoor environments

Among the numerous pollutants that are emitted by a diversity of indoor and outdoor sources, the common air pollutants that have received attention due to health significance are particulate matter (PM), carbon monoxide (CO), sulphur dioxide (SO_2), nitrogen dioxide (NO_2), ozone (O_3), lead (Pb), volatile organic compounds (VOCs), and polycyclic aromatic hydrocarbons (PAHs) (Han and Naeher, 2006). The physical and chemical characteristics of those that are significantly associated with health effects are discussed in the following sections.

2.2.1 Particulate matter

Particulate matter is a complex mixture of solid particles and liquid droplets, with varying physical and chemical characteristics. Primary particles are those that are directly introduced into the atmosphere in a solid or liquid state, while secondary particles are formed in the air by the transformation of gaseous precursors (Kulkarni et al., 2011). The particle size from combustion sources range from 0.001 – 100 μm (Chow et al., 2002), and largely explains its behaviour in the atmosphere and the associated health effects (Morawska and Zhang, 2002).

In many urban areas, PM_{10} and $PM_{2.5}$ are regularly measured for routine monitoring (Thorpe and Harrison, 2008). PM_{10} (coarse particles) constitutes those with aerodynamic diameter less than 10 μm (Chow et al., 2002, Morawska and Zhang, 2002, Kulkarni et al., 2011). In the outdoor air, coarse particles mostly originate from construction activities,

farming, mining, wind storms and resuspension of dusts by wind and traffic, while indoor sources include resuspension of floor dust and handling textiles (Jantunen et al., 1999, Pope III and Dockery, 2006). Natural sources of coarse particles includes pollens, mould spores and other biogenic particles (D'Amato et al., 2002). The chemical composition of coarse particles mostly includes soil minerals, non-volatile organics and fibres from textiles (Jantunen et al., 1999, Morawska and Zhang, 2002). The average lifetime of coarse particles in the atmosphere varies typically from minutes to hours and can travel hundreds of kilometres in the air (Jantunen et al., 1999).

$PM_{2.5}$ (fine particles) constitute those with aerodynamic diameter less than 2.5 μm (Chow et al., 2002, Morawska and Zhang, 2002, Kulkarni et al., 2011). The fine particles in the outdoor environments are introduced by combustion activities such as fossil fuels (by vehicles, power plants and other industries), and biomass, while indoor sources include cooking and heating fuels (e.g. wood, kerosene, gas) and tobacco smoke (Tuckett et al., 1998, D'Amato et al., 2002, Pope III and Dockery, 2006). The chemical composition of fine particles can range from soot to acid condensates, sulphates, nitrates and PAHs (Jantunen et al., 1999, Morawska and Zhang, 2002). The average lifetime of fine particles in the atmosphere ranges from a few minutes to weeks, and can travel thousands of kilometers (Jantunen et al., 1999).

Many urban centres around the world have started measuring submicrometre particles (< 1.0 μm) and UFP since the 1990s (Mejía et al., 2008). While UFP make only negligible contribution to particle mass, they can constitute up to 90% of total PN concentrations (Morawska et al., 2008). UFP are generated by combustion activities in both outdoor and indoor air, and atmospheric transformations of various precursors, for example, NO_x, SO_2, NH_3 and organic vapours, and typically contain "metals, organic compounds, carbon, and acid aerosols" (Westerdahl et al., 2005). The primary UFP have a relatively short lifetime

ranging from minutes to hours and they undergo rapid transformation through coagulation and condensation to larger aggregates, typically to $PM_{2.5}$ (Pope III and Dockery, 2006).

The penetration and deposition of PM_{10} in the respiratory pathway is limited to the thoracic region, while $PM_{2.5}$ can easily reach gas exchange regions (Peltier et al., 2011). Particulate matter has been associated with mortality and morbidity from respiratory and cardiovascular diseases, exacerbation of asthma, chronic bronchitis, and hospital admissions (D'Amato et al., 2002). UFP have high alveolar deposition rate with large surface area and potential to easily get into the blood stream (Buonanno et al., 2012a). In recent years UFP have been the focus of numerous research activities around the world because of their potentially greater health effects compared to larger particle mass fractions (Kam et al., 2012). However, while health effects of particle mass have been clearly established, there is very limited understanding of the health effects of UFP, even though toxicological studies have established greater effects compared to particle mass (WHO, 2005).

2.2.2 Carbon monoxide

Carbon monoxide (CO) is a colourless, odourless and tasteless gas, emitted during incomplete combustion of carbonaceous fuels (Spengler et al., 2001b). Because of these properties, CO cannot be detected by human senses (WHO, 2010). CO is relatively inert and is not absorbed by building materials and filter systems (Jantunen et al., 1999, WHO, 2010). Important indoor sources are heating and cooking devices using kerosene and biomass fuels, and indoor tobacco smoke (WHO, 2010). In urban areas street traffic is the major source of CO (Jantunen et al., 1999). It is also emitted by natural sources such as wild fires (Han and Naeher, 2006). When indoor sources are absent, the concentration of CO is generally lower than the outdoor concentration (Jones, 1999). The toxic nature of CO comes from its affinity to haemoglobin in the blood (~ 250 times more than oxygen), resulting in the formation of carboxyhaemoglobin (COHb) and cuts oxygen supply to tissues (Spengler et al., 2001b). COHb levels as low as 5.1 – 8.2% can result in transient neurological impairments, while

levels above 25 – 35% can result in unconsciousness, and eventually death above 60% (WHO, 2010). Such high levels of COHb are not expected in urban ambient air but can easily reach these levels inside motor vehicles and indoor spaces with unvented fire sources (Jantunen et al., 1999).

2.2.3 Nitrogen dioxide

Nitrogen dioxide (NO_2) has a characteristic pungent odour, which is reddish brown and water soluble (Jones, 1999). It is an important precursor for photochemical smog and ground level O_3 formation in presence of VOCs and sunlight (Han and Naeher, 2006). Major indoor sources are tobacco smoke, gas and wood based cooking and space heating devices, while road traffic is the principal outdoor source, along with power plants and other sources burning fossil fuels (D'Amato et al., 2002, WHO, 2010). NO (nitric oxide) and NO_2 are two principal oxides of nitrogen formed during high temperature combustion. Most NO is readily oxidised to NO_2 and therefore, NO_2 is used as an index of pollution among the oxides of nitrogen (Spengler et al., 2001b). In the ambient air, oxidation of NO to NO_2 is limited by availability of ozone (O_3), and while NO exhibits large temporal and spatial variability in the urban environment, NO_2 is more uniformly distributed over a large area (Jantunen et al., 1999). In absence of sources, NO_2 levels are generally lower in the indoor environment compared to outdoor concentrations (Spengler et al., 2001b). Wheezing and acerbation of asthma, respiratory infection and reduced lung functions are some of the health effects associated with exposure to NO_2 (Kattan et al., 2007, Kim et al., 2011). It also contributes to the acidification of ecological systems (Demirel et al., 2014).

2.2.4 Volatile organic compounds

Volatile organic compounds (VOCs) are a class of organic compounds with boiling points ranging from 50 – 100 °C, lower limit and 240 – 260 °C, upper limit (WHO, 1989), and a high volatility in ambient air and room temperature (Han and Naeher, 2006, Barro et al., 2009). Because of this property, many VOCs off-gas readily, leading to a rapid reduction

in concentrations over a short time period (Jones, 1999). The primary sources of VOCs in the outdoor air are fossil fuel combustion by the transportation sector and industrial processes (Demirel et al., 2014), and gasoline loss due to evaporation from fuel stations (Jantunen et al., 1999, Lan et al., 2013). The gasoline vapor mainly contains n-butane, isopentane, n-pentane and isobutene, while tailpipe emissions consists of methane, toluene, ethylene, xylene, n-butane and benzene (Jantunen et al., 1999). The indoor sources include combustion processes such as tobacco smoke and kerosene heaters, and hosts of household products and items such as paints, adhesives, furnishing and clothing, building materials, and cleaning products, which emit a range of organic compounds (Zabiegala, 2006, Barro et al., 2009, Demirel et al., 2014). Due to their good insulation property, durability and economy associated with materials containing VOCs, they are widely used for construction (Jones, 1999), making it the dominant source, especially for modern built environments. There can be several dozen individual compounds since essentially all materials found indoors contain VOCs, including inorganic materials due to surface accumulation through the sink effect (Spengler et al., 2001d). VOCs are also emitted during cooking activities involving high temperature frying and grilling (Huang et al., 2011). On average, indoor VOCs level was found to be 2 – 5 times higher than the outdoor levels (Barro et al., 2009).

Benzene, toluene, ethylbenzene and xylene (BTEX) are classified as hazardous air pollutants and more commonly discussed from health effects point of view than other VOCs (Han and Naeher, 2006, Demirel et al., 2014). The International Agency for Research on Cancer has listed benzene as Group-I human carcinogen (Demirel et al., 2014). Therefore, the WHO and the US Environmental Protection Agency have not proposed any threshold level for benzene (WHO, 2010, Lan et al., 2013). In general, VOCs can result in both acute and chronic health effects, and most have the potential to cause sensory irritation, allergic skin reactions, fatigue and dizziness, and impairment of the central nervous system (Zhang and Smith, 2003, Barro et al., 2009). Besides human health effects, VOCs are key precursors for

tropospheric ozone and secondary aerosol formation (Król et al., 2010).

2.2.5 Aldehydes

Aldehydes belong to a class of organic compounds called carbonyls consisting of a large number of compounds with varying physical and chemical properties, but only a few compounds (e.g. formaldehyde, acrolein, acetaldehyde and glutaraldehyde) are widely used for industrial and commercial purposes (Spengler et al., 2001a). Anthropogenic sources of aldehydes include incomplete combustion of fossil and biomass fuels, with vehicles being the principal outdoor source and biomass cookstoves the principal indoor sources (Zhang and Smith, 1999, Feng et al., 2005). Besides, furniture and building materials also present a major source of indoor carbonyls (Barro et al., 2009). Carbonyls are also emitted during high temperature cooking processes involving frying and grilling (Huang et al., 2011). They are also formed as a result of secondary atmospheric oxidation of anthropogenic hydrocarbons and VOCs (Carlier et al., 1986, Feng et al., 2005). The predominant carbonyls in both urban and indoor air are formaldehyde and acetaldehyde, and are mostly associated with photochemical smog formation in urban air (Cavalcante et al., 2006, Barro et al., 2009).

Like VOCs, most carbonyls are volatile and they readily off-gas during summer and disperse in the atmosphere (Feng et al., 2005). Because they are water soluble and highly reactive, exposure to carbonyls results in sensory irritation of eyes and mucous membranes of the upper respiratory tract (Spengler et al., 2001a, Pegas et al., 2011). Formaldehyde is a carcinogen and causes nasopharyngeal cancer in humans, and therefore many countries have regulations in place for its use and production (Cavalcante et al., 2006), while acetaldehyde, benzaldehyde and acrolein are suspected to possess carcinogenic and mutagenic properties (Barro et al., 2009).

2.3 Human exposure to air pollution

Human exposure was first defined by Ott (1982) as an event when a person comes in contact with the pollutant. The further elaboration of this definition states a need to account

for the duration for which a person comes in contact with the pollutant. However, many past studies have treated measured pollutant concentrations as human exposures (Morawska et al., 2013). This approach is incorrect since it doesn't account for the duration of contact with the pollutant, which actually drives the health effects. A clear distinction between 'concentration' and 'exposure' have been provided by Morawska et al. (2013). The pollutant concentration is defined as a "numerical value of the amount of an individual pollutant per unit volume of air at a particular point in time or averaged over a period of time", while exposure is a "product of the pollutant concentration and the time over which a person is in contact with that pollutant". When only concentration data is available, to avoid confusion, Morawska et al. (2013) recommend describing it as "concentrations to which a person is/would be exposed" rather than exposure. Further, 'dose' is a different metric and should not be mistaken with 'exposure'. Dose quantifies how much of an ingested pollutant is actually delivered to the target organs leading to physiological effects, which is affected by several factors, for example inhalation rate and size of particles (Smith, 1993, Morawska et al., 2013). Thus, there can be exposure without dose, but there cannot be dose without exposure (Ott, 1982). Health effects in turn depend on the dose received, which is affected by the exposure experienced in various microenvironments (Weijers et al., 2004). The actual pathway for health effects resulting from presence of a pollutant in the environment is outlined in Figure 2.1.

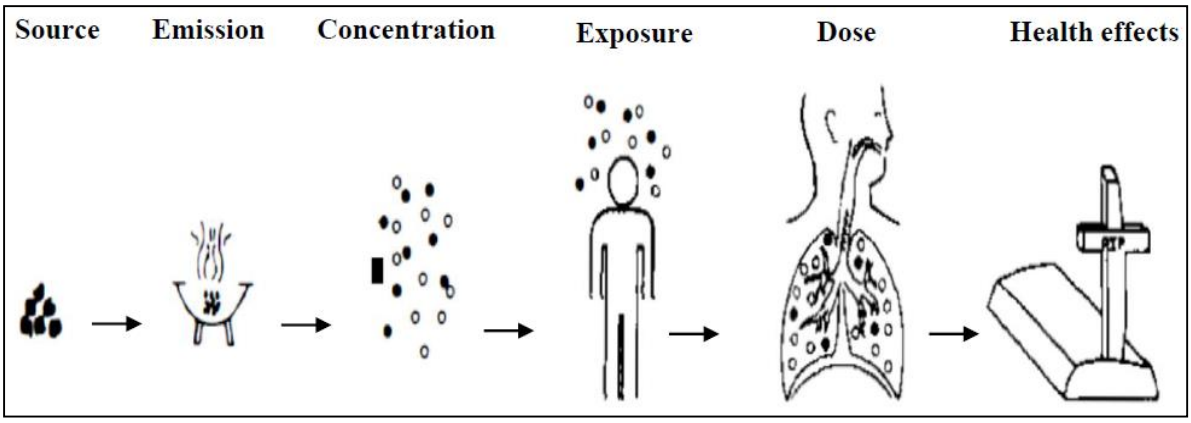

Figure 2.1: A typical pathway for health effects resulting from presence of a pollutant in the environment.
Source: (Smith, 1993).

2.3.1 Exposure assessment methods

Exposure assessment is a process of measuring and evaluating the magnitude and frequency of exposure to pollutants in various microenvironments, along with the characteristics of population, for example individual versus total population, and age and gender (Ott, 1982, Clark et al., 2013, Morawska et al., 2013).

Human exposure studies have relied on both direct and indirect assessment methods. The broadest of the indirect methods is the use of data from fixed monitoring stations to estimate exposure to outdoor pollutants (Kaur et al., 2007, Cattaneo et al., 2010). In relation to indoor exposure, indirect methods have been based on house and occupant activities as proxies for exposure estimation, such as type of fuels used (solid, liquid or gas), time spent near the fire and whether or not people were present while cooking (Ezzati and Kammen, 2002a, Parikh et al., 2003, Zhang and Smith, 2007). Yet again, some exposure studies have used microenvironment measurements, both indoors and outdoors, and time-activity patterns to estimate exposure indirectly (Balakrishnan et al., 2004, Devakumar et al., 2014). Indirect methods have the advantage when the focus is on larger population exposure, which can be done at a shorter time period and with minimal resources (Cattaneo et al., 2010). However, because indirect methods have to rely on point measurements, this approach doesn't account for the spatial variations of pollutant concentrations (Sarnat et al., 2005, Buonanno et al.,

2012a, Hinwood et al., 2014, Mazaheri et al., 2014). The degree of variability over time and space is even more pronounced for UFP, which have been shown to vary by orders of magnitude between different indoor and outdoor environments (Buonanno et al., 2011). Exposure estimated from indirect methods, especially using data from fixed monitoring stations assume that each person is exposed to the same level of a given pollutant, based on the mean values obtained (Buonanno et al., 2013b). Therefore, indirect methods can result in a significant miscalculation of exposure (Saarela et al., 2003, Kaur et al., 2007, Dionisio et al., 2012, Buonanno et al., 2013b, Clark et al., 2013).

In contrast, direct methods have the advantage of providing the real exposure experienced by people. Exposure studies in the past have been limited by the complexity of instrumentation required for measurements. Light portable monitors are now available, which can be conveniently attached to peoples' clothing or the body, enabling direct exposure measurements from various microenvironments where people spend time (Jantunen et al., 2002, Morawska et al., 2013). Personal sampling is the term used when exposure is measured directly from the person's breathing zone (Cattaneo et al., 2010). Depending on the instrumentation, time-integrated and real-time approaches can be used to assess the personal exposure (Clark et al., 2013). The time integrated method provides average exposure for the duration of measurement, without accounting for peak concentrations and temporal distributions (Bruce et al., 2004, Baumgartner et al., 2011). Real-time methods coupled with time-activity data have been used to assess short-term peaks, contributions from different activities/microenvironments, and temporal variations in exposure (Buonanno et al., 2012a, Mazaheri et al., 2014). However, direct exposure assessments are time consuming and expensive, and they are also not feasible for measuring more than one pollutant due to the inconvenience of attaching several samplers close to person's breathing zone (Cattaneo et al., 2010).

2.4 Relationship between indoor-outdoor pollution

Indoor pollution levels are significantly influenced by outdoor sources (Lee and Chang, 2000, Massey et al., 2012, Amato et al., 2014). This is because outdoor pollutants are easily transported indoors through building ventilation systems (Chithra and Shiva Nagendra, 2012, Rivas et al., 2014). Three common types of building ventilation systems: mechanical, natural, and infiltration were described in Chen and Zhao (2011). Mechanical ventilation involves forced supply of fresh outdoor air into the indoor space. This process can promote ingress of outdoor particles, since filters used in mechanical systems are not necessarily efficient to particles entry. Natural ventilation refers to free movement of outdoor air through open doors and windows, resulting in large air exchange rates and consequently drawing in more outdoor pollutants inside. Infiltration involves air flow through structural defects associated with buildings (gaps and cracks), and since the air exchange rate under such conditions is relatively low, the flow transports relatively less pollutants (Chen and Zhao, 2011).

Kang et al. (2006) found consistent variations in indoor concentrations of particles with variation of outdoor levels. Mullen et al. (2011a) found higher PN concentrations indoors when ventilation rates were high, corresponding to higher outdoor concentrations. Likewise, Lee and Chang (2000) have linked higher PM_{10} concentrations indoors (with natural ventilation) to infiltration from outdoors. Therefore, studies investigating indoor pollution levels should examine the extent to which outdoor sources influence indoor concentrations (Koutrakis et al., 1992, Morawska et al., 2013).

2.4.1 Indoor-outdoor ratio

Indoor-outdoor (I/O) ratio is a useful metric for characterizing the relationship between indoor and outdoor pollution levels. I/O ratio quantifies how much of the variations in indoor concentrations can be explained on the basis of concentration observed in the

outdoor air (Massey et al., 2012). This relationship is defined by the following equation (Chen and Zhao, 2011):

$$I/O = \frac{C_{in}}{C_{out}} \quad (2.1)$$

Where C_{in} = indoor pollutant concentration and C_{out} = outdoor pollutant concentration. Thus, I/O ratio >> 1 indicates dominant indoor source, I/O ratio = 1 indicates similar contribution from indoor and outdoor sources, and I/O ratio << 1 indicates dominant outdoor source (Zabiegala, 2006, Massey et al., 2012).

Some of the earliest studies on indoor-outdoor relationships conducted in developed countries were reviewed by Andersen (1972). In the 14 studies that Anderson reviewed, I/O ratios ranging from 0.2 – 1 were reported. But in general, the range of I/O ratios in developed countries are between 0.7 – 4 (Zabiegala, 2006). Subsequent studies describing I/O ratios have reported good correlation between indoor and outdoor concentrations, for example (Morawska et al., 2001, Lawrence et al., 2005, Kang et al., 2006, Massey et al., 2009, Massey et al., 2012). Morawska et al. (2001) reported I/O ratios ranging from 0.78 – 1.07 for submicrometer particles, 0.95 – 1.0 for supermicrometer particles (0.54 – 19.81 μm), and 1.01 – 1.08 for $PM_{2.5}$, respectively for residential houses in Brisbane. Based on these I/O ratios, the authors concluded that outdoor particle concentrations could be used to predict the indoor concentrations under normal ventilation, while air exchange rate should be known to predict the same during minimum ventilation. In India, average I/O ratios for different particle mass fractions were found to be close to or above 1 for roadside and rural houses, and less than 1 for urban houses (Massey et al., 2009).

2.4.2 Factors affecting indoor-outdoor relationships

The I/O ratio described by equation (1.2) does not take into account important factors affecting the indoor-outdoor relationship such as emission rates from indoor sources, pollutant loss due to diffusion and deposition, penetration factors, and air exchange rates

(Morawska and Salthammer, 2003). These factors in turn are influenced by other parameters. Air exchange rate is influenced by ventilation type, number of doors and windows, cracks and gaps associated with building structure, and meteorological conditions (discussed further in Section 2.4.3). The deposition rate and penetration factors largely depend on particle size (Manisha and Sioutas, 2004). An estimation of indoor pollutant levels by considering the above parameters is demonstrated in equation 2.2 (Koutrakis et al., 1992):

$$C_{in} = \left(\frac{P\ a\ C_{out} + \frac{Q_{is}}{V}}{a + k} \right) \tag{2.2}$$

Where, C_{in} = indoor concentration, P = penetration factor (dimensionless), a = air exchange rate, C_{out} = outdoor concentration, Q_{is} = mass flux generated from indoor sources (indoor particle generation rate), V = volume of the house, k = decay rate due to diffusion and sedimentation.

When there are multiple indoor sources, equation (2.2) can be rearranged to give the following equation (Morawska and Salthammer, 2003):

$$C_{in} = \frac{P\ a\ C_{out} + V^{-1} \sum_{i=1}^{n} Q_{is}^{i}}{a + k} \tag{2.3}$$

Where n = number of investigated indoor sources, and Q_{is}^{i} = mass flux generated by the indoor source i.

For houses with natural ventilation, when no indoor sources are present, the main factors affecting the indoor particle concentrations are penetration factors, deposition rates, and air exchange rates (Morawska and Salthammer, 2003). The equation for the same can be written as:

$$C_{in} = \frac{P\ a\ C_{out}}{a + k} \tag{2.4}$$

The equation 2.4 has been used to determine *P* and *k* in experimental studies in which indoor and outdoor concentrations have been measured (Morawska and Salthammer, 2003). Further, the above equation can be rearranged to provide the I/O ratio, which is equivalent to infiltration factor (defined as equivalent fraction of ambient pollutant penetrating indoors). Thus equation 2.4 can be expressed as:

$$F_{INF} = \frac{C_{in}}{C_{out}} = \frac{P\,a}{a+k} \tag{2.5}$$

These equations do not take into account chemicals processes such as condensation, evaporation or coagulation that may be associated with pollutants, since these are not significant, particularly in residential houses (He et al., 2004, Buonanno et al., 2009).

2.4.3 Air exchange rate

Air exchange rate (AER) describes the rate at which indoor air is replaced by the outdoor air (Guo et al., 2008). Therefore, AER affects the concentration of indoor pollutants and the peak concentrations, including its residence time after the source has ceased (Zhang and Morawska, 2002). AER in turn is influenced by the dwelling structure and weather conditions, and also by operation of combustion sources within the house (Gupta et al., 1998). Generally, if pollution levels are lower outdoors, indoor air quality (IAQ) can be improved by increasing the AER, i.e. by opening doors and windows or by operating ceiling fans, while on the other hand, when outdoor pollution levels are higher, IAQ can be improved by reducing natural ventilation or AER (Guo et al., 2008).

AER is particularly important for the determination of source emission rates in actual houses (He et al., 2004). It is generally determined by introducing a tracer gas into the indoor environment and measuring its decay over time. CO_2 is most commonly used for determination of AER since it is cheaper than other tracer gas like SF6 (He et al., 2004). However, SF6 use has been banned in many countries since it is a potent greenhouse gas. The following conditions are important when tracer gas is used to compute AER (Guo et al.,

2008): (i) constant exfiltration rates of the tracer gas used, (ii) uniform mixing, (iii) negligible chemical interactions between the tracer gas and other chemicals in the indoor space, and (iv) no indoor source should be operating.

The following equation can be used to compute the AER (He et al., 2004):

$$AER = \frac{1}{t}\ln\frac{C_t}{C_0} \tag{2.6}$$

Where t = time, C_t = CO_2 concentration at time t, and C_0 = CO_2 concentration at time 0.

2.5 World Health Organisation air quality guidelines

WHO has developed a series of guidelines for both outdoor and indoor air quality, synthesising the available scientific information from all over the world. The guidelines contain an in depth review of literature and health risks of the relevant air pollutants, and provide recommendations to assist countries in reducing the health impacts of air pollution. The concentrations specified in the guidelines are based "on no observed adverse effect levels" (NOAEL) or "lowest observed adverse effect levels" (LOAEL). However, guidelines are only recommendations and do not constitute legally binding regulations. The second edition of the WHO air quality guidelines for Europe published in 2000 has recommended threshold levels for several organic and inorganic pollutants found in outdoor air, but not for PM_{10} and $PM_{2.5}$ due to insufficient epidemiological information (WHO, 2000). Subsequently, based on the new scientific evidences, an update of air quality guidelines for PM, O_3, NO_2 and SO_2 was published in 2005 (WHO, 2006).

In recent years, in view of serious indoor air pollution problems, WHO has developed a series of guidelines for IAQ. The first of these series was on dampness and mould published in 2009 (WHO, 2009), followed by guidelines for selected common indoor pollutants, published in 2010 (WHO, 2010). The latest in the series of guidelines is on household fuel combustion, published in 2014 (WHO, 2014a). This new guideline features the current evidence of exposure levels and health risks from household fuel combustion, and provides

practical recommendations for reducing the health burden. In particular, the new guideline has recommended emission rate targets to help countries assess how well interventions can achieve concentrations recommended by the guidelines. Table 2.1 presents the WHO guidelines for the selected indoor and outdoor pollutants. A point to note is that even when pollutant levels are below NOAEL or LOAEL, it could still result in health effects due to other conditions. For example, many pollutants coexist or come from the same source which can result in additive or synergistic effects (Han and Naeher, 2006).

Table 2.1: WHO air quality guidelines for selected outdoor and indoor pollutants

Pollutants	**Outdoor mean guidelines**	**Indoor mean guidelines**
PM_{10}	50 µg/m^3: 24 hour 20 µg/m^3: annual	50 µg/m^3: 24 hour 20 µg/m^3: annual
$PM_{2.5}$	25 µg/m^3: 24 hour 10 µg/m^3: annual Source: WHO (2006)	25 µg/m^3: 24 hour 10 µg/m^3: annual Source: WHO (2010)
Carbon monoxide	100 mg/m^3: 15 minutes 60 mg/m^3: 30 minutes 30 mg/m^3: 1 hour 10 mg/m^3: 8 hours Source: WHO (2000)	100 mg/m^3: 15 minutes 35 mg/m^3: 1 hour 10 mg/m^3: 8 hours 7 mg/m^3: 24 hours Source: WHO (2010)
Nitrogen dioxide	200 µg/m^3: 1 hour 40 µg/m^3: annual Source: WHO (2006)	200 µg/m^3: 1 hour 40 µg/m^3: annual Source: WHO (2010)
Benzene	No safe level recommended Source: WHO (2000)	No safe level recommended Source: WHO (2010)
Formaldehyde	0.1 mg/m^3: 30 minutes Source: WHO (2000)	0.1 mg/m^3: 30 minutes Source: WHO (2010)
$PM_{2.5}$: ERT		0.23 mg/min (unvented stove) 0.80 mg/min (vented stove)
CO: ERT		0.16 g/min (unvented stove) 0.59 g/min (vented stove) Source: WHO (2014a)

ERT: Emission rate targets to meet the indoor air quality guidelines: $PM_{2.5}$ - annual (10 µg/m^3) and interim target-1 (IT-1, 35 µg/m^3), and CO 24 hours average.

In summary, diverse pollutants are found in indoor and outdoor environments resulting from both combustion and non-combustion sources. By far, particles constitute the most complex pollutants both in terms of their physical and chemical characteristics. In

recent years, ultrafine particles have been the research priority both in the field of toxicology and epidemiology. Several pollutants have been listed as carcinogens or have been suspected to possess carcinogenic property, for example benzene and formaldehyde. Health effects result from exposure to pollutants. It is important to note that exposure has a time dimension in addition to the pollutant concentration. Exposure can be measured using direct and indirect methods, however, direct measurements are more useful in assessing exposure. Indoor pollution levels are significantly affected by outdoor sources since building ventilation systems readily promote ingress of outdoor pollutants. I/O ratios have been used by studies to quantify the extent of the influence that outdoor sources have on indoor levels.

2.6 Household air pollution

Household air pollution refers to "air pollution generated by household fuel combustion, leading to indoor air pollution, and contributing to ambient air pollution" (WHO, 2014a). From Table 2.2 (which presents the common indoor pollution sources), it can be seen that household fuels are principal sources of many pollutants found indoors. Household fuels used for cooking and heating range from solid fuels (coal and biomass) to refined fuels kerosene and liquefied petroleum gas (LPG), and electricity. The use of solid fuels in inefficient combustion devices (traditional stoves) promotes incomplete combustion resulting in severe HAP from combustion products. This is specially the case in rural areas in many developing countries.

Table 2.2: Pollutants generated by indoor sources

Pollutants	Major Indoor Sources
Fine particles	Fuel/tobacco combustion, cleaning, fumes from food being cooked, e.g. from cooking oil
Carbon monoxide	Fuel/tobacco combustion
Polycyclic aromatic hydrocarbons	Fuel/tobacco combustion, fumes from food being cooked, e.g. from cooking oil
Nitrogen oxides	Fuel combustion
Sulphur oxides	Coal combustion
Arsenic and fluorine	Coal combustion
Volatile and semi-volatile organic compounds	Fuel/tobacco combustion, consumer products, furnishings, construction materials, fumes from food being cooked, e.g. from cooking oil
Aldehydes	Furnishing, construction materials, cooking
Pesticides	Consumer products, dust from outside
Asbestos	Remodelling/demolition of construction materials
Lead	Remodelling/demolition of painted surfaces
Biological pollutants	Moist areas, ventilation systems, furnishings
Free radicals and other short-lived, highly reactive compounds	Indoor chemistry
Radon	Soil under building, construction materials

Source: (Zhang and Smith, 2003).

2.6.1 Solid fuel usage and household air pollution problems in developing countries

Fuels of either plant or animal origin that are used as household energy sources are called biomass fuels (Bruce et al., 2000). The most common biomass fuels used in developing countries are wood, charcoal, crop residues and dung (Balakrishnan et al., 2002). While biomass fuels contain minimal intrinsic contaminants, substantial pollutants are emitted due to incomplete combustion. On the other hand, coal is naturally associated with several inherent constituents such as sulphur, mercury, arsenic, fluorine, and lead, depending on the type of coal (Zhang and Smith, 2007, Bonjour et al., 2013). During coal combustion, these inherent constituents are released as contaminants into the environment either in its free form or as an oxidized products (Zhang and Smith, 2007).

A conceptual framework called ‘energy ladder’ has been used to describe the trend in household fuel use in developing countries (Smith et al., 1994). A typical energy ladder for South Asia is presented in Figure 2.2. The fuel types placed on lower levels of the energy ladder are more polluting than those occupying the higher levels. Moving up the energy ladder is characterized by health benefits and convenience but with increasing costs. It has

been found that society generally moves up the ladder when they have sufficient resources (Smith et al., 1994). However, for people in rural areas of developing countries, clean fuels such as LPG, kerosene and electricity are beyond the means of the majority of households (Dasgupta et al., 2009).

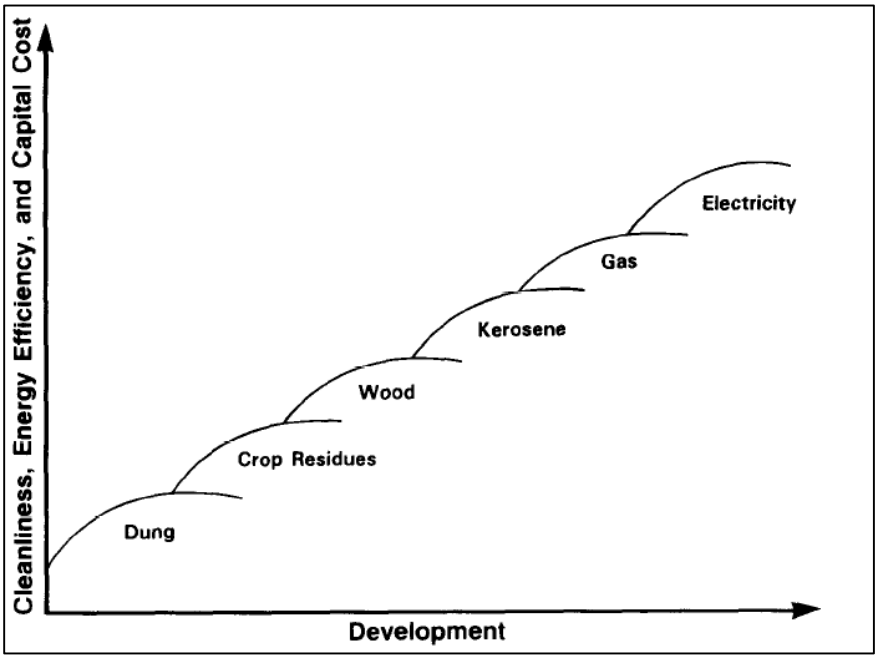

Figure 2.2: A typical energy ladder for household energy sources in South Asia.
Source: (Smith et al., 1994).

It is estimated that almost three billion people living in the world's poorest regions still depend on solid fuels for cooking (about 2.6 billion on traditional biomass and 0.4 billion on coal) (Legros et al., 2011). For instance, 90% of the rural households in India use biomass as the primary energy source (Parikh and Laxmi, 2000, Saud et al., 2011). In China, biomass fuels constitute nearly 80% of the domestic energy consumed in rural areas (Zhang and Smith, 2007). Thus, people in many developing countries face severe energy poverty and therefore live in highly polluted homes. Further, solid fuels are mostly used in traditional stoves, without any smoke venting system, and often in poorly ventilated kitchens (Rehfuess et al., 2006). In most developing countries an open fire is the only option for cooking (Mitra et al., 2002). It is this inefficient energy conversion technology that exposes large number of

people to high levels of combustion products (Foell et al., 2011). As a result, the highest exposures to air pollution are generally experienced in rural homes of developing countries (Smith, 1993), and as presented in Figure 2.3, a number of variables have been found to influence the exposure to HAP.

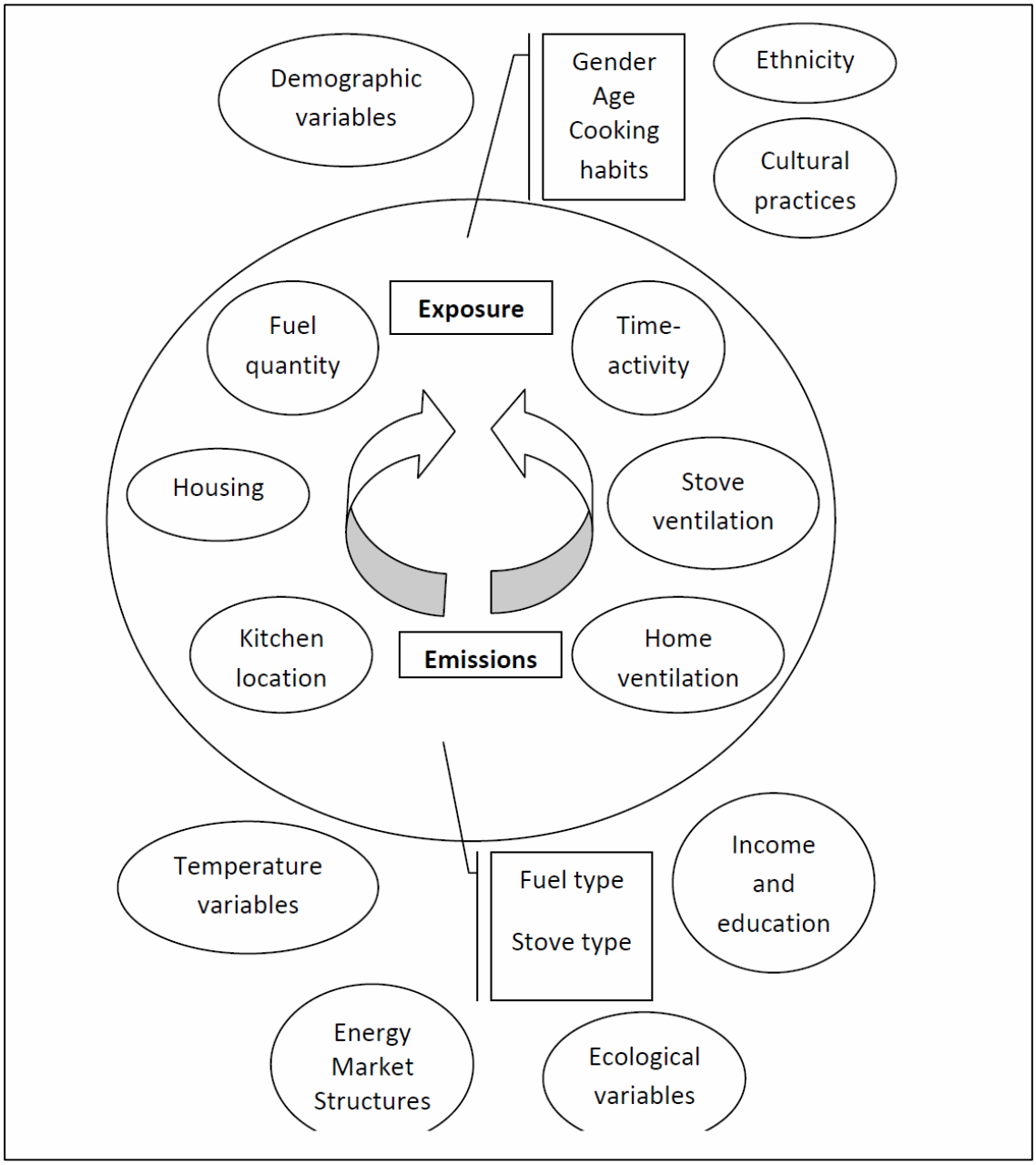

Figure 2.3: Factors influencing HAP exposures at individual, household or community levels. Source: (Balakrishnan et al. 2014).

Although between 1980 and 2010 there has been a 21% decrease globally in the number of households relying on solid fuels for cooking, the number of people exposed to emissions have remained the same (around 2.8 billion) due to growth in population (Bonjour et al., 2013). South East Asia has one of the highest rates of solid fuel use in 2010 at 61% (Bonjour et al., 2013). Therefore, use of solid fuels still presents significant health risks in developing countries (Balakrishnan et al., 2004). 99% of the almost two million deaths that occur annually from pneumonia, chronic lung disease and lung cancer from exposure to HAP are in developing countries (Legros et al., 2011).

Many studies have consistently demonstrated a strong relationship between HAP and human health in developing countries (Ellegard, 1996, Rumchev et al., 2007, Kumar et al., 2008a, Lakshmi et al., 2010, Verma et al., 2010, Mengersen et al., 2011, Murray et al., 2012). However, awareness about the issue is generally lacking in many developing countries with only limited policy initiatives existing for effective management of HAP (Quagraine and Boschi, 2008).

In most developing countries, women and children generally spend a majority of their time indoors and are more exposed to extreme concentration of household combustion products (Siddiqui et al., 2009). This is because most women are engaged in cooking for the family which requires them to be indoors and mostly in close proximity to the fire source (Verma et al., 2010). This would mean children also spending their time inside kitchens with adults. Further, cooking durations are relatively longer in rural areas, leading to extended exposure to peak emissions (Rumchev et al., 2007). Several studies have presented time activity patterns for women and children. For example, Barnes et al. (2005) reported that children in South Africa spend between 52 and 61% of their daily time inside kitchens and mostly within 1.5 meters from the fire. In Tibetan plateau, women and children spend up to seven hours more time inside their residential tents than other family members (Li et al., 2012). In rural Nepal, women cooks spend up to 20% of the day near stoves and more than

90% of that duration is spent around two meters from the stove (Reid et al., 1986). Similarly, in rural Mexico, women spend about 60% of their time (daytime) indoor, primarily inside kitchens (Brauer et al., 1995), while in South India, women spend as much as 87 – 90% of their day indoors (Andresen et al., 2005).

2.6.2 Household air pollution studies in developing countries

Tables 2.3 – 2.6 present a summary of the studies (reporting particle matter, CO, NO_2, VOCs and carbonyls) conducted in developing countries in residential environments. A comprehensive documentation of more recent studies providing an overview of methods and results of HAP measurements in developing countries is available in the WHO HAP database (WHO, 2016). Particle mass fractions have been the most widely investigated indoor pollutants in residential environments in developing countries. Most studies have quantified PM_{10} and $PM_{2.5,}$ while older studies have focused on Total Suspended Particles (TSP). It can be seen that concentrations reported were very high especially for solid fuels, exceeding the WHO guidelines by several orders of magnitude (Table 2.1). The PM concentrations reported for LPG and kerosene were also very high. Therefore, a shift to clean fuels alone does not completely resolve the HAP problems, since pollution levels indoor can be elevated by external factors such as infiltration of neighbourhood pollution and a host of other outdoor combustion activities.

Several review studies from developing countries have reported high indoor PM concentrations. For example, a literature review by Ezzati (2008) has reported 24-hour indoor PM_{10} concentrations ranging from 200 – 5000 $\mu g/m^3$ in houses using biomass fuels. A review study in China has reported mean indoor PM_{10} concentrations of 24.4 mg/m^3 for households using smoky coal, 22.3 mg/m^3 for wood users, and 1.8 mg/m^3 for anthracite coal users (Zhang and Smith, 2007).

Studies reporting PN level in homes in developing countries are scarce, especially for rural settings. Mullen et al. (2011b) have investigated UFP in high rise Beijing apartments,

where cooking was done using natural gas. They have reported mean indoor UFP concentrations ranging from $2.80 \times 10^3 - 2.91 \times 10^4$ particles/cm^3. Sze-To et al. (2012) measured PN inside kitchens and living rooms of 16 homes in Hong Kong during cooking using gas stoves. The mean concentration during cooking was nearly ten times higher in kitchens (3.42×10^5 particles/cm^3) compared to living rooms (1.35×10^5 particles/cm^3). However, concentrations in living rooms were also significantly elevated as result of indoor cooking. Another study in Hong Kong of 12 naturally ventilated homes during cooking using gas stoves found that UFP (14 – 100 nm) and accumulation mode (100 – 661.2 nm) number concentrations were elevated by 10 times inside living rooms and 20 – 40 times inside kitchens (Wan et al., 2011). Currently, there are no studies reporting PN concentrations in houses using solid fuels.

A good number of studies have also investigated CO and NO_2 levels in developing countries. In general, it can be seen that higher levels are associated with the use of biomass fuels (Table 2.4 and 2.5). A review study in China has reported the highest indoor CO concentration of 23 mg/m^3 for households relying on open fire solid fuels (Zhang and Smith, 2007). CO levels between 5 and 60 mg/m^3 are typical in indoor environments (Spengler et al., 2001b). A few studies have also characterized VOCs and carbonyls, mostly during cooking in houses (Table 2.6). For example, Huang et al. (2011) identified 45 VOCs and carbonyls from cooking emissions in Hong Kong apartments which used LPG. None of the carcinogenic compounds such as benzene, formaldehyde and acetaldehyde have been detected in the raw fuel, indicating that these compounds were primarily emitted during the cooking process. This study concluded that while concentrations were low, the potential health impacts to occupants were still significant enough. In Bangladesh, Khalequzzaman et al. (2011) reported significantly higher benzene, toluene, and xylene concentrations inside kitchens of households using biomass fuels (due to incomplete combustion) than those using fossil fuels. Further, concentrations were found to be significantly higher during winter than in summer.

2.6.3 Stove and fuel types

Two of the most important factors affecting HAP levels are fuel and stove types used for cooking and heating. Among fuel types, significantly higher concentrations of respirable particles were associated with cattle dung and agricultural residues compared with other fuels like wood, kerosene and gas (Balakrishnan et al., 2002, Balakrishnan et al., 2004). An example of respirable particle levels from different household fuels is presented in Figure 2.4. Dung was also found to emit higher concentrations of particles with smaller diameters compared to other solid and gaseous fuels (Verma et al., 2010). A comparative study of different fuels showed that indoor CO concentration from coal was 155% and 45% higher than LPG and kerosene, respectively (Kandpal et al., 1995). However, LPG was found to emit higher level of NO_2 (25 – 55% and 10 – 15% higher than coal and kerosene, respectively). Khalequzzaman et al. (2010) reported higher particles, CO, CO_2, NO_2 and VOCs in houses using biomass fuels compared to those using fossil fuels.

Stove types present a unique situation to investigate the effectiveness of technological intervention to reduce HAP. In developing countries, often the division is between households using traditional open fire stoves versus improved stoves with chimney (Figures 2.5 and 2.6). For example, Li et al. (2012) investigated $PM_{2.5}$ and CO concentrations in dwellings which used traditional open fire and improved cookstoves with chimney. They reported mean (24-hours) indoor $PM_{2.5}$ and CO concentrations of 1.42 ± 3.26 and 6.69 ± 9.11 mg/m^3 for dwellings using open fire stoves, and 0.14 ± 0.65 and 0.12 ± 1.01 mg/m^3, respectively for dwellings using improved chimney stoves. Likewise, Singh et al. (2012) found mean (24-hours) $PM_{2.5}$ and CO concentrations of 2.07 mg/m^3 (95% CI: 1.42 – 2.71) and 21.5 ppm (95% CI: 14.5 – 28.6) for houses using traditional cookstoves, and 0.76 mg/m^3 (95% CI 0.521 – 1.00) and 8.62 (95% CI: 6.18 – 11.1) ppm for houses using improved cookstoves. A significantly higher PM_{10} concentration for houses using open fire stoves (1267 ± 74 $\mu g/m^3$) than those using chimney stoves (348 ± 145 $\mu g/m^3$) was also reported by

Morawska et al. (2011). While improved cookstoves presented a significant reduction in pollutant concentrations, the levels were still higher than the WHO guidelines for PM_{10}, $PM_{2.5}$ and CO. Similar results with lower particle mass and CO concentrations in houses with improved chimney stoves, albeit still higher than the WHO guidelines, were reported by several studies (Edwards et al., 2007, Tian et al., 2009, Clark et al., 2010).

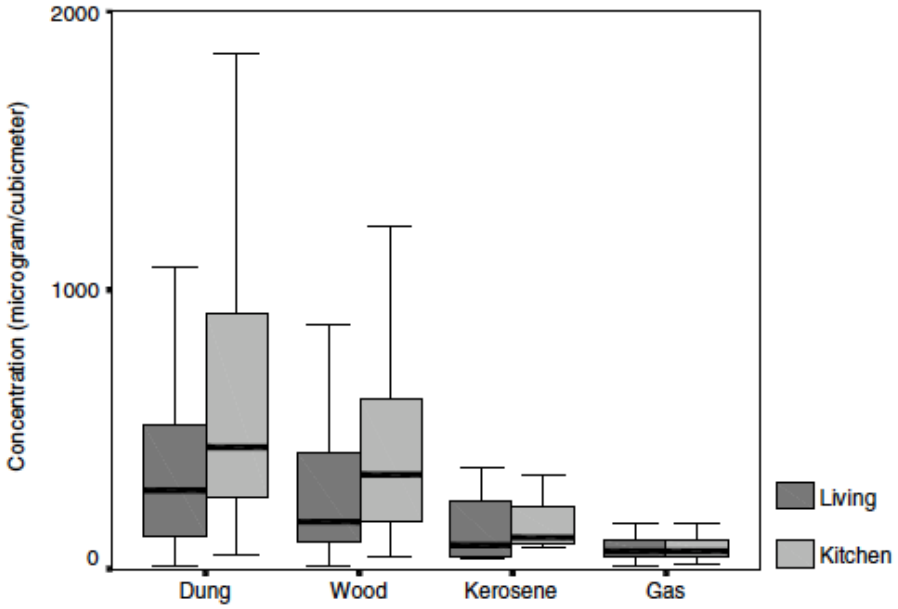

Figure 2.4: Distribution of 24-hours mean respirable particle concentrations from different household fuels measured inside living rooms and kitchens.
The box plot presents the minimum, first quartile, median (middle dark line), third quartile and maximum. Source: (Balakrishnan et al., 2004).

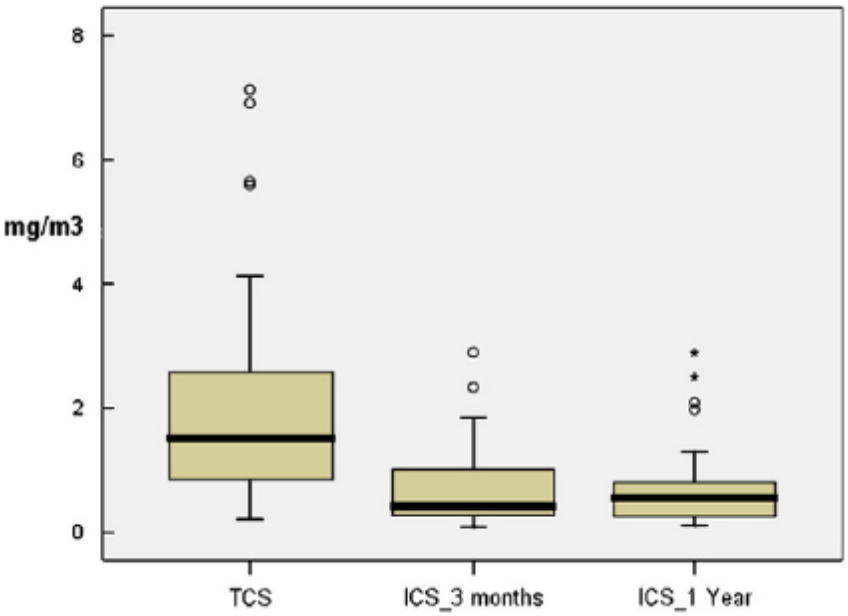

Figure 2.5: Distribution of 24-hours median $PM_{2.5}$ concentrations in houses using traditional stoves (TCS) and improved cookstoves (ICS).
ICS_3 months and ICS_1 year: HAP monitoring conducted 3 months and 12 months post ICS installation. Source: (Singh et al., 2012).

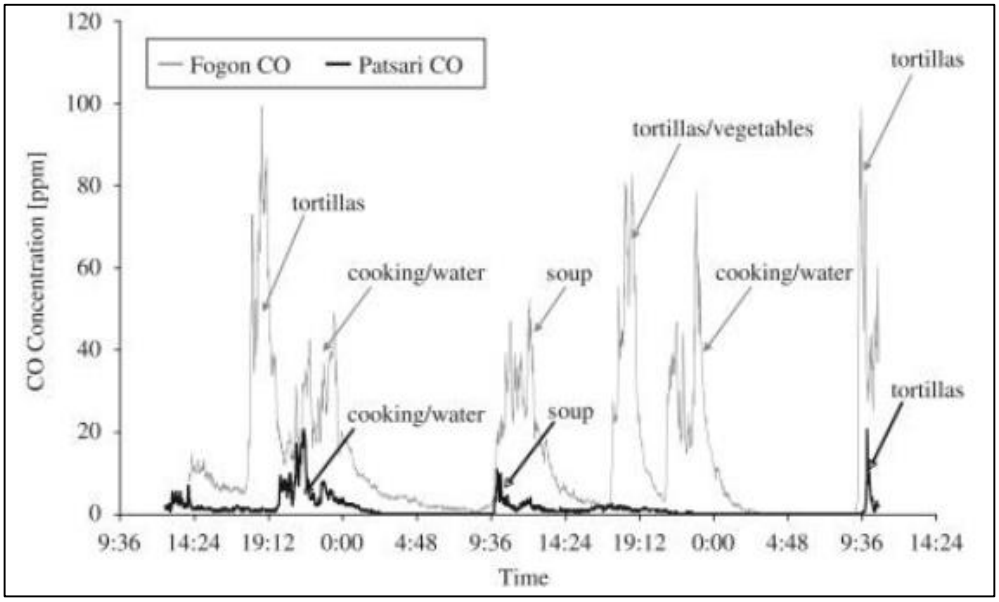

Figure 2.6: A typical CO concentrations during cooking using traditional Fogon and improved Patsari stove.
Source: (Balakrishnan et al. 2014).

2.6.4 Characterization of emissions from household combustion devices

Characterization of pollutant emissions from household combustion devices is important for source apportionment and assessment of exposure levels (Zhang and Morawska, 2002). The common combustion devices found in residential environments

include stoves used for cooking and heating, and lighting lamps. Although small in size, their numbers can add to billions, which can cumulatively contribute significantly to emission levels of various particulate and gaseous pollutants (Zhang et al., 2000, Morawska and Zhang, 2002). Unlike gas and electric stoves the fuel combustion process in traditional stoves is difficult to control and leads to different emission profiles during the different stages of the combustion cycle. While household stoves have been known to present different emission characteristics than larger combustion devices, emissions from small household combustion sources have not been sufficiently characterized (Zhang et al., 1999, Zhang and Morawska, 2002).

2.6.4.1 Emission factors

Emission factor is defined as the "amount of a concerned pollutant emitted per unit mass of fuel burned or per defined task performed" (Zhang and Morawska, 2002). Thus, emission factors can be measured: (i) based on fuel mass analysis (mass-based emission factor), which has a unit of g/kg of fuel (Zhang et al., 2000) and (ii) based on a specific task performed (task-based emission factor), with units of either g/mile or g/MJ, depending on the type of tasks investigated (Zhang and Morawska, 2002). Emission factors are influenced by types of fuel and stoves used (Zhang et al., 1999, Arinto et al., 2006).

The emission factors for some of the commonly used fuels in rural houses in developing countries have been investigated by several studies, e.g. fuel wood (*Acacia nilotica*) (Ahuja et al., 1987), and dung cakes and crop residues (Joshi et al., 1989). CO and TSP emission factors for *Acacia nilotica* were found to be in the range of 13 – 68 g/kg and 1.1 – 3.9 g/kg, respectively, while estimated emission factors for dung cakes and crop residues ranged from 26 – 67 g/kg and 20 – 114 g/kg for CO, and 4.1 – 7.8 g/kg and 2.1 – 12.0 g/kg for TSP, respectively. Chen et al. (2005) determined emission factors of particles, black carbon (BC), organic carbon (OC), and PAHs for five types of coal used as household fuels. The estimated emission factors of all the combustion products were the highest for

bituminous coal, while the lowest were for the anthracite coal. A comprehensive study of emission factors for a range of fuel/stove combinations (all kinds of traditional and improved stoves made from mud, brick, and metal, with and without chimney) and fuel types (dung, crop residues, wood, charcoal, kerosene, coal and gases), commonly used in developing countries have been conducted in simulated houses in India and China (Zhang et al., 1999, Zhang et al., 2000). The result showed significantly higher emissions per unit of energy delivered from solid fuels than liquid and gas fuels, which was due to poor thermal and combustion efficiency of solid fuels/stove combinations. Likewise, other studies also have reported emission factors from cookstoves estimated under laboratory conditions (Roden et al., 2009, Jetter et al., 2012).

2.6.4.2 Emission rates

Pollutant emission rate, also referred to as source strength is defined as the amount of particular pollutant emitted per unit time, and has a unit of g/h or Tg/year (Mitra et al., 2002). Emission rate differs from emission factor in that the former considers the time dimension, i.e., amount of pollutant emitted per unit time (Zhang and Morawska, 2002).

Similar to emission factors, emission rates also depend on fuel-stove types and conditions under which combustion is carried out. For example, Schare and Smith (1995) compared PM emission rates from two commonly used lamps in highland Guatemala (traditional can lamp and glass chimney lamp), both fuelled with kerosene. The PM emission rates were 535 ± 155 mg/h for a can lamp and 295 ± 120 mg/h for a glass lamp. Similarly, Fan & Zhang (2001) conducted a laboratory estimation of emission rates for portable combustion devices which included kerosene lamp, oil lamp and kerosene space heater. The estimated particle mass emission rates ranged from 5.6 ± 0.1 – 142.3 ± 40.8 mg/h, while CO emission rates ranged from 5,500 ± 700 – 21,000 ± 90,000 mg/h, respectively. To date no studies have reported emission characteristics for PN from household combustion devices using solid fuels.

In summary, solid fuels use is the major cause of severe HAP in developing countries, with a large rural population depending on it for cooking and heating. Women and children are particularly affected the household combustion products, since they spend the majority of their time indoors at home, and mostly inside kitchens. The option for movement up the energy ladder is limited for rural population in developing countries with the pricing structure of clean fuels being beyond the financial capacity of the people. While there have been several studies focussing on HAP levels in developing countries, studies have been limited to conventional pollutants. The PN emissions, from use of solid fuels have never been investigated. Hardly any study has reported emission rates from biomass stoves. The majority of the studies on emission characteristics have been conducted under laboratory or simulated conditions. Such studies are not very useful to real situations as many confounding factors influence emission rates under real-world conditions.

Table 2.3: Indoor particle matter concentrations in residential environment in developing countries

Study	**Location**	**Rural/Urban**	**Particles**	**Fuel**	**AM ($\mu g/m^3$)**	**GM ($\mu g/m^3$)**	**[SE](SD)**	**Range ($\mu g/m^3$)**	**Sampling Duration**
(Brauer et al., 1995)	San Jose ´de Solis, Mexico	Rural	$PM_{2.5}$	Biomass	554.7		(492.9)	30.1 – 1492.9	9 hr
			PM_{10}	Biomass	767.9		(540.5)	49.3 – 1654.9	
(Ellegard, 1996)	Maputo, Mozambique	Sub-urban	PM < 7.1µm	Wood Charcoal LPG Kerosene Coal	1200 540 200 760 940		[131] [80] [110] [270] [250]		Average cooking time for one meal (1.5 hr)
(Lodhi and Zain-al-Abdin, 1999)	Malaysia	Rural	SPM	Wood Gas	300 50				Cooking hours
(Albalak et al., 1999)	Cantuyo & Taipillanga, Bolivia	Rural	PM_{10}	Biomass		1830[a]	(2990)		6 hr
(Naeher et al., 2000)	Xela, Guatemala	Rural	$PM_{2.5}$	Biomass	5310		(4750)	260 – 13800	Cooking time
(Balakrishnan et al., 2002)	Tamil Nadu, India	Rural	PM_4	Wood Wood chips Agriculture produce	847[b] 269[b] 245[b]	498[b] 246[b] 221[b]	[50] [376] [23]		Cooking hours
(Balakrishnan et al., 2004)	Andhra Pradesh, India	Rural	PM_4	Dung Wood	732[a] 362[b] 500[a] 345[b]	470[a] 235[b] 340[a] 204[b]	[88] [37] [30] [26]		24 hr
(Andresen et al., 2005)	Mysore, India	Urban	$PM_{2.5}$	Kerosene LPG	98[c] 155[d] 71[c] 73[d]	86[c] 137[d] 54[c] 59[d]	[9](50) [13](71) [9](48) [6](34)	20 – 230 56 – 264 16 – 150 6 – 142	24 hr
(Rumchev et al., 2007)	Gwanda District, Zimbabwe	Rural	RSP	Biomass				230 – 2300	8hr
(Fischer and Koshland, 2007)	Jilin province, China	Rural	RSP	Mostly multiple fuels	312[a] 1880[a]	233[a] 1126[a]		323 – 1440 159 – 6200	42 hr 1 hr peak
(Kumar et al., 2008a)	Ashok Vihar, India	Urban	SPM	Biomass/LPG	960		(360)	80 – 1530	8 hr

Table 2.3: Continued

Study	Location	Rural/Urban	Particles	Fuel	AM(μg/m^3)	GM (μg/m^3)	[SE](SD)	Range (μg/m^3)	Sampling Duration
(Begum et al., 2009)	Savar, Dhaka District, Bangladesh	Rural	PM_{10}	LPG Biomass	178[a] 197[b]			104 – 332 592 – 1177	4 hr 8 hr
(Siddiqui et al., 2009)	Rehri Goth, Pakistan	Semi-rural	$PM_{2.5}$	Wood	2740		(2060)		8 hr
(Kang et al., 2009)	Nam Co region, Tibetan	Rural	TSP	Yak dung	3160				24 hr
(Tian et al., 2009)	Xuan Wei, China	Rural	PM_{10}	Bituminous coal	1600			200 – 4200	24 hr
(Clark et al., 2010)	Santa Lucia & Suyapa, Honduras	Semi-urban, Rural	$PM_{2.5}$	Wood	1002.3	650.2	(1089.4)	59.8 – 4835.4	8 hr
(Khalequzzaman et al., 2010)	Moulvibazar & Dhaka, Bangladesh	Urban	Dust particles	Biomass	821[e] 454[f] 633[g] 208[h]		(1011) (306) (727) (153)		Mean of five 1-min measurements
(Morawska et al., 2011)	Vientiane & Bolikhamxay, Laos	Rural	PM_{10}	Mostly wood	1183[i] 1275[j]		[99] [98]	147 – 3030 43 – 4577	12 hr
(Li et al., 2012)	Nam Co & Anduo region, Tibet	Rural	$PM_{2.5}$	Dung	1420[k]		(3260)		24 hr
(Singh et al., 2012)	Dang, western region; Dolakha, central region; Ilam, eastern of Nepal	Rural	$PM_{2.5}$	Wood, dung & agricultural residue	2070[a]		95 % CI 0.521–1.00		24 hr
(Massey et al., 2012)	Agra, India	Roadside houses Urban houses	PM_{10} $PM_{5.0}$ $PM_{2.5}$ $PM_{1.0}$ PM_{10} $PM_{5.0}$ $PM_{2.5}$ $PM_{1.0}$		247 211 161 111 181 145 123 99		(78) (59) (62) (32) (84) (62) (45) (41)		Annual-sampling from Oct 2007 to March 2009

AM: Arithmetic Mean, GM: Geometric Mean, [SE]: Standard Error, (SD): Standard Deviation, TSP: Total Suspended Particles, SPM: Suspended Particulate Matter, RSP: Respirable Particulate Matter, [a] Kitchen concentration, [b] Living area concentration, [c] Summer concentration, [d] Winter concentration, [e] Winter cooking kitchen concentration, [f] Winter non-cooking kitchen concentration, [g] Summer cooking kitchen concentration, [h] Summer non-cooking kitchen concentration, [i] In Bolikhamxay Province, [j] In Vientiane Province, [k] Residential tent.

Table 2.4: Indoor CO concentrations in residential environment in developing countries

Study	Location	Rural/Urban	Fuel	AM	GM	[SE](SD)	Range	Sampling Duration
(Lodhi and Zain-al-Abdin, 1999)	Malaysia	Rural	Gas Wood	0.1 ppm 2.9 ppm				Cooking hours
(Naeher et al., 2000)	Xela, Guatemala	Rural	Open fire	22.9 ppm		(28.1)	0 – 250 ppm	Cooking time
(Lawrence et al., 2005)	Agra, India	Rural	Biomass/LPG	1150 ppb		(370)		8 hr
(Fischer and Koshland, 2007)	Jilin Province, China	Rural	Mostly multiple fuels	4.20 ppm 20.5 ppm	2.61 ppm 11.3 ppm		0.26 – 40 ppm 1.1 – 169 ppm	24 hr 1 hr peak
(Rumchev et al., 2007)	Gwanda District, Zimbabwe	Rural	Biomass				10 – 60 mg/m^3	8 hr
(Kulshreshtha et al., 2008)	Nizamudin, New Delhi, India	Urban slum	LPG/kerosene	5[a] µg/m^3 6[b] µg/m^3 1[c] µg/m^3				6hr/day × 3 repeats
(Padhi and Padhy, 2008)	West Bengal, India	Rural	Biomass LPG	837.8 ppb 247 ppb	340.5 ppb 120 ppb			24 hr
(Siddiqui et al., 2009)	Rehri Goth, Pakistan	Semi-rural	Wood	29.5 ppm		(16.2)		8 hr
(Khalequzzaman et al., 2010)	Moulvibazar & Dhaka, Bangladesh	Urban	Biomass	6.3[d] ppm 2.8[e] ppm 19.6[f] ppm 3.6[dg] ppm		(9.8) (4.0) (15.1) (7.6)		Mean of five 1-min measurements
(Smith et al., 2010)	San Marcos, Guatemala	Rural	Open wood fire	10.2[h] ppm	8.4[h] ppm			48 hr
(Morawska et al., 2011)	Vientiane & Bolikhamxay, Laos	Rural	Mostly wood	0.490[i]ppm 0.430[j] ppm		[0.059] [0.032]	0.045 – 1.976 ppm 0.002 – 1.667 ppm	12 hr
(Li et al., 2012)	Nam Co & Anduo region, Tibet	Rural	Dung	6.6 mg/m^3		(9.1)		24 hr
(Singh et al., 2012)	Eastern Nepal	Rural	Wood, dung & agricultural residue	21.5[h] ppm		95 % CI: 14.5 – 28.6		24 hr

AM: Arithmetic Mean, GM: Geometric Mean, [SE]: Standard Error, (SD): Standard Deviation, [a] Rainy season, [b] Summer season, [c] Winter season, [d] Winter cooking kitchen concentration, [e] Winter non-cooking kitchen concentration, [f] Summer cooking kitchen concentration, [g] Summer non-cooking kitchen concentration, [h] kitchen concentration, [i] In Bolikhamxay Province, [j] In Vientiane Province

Table 2.5: Indoor NO_2 concentrations in residential environment in developing countries

Study	**Location**	**Rural/Urban**	**Fuel**	**AM**	**GM**	**[SE](SD)**	**Range**	**Sampling Duration**
(Lawrence et al., 2005)	Agra, India	Rural	Biomass/LPG	230.75 ppb		(85)		8 hr
(Kulshreshtha et al., 2008)	Nizamudin, New Delhi, India	Urban slum	LPG/kerosene	199[a] µg/m^3 140[b] µg/m^3 73[c]µg/m^3				6hr/day × 3 repeats
(Kumar et al., 2008a)	Ashok Vihar, India	Urban	Biomass/LPG	19.02 µg/m^3		(12.79)	1.46 – 81.46 µg/m^3	8 hr
(Kumar et al., 2008b)	New Delhi, India	5 residential areas, 2 industrial areas, 3 villages,	LPG Biomass	30.26 µg/m^3 31.80 µg/m^3		(18.60) (33.94)		> 6 hr
(Kumie et al., 2008)	Butajira, Ethiopia	Rural	Biomass	97 µg/m^3		(91.4)	0 – 978 µg/m^3	24 hr
(Padhi and Padhy, 2008)	West Bengal, India	Rural	Biomass LPG	71.7 ppb 37.7 ppb	31.8 ppb 10 ppb			24 hr
(Khalequzzaman et al., 2010)	Moulvibazar & Dhaka, Bangladesh	Urban	Biomass	60.1[d] µg/m^3 45.2[e] µg/m^3		(2.0) (1.9)		24 hr
(Morawska et al., 2011)	Vientiane & Bolikhamxay, Laos	Rural	Mostly wood	561[f] µg/m^3 1210[g] µg/m^3		[45] [94]	142 – 1837µg/m^3 68 – 4904 µg/m^3	12 hr

AM: Arithmetic Mean, GM: Geometric Mean, [SE]: Standard Error, (SD): Standard Deviation, [a] Rainy season, [b] Summer season, [c] Winter season, [d] Winter kitchen concentration, [e] Summer kitchen concentration, [f] In Bolikhamxay Province, [g] In Vientiane Province.

Table 2.6: Indoor VOCs and carbonyl concentrations in residential environment in developing countries

Study	**Location**	**Rural/Urban**	**Fuel**	**Compounds**	**AM ($\mu g/m^3$)**	**GM ($\mu g/m^3$)**	**[SE](SD)**	**Range ($\mu g/m^3$)**	**Sampling Duration**
(Pandit et al., 2001)	Trombay, Mumbai, India	Urban	Kerosene	Benzene Toluene *p*-Xylene *o*-Xylene		103.4[a] 61.1[a] 23.7[a] 9.9[a]		43.9 – 166.4[a] 36.3 – 88.1[a] 17.6 – 33.5[a] 6.0 – 14.2[a]	Cooking time
(Huang et al., 2011)	Hong Kong	Urban Apartment,	LPG	Benzene Toluene Ethylbenzene m-Xylene p-Xylene o-Xylene Formaldehyde Acetaldehyde Acrolein Hexaldehyde	1.3 27.6 3.1 2.7 1.8 2.6 151 4.5 17.7 15.7		(0.5) (3.2) (2.2) (1.8) (1.0) (0.7) (45) (3.5) (6.7) (3.2)		Cooking time (90 min)
(Khalequzzaman et al., 2010)	Moulvibaza & Dhaka, Bangladesh	Urban	Biomass/ fossil fuel	Benzene Toluene Xylene Formaldehyde		54.2[b] 31.4[c] 34.2[b] 26.8[c] 18.9[b] 9.3[c] 60.1[b] 45.2[c]	(2.5) (2.7) (5.1) (2.8) (2.9) (3.0) (2.0) (1.9)		24 hr

[a] Kitchen concentration, [b] Winter kitchen concentration, [c] Summer kitchen concentration

2.7 Air quality in school environments

The health effects of air pollution are disproportionately higher for children than adults. This is because they breathe more air relative to their body size compared to adults, and their developing system presents less resistance to pollutants (Buonanno et al., 2012b, Zhang and Zhu, 2012, Demirel et al., 2014). Further, breathing through the mouth is very common for children, which leads to direct ingestion of contaminants in the air (Pegas et al., 2011). These vulnerabilities make children the sub-population most sensitive to air pollution (Raysoni et al., 2013).

Schools have distinctive indoor and outdoor facilities resulting from specific building structures, ventilation systems and activities conducted (Lee and Chang, 2000). Given the importance of education, schools host the highest population density of children at any given time, compared to any other environments. Children spend between 28% and 35% of their daily time at schools (both indoors and outdoors), making it the second most important environment, after homes (Sofuoglu et al., 2011, Buonanno et al., 2012a, Mazaheri et al., 2014). A significant portion of their day is spent inside classrooms, where exposure patterns can be different from ambient exposure (Raysoni et al., 2013). However, classroom environments and other indoor facilities in schools have received very little attention as important zones of children's exposure to air pollution (Fromme et al., 2007, Mullen et al., 2011a). The time children spend outdoors at school is characterized by physically active engagements. The inhalation rate for children is reported to be the highest during playing and sporting, which leads to higher pollutant intake (Buonanno et al., 2011). Therefore, in a polluted school environment, it is likely that health problems arising from higher pollutant intake may outdo the benefits of outdoor activities for children's learning and development (Mejía et al., 2008).

Several studies have highlighted air quality problems in classrooms and other indoor and outdoor facilities in schools. For example, two recent literature reviews have concluded that a substantial portion of children's daily exposure to particles occurs in school environments (Mejía et al., 2011, Morawska et al., 2013). PM_{10} concentrations exceeding the local indoor and outdoor standards, and CO_2 exceeding American Society of Heating, Refrigerating, and Air-Conditioning Engineers (ASHARE) standard, indicating congested classrooms and poor ventilation have been reported for schools in Hong Kong (Lee and Chang, 2000). ASHRAE Standard 62.1 specifies 1000 ppm standard for CO_2 as a comfort parameter, exceedance of which indicates inadequate building ventilation (ASHRAE, 2013). Average CO_2 levels exceeding 1000 ppm and often reaching 3000 ppm was observed in 64 classrooms in Michigan (Godwin and Batterman, 2007). Zwoździak et al. (2013) reported fine particles concentrations exceeding the short-term WHO guideline for a classroom in Poland. A health risk assessment in three schools in Turkey has identified formaldehyde and benzene as compounds of concern since their carcinogenic risks were found to be higher than the threshold levels (Sofuoglu et al., 2011).

For school children, adverse health outcomes from exposure to air pollution leads to absenteeism from schools, and consequently, poor academic performance. For example, Gilliland et al. (2001) found that diurnal changes in ozone concentrations in a school substantially increased children's absenteeism due to respiratory illnesses. The absence rate was found to be higher 2 – 3 days after the exposure, reaching the highest rate on day 5. Apart from affecting their overall development, poor air quality in schools could seriously affect children's future career and also the society at large (Mendell and Heath, 2005). Despite all these concerns, there are very minimal regulations even in the United States to protect children from environmental exposure

in schools (Mendell and Heath, 2005). Therefore, characterization of air pollution in schools is important to assess exposure and enable a reduction in health risks (Morawska et al., 2013, Raysoni et al., 2013).

2.7.1 Air quality characterization methods in schools

School settings, with a range of indoor and outdoor pollution sources, present a unique environment for air quality investigations. Generally, air quality characterization in schools are done at three spatial scales: (i) city scale, (ii) school scale and (iii) personal scale (Mejía et al., 2011). The city scale is the broadest approach involving characterization of air quality in schools based on fixed site monitoring across several city blocks. School scale involves in-situ characterization of air quality at a ground level by installing monitors in different indoor and outdoor school facilities. Personal scale involves direct measurement from children by using light portable instruments attached to their outer body/clothing to measure personal exposure (Morawska et al., 2013). While city scale measurements have advantages of not interfering with the school system (Mejía et al., 2011), in-situ measurements done within school premises provide a better assessment of local air quality (Rivas et al., 2014).

2.7.2 Sources of air pollution in the school

In the last decade many air quality studies based on measurements in schools have been reported from around the world, for example see Table 2.7. More than 70 epidemiological publications, exploring air pollution exposure in schools and associated health impacts have been recorded as of 2011 (Mejía et al., 2011). The pollutants commonly found in schools include all kinds of biological contaminants, particulate matter, lead, and gases (VOCs, formaldehyde, CO, and NO_2) (Jovanović et al., 2014), originating from hosts of indoor and outdoor sources.

The sources inside classrooms include emissions from building materials, furniture and products used for cleaning (Jovanović et al., 2014). Teaching activities themselves can be an important source of particles (Polednik, 2013). School facilities such as swimming pools, science laboratories, computer rooms, and activities associated with these resources present additional sources of pollution (Godwin and Batterman, 2007). In 25 schools in Brisbane, over 80% of the indoor VOCs concentration was contributed by three indoor sources which includes cleaning products, air fresheners, and art and craft activities (Mishra et al., 2015).

Air quality problems in schools are exacerbated by poor ventilation systems, uncleaned or improperly cleaned indoor surfaces, and crowded classrooms with potential for continuous re-suspension of particles (Almeida et al., 2011). Additionally, delaying routine maintenance and servicing of indoor facilities and ventilation systems can potentially lead to IAQ problems (Guo et al., 2008). Pollution problems in schools also arise due to cheap materials being used for construction, inadequate funding leading to environmental deficiencies from poor maintenance and servicing of facilities, and the lack of proper landscaping (Mendell and Heath, 2005, Godwin and Batterman, 2007).

Re-suspension of particles from classroom floors is the major source of coarse particles in schools (Polednik, 2013). An example of particle mass and CO levels inside the classroom and school outdoor environment is presented in Figure 2.7. Among the range of particles and gaseous pollutants investigated, Chithra and Shiva Nagendra (2012) reported the highest I/O ratio of 2.5 for PM_{10}, indicating dominant indoor source (as a result of re-suspension) for a school in India. Children's movement and cleaning activities have been identified as the factors resulting in elevated PM_{10} and $PM_{2.5}$ concentrations inside classrooms in Poland (Zwoździak et al., 2013). Air

quality evaluation in 64 schools in Munich found a good correlation between particle mass concentrations inside classrooms with school characteristics such as size of the classrooms, number of children, and frequency of classes (Fromme et al., 2007). Further, this study also found better air quality in classrooms with a mechanical ventilation system than those with natural ventilation. In Belgium, classroom $PM_{2.5}$ was not only higher than outdoor, its elemental compositions were also reportedly different from that of outdoor environment (Stranger et al., 2008).

Laiman et al. (2014) characterized a number of sources of PN inside classrooms in 25 schools in Brisbane, Australia. During school hours, their results showed that PN concentrations were elevated by a factor of more than four when indoor sources were operated, which included grilling events at the school tuckshop, heating and printing in the classrooms. During non-school hours, cleaning activities constituted the dominant source, elevating the PN concentration by a factor of five, presenting an average emission rate of $(2.09 \pm 6.30) \times 10^{11}$ particles/min. Likewise, Morawska et al. (2009) found higher PN concentrations in the classroom coinciding with the cleaning time and when art activities were conducted compared to other classroom activities. In particular, art activities such as painting, gluing and drawing resulted in the highest PN concentrations (over 1.4×10^5 particles/cm^3) in the classroom, which was approximately one order magnitude higher than the outdoor levels. Mullen et al. (2011a) linked higher PN concentrations inside classrooms during occupancy time to higher ventilation rates, which elevated infiltration from outdoors more than operation of indoor sources. For 9 schools in Michigan, dominant VOCs inside classrooms were benzene, ethylbenzene, toluene, xylene, α-pinene, and limonene, with higher indoor concentrations than the outdoor (Godwin and Batterman, 2007).

In many of the investigated schools, local traffic presented the primary outdoor source in school environments. As expected, pollution levels were always higher in schools located near busy traffic intersection or urban areas than schools located away from the urban setting and a major traffic source. For example, in Lisbon primary schools, NO_2 concentrations were always higher outdoors than indoors, which was due to higher vehicular emissions in outdoor environments (Pegas et al., 2011). Raysoni et al. (2013) characterized multiple pollutants (PM, NO_2, BC and BTEX) in three schools located in an area of high traffic density and one school located in a low traffic density area. They found similar outdoor concentrations in three schools located in the high traffic zones, which were higher than concentrations found in the school located in a low traffic zone. Likewise, Demirel et al. (2014) found higher outdoor BTEX and NO_2 concentrations for a school located in the urban centre than one located in a sub-urban area. Similarly, other studies have also reported higher PN concentrations in urban schools compared to those located in rural areas, due to influence from traffic emissions (Buonanno et al., 2013a, Fonseca et al., 2014). Studies have also observed a seasonal air pollution trend in schools, with air quality being poorer during winter than summer and autumn for particulate matter (Fromme et al., 2007, Chithra and Shiva Nagendra, 2012, Zwoździak et al., 2013), and VOCs and formaldehyde, highest in winter followed by spring and fall seasons (Sofuoglu et al., 2011).

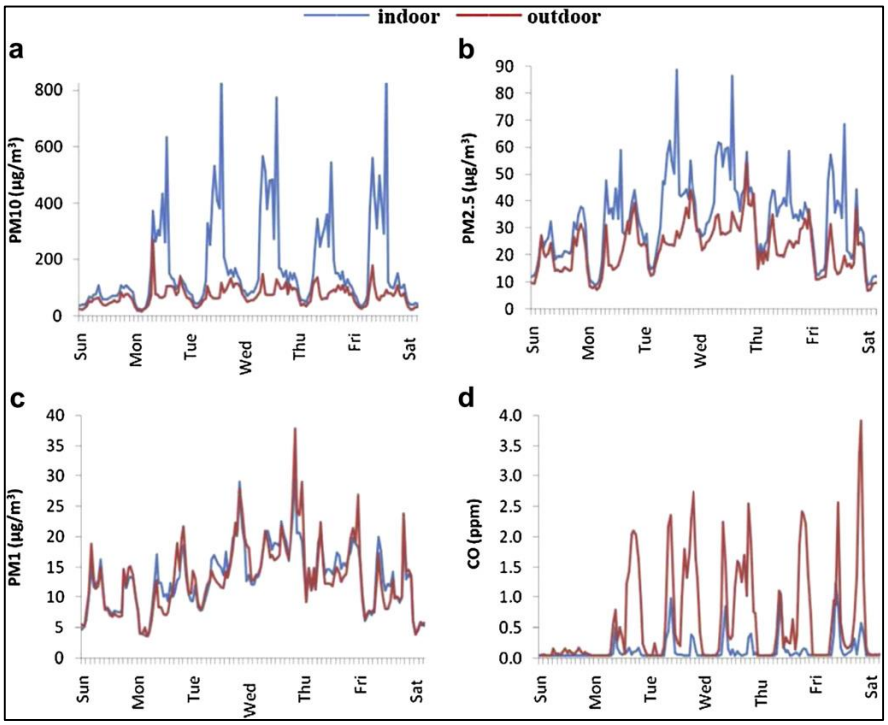

Figure 2.7: PM_{10}, $PM_{2.5}$, $PM_{1.0}$ and CO concentrations inside the classroom and school outdoor environment.
Source: (Chithra and Shiva Nagendra, 2012).

2.7.3 Influence of local outdoor sources on indoor level in schools

Many studies have reported that pollution levels inside classrooms are largely influenced by outdoor sources, for example (Lee and Chang, 2000, Mullen et al., 2011a, 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 (as discussed in Section 2.4).

One of the most comprehensive assessments of air quality in schools came from Stranger et al. (2008), where they investigated multiple pollutants in 27 urban and suburban schools in Belgium. The investigated pollutants included NO_2, SO_2, O_3, BTEX, and $PM_{2.5}$ and its elemental composition. I/O ratios for NO_2, SO_2 and O_3 were below 1, indicating influence of outdoor pollution. 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. 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 school in India was 0.51 (Chithra and Shiva Nagendra, 2012).

2.7.4 Air quality investigations for schools in developing countries

While school environments contribute significantly to children's daily exposure, only a handful of studies have been done in developing countries focussing on schools located in urban areas. For example, Zhao et al. (2008) examined the relationship between pollutant concentrations (SO_2, NO_2 and O_3) and exposure symptoms in ten schools in Taiyuan, China. They have reported that indoor exposure to pollutants, primarily originating from outdoors, was associated with asthmatic symptoms for school children. Lee and Chang (2000) investigated indoor and outdoor air quality in five schools in Hong Kong located in residential, industrial and rural areas. The parameters measured included CO_2, SO_2, NO, NO_2, PM_{10}, formaldehyde and bacterial counts. The major air quality problems arose due to high PM_{10} and CO_2 levels inside classrooms. Tippayawong et al. (2009) conducted indoor and outdoor size-resolved particle number concentration (0.3 – 5.0 μm) measurements in a naturally ventilated school located in a busy commercial area in Chiang Mai, Thailand. Median indoor concentrations ranging from 4.1×10^5 particles/m^3 for 2.5 – 5.0 μm to 1.6×10^8 particles/m^3 for 0.3 – 0.5 were observed during the daytime. Unlike other studies, they found higher concentrations on weekends than weekdays, and also higher

concentration at night than daytime. While results reported significant penetration of outdoor particles to indoor, classroom concentrations were not associated with children's activities. In India, three longitudinal studies covering different seasons and up to a yearlong monitoring for naturally ventilated schools located in metropolitan areas and heavy industrial zones were conducted, in Delhi (Goyal and Khare, 2009), Chhattisgarh (Gadkari, 2010), and in Chennai (Chithra and Shiva Nagendra, 2012). The parameters measured included particle mass (PM_{10}, $PM_{2.5}$, $PM_{1.0}$) and CO, and all the studies have reported significant air quality problems with concentrations exceeding the Indian National Ambient Air Quality Standards, even on weekend days (Figure 2.8). So far, no study has reported air quality investigations for schools located in rural areas. The pollution sources in rural schools can be very different from that of urban schools, thereby presenting different risks to children. Despite the potentially severe toxicity of ultrafine particles for children, currently there is no data available for schools in developing countries.

In summary, good air quality in schools is important since children spend a significant portion of the day in school environments. However, many studies have reported substantial air quality problems in schools resulting from numerous sources found in indoor and outdoor environments. Particle re-suspension and VOCs emission from building materials and furniture are major air pollutants, common in classrooms. In outdoor environments, traffic emissions are primary sources of particulate and gaseous pollutants. Further, outdoor pollutants easily infiltrate into classrooms and other indoor spaces in schools. There are only limited air quality investigations in schools in developing countries. In particular schools located in rural areas have never been investigated and their exposure and health effects are not known.

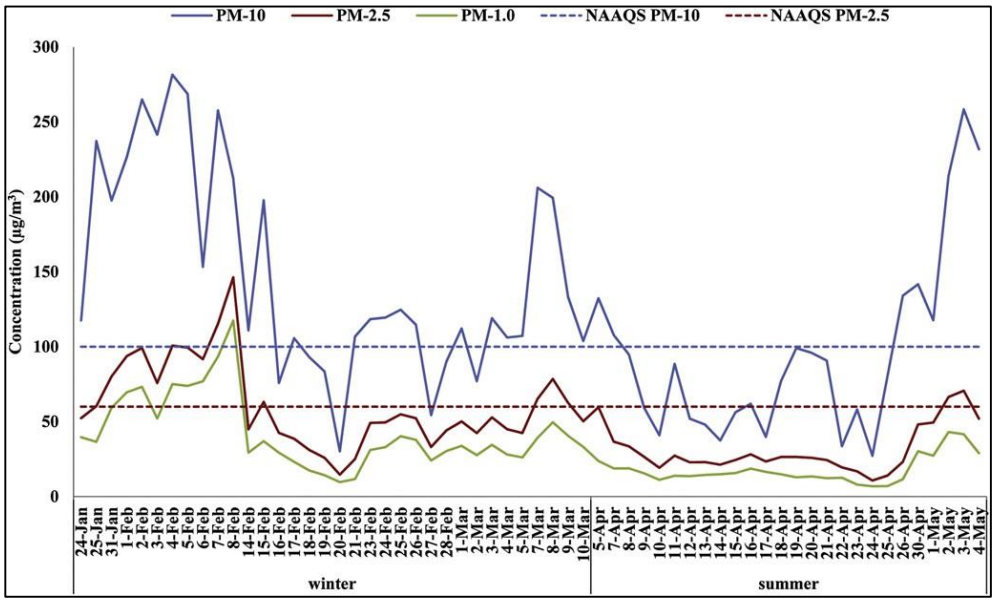

Figure 2.8: 24-hours PM_{10}, $PM_{2.5}$, $PM_{1.0}$ concentrations inside the classroom in India.
NAAQS: National Ambient Air Quality Standard for India.
Source: (Chithra and Shiva Nagendra, 2012).

Table 2.7: Example of air quality studies in schools

Study	**Location**	**Pollutant**	**Indoor Mean (SD), [95% CI]**	**Outdoor Mean, (SD) [95% CI]**	**Mean I/O (SD)**
(Lee and Chang, 2000)	Hong Kong - 5 schools	PM_{10} (μg/m^3)	21 – 617		
(Godwin and Batterman, 2007)	Michigan, USA 9 schools	Benzene (μg/m^3) Toluene (μg/m^3) Ethylbenzene (μg/m^3) o-Xylene (μg/m^3) Styrene (μg/m^3) 1,2,4,Trimethylbenzen (μg/m^3) α-Pinene (μg/m^3) Limonene (μg/m^3)	0.09 2.81 0.24 0.24 0.04 0.14 1.35 4.41	0.06 0.52 <0.01 <0.01 <0.01 0.01 0.11 0.29	1.4 – 10 >4 >10 >10 >4 >10 >10
(Stranger et al., 2008)	Antwerp, Belgium 27 schools	Benzene (μg/m^3) Toluene (μg/m^3) Ethylbenzene (μg/m^3) o-Xylene (μg/m^3) (m+p)-Xylene (μg/m^3)	1.54[a] 0.41[b] 5.23[a] 3.64[b] 1.46[a] 0.68[b] 0.57[a] 0.37[b] 3.58[a] 1.70[b]	1.87[a] 0.40[b] 4.29[a] 1.70[b] 0.42[a] 0.57[b] 1.19[a] 0.56[b] 2.13[a] 0.88[b]	0.9 1.0 1.2 2.5 1.6 2.6 1.6 2.5 1.9 2.6
(Weichenthal et al., 2008)	Ontario. Canada 2 schools	$PM_{2.5}$ (μg/m^3) PN (particles/cm^3)	10.9 (10) – 22.6 (22.7) 4.6×10^3 (2.2×10^3) – 5.4×10^3 (3.2×10^3)	- 1.3×10^4 (8.5×10^3) – 1.5×10^4 (1.1×10^4)	
(Zhao et al., 2008)	Taiyuan, China 10 schools	SO_2 (μg/m^3) NO_2 (μg/m^3) O_3 (μg/m^3) Formaldehyde (μg/m^3)	264.8 (139.0) 39.4 (9.5) 10.1 (10.4) 2.3 (1.1)	712.8 (189.3) 52.3 (9.5) 12.4 (3.3) 5.8 (0.6)	0.38 (0.17) 0.78 (0.22) 0.91 (0.93) 0.39 (0.18)

Table 2.7: Continued

Study	Location	Pollutant	Indoor Mean (SD), [95% CI]	Outdoor Mean, (SD) [95% CI]	Mean I/O (SD)
(Wichmann et al., 2010)	Stockholm, Sweden 6 schools	$PM_{2.5}$ ($\mu g/m^3$) NO_2 ($\mu g/m^3$)	8.1 (2.9) 17.3 (12.5)	9.7 (4.5) 20.6 (16.0)	0.94 (0.47) 0.96 (0.36)
(Almeida et al., 2011)	Lisbon, Portugal - 3 schools	$PM_{2.5}$ ($\mu g/m^3$) $PM_{2.5-10}$ ($\mu g/m^3$)	10 73	3 – 10 8 – 47	
(Mullen et al., 2011a)	Northern California, USA - 6 schools	PN (particles/cm^3)	6.9×10^3	1.5×10^4	0.47
(Pegas et al., 2011)	Lisbon, Portugal - 14 schools	NO_2 ($\mu g/m^3$)	14.6 – 37.4[c] 12.4 – 45.8[d] 10.2 – 33.6[e]	35.7 – 45.9[c] 34.7 – 50.1[d] 22.8 – 41.3[e]	0.36 – 0.95 0.35 – 0.99 0.44 – 1.00
(Sofuoglu et al., 2011)	Izmir, Turkey	$TOVC_{51}$ ($\mu g/m^3$) Formaldehyde ($\mu g/m^3$)	10 (8) – 27 (11)[f] 51 (48) – 111(64)[e] 32 (16) – 66 (49)[c] 19 (4) – 55 (32)[f] 30 (17) – 49 (20)[e] 36 (13) – 44 (16)[c]	10 (4) – 22 (2)[f] 16 (9) – 41 (19)[e] 13 (8) – 25 (2)[c] 2.3 (1.5) – 8.8 (2.1)[f] 5.9 (3.4) – 7.6 (0.4)[e] 0.6 (0.03) – 7.6 (1.3)[c]	
(Chithra and Shiva Nagendra, 2012)	Chennai, India 1 school	PM_{10} ($\mu g/m^3$) $PM_{2.5}$ ($\mu g/m^3$) PM_1 ($\mu g/m^3$) CO (ppm)	149 (69)[e], 95 (61)[g] 61 (29)[e], 32 (16)[g] 43 (24)[e], 18 (9)[g] 0.10 (0.18)[e], 0.11 (0.14)[g]		2.52 (2.71) 1.44 (0.67) 0.97 (0.18) 0.51 (0.38)
(Buonanno et al., 2013a)	Cassino, Italy - 3 schools	PN (particles/cm^3)	$2.0 \times 10^4 – 3.5 \times 10^4$	$2.8 \times 10^4 – 4.7 \times 10^4$	0.63 – 0.74
(Raysoni et al., 2013)	El Paso, Texas, USA - 4 schools	PM_{10} ($\mu g/m^3$) $PM_{2.5}$ ($\mu g/m^3$) BC ($\mu g/m^3$) NO_2 (ppb) Benzene ($\mu g/m^3$) Toluene ($\mu g/m^3$) Ethylbenzene ($\mu g/m^3$) (m+p)-Xylene ($\mu g/m^3$) o-Xylene ($\mu g/m^3$)	15 (5.3) – 37 (17) 8.2 (3.1) – 15 (6.0) 0.16 (0.24) – 0.38 (0.25) 3.29 (1.38) – 8.84 (2.68) 0.34 (0.15) – 0.96 (0.36) 0.92 (0.34) – 4.45 (4.20) 0.30 (0.15) – 0.84 (0.51) 0.56 (0.24) – 1.61 (0.65) 0.26 (0.09) – 0.62 (0.23)	21(19) – 35 (17) 9.8 (6.0) – 14.0 (6.1) 0.21 (0.24) – 0.53 (0.32) 3.58 (1.74) – 10.69 (3.09) 0.52 (0.21) – 1.51 (0.52) 1.16 (0.58) – 4.25 (1.81) 0.27 (0.13) – 0.86 (0.36) 0.68 (0.35) – 2.26 (0.94) 0.28 (0.12) – 0.88 (0.36)	0.49 – 1.12 0.68 – 1.00 0.32 – 1.42 0.70 – 1.02 0.64 – 0.74 0.98 – 1.22 0.81 – 1.19 0.73 – 0.91 0.76 – 1.00

Table 2.7: Continued

Study	Location	Pollutant	Indoor Mean (SD), [95% CI]	Outdoor Mean, (SD) [95% CI]	Mean I/O (SD)
(Zwoździak et al., 2013)	Wroclaw, Poland 1 school	PM_{10} ($\mu g/m^3$) $PM_{2.5}$ ($\mu g/m^3$) PM_1 ($\mu g/m^3$)	68.5 (21.8)[e], 43.1 (17.9)[g] 59.8 (21.6)[e], 13.5 (4.1)[g] 21.2 (5.3)[e], 8.5 (3.6)[g]	56.8 (17.3)[e], 24.7 (10.5)[g] 49.1 (15.6)[e], 16.0 (9.1)[g] 25.5 (7.8)[e], 8.9 (3.4)[g]	2.5 (1.7), 5.6 (2.9) 2.0 (0.8), 4.1 (2.9) 0.8 (0.2), 1.3 (0.4) - only day time I/O presented
(Fonseca et al., 2014)	Paranhos & Xisto, Portugal - 3 preschools	PN (particles/cm^3)	$1.2 \times 10^4 – 1.8 \times 10^4$	$1.0 \times 10^4 – 1.7 \times 10^4$	0.54 – 0.93
(Laiman et al., 2014)	Brisbane, Australia - 25 schools	PN (particles/cm^3)	7.7×10^4 [1.19, 25.89][h] 4.7×10^4 [0.64, 16.86][i] 4.3×10^4 [0.78, 14.36][j]	1.2×10^4 [0.23, 3.95] 1.2×10^4 [0.18, 3.64] 0.7×10^4 [0.15, 2.28]	6.26 4.29 5.74
(Jovanović et al., 2014)	Zajecar, Serbia 1 school	PM_{10} ($\mu g/m^3$) $PM_{2.5}$ ($\mu g/m^3$) TVOCs ($\mu g/m^3$) Formaldehyde ($\mu g/m^3$) Ozone ($\mu g/m^3$) NO_2 ($\mu g/m^3$)	70.63 (19.8) 43.58 (12.9) 48.67 (11.3) 63.74 (22.8) 15.51 (6.50) 15.02 (7.50)	- - 1.46 5.07 217.71 9.14	1.01 0.98 31.96 12.58 0.07 1.64
(Rivas et al., 2014)	Barcelona, Spain - 39 schools	$PM_{2.5}$ ($\mu g/m^3$) NO_2 ($\mu g/m^3$) PN (particles/cm^3)	37 (13) 30 (12) 1.6×10^4 (6.6×10^3)	29 (20) 47 (17) 2.3×10^4 (1.0×10^4)	

[a] Urban, [b] Suburban, [c] Spring, [d] Autumn, [e] Winter, [f] Fall, [g] Summer, [h] During printing, [i] During heating, [j] During cleaning

2.8 Air quality characterization in transportation microenvironments

Human exposures in transportation microenvironments, such as on-roads, roadsides, inside vehicles and public transport facilities are a growing concern due to higher pollution levels encountered in these environments compared to ambient and background levels (Kaur et al., 2007, Jayaratne et al., 2008, Wang et al., 2011). For example, Hitchins et al. (2000) evaluated the horizontal distribution of vehicle emissions at different distances from the major road. They found that roadside concentrations of fine particles and UFP measured at 15 meters from the road (the nearest sampling point from the road) decreased to half at 50 – 150 meters depending on the wind direction. Kittelson et al. (2004) also measured PN concentrations at different distances from the highway. They observed high PN concentrations of 4.03×10^5 particles/cm^3 on-road, with much lower concentration (3.80×10^4 particles/cm^3) at 10 meters, and significantly lower concentration (9.44×10^3 particles/cm^3) at 700 meters from the highway. The measurements done at 36 roadside locations showed mean concentrations two to three times higher for $PM_{2.5}$, PM_{10}, and $PM_{10\text{-}2.5}$, respectively compared to background levels (Colbeck et al., 2011). Inside the tunnel, average CO, NO_x, and $PM_{2.5}$ concentrations were found to be 17, 25, and 8 times higher than the background concentrations (Kirchstetter et al., 1999). Similarly, other studies have observed higher pollutant concentrations inside vehicles compared with simultaneously measured ambient levels (Gouriou et al., 2004, Apte et al., 2011, Colbeck et al., 2011).

Time spent in transportation microenvironments may represent only a fraction of the total daily time, for example 6% of the day inside vehicles in the United States (Klepeis et al., 2001), and 3 – 17% in Delhi depending on gender and occupation (Saksena et al., 2007). However, due to high pollution levels typical to such environments, it can contribute to a significant portion of the daily exposure, thereby accounting for the disproportionate fraction of exposure (Kaur et al., 2007, Fruin et al., 2008, Wallace and Ott, 2011, Wang et al., 2011). A review of 47 studies on commuter exposure to UFP, across different transport modes

indicated evidence of substantial daily exposure during transit with a mean of 3.4, 4.2, 4.5, 4.7, 4.9 and 5.7 × 10^4 particles/cm^3 for bicycle, bus, automobile, rail, walking and ferry modes, while mean in-vehicle exposure in tunnel routes was 3.0 × 10^5 particles/cm^3 (Knibbs et al., 2011).

2.8.1 Traffic emissions along roadways

Along the roadways, vehicle emissions are the primary source of air pollution (Peltier et al., 2011). Because of the low source height, traffic emissions can result in severe local air quality problems (Pirjola et al., 2006). Particulate matter in traffic emissions come from both combustion (tailpipe/exhaust emissions) and non-combustion (non-exhaust emissions) traffic activities (Hussein et al., 2008, Amato et al., 2011b). A number of factors such as engine type and size, fuel composition, combustion temperature and pressure, road conditions, age and maintenance of vehicles, traffic volume, and driving behaviour significantly affect emission levels (Zavala et al., 2006, Pant and Harrison, 2013). Besides particles, exhaust emissions contain several gaseous pollutants such as NO_x, CO, CO_2, VOCs and ozone (Pirjola et al., 2006, Rogers et al., 2006).

2.8.2 Exhaust emissions

Exhaust particulate emissions are of two modes: (i) primary particles or soot generated as a result of fuel combustion in the engine, and (ii) secondary particles formed as a result of nucleation in the atmosphere during dilution and cooling of exhaust (Pirjola et al., 2006). Exhaust emissions contribute to fine particles (Dahl et al., 2006, Kam et al., 2012, Kwak et al., 2014), and ultrafine particles (Westerdahl et al., 2005, Morawska et al., 2008, Pey et al., 2009). In general diesel vehicles were associated with higher emission of particles than gasoline vehicles. The size of particles emitted from the diesel engines range from 20 – 130 nm, while those from gasoline engines range from 20 – 60 nm (Morawska et al., 1998, Ristovski et al., 1998). Kirchstetter et al. (1999) observed that heavy-duty diesel trucks emitted 24, 37, and 21 times more $PM_{2.5}$, BC, and sulphate mass per unit mass of fuel burnt

than gasoline powered light-duty vehicles. Fruin et al. (2008) found that diesel powered vehicles, despite constituting only 6% of the total traffic contributed to the highest concentrations of UFP, BC and nitric oxide (NO). In high traffic density areas, Mejía et al. (2008) found that UFP constituted 82 – 90% of particle number concentrations, while Kittelson et al. (2004) observed majority of the particles in nanoparticle range (< 50 nm). Further, highest PN concentrations were associated with high traffic speed, which was due to the increase in engine load, exhaust temperature and flow at higher vehicle speed (Kittelson et al., 2004). Between the compressed natural gas (CNG) and ultra-low diesel buses, Jayaratne et al. (2009) reported higher PM_{10} emissions of at least two orders magnitude for diesel buses than CNG buses fitted with oxidation catalysts. However, particle number emissions were not statistically different between the two types of buses.

2.8.3 Non-exhaust emission

Non-exhaust particulate matter emissions result from resuspension of road dust, wear and tear of vehicle parts (such as brake, tyre and clutch), and road pavement abrasion, mainly contributing to coarse range ($PM_{2.5\text{-}10}$ μm) (Dahl et al., 2006, Kam et al., 2012, Kwak et al., 2014). However, re-suspension of accumulated road dusts and road wear particles constitute the major fraction of non-exhaust emissions, while particles from tyre and brake wear make minimal contributions (Keuken et al., 2010). Primary PM_{10} in urban areas mainly originate from non-exhaust traffic emissions (Dahl et al., 2006, Hussein et al., 2008). However, some studies have confirmed UFP presence in non-exhaust emissions, which were linked to tyre-road pavement interaction (Dahl et al., 2006, Kwak et al., 2014).

Understanding of non-exhaust emissions is essential since they are not only inherently toxic but also play an important role as a carrier medium for other species such as heavy metals and carcinogens (Amato et al., 2011a). While zero tailpipe emissions can be achieved with the adoption of cleaner fuels and emission standards, non-exhaust emissions are expected to remain the dominant vehicular emissions (Dahl et al., 2006, Kaur et al., 2007,

Rexeis and Hausberger, 2009). In fact, it was estimated that between 80 – 90% of total PM emissions from road traffic will come from dust suspension and other abrasion processes by 2020 (Rexeis and Hausberger, 2009). This is because there are currently no abatement strategies for non-exhaust emissions (Amato et al., 2011a). Besides, the detrimental health impacts, non-exhaust emissions largely contribute to non-conformity to air quality standards (Amato et al., 2011b). There has been a significant research on characterization of exhaust emissions, resulting in a large body of knowledge. However, understanding of non-exhaust emissions while important is currently limited, hence there is a need for more research in this field (Amato et al., 2011a, Pant and Harrison, 2013).

2.8.4 Mobile measurements on roadways

Characterization of on-road air quality is challenging given the long distances involved and the fact that conventional air pollution monitors are not suitable for measurements in such environments. Particularly, fixed monitors do not account for the actual pollutant concentrations found in the heart of the road traffic (Gouriou et al., 2004). This has led to the development of vehicle-based mobile sampling methods specifically aimed at quantifying on-road pollution during real-world driving (Gouriou et al., 2004, Knibbs et al., 2009).

Many on-road studies using vehicle based mobile measurements for roadways are described in the literature. One of the first such studies was designed by Kittelson et al. (2000), where they used a mobile emission laboratory (MEL) for continuous measurement of particle number, surface area and size distribution, along with CO_2 and NO_x from vehicle exhausts on highways. Following this, another vehicle based mobile platform was designed by Bukowiecki et al. (2002), where they assessed temporal and spatial variations of aerosols and gases along roadways. In subsequent and recent years several studies quantifying on-road pollution using mobile platforms have been reported. Around the world on-road mobile measurements (for example) have been used for both tunnel (Gouriou et al., 2004, Yao et al.,

2007, Knibbs et al., 2009) and above-ground urban road studies (Kittelson et al., 2004, Westerdahl et al., 2005, Pirjola et al., 2006, Zavala et al., 2006, Yao et al., 2007, Fruin et al., 2008, Wang et al., 2009, Westerdahl et al., 2009, Guo et al., 2014). All of these studies have highlighted the advantages of vehicle-based mobile quantification of on-road pollution levels.

2.8.5 Focus of vehicle based on-road measurements

Mobile platforms have been used for a range of on-road emission investigations. This included estimation of emission factors (Kittelson et al., 2004, Westerdahl et al., 2009, Guo et al., 2014), in-vehicle exposure assessment (Fruin et al., 2004, Gouriou et al., 2004, Zhu et al., 2008, Apte et al., 2011, Colbeck et al., 2011), quantification of non-exhaust vehicle emissions (Hussein et al., 2008, Kwak et al., 2014) and evaluation of air quality control measures (Wang et al., 2009, Westerdahl et al., 2009).

2.8.5.1 Emission factors

Using mobile platform, Guo et al. (2014) characterized fuel emission factors from 13 different buses during real-world driving along their daily routes. They have reported that CNG buses emitted more CO and total hydrocarbons but less NO_x and particulate matter compared with diesel buses. Kittelson et al. (2004) estimated fuel specific and particles/km emissions from gasoline dominated vehicle fleet on highways which were reported to be 2.2 – 11 × 10^{15} particles/(kg-fuel) and 1.9 – 9.9 × 10^{14} particles/km. Similarly, Westerdahl et al. (2009) characterized on-road emission factors for light and heavy duty vehicles, the CO and PN values were reported to be 95 g/kg-fuel and 1.8 × 10^{15} particles/(kg-fuel) for light duty vehicles, and 50 g/(kg-fuel) and 1.1 × 10^{16} particles/(kg-fuel), respectively for heavy duty vehicles.

2.8.5.2 In-vehicle exposure

Studies have also taken the advantage of the versatility of mobile platforms for assessing in-vehicle exposure during real-life driving along roadways. Such direct measurements provide accurate quantification of exposure acquired during commuting on

roads. For example, Gouriou et al. (2004) used mobile platform to measure exposure of car passengers to PN. They reported that passengers were exposed to concentrations up to 10^6 particles/cm^3 during the road journey. Apte et al. (2011) conducted exposure measurement inside the auto-rickshaw (a semi enclosed vehicle which is a common transport mode in South Asian urban areas) in New Delhi. The measured mean $PM_{2.5,}$ BC and PN exposure of 200 µg/m^3, 43 µg/m^3, and 2.9 × 10^5 particles/cm^3, respectively were reported to be higher than in-vehicle exposure reported for megacities around the world. Further, this study reported that in-vehicle concentrations were 1.5 – 8.4 times higher than simultaneously measured ambient levels and that exposure during daily commuting in Delhi was as large as the total daily exposures for urban residents in many developed countries. Similarly, in-vehicle exposures in Jakarta were higher than other environments by 30 – 200% for $PM_{2.5}$, 30 – 300% for PN, and 180 – 700% for, CO, respectively (Both et al., 2013). In contrast, Colbeck et al. (2011) reported lower in-vehicle concentrations for different particle mass fractions (PM_{10}, $PM_{2.5}$, PM_1, $PM_{10-2.5}$) compared to simultaneously measured (fixed site) roadside concentrations. However, in-vehicle concentrations of all particle fractions were still very high.

Several factors are likely to influence in-vehicle exposure levels such as the mode of transport, features and characteristics of vehicles, vehicle speed, ventilation and meteorological conditions (wind speed and wind direction) (Kaur et al., 2007). For example, between different transport modes, Chan et al. (2002) found the highest in-vehicle PM_{10} exposure (203 µg/m^3) for a non-air conditioned bus, while the highest CO exposure (28.7 ppm) was observed for an air conditioned taxi. Likewise, relative exposure assessment for different modes of transport done along the same route found that car passengers were exposed to higher concentrations of CO than those commuting by bus and cycle, while UFP exposure was higher for car and bus passengers than for cyclists (Kingham et al., 2013). A lower exposure for cyclists is expected since they are able to avoid busy traffic by commuting

along bike routes and pedestrian walkways, unlike vehicle commuters who need to align directly behind the emission source (Kaur et al., 2007). Additionally, poor vehicle maintenance causes higher in-vehicle particle mass and CO exposure (Chan et al., 2002). Further, due to high air exchange rates typical of moving vehicles, in-vehicle pollutant concentrations are likely to be close to on-road concentrations (Fruin et al., 2008).

2.8.5.3 Non-exhaust traffic emissions

Hussein et al. (2008) investigated factors affecting non-exhaust particle emissions on roads using a mobile measurement system. One of the significant findings of this study was that road dust emissions depended on tyre types with higher emissions from the studded tyre, 2 – 6.4 times the friction tyre and 4.4 – 17 times the summer tyre, respectively. Further, it was found that main cause of emission from the friction tyre was due to the suspension of road dusts, while road wear was the main cause for the studded tyre. A similar approach was used by Kwak et al. (2014) to quantify non-exhaust UFP concentrations from tyre-road surface interactions. A significant increase in UFP concentrations was observed during braking and cornering compared to driving under constant speed. The reasons attributed for the increase in UFP concentrations during braking were volatilization of tyre components, extender oils, and bitumen from the road pavement.

2.8.5.4 Evaluation of air quality regulations

Mobile platforms have been successfully used to evaluate the effectiveness of air quality control measures in Beijing during the 2009 Summer Olympics. In response to severe air quality problems and in preparation for the Olympics, Beijing introduced several short and long-term emissions reduction regulations both before and during the Olympics. Wang et al. (2009) conducted mobile on-road measurements along the major road in Beijing to evaluate the effectiveness of administrative regulations by measuring the temporal and spatial variations of multiple pollutants. They found significant reductions in concentrations during the control periods (up to 71%) compared to the pre-control periods. Concentrations

gradually increased after the control measures had been lifted. A parallel investigation was also conducted in Beijing by Westerdahl et al. (2009) using a mobile method. They also reported significant reduction in on-road concentrations, thus proving the usefulness of mobile methods in evaluating the effectiveness of control measures.

2.8.6 On-road air quality investigations in developing countries

Despite air quality generally being poor on roads in developing countries, where there is also a high population density living near roadsides, there have been very limited studies focussing on road exposures. A literature review on commuter exposure has noted the scarcity of studies from developing countries (Knibbs et al., 2011). While a few on-road studies were reported from Asian countries, investigations were done along busy metropolitan roads where traffic volume was the important determining factor (Saksena et al., 2007, Yao et al., 2007, Wang et al., 2009, Westerdahl et al., 2009, Apte et al., 2011, Colbeck et al., 2011, Guo et al., 2014, Kwak et al., 2014). On-road studies have not been extended to major roads traversing rural areas and in a range of geographical conditions. While traffic volume is an important determinant of air quality along urban roads, a host of potential sources are likely to be present along rural roads. In addition to traffic contributions, local combustion activities such as agricultural and residential wood burning are likely to contribute to on-road pollution levels for rural roads in developing countries, including long-range transport of regional and urban pollutants. Therefore, the contributions of both traffic and non-traffic activities to on-road pollution as well as exposure for commuters and roadside residents outside urban and metropolitan areas are not known. In fact, the main focus of air quality studies in developing countries has been on HAP and the health effects of occupants exposed to it. While health effects of exposure to HAP and traffic emissions, and other sources found on roadways are likely to vary, there are no studies focussing on roadway exposure in rural areas.

In summary, air quality characterization in transportation microenvironments is important since people spend a significant portion of their time in these environments. Studies have shown that on-road and roadside pollutant concentrations were significantly higher than other environments. Besides commuting, many people live near busy roads, exposed to high concentration of pollutants. Fixed site monitoring is not suitable for on-road measurements, since large network of monitoring sites would be required to capture the temporal and spatial distribution of pollutants. Several studies have successfully used mobile platforms for on-road characterization of pollutants during real-world driving. The focus of the mobile platform measurements varied from estimation of vehicle-fuel specific emission factors to in-vehicle exposure and for evaluation of air quality control measures. There have been a few on-road studies in the developing world, focussing on busy urban roadways. So far no studies have been conducted for major roadways in rural areas. This is important because exposure patterns and therefore health risks can be different from that of urban roads, given the existence of different pollution sources in rural areas.

2.9 Air quality concerns in Bhutan

Bhutan (population ~700,000) is an eastern Himalayan nation with an area of 38,394 square kilometres, landlocked and bordered by India and China. Nearly 70% of the population live in rural areas and are subsistence farmers, depending mainly on farming, livestock rearing and a range of natural resources. 70.46% of Bhutan's total land area is under forest cover (RGoB, 2010). The country is rich in water resources with enormous potential for hydropower development. Of the estimated 30,000 megawatt potential, only about 1500 megawatt has been tapped so far through large and small scale hydropower plants (DoE, 2009).

2.9.1 Outdoor air quality problems in Bhutan

There is a growing concern about outdoor air pollution and its effects on health and the environment in Bhutan, especially in urban centres from vehicular emission. As of 2013,

Bhutan had a total of 76,926 vehicles, with 53% registered in the western region, 36% in the southern region, 6% in the central region, and 5% in the eastern region, and with a sharp increase being reported each year (NSB, 2014). Most of these vehicles are concentrated in a few urban centres, resulting in localised air quality problems. In response to this, the Bhutanese Government has implemented several strategies to curb vehicular pollution by banning leaded gasoline, import of used cars and regular emission tests for all kind of vehicles (Phuntsho and Kanitpong, 2010).

National Environment Commission (NEC) is an official organisation in Bhutan mandated with overseeing the environmental health in the country. A few years ago NEC started monitoring PM_{10} levels, using high volume samplers in selected places, which includes both urban and rural areas. Table 2.8 shows the National Ambient Air Quality Standards for Bhutan. From the available limited data, there were some indications of particulate matter levels exceeding WHO guidelines in urban centres (NEC, 2010). NEC is also monitoring industrial point source emissions annually from major industries.

Currently, effective air quality monitoring in Bhutan is challenged by a number of limitations, such as lack of adequate technical capacity, instrumentation and financial capacity to undertake regular and comprehensive monitoring. However, Bhutan has good leadership and a government which is keen on solving ambient air quality problems in the country. Plans are underway to expand and strengthen monitoring stations across the country in collaboration with academic institutions and other relevant agencies. Bhutan is already a signatory to a regional declaration on Control and Prevention of Air Pollution and its Likely Transboundary Effects for South Asia, The Male Declaration. This declaration requires member countries to establish and strengthen existing air quality monitoring systems. Recently, the current Government went a step ahead by declaring Bhutan as a hub for electric cars. Under this scheme, the conventional cars are heavily taxed, over 100%, while zero tax is levied for electric cars.

Table 2.8: Ambient Air Quality Standards for Bhutan (µg/m^3)

Parameter	Averaging Time	Industrial Area	Mixed Area	Sensitive Area
Total Suspended Particulate Matter	24 hours	500	200	100
	Annual average	360	140	70
Respirable Particulate Matter (PM_{10})	24 hours	200	100	75
	Annual average	120	60	50
Sulphur Oxide	24 hours	120	80	30
	Annual average	80	60	15
Nitrogen Oxides	24 hours	120	80	30
	Annual average	80	60	15
Carbon Monoxide	8 hours	5000	2000	1000
	1 hour	10,000	4000	2000

Mixed areas refer to residential, commercial or both, while sensitive areas include places where hospitals, schools, or important ecosystems are located. Source (NEC, 2010).

2.9.2 Indoor air quality problems in Bhutan

The primary energy source for the majority of Bhutanese population is biomass which includes firewood, woodchips and animal dung (DoE, 2009). The residential sector is the highest consumer of energy with 48.7%, and 91% of this constituted biomass (DoE, 2009). At the household level, energy is used for cooking meals, space heating and lighting, fodder preparation for cattle, and liquor distillation. Generally, multiple energy sources are used for cooking, while many houses, particularly in rural areas use traditional stoves for cooking and space heating. There is a significant difference in energy usage pattern between urban and rural areas (Table 2.9). While urban households mostly rely on electricity, LPG and kerosene, rural areas primarily depend on firewood. The amount of firewood used is significant in Bhutan, and country has the highest global per capita firewood usage (DoE, 2009).

A record with Bhutan Electricity Authority (BEA) showed that 68,590 rural households of the total 88,642 have been electrified as of December 2010 (Personal Communication, BEA, 23 March 2012). However, except for lighting where electricity has become the primary source, multiple fuels are still used for cooking in rural areas. The electrified rural households were found to consume only about 25% less firewood than un-

electrified houses, with per capita wood consumption of 10.4 and 7.8 tonnes per annum for rural un-electrified and electrified houses, respectively (DoE, 2005). This is because many household activities such as fodder preparation, brewing and distilling local liquor, which are energy and time intensive cannot be done using standard gas or electric stoves. The fodder preparation alone accounted for 26% of the residential energy consumption in rural areas (DoE, 2010).

Table 2.9: Energy sources for lighting and cooking in urban and rural areas in Bhutan

Energy Source	Lighting (%)		Cooking (%)	
	Urban HH	Rural HH	Urban HH	Rural HH
Electricity	96.3	40.0	46.5	21.4
Kerosene	2.6	51.2	5.4	6.6
Firewood	0.2	3.7	4.3	56.4
Solar	0.3	3.5	0.0	0.1
LPG	0.1	0.1	43.6	15.2
Others	0.5	1.5	0.2	0.3

HH: Households, Source: (RGoB, 2006).

To date, quantitative information related to HAP does not exist in Bhutan. However, fuels used for cooking and space heating could seriously compromise the IAQ, even for the urban households. Use of metal combustion stoves, where wood is burnt for space heating (usually not used for cooking), locally known as *bukhari* (Figure 2.9) is increasingly used in both urban and rural areas. Although this device has a chimney, during initial lighting, the door of the combustion chamber has to be left open and usually a small amount of kerosene is added to start the fire. This activity generates a thick smoke, which readily disperses into the indoor space. Also, during the windy days, a lot of smoke is forced back through the chimney into the indoor environment.

In rural un-electrified houses, cooking is normally done on traditional stoves, without a chimney (Figure 2.10). It consists of one or two openings through which firewood is fed and ash is removed, and two or three potholes, with raised lumps where pots can rest. The thermal efficiency of traditional stoves were found to range from 8 – 18% (DoE, 2005).

Efforts have been made in the past to reduce the HAP and pressure on forests by promoting improved cookstoves. The National Women Association of Bhutan was one of the first organizations to disseminate this program with assistance from the UNICEF and the government. In early 1990s, an estimated 14000 – 15000 improved stoves were installed free of costs in different parts of the country (Palit and Garud, 2010).

Some qualitative reviews to assess the benefits of improved stoves following its successful dissemination have been done in the past. In two of the projects funded by the United Nation Development Programme (UNDP) in Bhutan, a review has found that wood consumption was lowered by about 50%, and fuel collection time lowered significantly (Ugyen and Giri, 2004). Further, this report also highlighted perceived health benefits enjoyed by the people with less smoke exposure. However, several factors have limited the sustained use of improved stoves. Overtime stoves became non-operational and people eventually switched to traditional stoves. The major drawback perceived by the users was that stoves had poor heat radiation capacity, thereby limiting room heating ability and extending the cooking time (Ugyen and Giri, 2004, Palit and Garud, 2010). Thus, users ended up modifying the pothole dimensions, inflicting structural damage to stoves. Further, chimneys were removed by people due to perceived fire hazards, and also traditionally rural people need smoke to dry chilli and grain in the kitchen. While the burden of respiratory diseases in the country is substantial (MoH, 2007), to date no quantitative data on HAP is available in Bhutan.

In summary, there is a growing concern about air quality problems in Bhutan due to rapid urbanisation and public infrastructure development. For nearly 70% of the population living in rural areas and depending mainly on solid fuels, HAP is the major concern. This makes them very vulnerable to health effects of household combustion products. Yet, no quantitative air quality study has been done till date. Therefore, air quality studies are clearly

needed in Bhutan to support the formulation of policies to aid the reduction of health risks and the promotion of a healthy lifestyle.

Figure 2.9: Metal chimney stove (bukhari) used for heating and usually not for cooking.

Figure 2.10: Traditional mud cookstove used for cooking and space heating.

2.10 Knowledge gaps

The literature review has identified several knowledge gaps in the current literature:

- Particulate matter is by far the most common and complex combustion product found in indoor and outdoor environments, and the magnitude of its health effects is depended on particle size. Currently there is no data available for ultrafine particles from houses relying on solid fuels (Section 2.6.2), as well as in school environments (Section 2.7.4) in developing countries.
- Although emission characteristics from household stoves used for cooking and heating have been conducted, there are still limited data from the field during actual cooking and heating activities. In particular no data is available for ultrafine particles from household use of solid fuels (Section 2.6.4).
- Air quality investigations in schools have focussed on pollution sources present in the immediate indoor and outdoor environments. There are no data for long-range transport of regional and urban pollutants and their influence on school air quality (Section 2.7.2 and 2.7.3).
- In developing countries, a handful of available air quality studies for schools were done in big cities and near busy traffic areas. Currently, there are no data available for air quality in schools located in rural areas, where pattern of exposure and health risks can be different due to existence of different sources compared with urban schools (Section 2.7.4).
- Both exhaust and non-exhaust traffic emissions contribute to high pollutant concentrations in transportation microenvironments. The extent of non-exhaust emissions and their health effects have not been sufficiently characterized (Section 2.8.1).
- In developing countries, human exposure in transportation microenvironments is less known. The available studies were done in relation to commuters along urban roads.

The pollution levels and exposure for major roads extending to rural areas are currently not known, where large number of people commute and live near the roadsides (Section 2.8.6).

- Deteriorating air ambient quality has become a major concern for Bhutan. 70% of the population live in rural areas and heavily rely on biomass fuels for cooking and space heating. Currently, no scientific air quality information is available for both ambient and household air pollution in Bhutan (Section 2.9).

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Chapter 3: A Pilot Study of Traditional Indoor Biomass Cooking and Heating in Rural Bhutan: Gas and Particle Concentrations and Emission Rates

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Statement of Contribution

Statement of Contribution

The authors listed below have certified* that:

- they meet the criteria for authorship in that they have participated in the conception, execution, or interpretation, of at least that part of the publication in their field of expertise;
- they take public responsibility for their part of the publication, except for the responsible author who accepts overall responsibility for the publication;
- there are no other authors of the publication according to these criteria;
- potential conflicts of interest have been disclosed to (a) granting bodies, (b) the editor or publisher of journals or other publications, and (c) the head of the responsible academic unit, and
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In the case of this chapter: *A Pilot Study of Traditional Indoor Biomass Cooking and Heating in Rural Bhutan: Gas and Particle Concentrations, and Emission Rates*

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Tenzin Wangchuk QUT Verified Signature Date: 24/08/2015	Conducted measurements, performed data analysis and wrote the manuscript.
Congrong He*	Contributed to data analysis and interpretation, and reviewed the manuscript.
Luke D. Knibbs*	Contributed to data analysis and reviewed the manuscript.
Mandana Mazaheri*	Contributed to data analysis and reviewed the manuscript.
Lidia Morawska*	Supervised the project, contributed to data analysis and interpretation, and reviewed the manuscript.

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Abstract

Although many studies have reported the health effects of biomass fuels in developing countries, relatively few have quantitatively characterized emissions from biomass stoves during cooking and heating. The aim of this pilot study was to characterize the emission characteristics of different biomass stoves in four rural houses in Bhutan during heating (metal chimney stove), rice cooking (traditional mud stove), fodder preparation (stone tripod stove) and liquor distillation (traditional mud stove). Three stage measurements (before, during and after the activity had ceased) were conducted for $PM_{2.5}$, particle number (PN), CO and CO_2. When stoves were operated, the pollutant concentrations were significantly elevated above background levels, by an average of 40 and 18 times for $PM_{2.5}$ and CO, respectively. Emission rates (mg/min) ranged from 1.07×10^2 ($PM_{2.5}$) and 3.50×10^2 (CO) for the stone tripod stove during fodder preparation to 6.20×10^2 ($PM_{2.5}$) and 2.22×10^3 (CO) for the traditional mud stove during liquor distillation. Usable PN data was only available for one house, during heating using a metal chimney stove, which presented an emission rate of 3.24×10^{13} particles/min. Interventions to control household air pollution in Bhutan, in order to reduce the health risks associated with cooking and heating are recommended.

Keywords: Bhutan, rural, emission rate, firewood, cooking, heating

Practical Implications

Household air pollution resulting from the use of solid fuels for cooking and heating is the leading cause of premature death in many developing countries. In order to assess the health risks, quantitative characterization of the emissions from stoves and cooking activities under real-world conditions is crucial. Our study adds to a small

number of studies that estimated emission rates from biomass stoves under real-world operating conditions, the findings of which will be useful for agencies responsible for public health care system and those working towards the improvement of indoor environments.

3.1 Introduction

Although there has been a 21% decrease globally in the number of households relying on solid fuels for cooking between 1980 and 2010, the number of people exposed to emissions has remained the same (around 2.8 billion) due to population growth (Bonjour et al., 2013). Household air pollution (HAP) is the single most significant global environmental risk factor, accounting for nearly 5% of the global burden of disease (expressed as disability adjusted life-years (DALYs)) (Smith et al., 2014). This presents significant health risks in developing countries (Balakrishnan et al., 2004), with an estimated 2.6 billion people depending on biomass fuels (i.e. wood, crop residues and cattle dung) for cooking (Legros et al., 2011). In rural areas, traditional stoves are often used in poorly ventilated kitchens (Rehfuess et al., 2006), and for relatively long durations (Rumchev et al., 2007). Unlike gas and electric stoves, the fuel combustion process in traditional stoves is very difficult to control and leads to different emission profiles during different stages of the combustion cycle. Women and children spend the greatest amount of time inside kitchens and therefore incur the highest pollutant exposures (Andresen et al., 2005, Barnes et al., 2005, Li et al., 2012). Particles emitted from cooking also readily infiltrate into living rooms, thereby extending the risks to all occupants (Dasgupta et al., 2009, Wan et al., 2011). Studies in developing countries have consistently demonstrated the adverse health effects of particulate matter (PM) and gaseous pollutants emitted during the combustion of biomass fuels (Rumchev et al., 2007, Kulshreshtha et al., 2008, Kumar

et al., 2008a, Lakshmi et al., 2010, Verma et al., 2010, Mengersen et al., 2011, Murray et al., 2012). A recent World Health Organization (WHO) report has attributed 4.3 million premature deaths in 2012 to HAP, with the majority in low and middle income countries (WHO, 2014b).

Despite the large number of studies highlighting the effects of biomass fuels on health in developing countries, relatively few studies have quantitatively characterized biomass stove emissions during cooking and heating within indoor environments. Most studies relied on measurements that provided time integrated average concentrations from which time resolved peak concentrations could not be obtained. Therefore, detailed quantification of emission rates, which are crucial for exposure assessment and for identifying the major contributors to emissions, cannot be determined. The high intensity peak emissions during cooking can account for 31 – 61% of the total exposure to particles, and neglecting the contribution of peak emissions, can lead to underestimation of exposure (Ezzati et al., 2000, Mazaheri et al., 2014). Good quality emission rate data collected under real-world conditions are important inputs for health impact assessment, and when comparing the effects of different stoves and cooking practices.

Bhutan (population ~700,000) is a small eastern Himalayan country with an area of 38,394 km^2, and is bordered by India and China. In general, the environmental conditions, as well as social characteristics of Bhutan, are largely comparable with the rest of the Himalayan region. Nearly 70% of the population live in rural villages and are subsistence farmers. Although most villages have access to electricity, the use of firewood in traditional stoves is very common for cooking and heating. Rural areas accounted for 96% of the total firewood consumption in Bhutan, and rural households with electricity consumed only 25% less firewood than those without (DoE, 2005).

This is largely because cooking activities, such as fodder preparation for cattle (usually a mix of vegetable wastes, rice husk and corn flour) and distilling home liquor (the primary cooking activities in village homes), cannot be done using standard electric or gas stoves due to the size of the pots needed for such activities. Also, refined fuels (e.g., liquefied petroleum gas (LPG) and kerosene) are expensive and in short supply in the villages, while wood is plentiful and can be collected freely. Bhutan has one of the world's highest rates of firewood consumption per capita (DoE, 2009). This large-scale use of firewood for cooking and heating in Bhutan is likely to result in poor indoor air quality (Bruce et al., 2013). Moreover, the burden of respiratory disease in Bhutan is substantial (MoH, 2007). However, there have been no studies on household air pollution exposure in Bhutan, which has made it difficult to estimate the health impacts of household biomass combustion.

As a part of a larger study investigating air quality in different microenvironments in rural villages in Bhutan, we sought to perform measurements of indoor biomass combustion products in a representative selection of rural village houses during cooking and space heating. The primary objectives were to estimate emission rates from biomass stoves, to compare the results with those derived from other studies and to determine if pollutant levels and emission rates in Bhutan differ from those reported in other economically developing countries.

3.2 Materials and Methods

3.2.1 Study design

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. Four houses (H1-H4) located in different villages 5-10 km apart were selected to represent the most common stove types and cooking

activities. The general characteristics of the four houses investigated are presented in Table 3.1. All houses were traditional structures built from mud, wood and stone, except for the walls of the H1, which were strengthened with concrete. H1 was occupied by a family of a government employee stationed in the village and used LPG and electricity for cooking meals in an indoor kitchen. H1 had a metal chimney stove (locally called bukhari) for space heating in the living room (Figure 3.1 (a)). This stove had a combustion chamber with a door through which wood is added and a separate chamber with a drawer for ash collection and removal. H2-H4 belonged to farmers and cooking was mostly done using traditional stoves in a separately structured kitchen outside the main house. H2 and H4 had traditional stoves, built mostly from mud (Figure 3.1 (b) and (d)). H3 had a stone tripod stove, the simplest open fire stove, where firewood can be fed from more than one direction (Figure 3.1 (c)).

This study involved no direct human participation. The Trashigang District Administration, Royal Government of Bhutan, provided approval of the study through letter no DAT/ADM(3)2012/6990 and a verbal consent was obtained from the head of the family for conducting measurements in the selected houses.

Table 3.1: General characteristics of the houses investigated

House	Stove	Activity investigated	Location of the measurement setup	Ventilation status	Volume (m^3)
H1	Metal chimney stove	Heating	Living room	Windows and door closed	80
H2	Traditional mud stove	Rice cooking	Kitchen	Windows and door opened	82
H3	Stone tripod stove	Fodder preparation	Kitchen	Windows and door opened	21
H4	Traditional mud stove	Liquor distillation	Kitchen	Windows closed, door opened	42

Figure 3.1: Stove types and cooking activities: (a) chimney stove/heating in H1, (b) traditional mud stove/rice cooking in H2, (c) stone tripod stove/fodder preparation in H3, and (d) traditional mud stove/liquor distillation in H4.

3.2.2 Measurements

$PM_{2.5}$, PN, CO and CO_2 measurements were done in January 2013 for H1, H2 and H3 (during winter) and in April 2013 for H4 (during spring). The measurements were conducted in three stages, with 45 to 60 minutes of background measurement before the activity, followed by the activity, and at least an hour after the activity has ceased. A single measurement per activity was conducted in each house on different days. Instruments were placed 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 instrument location with respect to combustion source is important given the potential for spatial

gradients in concentration. However, for the real-world measurements performed in homes, we did not want to unduly inconvenience the occupants. Therefore, the location of the instrumentation setup, while mostly consistent, was not standardized in the present study. As is common with all traditional structures in rural villages in Bhutan, all houses used natural ventilation by opening doors or windows, and all the houses had substantial gaps in their structure that would aid ventilation. Measurements were conducted under normal ventilation conditions.

Except for H1, where sampling was done during heating in the family's living room, the sampling for other houses was done inside kitchens. At the time of measurements, the stove in H2 was used for cooking rice, fodder preparation in H3, and liquor distillation in H4 (see Figure 3.1). It should be noted that while stove types differed, all houses used firewood during the measurements, as they always do. The chimney of the heating stove was projected outside through the window in H1, while no venting system was present in the other houses. Also, during the measurements, any potential outdoor source of pollution, which could result in emission infiltration, was noted.

The duration of activities was 5 hours for heating in H1 (from 17:00 to 22:00, until occupants went to bed), 34 minutes for cooking in H2 (cooking rice for a family of 5), 50 minutes for fodder preparation in H3, and 2 hours for liquor distillation in H4. All the cooking activities involved boiling and steaming, and no frying or grilling was involved.

Additionally, we conducted outdoor background CO_2 measurements for five consecutive days (on different days from measurement in houses) to account for the air exchange rate (AER) estimation (see Section 2.4) at the Sherubtse College

campus. The background site was located at a distance ranging from 2 km from H1 to 7 km from H4.

3.2.3 Instrumentation

Details of the instrumentation used were reported in our previous paper from Bhutan (Wangchuk et al., 2015). But briefly, $PM_{2.5}$ was measured by using a DustTrak aerosol photometer (TSI Model 8520, TSI Inc., St. Paul, MN, USA), that operates based on a light scattering technique, where the amount of scattered light is proportional to the mass concentration of the aerosol. PN was measured by using a NanoTracer (NT, Philips Aerasense, Netherlands), which works by diffusion charging and measures PN concentrations up to 1×10^6 particles/cm^3 in the size range of 10-300 nm. Temperature, relative humidity, CO and CO_2 concentrations were measured using indoor air quality (IAQ)-CALC (Model 7545, TSI Incorporated, St. Paul, MN, USA).

All the instruments 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.

DustTrak was tested and calibrated for ambient urban concentrations against 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 emissions, therefore, the measured $PM_{2.5}$ concentrations represent approximations of the actual values. The DustTrak data were considered relevant for the scope of this study, as the recorded $PM_{2.5}$ concentrations were used to analyze the relative differences in the three monitoring scenarios. For simplicity, the DustTrak results

discussed in this paper are referred to as $PM_{2.5}$ from now on (omitting the term 'approximation'). A correction factor for NT measurements of PN concentrations is described in the Supporting Information (SI) file.

All the instruments were set to a 10 second averaging interval. The DustTrak was zero calibrated and the flow rate checked prior to each sampling trip. Instrument time stamps were synchronized with the local time.

3.2.4 Estimation of air exchange rate

An estimate of air exchange rates (AER) in each house was obtained based on CO_2 decay, once the stove was ceased, as demonstrated in equation (1) (Thatcher and Layton, 1995, He et al., 2004):

$$\alpha = \frac{1}{t} ln \frac{C_t}{C_0} \quad (1)$$

Where α = air exchange rate, t = duration of CO_2 decay, C_t = CO_2 concentration at time t, and C_0 = CO_2 concentration when the source was discontinued. It should be noted that outdoor CO_2 can influence the indoor CO_2 concentration variations through penetration indoors. Thus, the estimated AER by equation (1) will be lower than the true value if the outdoor or background CO_2 value was not subtracted from the indoor value before taking the natural log of the data and plotting it against time. In order to account for this we have subtracted the outdoor background CO_2 concentration from the measured indoor concentrations.

3.2.5 Estimation of emission rates

The main factors influencing indoor particle concentrations are indoor and outdoor sources, particle loss due to deposition on indoor surfaces, penetration of outdoor particles, and air exchange rates (Morawska and Salthammer, 2003). The

equation used to relate these parameters has been reported in previous studies (Koutrakis et al., 1992, Thatcher and Layton, 1995, Chen et al., 2000):

$$\frac{dC_{in}}{dt} = P * \alpha * C_{out} + \frac{Q_s}{V} - (\alpha + k) * C_{in} \quad (2)$$

Where C_{in} and C_{out} are indoor and outdoor particle concentrations, respectively, P is the penetration efficiency, k is the deposition rate, α is the air exchange rate, Q_s is the indoor particle generation rate, t is time and V is the effective volume of the space. The average emission rate can be estimated by simplifying equation (2), using average values and assuming that the effects of particle dynamics such as condensation, evaporation and coagulation are negligible under the conditions usually encountered in residences, as described in He et al. (2004). The average emission rate, Q_s, is therefore given by equation (3):

$$\overline{Q_s} = V \left[\frac{C_{int} - C_{in0}}{\Delta T} + \overline{(\alpha + k)} * \overline{C_{in}} - \bar{\alpha} * C_{in0}\right] \quad (3)$$

Where C_{int} and C_{in0} are the peak and initial indoor particle concentrations respectively, $\overline{(\alpha + k)}$ is the average total removal rate (which can be calculated using average decay rate after the source ceased), $\bar{\alpha}$ is the average air exchange rate, and ΔT is time difference between the initial and peak concentrations.

3.2.6 Data processing and 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 of the student's t-test) was used to test the mean differences.

Extremely high PN concentrations, exceeding 1×10^6 particles/cm^3 (which was beyond the upper detection limit of the NT) were observed when the stoves were operated. This resulted in a failure of the instrument during measurements in H2. There was no way to deal with this issue in the field, as the instrument required manufacturer servicing. Therefore, no usable PN data were available for H2, H3 and H4. For heating in H1, concentrations exceeding the upper limit of the instrument were left as 1×10^6 particles/cm^3 and the analysis involving this data represents a lower bound.

3.3 Results

The results of temperature and relative humidity in the houses during the measurements are presented in the SI file.

3.3.1 $PM_{2.5}$, PN and CO concentrations

Figures 3.2 and 3.3 present the mean $PM_{2.5}$, PN and CO concentrations for the background, during and after the activities have ceased. In all the houses, concentrations of all the pollutants were significantly higher during the activity than the background levels (on average by a factor 40 and 18 for $PM_{2.5}$ and CO, respectively) and after activities have ceased ($p < 0.001$). It should be noted that background concentrations were, at times, influenced by neighborhood activities. For example, before the measurements in H1, smoke from the next-door neighbor had already infiltrated into the house, leading to a high background level. The measurements in H4 were started in the early morning hours, which was associated with a relatively low background level. Further descriptions of concentration variations are presented in the SI file.

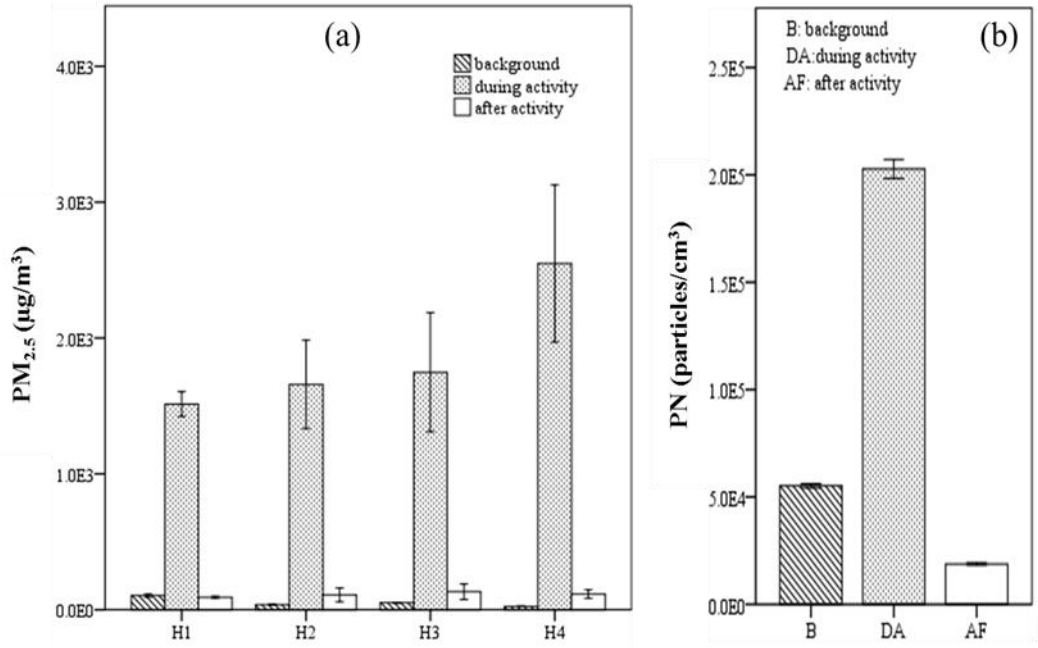

Figure 3.2: (a) $PM_{2.5}$ concentrations for all the studied houses (H1-H4) and (b) PN concentrations for H1 only during the 3 stages of the measurements: prior to activity (background), during and after the activity.
(Note: H1- heating/metal chimney stove, H2- cooking rice/traditional mud stove, H3- fodder preparation/stone tripod stove, H4- liquor distillation /traditional mud stove).
Error bars show 95% confidence interval.

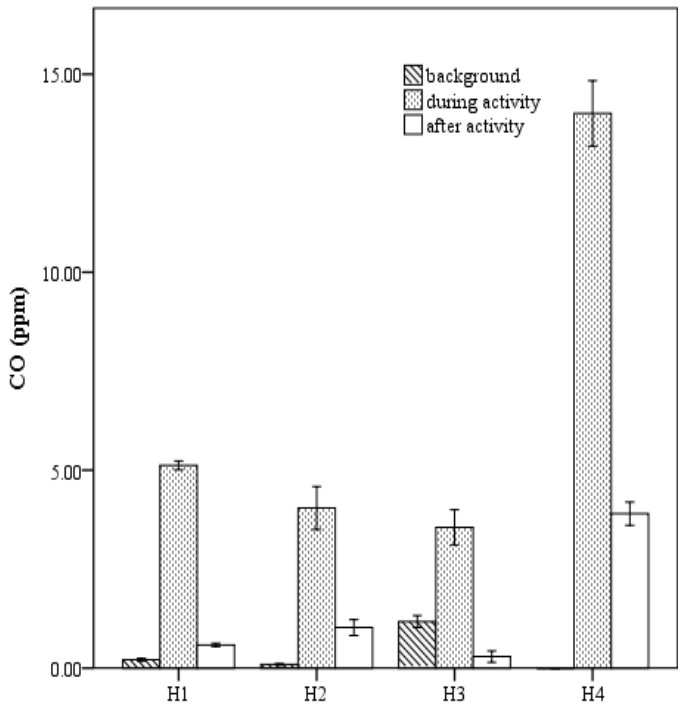

Figure 3.3: CO concentrations prior to the activity (background), during and after the activity (H1-H4).
Error bars show 95% confidence interval.

3.3.2 Emission rates from stoves

The CO_2 decay after the heating and cooking activities have ceased presented a mean (sd) air exchange rate of 1.75 ± 0.50 h^{-1}. Figure 3.4 and SI Table S3.2 show the summary statistics of emission rates for $PM_{2.5}$, PN and CO. The $PM_{2.5}$ emission rates during startup lighting ranged from 1.13×10^2 mg/min for the stone tripod stove during fodder preparation in H3, to 1.67×10^3 mg/min for the chimney stove during heating in H1. The CO emission rates during startup lighting ranged from 3.76×10^2 mg/min for the stone tripod stove in H3 to 3.09×10^3 mg/min for the traditional mud stove during liquor distillation in H4. The PN emission rate during startup lighting of the chimney stove in H1 was 7.71×10^{13} particles/min. As explained before, PN data were not available for other houses due to the instrument malfunction. However, based on the measurements in H1, it is expected that emission rates would be extreme in other houses, with peak concentrations exceeding 1×10^6 particles/cm^3, as observed in H1. Overall, the highest mean (arithmetic) $PM_{2.5}$ emission rate was observed during liquor distillation (traditional mud stove) in H4, followed by heating (metal chimney stove) in H1, rice cooking (traditional mud stove) in H2, and fodder preparation (stone tripod stove) in H3. Likewise, the highest mean CO emission rate was during liquor distillation in H4 and the lowest during fodder preparation in H3. Heating in H1 presented a mean (sd) PN emission rate of 3.24×10^{13} (2.48×10^{13}) particles/min.

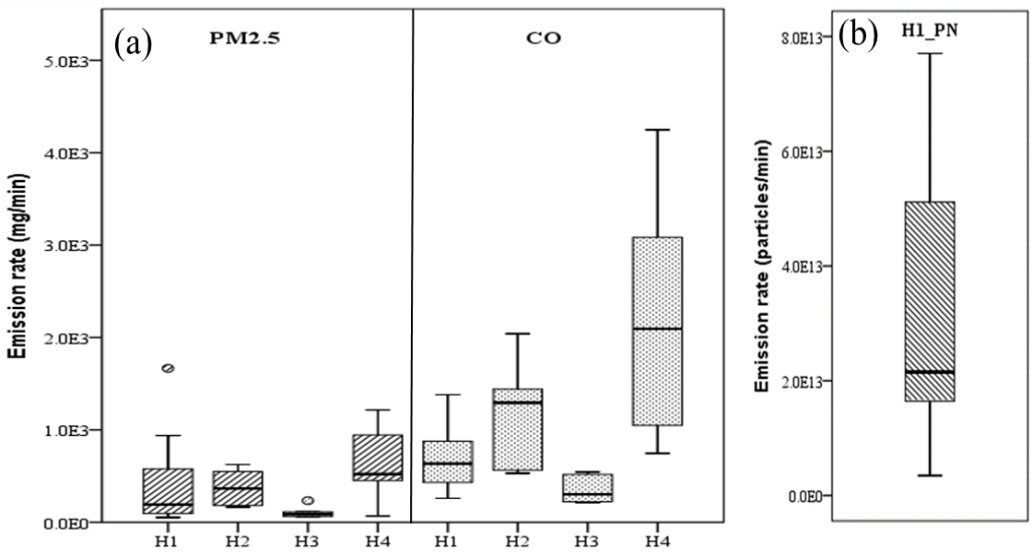

Figure 3.4: Emission rates (a) $PM_{2.5}$ (mg/min) and CO (mg/min) for H1-4, (b) PN (particles/min) for H1 only. The boxplot presents minimum, first quartile, median (middle dark line), third quartile and maximum values.

3.4 Discussion

While our study was a pilot investigation performed in a limited number of houses, it is one of only a small number of studies to estimate pollutant emission rates from biomass stoves during cooking and heating under real-world operating conditions. Moreover, despite the limited number of measurements, our group had extensive local knowledge regarding typical heating and cooking practices in the study region. This allowed us to specifically target activities that were representative of those occurring among the wider population.

Among the activities we found that liquor distillation resulted in the highest emission rates for both $PM_{2.5}$ and CO than other activities. This is because liquor distillation requires a longer time, more fuel and intense heat in comparison to other cooking types, which is reflected in the high pollutant concentrations during this process. Therefore, emission rates were more influenced by the cooking activities than the type of biomass stoves used. Despite H2 and H4 using a similar stove (traditional mud stove), both $PM_{2.5}$ and CO emission rates were two times higher

during liquor distillation in H4 than rice cooking in H2. Also among the houses/activities, the mean $PM_{2.5}$ and CO emission rates were the lowest for H3 during fodder preparation, using a stone tripod stove. Unlike liquor distillation, fodder preparation did not require intense heat and proper cooking. However, it should be noted that our results are based on limited data with single measurement per activity. Future studies should conduct multiple measurements and by recruiting more houses. Additionally, it is possible that our estimated emission rates may have been influenced by the type of wood used, as households had access to different wood species for fuels.

Several studies have investigated emission rates when cooking was done using gas and electricity. The comparison of our results with these studies showed that emission rates from biomass stoves were orders of magnitude higher. For example, He et al. (2004) reported the highest median $PM_{2.5}$ and PN emission rates of 2.68 and 2.78 mg/min, and 4.57×10^{11} and 7.34×10^{11} particles/min, respectively during frying and grilling using gas and electric stoves. Similarly, Wallace and Ott (2011) reported mean PN emission rate of 5.11×10^{12} particles/min during cooking using gas stoves. A cooking test conducted in a simulated kitchen using a gas stove reported the highest $PM_{2.5}$ and PN emission rates ranging from 5.2×10^{2} to 1.0×10^{4} μg/min and 2.6×10^{12} to 3.5×10^{12} particles/min, respectively (Buonanno et al., 2009). As expected, these values were 1-2 orders of magnitude lower than the $PM_{2.5}$ and PN emission rates obtained for biomass stoves in the present study.

The recent WHO guidelines for household fuel combustion recommend emission rate targets (ERT) in order to assess how well various interventions can help meet the guidelines (WHO, 2014a). The ERT are based on stove types: for unvented stoves ($PM_{2.5}$ = 0.23 mg/min, CO = 0.16 g/min) and for vented stoves ($PM_{2.5}$ = 0.80

mg/min, CO = 0.59 g/min), respectively, to meet annual and interim target-1 (IT-1) for $PM_{2.5}$, and 24-hour guideline for CO. Our estimated $PM_{2.5}$ and CO emission rates during heating (which constituted emissions from the stove only) exceeded the WHO recommended ERT by several orders of magnitude. The degree of exceedance is likely to be even higher during cooking activities due to contributions from cooking emissions. Improvements are clearly required in both stove efficiency and emission reduction technologies in order to increase the likelihood of these targets being met. Alternatively, a large shift in energy sources towards cleaner fuels may offer the greatest benefit, albeit with many associated logistical and economic challenges.

The WHO guidelines for household fuel combustion also provides practical recommendations for countries to minimize HAP (WHO, 2014a). Recommendations range from the promotion of improved stove design, shifting to clean fuels, improved ventilation, behaviour change, and user education and training. Although improved stove design does not consistently bring down emission levels to the recommended WHO guidelines (for example, $PM_{2.5}$ and CO), past studies in the Himalayan region have shown that it results in a significant reduction of HAP (Li et al., 2012, Singh et al., 2012). This means that the promotion of improved stove design will result in substantial health benefits. For a rural community in Bhutan, shifting to clean fuels (LPG or electricity) will remain a challenge. The prevalence of customary cooking practices, such as liquor distillation and fodder preparation, limit their use as a primary fuel. More importantly, costs associated with electricity and LPG will force people to continue using firewood, which is in abundance and can be collected freely.

Since cooking and heating using firewood are primary indoor activities among rural homes in Bhutan, residents are chronically exposed to extremely high concentrations of combustion products. Concentrations remain elevated above the

background levels even after the activities have ceased. Moreover, it was observed that after the cooking, unburnt woods were withdrawn from the combustion chamber and buried in ash within the hearth to save the ember for subsequent cooking. While this procedure sustains the fire source in the kitchen and saves the residents from buying matches, it extends the source emission time after the activity. This is one of the major differences between the use of traditional wood stoves, and gas or electric stoves. Cooking tests in houses in Hong Kong using gas stoves found that one hour after the cooking was sufficient for $PM_{2.5}$ and PN to decay to background levels (Sze-To et al., 2012). Similarly, cooking tests conducted in Brisbane houses using gas and electricity found that $PM_{2.5}$ and PN concentrations declined to background levels within 15 minutes of the conclusion of the activity under normal ventilation and 45 minutes under minimum ventilation conditions (He et al., 2004).

Further, during the investigation, it was observed that women and children who were at home spent their time close to stoves during cooking. Further, the person cooking had to position themselves directly over the fire to access the stove top. This point towards a need for systematic interventions to reduce HAP in Bhutan. The development of improved stoves will need to be backed by adequate training and education for sustained adoption of the device, and to change peoples' behaviour and perceptions. In the past, the promotion of mud chimney stoves in Bhutan was not successful, among other reasons, due to poor maintenance, perceived fire risks, and because traditionally, smoke is needed to dry chilli and grains in the kitchen (DoE, 2005). In Nepal and China, it was found that improved stove design was an appropriate intervention to reduce HAP, but proper education in relation to their operation and maintenance by users was also necessary (Baumgartner et al., 2011,

Singh et al., 2012). Additionally, interventions should target the larger community rather than isolated households.

3.5 Conclusion

As a part of larger air quality investigation in different microenvironments in Bhutan, this study quantitatively characterized biomass stove emissions during cooking and heating in four rural homes during real-world operations. Emission rates of both $PM_{2.5}$ and CO were highest during liquor distillation and lowest during fodder preparation. Notwithstanding the limited data, we found that emission rates were more dependent on the type of activity than on stove type. Despite H2 and H4 using a similar stove, estimated emission rates for both $PM_{2.5}$ and CO were two times lower during rice cooking in H2 than liquor distillation in H4. Compared with gas and electric stoves, emission rates from biomass stoves were a few orders of magnitude higher.

The results of this study highlight a serious public health issue for people living in the villages in Bhutan, and the need for interventions to reduce health risks. Making a shift to clean energy sources in rural areas in developing countries present a significant challenge due to its associated costs and accessibility. Improved stove design, supported by proper education on maintenance and adaption with traditional household practices, would offer substantial reduction in exposure levels.

Further research should ideally: (1) investigate more houses and contributions from other indoor activities, such as incense combustion and lighting oil lamps as a part of religious ceremonies; (2) assess personal exposure for cooks and other family members; and (3) assess short- and long-term health outcomes associated with exposure to combustion products in such indoor environments.

3.6 Acknowledgements

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Supporting Information (SI)

A pilot Study of Traditional Indoor Biomass Cooking and Heating in Rural Bhutan: Gas and Particle Concentrations and Emission Rates

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Correction factor for NT

A correction factor for NT measurements of PN count was computed by running the instrument side-by-side with a condensation particle counter (CPC) TSI model 3787 as described by Mazaheri et al. (2013):

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

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 our results could be underestimates.

Temperature and relative humidity

The mean indoor temperature and relative humidity for three stage measurements (background, during and after the activity) in the four investigated houses are presented in Figure S3.1. There was only a small variation of both the temperature and relative humidity among the houses (H1, H2 and H3), where measurements were done during the winter. Both the temperature and relative humidity were higher in H4, where measurement was done in the spring. The range of temperature and relative humidity variations in the houses corresponded to the mean outdoor ranges for winter and spring in the study area. It should be noted that gaps associated with the traditional structures readily facilitated infiltration of outdoor air, thereby influencing the indoor temperature and humidity. It can be seen from Figure S3.1 that temperature after the activity has ceased in H4 was higher than during the activity. As mentioned in the manuscript, the measurement in H4 was started in the early morning hours. When the measurement was concluded after nearly 4 hours of sampling, an increase in outdoor temperature has accordingly influenced the indoor temperature. Also there was only a marginal increase in temperature as a result of heating and cooking in the houses, ranging from 0.3 °C in H2 to 2.2 °C in H3. The marginal increase in temperature was mainly because the stoves being used were small, and only one operated in each of the houses at the time of this measurement. It should be mentioned here that traditional cookstoves are also used as a heating device during cold days in village homes. This leads to the conclusion that traditional cookstoves are poor radiators of heat, which means occupants will need to be closer to the stove for acquiring optimal warmth when used as a heating source, thereby incurring higher exposure in the process.

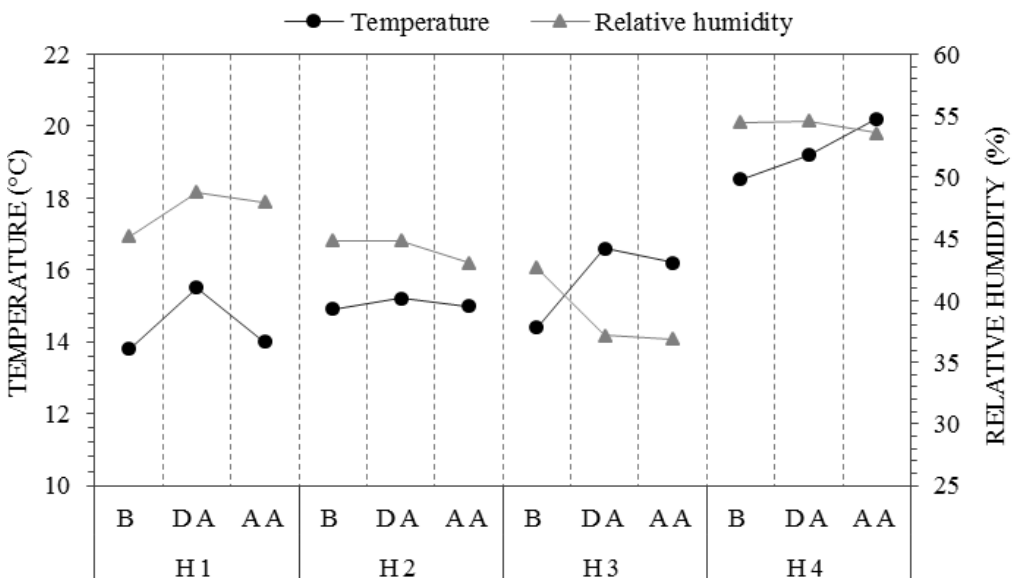


Figure S3.1: Mean indoor temperature and relative humidity variations in H1 (living room), H2 – H4 (kitchens) during the measurement. B: background, DA: during the activity, AA: after the activity.

$PM_{2.5}$, PN and CO concentrations

Table S3.1 presents the summary statistics of pollutant concentrations in the investigated houses. The mean (arithmetic) $PM_{2.5}$ concentrations ranged from 1.51 × 10^3 μg/m^3, during heating in H1 (metal chimney stove) to 2.55 × 10^3 μg/m^3, during liquor distillation in H4 (traditional mud stove). The mean CO concentrations ranged from 3.56 ppm, during fodder preparation in H3 (stone tripod stove) to 14 ppm, during liquor distillation in H4 (traditional mud stove). Statistical tests indicated that only mean $PM_{2.5}$ concentrations in H4 and H1 were significantly different from each other, while mean CO concentrations in H4 was significantly different than all other houses ($p < 0.001$). The heating in H1 presented a mean PN concentration of 2.03 × 10^5 particles/cm^3, which was very high even though it presented the lowest mean $PM_{2.5}$ concentration among the houses/activities. Due to the instrument malfunction PN data were not available for other houses. However, based on the measurement in H1, it is expected that PN concentrations would be extreme in other houses, with peak concentrations exceeding 1 × 10^6 particles/cm^3, as observed in H1. Therefore, open

wood fire source inside kitchens present an extreme environment for the instrumentation. Based on our experience we recommend the use of a simple dilution system.

The mean $PM_{2.5}$ concentration during heating with chimney stove in H1 was lower than cooking with un-vented traditional stoves in other houses, while it presented comparable CO concentrations to those in H2 and H3. The time-series concentrations during heating and cooking exhibited different emission patterns during different stages of the processes (for example, Figure S3.2 and Figure S3.3). With the heating stove, a distinctly sharp peak was observed during the startup lighting, while the subsequent peaks were markedly lower. In contrast, the traditional mud stove presented peaks with similar magnitude throughout the combustion process. The initial lighting of the chimney stove was done using softer and easily combustible wood along with some kerosene. The door of the combustion chamber was left open for that purpose until the fire has sufficiently built up. This not only resulted in emissions of thick smoke, but also readily dispensed into the indoor environment. It should be noted that, although chimney stove is considered an improved device compared with un-vented stoves, there are no recommended operational procedures for achieving lower emissions. The stove is produced by local fabricators and has not been scientifically evaluated for its efficiency and operational procedures. The initial lighting with traditional mud and stone tripod stoves in H2 to H4 were done from the ember saved from previous fire using softer wood, which did not produce a distinctly sharp peak like the chimney stove.

The use of different fuels for starting the fire seems to be a common trend with user of biomass stoves. For example, in Nepal, Devakumar et al. (2014) have reported occupants using straw and kerosene for initial lighting of biomass cookstoves, but

these were not used as main cooking fuels. Also, multiple episodic peaks during the combustion cycle were reported by other studies, which were associated with the startup lighting of stoves, and feeding and removal of fuels (Ezzati et al., 2000, Li et al., 2012, Maddalena et al., 2014). However, Li et al. (2012) pointed out difficulties in distinguishing emissions originating from food being cooked and those coming from stoves, due to peak emissions from stoves. For the present study, except for the initial peak produced by the startup lighting of stoves, we have not assessed the activities that resulted in subsequent peaks. However, since none of the cooking activities involved frying, grilling or roasting and based on field observations there was nothing to indicate that peaks were not the result of feeding wood into the fire.

The mean $PM_{2.5}$ concentrations ranging from 1.51×10^3 to 2.55×10^3 $\mu g/m^3$ during heating and cooking were 60 to 102 times higher than the WHO 24-hour indoor air quality guideline value (25 $\mu g/m^3$) (WHO, 2010). The mean CO concentrations ranging from 4.01 ppm (~ 4.98 mg/m^3) to 14.01 ppm (~ 17.28 mg/m^3), while mostly below the WHO 24-hours guideline value (7 mg/m^3), during liquor distillation, it was over two times higher than the guideline value. However, it should be noted that we compared our peak values with the WHO guidelines which are mean concentrations specified for 24-hours. The major drawback of WHO guidelines is it doesn't account for short-term peak concentrations, which as mentioned before is very important metric for exposure assessment.

The mean $PM_{2.5}$ concentration from the chimney stove (improved stove) although lower than traditional stoves was still very high. Similar results have been reported elsewhere: in the tents of nomadic Tibetans, Li et al. (2012) reported 24-hour $PM_{2.5}$ and CO concentrations of 1.42 and 6.69 mg/m^3 with occupants using open fire stoves, and 0.14 and 0.12 mg/m^3 with occupants using chimney stoves, respectively.

The $PM_{2.5}$ concentration was nearly 7 times higher than the 24-hour WHO guideline even in tents with chimney stoves. In Nepal, Singh et al. (2012) conducted a longitudinal study involving before and after investigations of HAP in houses which had improved cookstoves. They reported that $PM_{2.5}$ and CO reductions of up to 60% after the installation of improved cookstoves. However, concentration levels were still above the WHO guidelines. Similar results with lower particle mass and CO concentrations in houses with improved chimney stoves, albeit still higher than the WHO guideline, were reported in China (Edwards et al., 2007, Tian et al., 2009), Honduras (Clark et al., 2010) and Laos (Morawska et al., 2011).

Table S3.1: Summary statistics of $PM_{2.5}$ (µg/m^3), PN (particles/cm^3) and CO (ppm) prior to activity (background), during and after the activity

House*	Pollutant	Background			During Activity			After Activity		
		Median	AM GM	SD GSD	Median	AM GM	SD GSD	Median	AM GM	SD GSD
H1	$PM_{2.5}$	70	1.04×10^2 89	75 1.6	1.20×10^3	1.51×10^3 1.29×10^3	1.36×10^3 1.7	28	92 45	1.55×10^2 2.7
	PN	5.56×10^4	5.52×10^4 5.36×10^4	1.34×10^4 1.3	1.72×10^5	2.03×10^5 1.79×10^5	1.32×10^5 1.6	1.01×10^4	1.88×10^4 1.34×10^4	1.78×10^4 2.2
	CO	0.10	0.21	0.24	4.70	5.12	1.69	0.10	0.59	0.74
H2	$PM_{2.5}$	38	37 37	5 1.1	1.07×10^3	1.67×10^3 7.83×10^2	1.76×10^3 4.4	50	1.10×10^2 62	3.25×10^2 2.0
	CO	0.00	0.10	0.17	2.90	4.04	4.17	0.10	1.03	1.83
H3	$PM_{2.5}$	50	52 50	20 1.3	7.25×10^2	1.75×10^3 8.03×10^2	2.75×10^3 3.5	85	1.33×10^2 95	3.91×10^2 1.6
	CO	0.70	1.18	1.54	240	3.56	3.92	0.00	0.29	1.41
H4	$PM_{2.5}$	26	25 25	3 1.1	7.36×10^2	2.55×10^3 6.50×10^2	4.63×10^3 6.2	52	1.15×10^2 65	170 2.7
	CO	0.00	0.00	0.00	11.80	14.01	11.45	3.10	3.90	2.72

*: H1: heating/metal chimney stove, H2: rice cooking/traditional mud stove, H3: fodder preparation/stone tripod stove, H4: liquor distillation/traditional mud stove. AM: Arithmetic Mean, SD: Standard Deviation, GM: Geometric Mean, Geometric SD. GM and GSD were not computed for CO due to presence of zero concentrations.

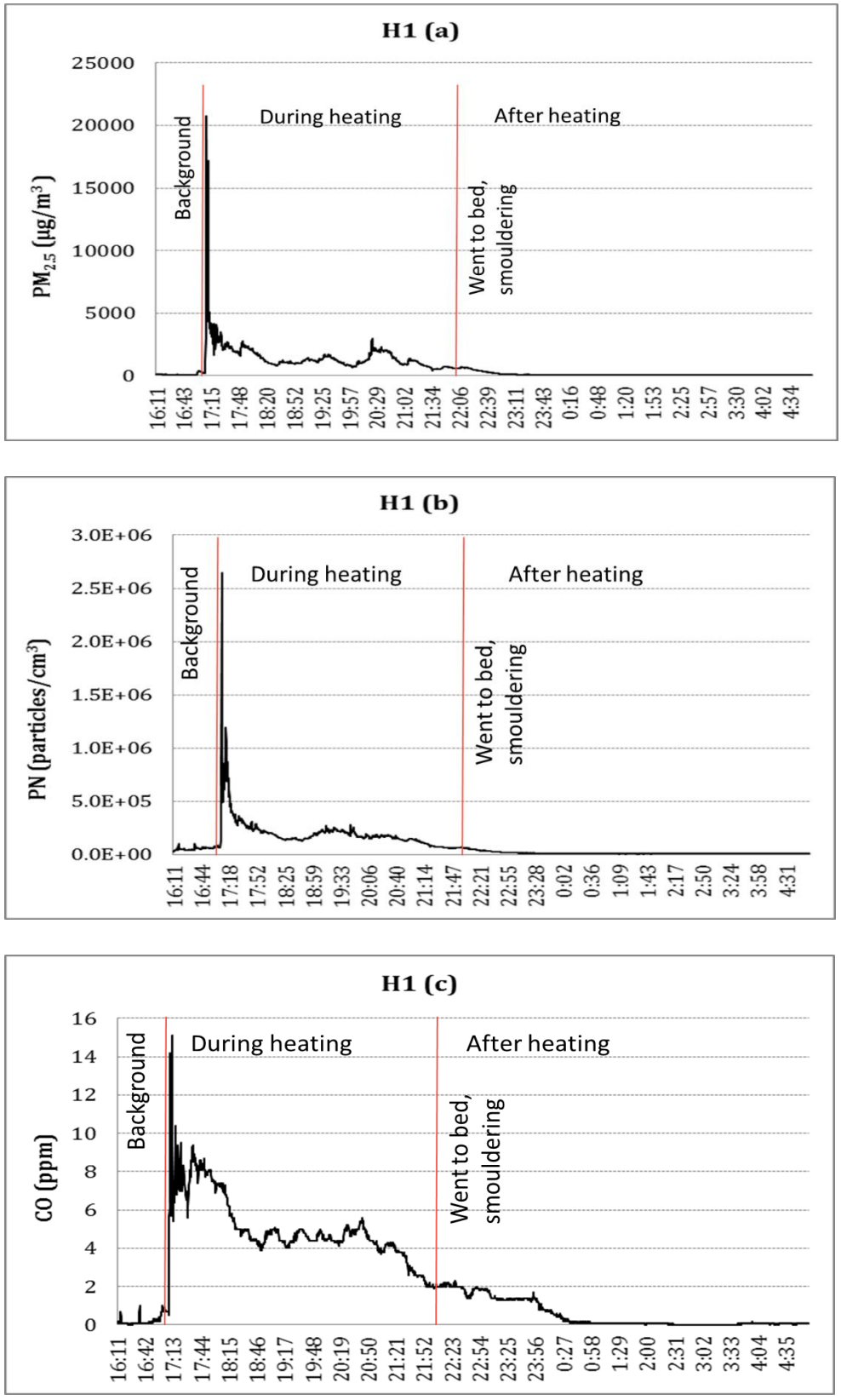
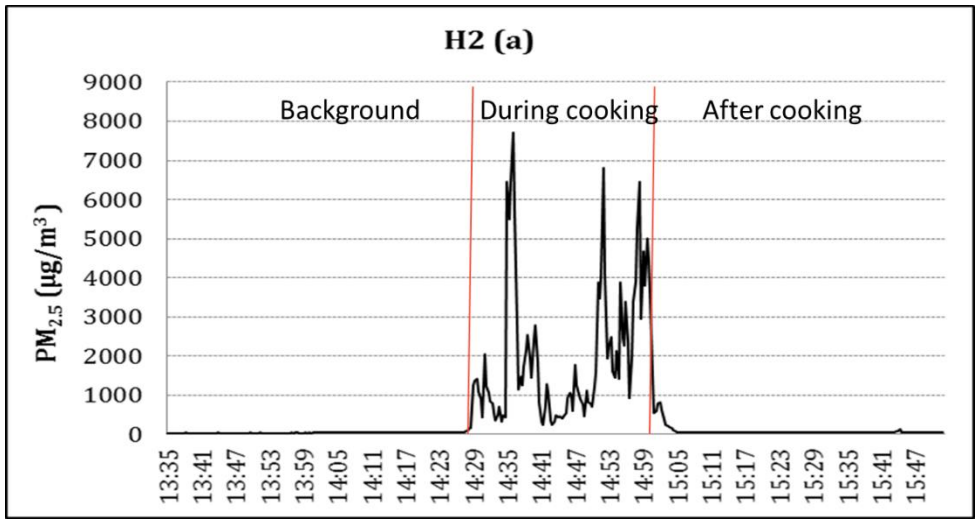


Figure S3.2: Time-series $PM_{2.5}$, PN and CO concentrations for chimney stove during heating in H1.



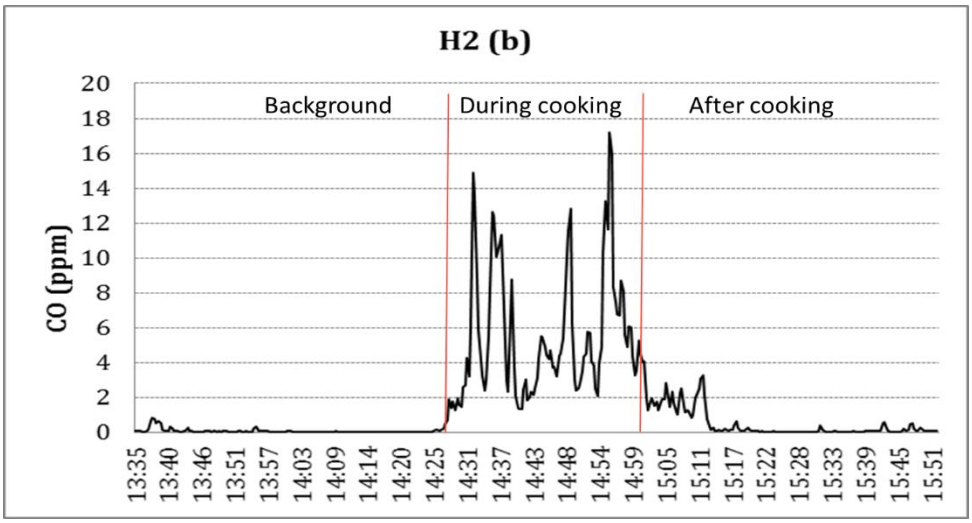


Figure S3.3: Time-series $PM_{2.5}$ and CO concentrations for traditional mud stove during rice cooking in H2.

Table S3.2: Summary statistics of peak values ($PM_{2.5}$: µg/m^3, PN: particles/cm^3, CO: ppm) and emission rates ($PM_{2.5}$: mg/min, PN: particles/min, CO: mg/min) in H1 – H4

Statistics	**H1**	**H2**	**H3**	**H4**
$PM_{2.5}$ Peak values (µg/m^3)				
Min	7.20×10^2	2.04×10^3	2.79×10^3	1.60×10^3
Max	2.08×10^4	7.71×10^3	1.11×10^4	2.87×10^4
AM	6.22×10^3	4.94×10^3	5.13×10^3	1.52×10^4
SD	7.98×10^3	2.3×10^3	3.13×10^3	9.55×10^3
GM	3.15×10^3	4.42×10^3	4.53×10^3	1.13×10^4
GSD	3.3	1.7	1.7	2.8
$PM_{2.5}$ Emission rate (mg/min)				
Min	5.30×10^1	1.66×10^2	5.70×10^1	6.70×10^1
Max	1.67×10^3	6.25×10^2	2.33×10^2	1.22×10^3
AM	4.31×10^2	3.75×10^2	1.07×10^2	6.20×10^2
SD	5.75×10^2	2.03×10^2	6.67×10^1	4.06×10^2
GM	2.22×10^2	3.26×10^2	9.35×10^1	$4.59\ 10^2$
GSD	3.2	1.8	1.7	2.8
PN Peak values (particles/cm^3)				
Min	2.80×10^5			
Max	1.00×10^6			
AM	6.64×10^5			
SD	3.61×10^5			
GM	5.70×10^5			
GSD	1.8			
PN Emission rates (particles/min)				
Min	3.46×10^{12}			
Max	7.71×10^{13}			
AM	3.24×10^{13}			
SD	2.48×10^{13}			
GM	2.33×10^{13}			
GSD	2.6			
CO Peak values (ppm)				
Min	5.00	6.00	8.00	21.00
Max	15.00	21.00	21.00	82.00
AM	9.91	12.77	13.77	48.20
SD	3.62	5.63	5.74	26.73
GM	9.28	11.60	12.80	41.69
GSD	1.5	1.7	1.5	1.8
CO Emission rates (mg/min)				
Min	2.59×10^2	5.30×10^2	2.11×10^2	7.47×10^2
Max	1.38×10^3	2.04×10^3	5.45×10^2	4.25×10^3
AM	6.90×10^2	1.19×10^3	3.50×10^2	2.22×10^3
SD	3.57×10^2	5.72×10^2	1.53×10^2	1.31×10^3
GM	6.14×10^2	1.07×10^2	3.23×10^2	1.88×10^3
GSD	1.7	1.7	1.5	1.9

AM: Arithmetic Mean, SD: Standard Deviation, GM: Geometric Mean, Geometric SD.

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Chapter 4: Seasonal Variations of Outdoor Air Pollution and Factors Driving them in the School Environment in Rural Bhutan

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Statement of Contribution

The authors listed below have certified* that:

- they meet the criteria for authorship in that they have participated in the conception, execution, or interpretation, of at least that part of the publication in their field of expertise;
- they take public responsibility for their part of the publication, except for the responsible author who accepts overall responsibility for the publication;
- there are no other authors of the publication according to these criteria;
- potential conflicts of interest have been disclosed to (a) granting bodies, (b) the editor or publisher of journals or other publications, and (c) the head of the responsible academic unit, and
- they agree to the use of the publication in the student's thesis and its publication on the QUT ePrints database consistent with any limitations set by publisher requirements.

In the case of this chapter: *Seasonal Variations of Outdoor Air Pollution and Factors Driving them in the School Environment in Rural Bhutan*

Contributor	Statement of contribution*
Tenzin Wangchuk QUT Verified Signature Date: 24/08/2015	Conducted measurements, performed data analysis and wrote the manuscript.
Congrong He*	Contributed to data analysis and interpretation, and reviewed the manuscript.
Marzenna R. Dudzinska*	Contributed to data analysis and interpretation, and reviewed the manuscript.
Lidia Morawska*	Supervised the project, contributed to data analysis and interpretation, and reviewed the manuscript.

Principal Supervisor Confirmation

I have sighted email or other correspondence from all Co-authors confirming their certifying authorship.

Professor Lidia Morawska	QUT Verified Signature	24/08/2015
Name	Signature	Date

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 ($\mu g/m^3$) of 2.37 and 2.41 for the wet, and 6.22 and 0.34 for the dry season, respectively. Mean NO_2 concentration for the dry season was 1.7 $\mu g/m^3$, 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

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

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

4.2 Methods

4.2.1 Study site

Bhutan is a small eastern Himalayan country bordered by India and China. Nearly 70% of Bhutan's population live in rural areas and are subsistence farmers (RGoB, 2006). In general, the environmental conditions, as well as social characteristics are largely comparable with the rest of the Himalayan region. As of 2013, Bhutan had 348 primary schools with a gross enrolment of 47,511 children (MOE, 2013). Most of them were day schools and children spent nearly eight hours

each day at these schools, for nearly eight months a year, making it one of the most important microenvironments for exposure after homes. Currently, no air quality data are available for schools in Bhutan.

This study was conducted in a rural primary school (hereafter called school) located in Kanglung within the Trashigang district in eastern Bhutan (Supporting Information (SI Figure S4.1). The school (altitude 1900 m) was centrally located within the block and had the highest enrolment (around 500 children at the time of this study) among the five primary schools in the same block. It was established in 1974 and functioned as a day school, with school activities taking place between 8 am and 4 pm on weekdays and 8 am to 12 pm on Saturdays. All school buildings were two storey traditional structures, constructed in the last five to eight years, replacing the structures built in 1970s. The school did not use any heating or cooling systems and 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.

4.2.2 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.

4.2.3 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 – August) in

the study area is characterised by a warm and wet weather, while the dry season (October – 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.

4.2.4 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.

4.2.5 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 – 0.05 µg/m^3 depending on the compound, as well as 0.1 – 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).

4.2.6 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.

4.2.7 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.

4.2.8 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).

4.3 Results

4.3.1 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.

4.3.2 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 4.1). 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 – 64 $\mu g/m^3$ and 11 – 158 $\mu g/m^3$ for PM_{10}, and 3 – 49 $\mu g/m^3$ and 5 – 147 $\mu g/m^3$ for $PM_{2.5}$, respectively (Figure 4.2). The daily mean PN concentrations for the wet season campaign ranged from 1.28×10^3 – 4.35×10^3 particles/cm^3 (Figure 4.3).

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 S4.1. The overall mean PM_{10} and $PM_{2.5}$ concentrations were higher for the dry season than the wet season (Figure 4.4). 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.

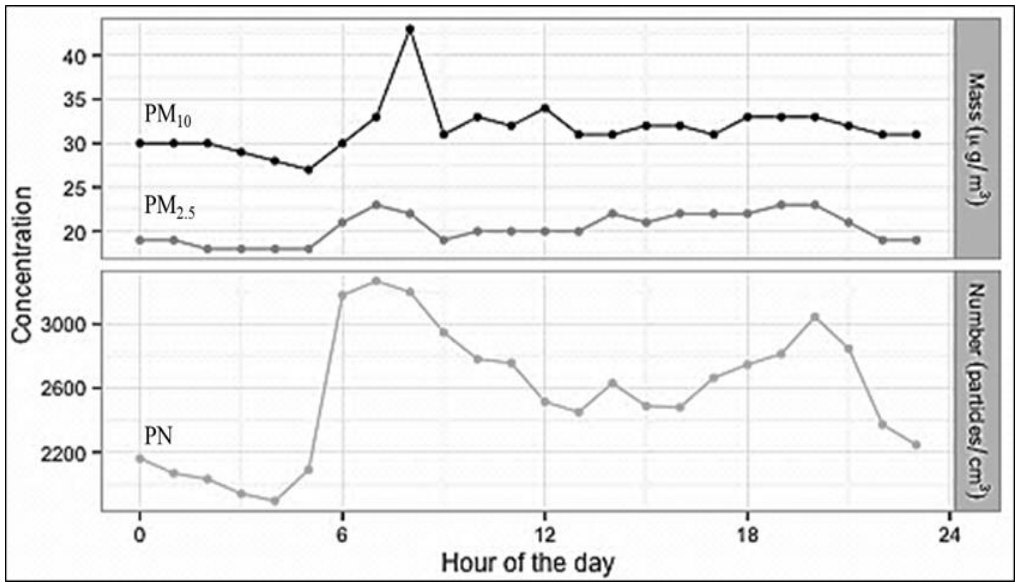

Figure 4.1: Mean hourly time-series concentrations of particle mass and PN for the entire measurement period.

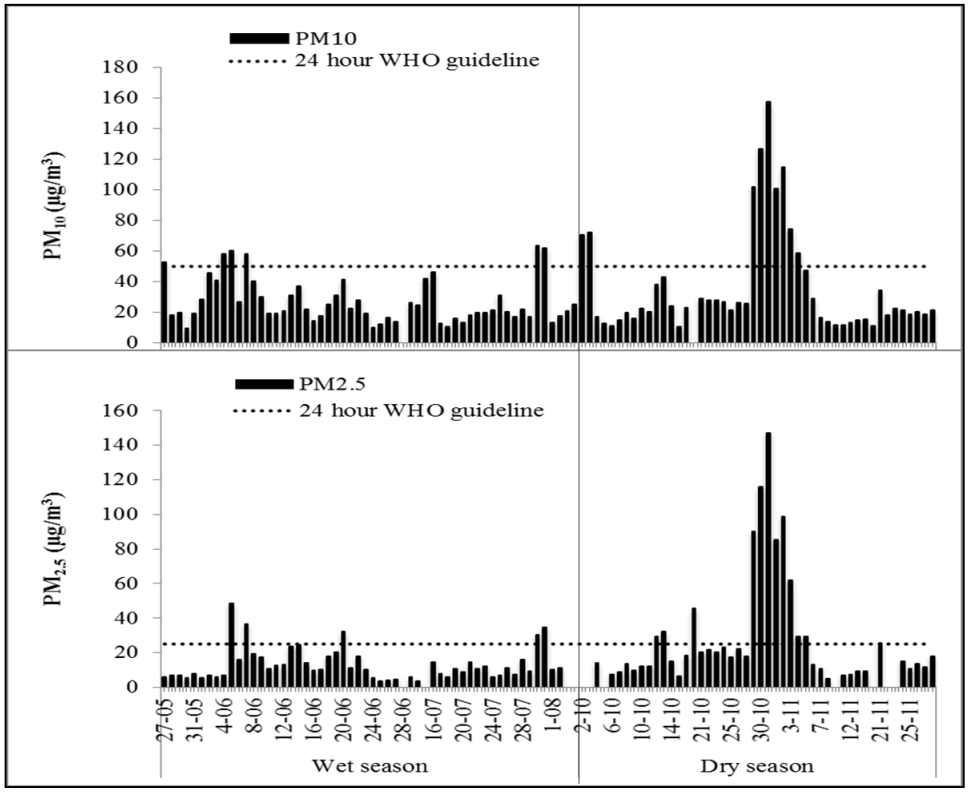

Figure 4.2: Variations of daily mean PM_{10} and $PM_{2.5}$ concentrations for the wet and dry seasons.

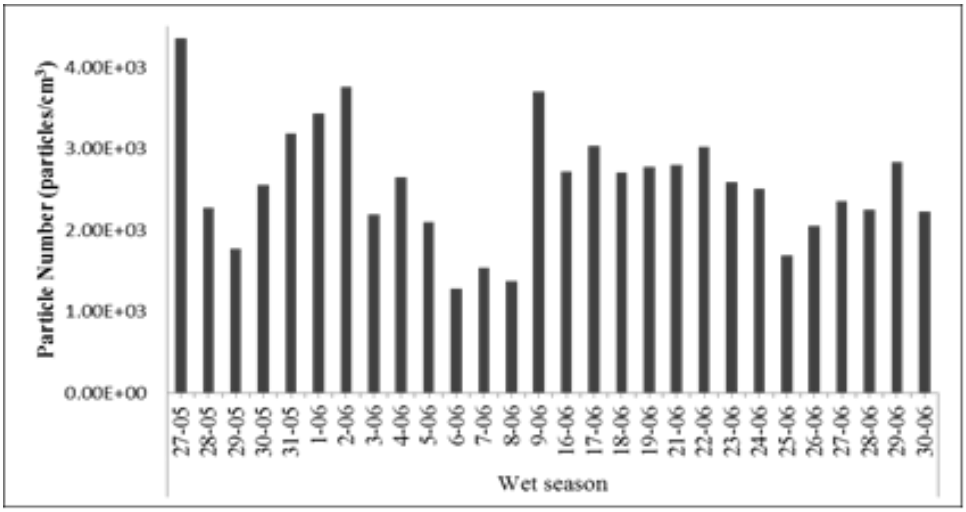

Figure 4.3: Variations of daily mean PN concentrations for the wet season.

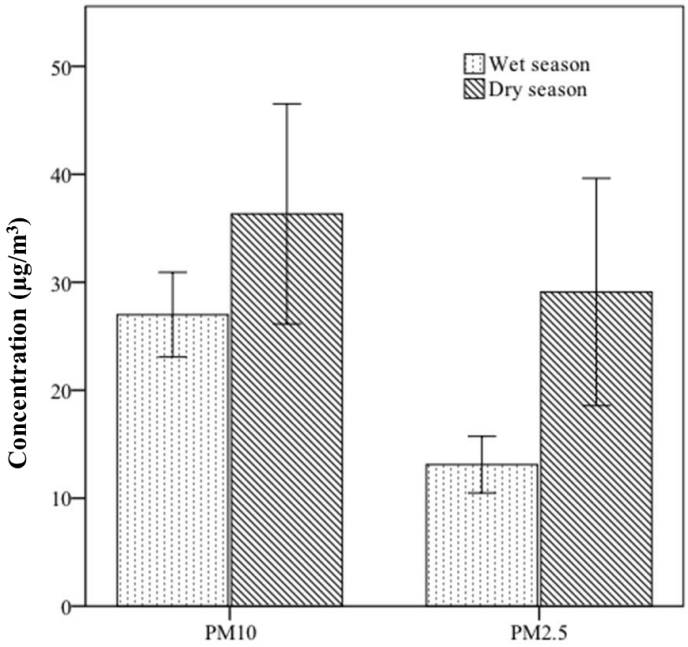

Figure 4.4: Overall mean concentrations for PM_{10} and $PM_{2.5}$ for the wet and dry seasons.
Error bars show 95% confidence interval.

Figure 4.5 and SI Table S4.1 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.

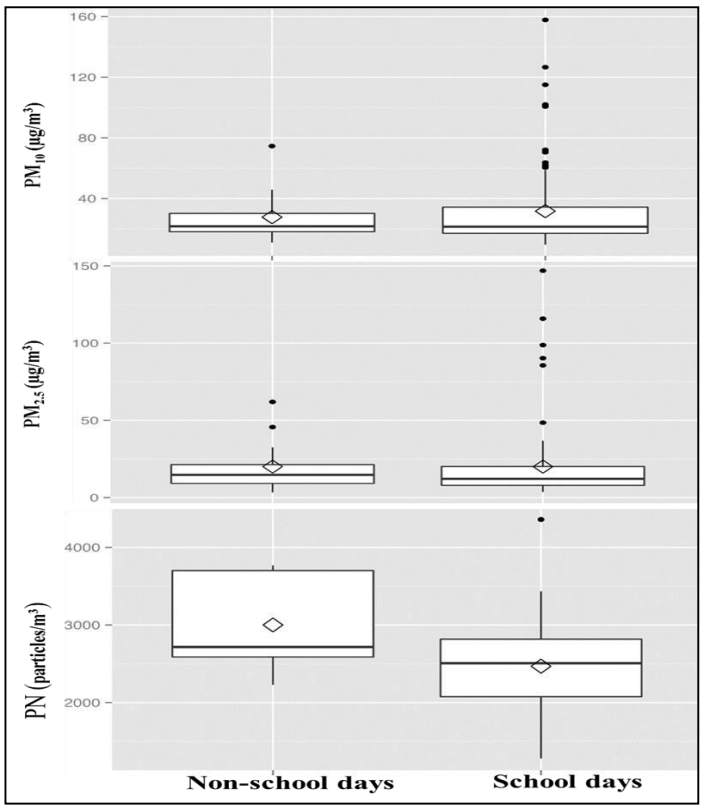

Figure 4.5: Box plot for PM_{10}, $PM_{2.5}$ and PN presenting the maximum, minimum, median (middle dark line), mean (square box), first and third quartile values for school and non-school days.

4.3.3 VOCs, Carbonyls and NO_2 concentrations

Figure 4.6 and SI Table S4.2 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.

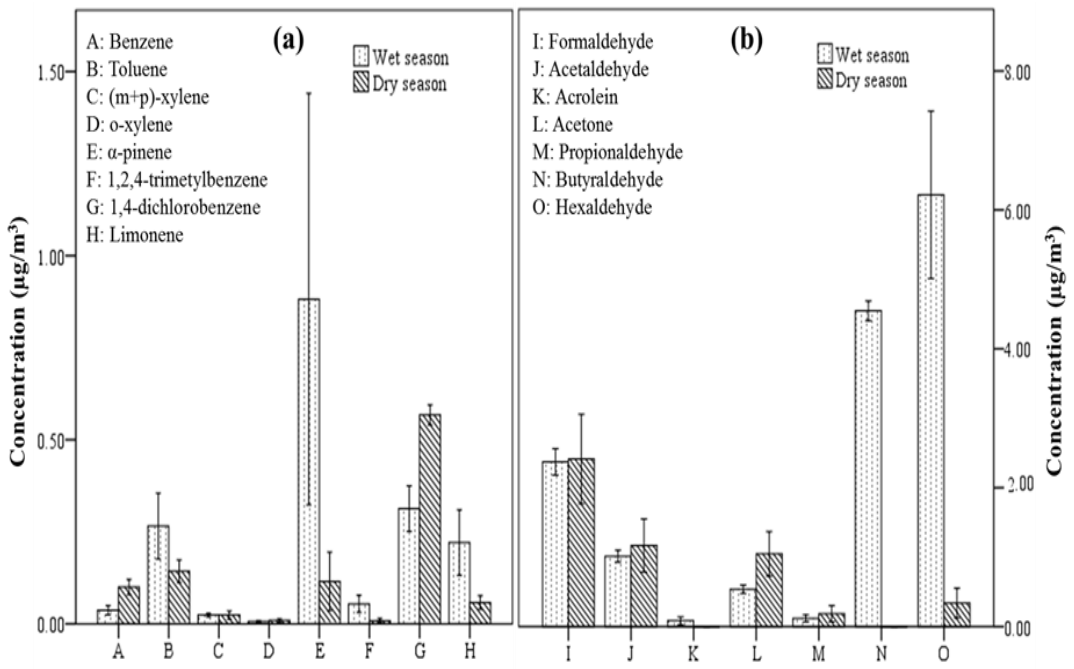

Figure 4.6: Average VOCs (a) and carbonyl (b) concentrations for the wet and dry seasons.
Error bars present 95% confidence interval.

4.3.4 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 – 434 µg/m^3 for $PM_{10,}$ 99 – 327 µg/m^3 for $PM_{2.5,}$ and 0.36 – 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 $\mu g/m^3$ for week 11 and 5.17 $\mu g/m^3$ 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 $\mu g/m^3$, 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.

4.3.5 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 S4.3). 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 S4.2 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.

4.4 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 4.1). 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 4.2). 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 S4.1).

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 4.5) 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 S4.2). 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 leaves and branches of certain plants are burnt). A statistically significant higher mean $PM_{2.5}$ concentration for the dry season than the wet season (but not for PM_{10}) (Figure 4.4) indicated that the contribution of combustion sources to fine particle concentrations was more pronounced for the dry season. It should be noted that while burning trash and incense are regular activities, burning crop residues, which was by far the dominant source, is usually done during the dry season. The higher mean and median concentrations of PN on non-school days (Figure 4.5) could be due to trash

burning in the vicinity of the school, as people have more time for cleaning on Sundays.

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

On four days when there was open wood fire cooking on the school grounds, children were actively engaged in different outdoor activities (no classroom lessons). The inhalation rate for children is generally highest during outdoor playing and sports (Buonanno et al., 2011). Therefore, open wood fire source in the school when children were engaged in outdoor activities was likely to subject them to a higher risk, by increasing their inhaled pollutant doses. Further, these events were noted to occur

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 S4.4). 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 S4.4). 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 received more rainfall than the dry season. Further studies quantifying PN levels in the dry season are needed.

In SI Table S4.5, the mean VOCs, formaldehyde and NO_2 concentrations of the current study are compared with other schools studies. Other carbonyls were not included, since we have not come across any such investigations in school outdoor environments. The mean VOCs concentration in this study were broadly comparable with a study in USA (Godwin and Batterman, 2007). However, school studies elsewhere have reported VOCs concentration several orders magnitude higher than the present study, for example in Turkey (Demirel et al., 2014), USA (Raysoni et al., 2013), Italy (Gennaro et al., 2013) and Belgium (Stranger et al., 2008). Although formaldehyde was one of the dominant carbonyls in the present study, its concentration was two times lower than in Turkey (Sofuoglu et al., 2011) and Serbia (Jovanović et al., 2014). The mean NO_2 concentration of the present study was 13–46 times lower compared to studies in Turkey (Demirel et al., 2014), Sweden (Wichmann et al., 2010), Spain (Rivas et al., 2014) and Belgium (Stranger et al., 2008).

4.5 Conclusions

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

Overall, a comparison with the studies conducted in developed countries showed comparable outdoor pollutant concentrations with a few of the studies, but in general, the pollutant levels were lower at the Bhutanese school environment. The major contributors were non-traffic sources such as dust resuspension (from bare ground), and biomass and trash combustion, as opposed to traffic emissions in developed countries. Even though the school is located next to the main road, traffic contribution was expected to be negligible because of the very low traffic volume. The Government of Bhutan is planning to develop the area around the school into an urban centre. In future, while there is likely to be a suppression of current dust sources with the installation of pavement and roads, the traffic volume is expected to increase, thereby leading to increased pollution.

4.6 Acknowledgements

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Sporting Information (SI)

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

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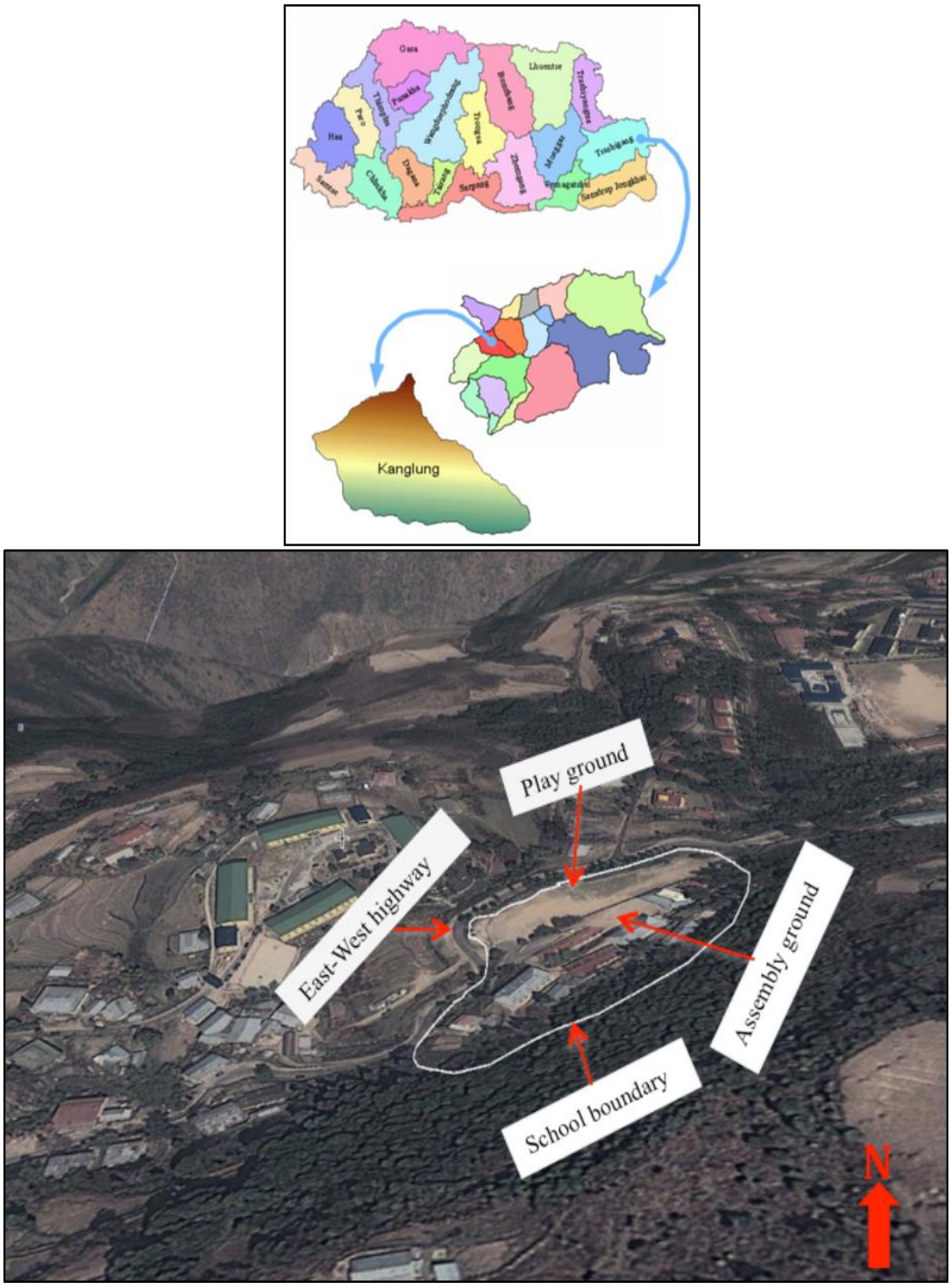


Figure S4.1: Map of Bhutan showing the study site.

Table S4.1: Summary statistics for PM_{10}, $PM_{2.5}$ ($\mu g/m^3$) and PN* ($particles/cm^3$) for the wet and dry seasons, and on school and non-school days.

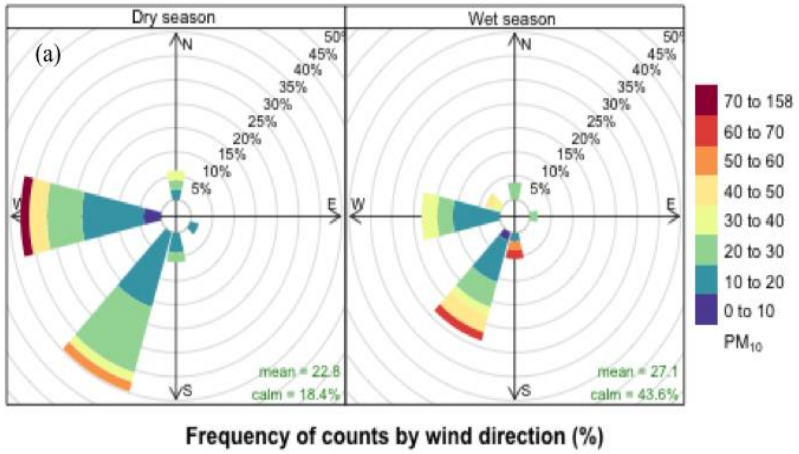
	Particles	**Minimum**	**Maximum**	**Median**	**Mean**	**SD**
Wet season	PM_{10}	10	64	21	27	15
	$PM_{2.5}$	3	49	10	13	9
	PN	1.28×10^3	4.35×10^3	2.57×10^3	2.56×10^3	7.25×10^2
Dry season	PM_{10}	11	158	22	36	34
	$PM_{2.5}$	5	147	17	29	33
School days	PM_{10}	10	158	21	32	27
	$PM_{2.5}$	4	147	12	20	25
	PN	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

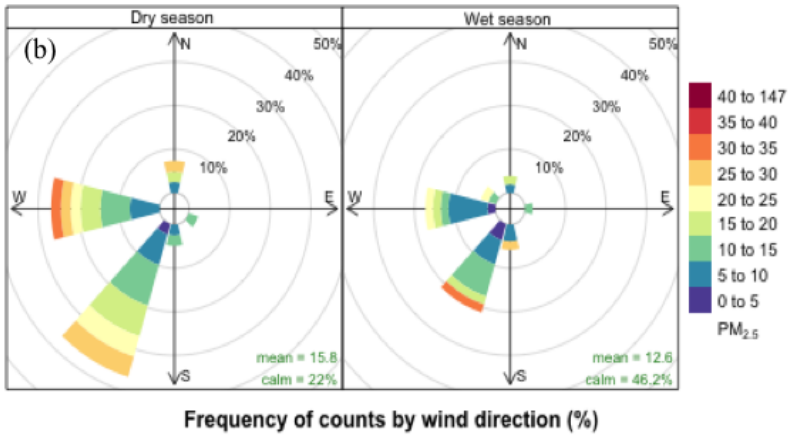
*No PN data for the dry season due to instrument malfunction. SD: standard deviation

Table S4.2: Summary statistics for VOCs, carbonyls and NO_2 ($\mu g/m^3$) for the wet and 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
Ethylbenzene	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-trimetylbenzene	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

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





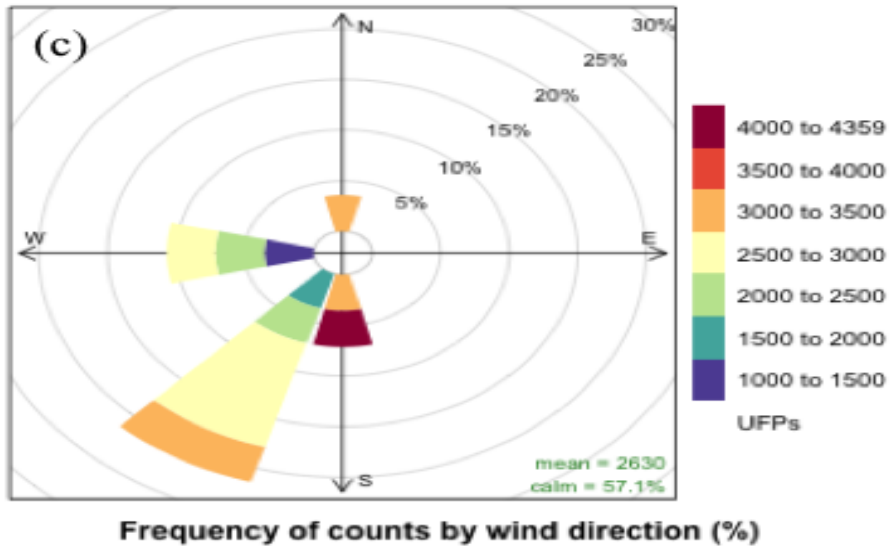


Figure S4.2: Pollution rose diagrams as function of wind direction (a) for PM_{10}, (b) for $PM_{2.5}$ and (c) for PN.

Table S4.3: 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

NM: No measurement, ND: Not detected
** $p < 0.01$, * $p < 0.05$

Table S4.4: 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

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

Table S4.5: 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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Chapter 5: Mobile Assessment of On-road Air Pollution and its Sources along the East-West Highway in Bhutan

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Statement of Contribution

The authors listed below have certified* that:

- they meet the criteria for authorship in that they have participated in the conception, execution, or interpretation, of at least that part of the publication in their field of expertise;
- they take public responsibility for their part of the publication, except for the responsible author who accepts overall responsibility for the publication;
- there are no other authors of the publication according to these criteria;
- potential conflicts of interest have been disclosed to (a) granting bodies, (b) the editor or publisher of journals or other publications, and (c) the head of the responsible academic unit, and

- they agree to the use of the publication in the student's thesis and its publication on the QUT ePrints database consistent with any limitations set by publisher requirements.

In the case of this chapter: *Mobile Assessment of On-road Air Pollution and its Sources Along the East-West Highway in Bhutan*

Contributor	Statement of contribution*
Tenzin Wangchuk QUT Verified Signature Date: 24/08/2015	Conducted measurements, performed data analysis and wrote the manuscript.
Luke D. Knibbs*	Contributed to data analysis and interpretation, and reviewed the manuscript.
Congrong He*	Contributed to data analysis and interpretation, and reviewed the manuscript.
Lidia Morawska*	Supervised the project, contributed to data analysis and interpretation, and reviewed the manuscript.

Principal Supervisor Confirmation

I have sighted email or other correspondence from all Co-authors confirming their certifying authorship.

QUT Verified Signature

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Name Signature Date

Abstract

Human exposures in transportation microenvironments are poorly represented by ambient stationary monitoring. A number of on-road studies using vehicle-based mobile monitoring have been conducted to address this. Most previous studies were conducted on urban roads in developed countries where the primary emission source was vehicles. Few studies have examined on-road pollution in developing countries in urban settings. Currently, no study has been conducted for roadways in rural environments where a substantial proportion of the population live. This study aimed to characterize on-road air quality on the East-West Highway (EWH) in Bhutan and identify its principal sources. We conducted six mobile measurements of PM_{10}, particle number (PN) count and CO along the entire 570 km length of the EWH. We divided the EWH into five segments, R1 – R5, taking the road length between two district towns as a single road segment. The pollutant concentrations varied widely along the different road segments, with the highest concentrations for R5 compared with other road segments (PM_{10} = 149 μg/m^3, PN = 5.74 × 10^4 particles/cm^3, CO = 0.19 ppm), which is the final segment of the road to the capital. Apart from vehicle emissions, the dominant sources were road works, unpaved roads and roadside combustion activities. Overall, the highest contributions above the background levels were made by unpaved roads for PM_{10} (6 times background), and vehicle emissions for PN and CO (5 and 15 times background, respectively). Notwithstanding the differences in instrumentation used and particle size range measured, the current study showed lower PN concentrations compared with similar on-road studies. However, concentrations were still high enough that commuters, road maintenance workers and residents living along the EWH, were potentially exposed to elevated pollutant concentrations from combustion and non-combustion sources. Future studies should

focus on assessing the dispersion patterns of roadway pollutants and defining the short- and long-term health impacts of exposure in Bhutan, as well as in other developing countries with similar characteristics.

Keywords

On-road, Bhutan, Particle Number, PM_{10}, CO

5.1 Introduction

Several studies have reported that air pollution levels in transportation microenvironments, such as on or near roadways and inside vehicles and tunnels, were higher than ambient concentrations (Hitchins et al., 2000, Zhu et al., 2002, Bae et al., 2007, Knibbs et al., 2009, Canagaratna et al., 2010, Li et al., 2013). For example, Kittelson et al. (2004) measured PN concentrations at different distances from the highway in Minnesota. They observed high on-road PN concentrations, ranging from 10^4 – 10^6 particles/cm^3. The concentrations were much lower at 10 – 20 meters, and significantly lower in areas 500 – 700 meters from the highway. Inside the tunnel, average CO, NO_x and $PM_{2.5}$ concentrations were found to be 17, 25 and 8 times higher than the background concentration in San Francisco (Kirchstetter et al., 1999). In Jakarta, CO concentrations during commuting for various transport modes were 180 – 700% higher than other microenvironments (Both et al., 2013). Similarly, in Lahore, Pakistan, roadside PM_{10} and $PM_{2.5}$ concentrations were 3 and 2 times higher than urban background levels (Colbeck et al., 2011). Therefore, fixed-site air quality monitoring does not adequately represent the pollution levels on and near roadways, and leads to an underestimation of exposure in these environments (Kaur et al., 2007, Apte et al., 2011).

Although time-activity surveys conducted in the United States have found that people spend only 6% of the day inside enclosed vehicles (Klepeis et al., 2001), this

can account for a disproportionate fraction of exposure to air pollutants like ultrafine (< 100 nm) particles (e.g. Fruin et al., 2008, Wallace and Ott, 2011). Characterizing air pollution in transport microenvironments is therefore important for understanding human exposure.

Characterization of on-road air quality is challenging, since a spatially dense network of fixed monitoring sites would be required to capture the spatial and temporal distribution of pollutant concentrations on and near the roads. This has led to the development of vehicle-based mobile sampling methods specifically aimed at quantifying on-road pollution during real-world driving (Gouriou et al., 2004, Knibbs et al., 2009). In recent years, several studies have been reported using mobile platforms to quantify on-road pollution. Around the world, on-road mobile measurements have been used for both tunnel (Gouriou et al., 2004, Yao et al., 2007, Knibbs et al., 2009) and above-ground urban road studies (Kittelson et al., 2004, Westerdahl et al., 2005, Pirjola et al., 2006, Zavala et al., 2006, Yao et al., 2007, Fruin et al., 2008, Wang et al., 2009, Westerdahl et al., 2009, Guo et al., 2014). The focus of these studies ranged from the estimation of emission factors (Kittelson et al., 2004, Westerdahl et al., 2009, Guo et al., 2014) to in-vehicle exposure assessment (Gouriou et al., 2004, Zhu et al., 2008, Apte et al., 2011, Colbeck et al., 2011), quantification of non-exhaust vehicle emissions (Hussein et al., 2008, Kwak et al., 2014) and evaluation of air quality control measures (Wang et al., 2009, Westerdahl et al., 2009).

While a few of the above listed on-road studies were conducted in Asian countries, they were based on measurements along busy metropolitan roads, where traffic volume was an important factor for determining road selection (Yao et al., 2007, Wang et al., 2009, Westerdahl et al., 2009, Apte et al., 2011, Colbeck et al., 2011, Guo et al., 2014, Kwak et al., 2014). On-road studies have not been extended to

major roads traversing rural areas or to include a range of geographical settings. This is important because, in addition to traffic contributions, rural areas can be affected by the long-range transport of urban pollutants, as well as local combustion sources, such as agricultural and residential wood burning in developing countries. Also, substantial numbers of people may live and work in the vicinity of both vehicle and non-vehicle sources of pollution near roads. Therefore, the contributions of both traffic and non-traffic activities to on-road pollution, as well as commuter and roadside exposure outside urban and metropolitan areas is not known.

The aim of the present study was to quantify and characterize on-road air quality for the East-West Highway (EWH) in Bhutan using a mobile platform method, in order to identify its principal sources. The primary objectives were to: (i) quantify PM_{10}, particle number (PN) count and CO, and relate these to activities along the road; and (ii) assess the contribution of different on-road and proximate sources to pollution levels.

5.2 Methods

5.2.1 Study location

Bhutan (population ~700,000) is a small eastern Himalayan nation bordered by India and China, with an area of 38,394 square kilometers. In general, the environmental conditions, as well as social characteristics, are largely comparable with the rest of the Himalayan region. In recent years, the road network in Bhutan has increased significantly, as more roads have been constructed, penetrating deeper into remote settlements, in order to enhance the local economy and the delivery of services. However, the road conditions are generally poor, with long unpaved stretches, sharp bends and steep slopes, even on the major road networks connecting different districts. As of 2013, Bhutan had a total of 67,926 registered vehicles (NSB, 2014).

The road under investigation, the EWH, is an economic lifeline for all of the eastern districts and some of the districts in central and western Bhutan. The present study covered the entire 570 km length of the EWH between Kanglung (X) in the east and Semtokha (Y), on the outskirts of the capital, Thimphu, in the west (Figure 5.1). For the purposes of this study, the entire EWH was divided into five segments, R1 – R5, with the road length between two district towns considered a single road segment (Figure 5.1). The distance of each segment varied from 68 km (R3) to 193 km (R2). Because the road is narrow, with lots of sharp turns, a one-way journey along the EWH requires two days of travel (approximately 18 hours), at an average speed of 30 to 40 km/hour. Except for R5, which had two lanes (one lane in both directions), the rest of the EWH has only one lane, with vehicles from both directions using the same lane. The road transverses diverse geographical features: deep valleys, mountaintops, wilderness areas, rugged terrain, and village and urban settlements (SI Figure S5.1). The altitude along the EWH ranges from 600 to over 3700 meters.

The four district towns (T1 – T4, Figure 5.1) connected by the EWH, where the road runs through the heart of the towns, vary in terms of area, degree of urbanization and resident population. Geographically, T1 is in the east, T2 and T3 are in the central and T4 in the west of the country. In terms of area and urbanization, T1 is the second largest town in eastern Bhutan and it is a gateway for commuters from the eastern districts travelling to central and western Bhutan. T2 and T3 are the largest and the smallest district towns in central Bhutan, respectively, while T4 is the second largest town in Western Bhutan. The 2005 Population and Housing Census of Bhutan showed a population of 6714 for T4, 4203 for T2, 3502 for T1 and 2695 for T3 (RGoB, 2006). While the road was wider through the towns, vehicle speed was much slower than between the towns, due to a higher traffic volume and pedestrians using the same road.

There are no town specific statistics of traffic composition and flow rate, other than the total number of registered vehicles in each region, which stands at 53% in the western region, 36% in the southern region, 6% in the central region and 5 % in the eastern region (NSB, 2014). Since the residents are mostly public servants and business people, most families own a car. In addition, T2 and T3 are important tourist destinations, and therefore, experience a transient increase in the number of vehicles and population, depending on the season. Also, most commuters travelling between Thimphu and eastern districts spend the night at T3. Further, since T3 is located at an altitude of ~3000 meters, the weather is cool all year round and therefore, the resident's burn wood for heating for most of the year. In recent years, urban activities have increased at T4 due to the construction of two mega hydropower plants a few kilometres downstream of the town. As a result, there has been a sharp increase in population and vehicles, which includes a substantial number of heavy-duty vehicles. Given this range of characteristics, the four towns connected by the EWH provide a good representation of urban road conditions and activities in Bhutan.

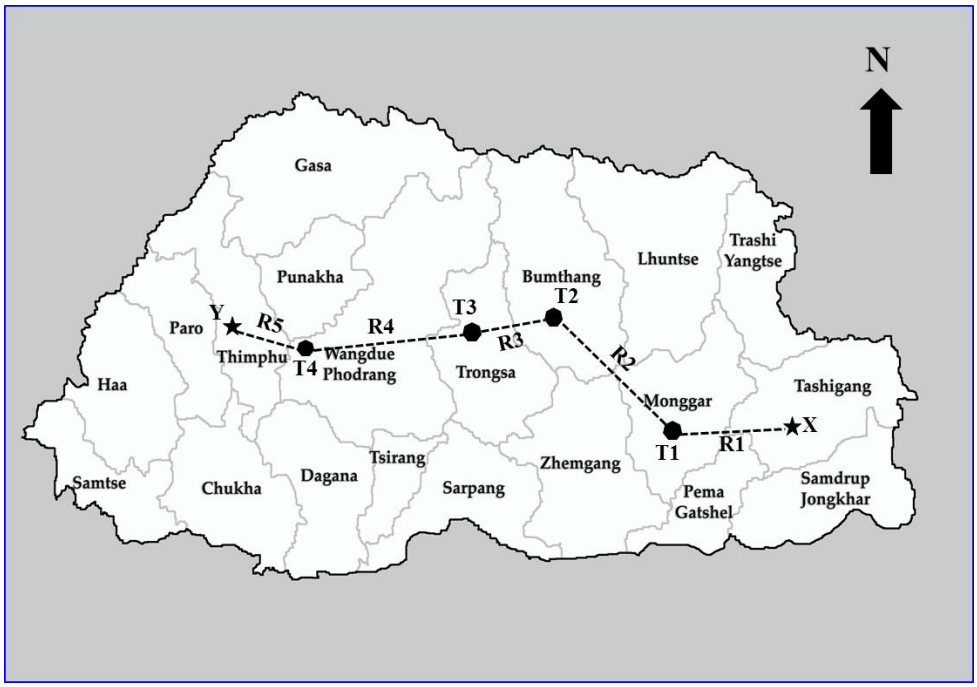

Figure 5.1: Map of Bhutan showing the East-West Highway where measurements were performed and the districts connected by the same road.
T1 – T4 = four district towns connected by the EWH. R1 = 110 km, R2 = 193 km, R3 = 68 km, R4 = 129 km and R5 = 70 km. Kanglung (X) is in the east and Semtokha (Y) is in the west. The route measured in this study joined these two places.

5.2.2 Instrumentation and quality assurance

PM_{10} was measured because the EWH had large stretches of unpaved roads and road works being undertaken at different locations during the measurement period, both of which are potential sources of coarse particles. PN and CO were chosen as an indicator of combustion activities, in order to capture emissions from both vehicles and biomass.

Details of the instrumentation used were discussed in our previous papers (Wangchuk et al., 2015a, Wangchuk et al., 2015b). Briefly, PM_{10} was measured using a DustTrak aerosol photometer (TSI Model 8520, TSI Incorporated, St. Paul, MN, USA), which operates based on a light scattering technique where the amount of scattered light is proportional to the mass concentration of the aerosol.

PN was measured using a NanoTracer (NT), Philips Aerasense, Netherlands, which works by diffusion charging and measures concentrations up to 1×10^6 particles/cm^3 in the size range of 10 – 300 nm. 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. *Fast* mode was used in this study due to the rapid changes in PN levels anticipated on the road.

A TSI indoor air quality meter (IAQ-CALC Model 7545, TSI Incorporated, St. Paul, MN, USA) was used to measure CO, as well as temperature and relative humidity. The device uses several sensors to monitor different parameters: an electro-chemical sensor for CO, a thermistor for temperature, and a thin-film capacitive sensor for relative humidity (TSI, 2004). The temperature and relative humidity results are not reported in this paper.

Prior to their shipment to Bhutan, all the instruments were tested and calibrated at the International Laboratory for Air Quality and Health, Queensland University of Technology, Brisbane, Australia. The measurements of PM_{10} obtained using the DustTrak instrument were not actual gravimetric values, as the instrument was not calibrated for the aerosol studied. The measurements are therefore approximations of PM_{10}, although relative differences between PM_{10} measurements were still captured. For simplicity, the DustTrak results discussed in this paper are referred to as PM_{10} (omitting the term 'approximation'). A correction factor for NT measurements of PN count was computed by running the instrument side-by-side with a condensation particle counter (CPC) TSI model 3787 as described by 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. It should be noted that the 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 our results could be underestimates.

All of the instruments were set to a 5 seconds averaging interval. The DustTrak was zero calibrated and their flow rates checked prior to each sampling trip. Instrument time stamps were synchronised.

5.2.3 Study design

A diesel powered Hyundai Santa Fe car (model year 2008) was used as the research vehicle. All of the instruments were secured on a rack, which was firmly secured to the rear passenger seat to minimise vibration and tilt while traversing the road. Sampling was done through a rear side window using conductive rubber tubes less than 1 m long (for PM_{10} and PN) to minimise particle loss and residence time. The tube inlets were oriented towards the direction of travel. The CO and temperature probe was placed outside the vehicle and secured in place. The instruments were powered by the research vehicle's battery via an uninterruptible power system (UPS).

5.2.3.1 PM_{10}, PN and CO measurements

A total of six on-road measurements were conducted on the EWH between 20/11/2012 and 08/01/2013. This was during winter, and characterized by cold and dry weather. Each measurement covered the entire EWH to the other end (Figure 5.1), and required two days of travel. The first day of the measurement/journey from either side ended in central Bhutan (T2, Figure 5.1).

5.2.3.2 Traffic data

Traffic information was collected concurrently with the air quality measurements. A visual count of traffic coming from the opposite direction was done in half hourly intervals throughout the campaign (we travelled along the EWH six times between 20/11/2012 and 08/01/2013). The procedure involved grouping vehicles into four categories, (1) two-wheeler (2) light (3) medium and (4) heavy, in line with Road Safety and Transport Authority (RSTA) of Bhutan's classification system. The two-wheeler category included motorbikes and scooters fuelled by petrol. Light vehicles included passenger cars, mostly fuelled by petrol, while medium and heavy vehicles included larger vehicles fuelled by diesel. A record of vehicles intercepted by the research vehicle during each half hourly interval was maintained in a field book. This did not include vehicles parked on the roadside or vehicles travelling through the town areas. Visual traffic counts were performed because, at the time of this campaign, no traffic data of any kind was available in Bhutan (Personal Communication, RSTA, Bhutan).

5.2.3.3 Characterization of pollution sources

Besides vehicles, several on-road and proximate pollution sources were observed along the EWH (SI Figure S5.2). These included several areas where road works were being undertaken, such as road widening works (earth excavation and drilling rocks) using heavy machinery and resurfacing work where bitumen was being heated on the road using open fire. There were also several stretches of unpaved (dirt) road at various locations along the EWH. Another important source was smoke plumes from nearby wood combustion activities, such as cooking and heating by roadside settlements, open fires lit by road workers and incense burning on mountain passes. It should be noted that Bhutan is predominantly a Buddhist country, where the burning

of incense is a ritual in many places. Mountain passes are considered sacred and most commuters along the EWH conduct this act at several locations, by burning of fresh alpine rhododendron branches.

The on-road pollution sources along R1 – R4 were grouped into four categories, (1) on-road vehicles, (2) road works (3) unpaved roads and (4) roadside combustion. It should be noted that at roadwork sites, the traffic was regulated, allowing vehicles to only travel at specified times, in order to expedite the work. Those vehicles arriving at the worksite outside the specified times were stopped until the road was open again. This resulted in vehicles travelling in convoy once the road was cleared for traffic. Source characterization was based on time-stamped voice recordings where the investigator noted all of the observed sources. Further, given the higher traffic volume and vehicle speed along R5, source characterization was not done for this segment, since it was deemed unsafe for the investigator to record their observations while simultaneously driving the research vehicle. However, because of the higher traffic volume, vehicle emissions are expected to be the dominant source for R5.

5.2.4 Data processing and analyses

Data were downloaded at the end of each day's measurements and inspected for any missing values and other anomalies. The PN concentrations were multiplied by the correction factor. The corrected data were aligned to the respective road segments, district towns and specific source events encountered along the road. During some events, the NT recorded PN concentrations exceeding 1×10^6 particle/cm^3, which was beyond the rated maximum of the instrument. These data were left unaltered and set at the maximum limit of the instrument.

5.2.4.1 Estimation of source contributions and on-road background concentrations

Source contributions were estimated from the time series concentrations and the voice recorded source characterization data. All of the peaks observed in the time series concentrations were aligned with the respective sources. The mean concentrations were computed by taking average of peak concentrations for the duration when an identified source was intercepted by the research vehicle. The background concentrations for each road segment were calculated by discounting the peaks contributed by the sources.

The extent to which on-road sources contributed to pollution levels can be estimated from the ratio (γ) of peak concentrations from the sources to background concentrations. We computed the trip average γ for each pollutant by subtracting the mean background level from the mean peak attributable to each of the on-road sources (as the peak already contained the background level) and then dividing it by the mean background concentrations for each of the road segments.

5.3 Results and Discussion

Details of the measurements conducted along the EWH are presented in Table 5.1. Between 20/11/2013 and 30/11/2013, instrument malfunction led to some data loss. For subsequent measurements, between 16/12/2012 and 08/01/2013, a complete set of data was captured over the entire length of the EWH. In total, a successful data capture was obtained for over 2900 km of driving along the EWH.

Table 5.1: Details of on-road measurements conducted along the EWH

Date	Start time	End time	Road segments	Direction of travel	Distance (km)	Pollutants
20-Nov-12	8:15	12:44	R1	X→T1	110	PM_{10}, PN
22-Nov-12	9:27	17:24	R3, R4	T2→T3→T4	197	PN
29-Nov-12	10:31	15:45	R4	T4→T3	129	PN
30-Nov-12	6:34	12:45	R2	T2→T1	193	PN
16-Dec-12	8:30	19:48	R1, R2	X→T1→T2	303	PM_{10}, PN, CO
17-Dec-12	8:37	18:46	R3, R4, R5	T2→T3→T4→Y	267	PM_{10}, PN, CO
22-Dec-12	9:38	18:33	R5, R4, R3	Y→T4→T3→T2	267	PM_{10}, PN, CO
23-Dec-12	7:29	18:21	R2, R1	T2→T1→X	303	PM_{10}, CO
01-Jan-13	7:40	18:06	R1, R2	X→T1→T2	303	PM_{10}, PN, CO
02-Jan-13	8:23	18:19	R3, R4, R5	T2→T3→T4→Y	267	PM_{10}, PN, CO
07-Jan-13	9:50	19:40	R5, R4, R3	Y→T4→T3→T2	267	PM_{10}, PN, CO
08-Jan-13	7:48	18:19	R2, R1	T2→T1→X	303	PM_{10}, PN, CO
Total road distance with successful data:					2909	

5.3.1 Traffic density

Figure 5.2 shows total traffic variations along the EWH during the campaign. As expected, the traffic density was highest for R5 and lowest for R2. R5 is the final segment of the EWH connecting the capital city to other districts. Although R2 is the longest segment (193 km) of the EWH, a major portion of the road passed through the wilderness and very few settlements were connected by this road segment. Overall, light vehicles constituted 75% of the total traffic observed during the campaign, while 14% were heavy, 8% medium and 2% were two-wheeler. The mean total half hourly traffic density during the campaign varied from 7 – 13 for R1, 5 – 6 for R2, 3 – 12 for R3, 16 – 20 for R4 and 28 – 63 for R5 (SI Table S5.1).

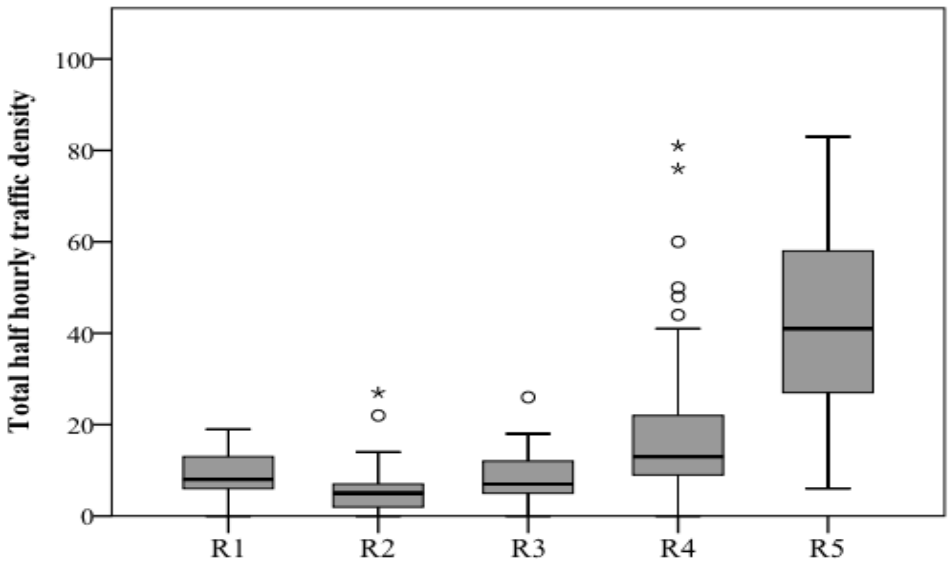

Figure 5.2: Total half hourly traffic variations along the EWH for the entire campaign. The boxplot presents minimum, first quartile, median (middle dark line), third quartile and maximum.

5.3.2 PM_{10}, PN and CO concentrations

Figure 5.3 and SI Table S5.2 present the summary statistics for on-road pollutant concentrations for different road segments of the EWH. In addition to the mean and median, the first and third quartiles have also been presented, since they are less affected by the extreme concentrations (Westerdahl et al., 2005) observed for this study (which is evident from the high standard deviations for all of the measured pollutants). It can be seen that the concentration of pollutants (PM_{10}, PN and CO) not only displayed substantial variability between different road segments (R1 – R5), but also between the measurement periods for individual road segments. However, the highest concentrations for all pollutants were observed for R5, except on 2 January 2013, where R3 and R4 presented higher CO than R5. The higher concentrations for R5 than other road segments were expected due to the higher traffic volume (Figure 5.2). A point to note is that R4 presented a comparable mean PM_{10} concentration with that of R5, which was due to the unpaved road, nearly 27 km in distance, towards the end of R4 when travelling from the eastern side. This stretch of R4 presented the highest PM_{10} concentrations throughout the campaign. Further, road widening work

was being conducted on the same stretch of R4. On 17 December 2012, the research vehicle arrived at the road widening site when the road was already closed (a full description of this event is provided in the Supplementary Information, SI file). When the road was opened for traffic, the research vehicle was caught in the middle of a long line of vehicles while driving through the same stretch of unpaved road. As a result, the high PM_{10} concentrations measured on that day elevated the overall mean concentration for R4 (see SI Table S5.2 D).

Summary statistics for on-road pollutant concentrations while driving through the four district towns are presented in Figure 5.3 and SI Table S5.3. While there were variations in minimum and maximum mean concentrations, the overall means were higher for T2 and T4 compared with T1 and T3. This points to an impact from the relative size of the towns, since T2 and T4 are bigger than T1 and T3, and also the relative degree of urban activities, as explained in Section 2.1. The overall mean for all the district town roads was PM_{10} = 128 ± 77 μg/m^3, PN = 4.00 × 10^4 ± 2.74 × 10^4 particles/cm^3 and CO = 0.29 ± 0.35 ppm, which were higher than the mean concentrations obtained for most of the road segments. Exceptions were found for PM_{10} for R4 and R5, and PN for R5, where higher concentrations were found for these road segments than the town roads. However, it should be noted that measurements for town roads were comparatively shorter than for R1 – R5, given their very short road lengths. Depending on the size of the towns, the sampling duration for town roads varied from 6 minutes for T3 to 17 minutes for T4, whereas for R1 – R5, sampling was undertaken for several hours. Further, concentrations in urban areas can vary greatly depending on the time of day, which is difficult to capture using mobile measurements. This problem can be addressed by complementing mobile measurements with simultaneous fixed site monitoring. Such results have been

reported by other studies. For example, Westerdahl et al. (2009) conducted concurrent on-road, and fixed site ambient and roadside measurements, using the same set of instrumentation. They reported daytime ratios of 2.9 (CO) and 2.1 (UFP) for on-road to roadside, and 13.3 (CO) and 3.0 (UFP) for on-road to ambient, respectively. Similarly, Apte et al. (2011) conducted simultaneous in-vehicle measurements inside an auto-rickshaw (a semi-enclosed three-wheeled vehicle, where in-vehicle concentration actually reflects on-road levels) and continuous urban background measurements using fixed monitors. They reported that, on average, in-vehicle PN concentration was 8.4 times higher than urban background levels. However, it should be noted that mobile measurements provide less data and that concentrations are related to the time of the day when measurements are conducted, compared to diurnal variations from fixed site measurements. Nevertheless, values obtained from mobile measurements give a good picture of the concentrations to which commuters are potentially exposed while travelling on roads, relative to urban background or ambient levels. In the current study, we could not conduct any fixed site measurements along any of the road segments or in the district towns, in order to adjust for temporal variations, due to limited number of instruments. Also, there were no monitoring stations near the routes travelled, including in the district towns.

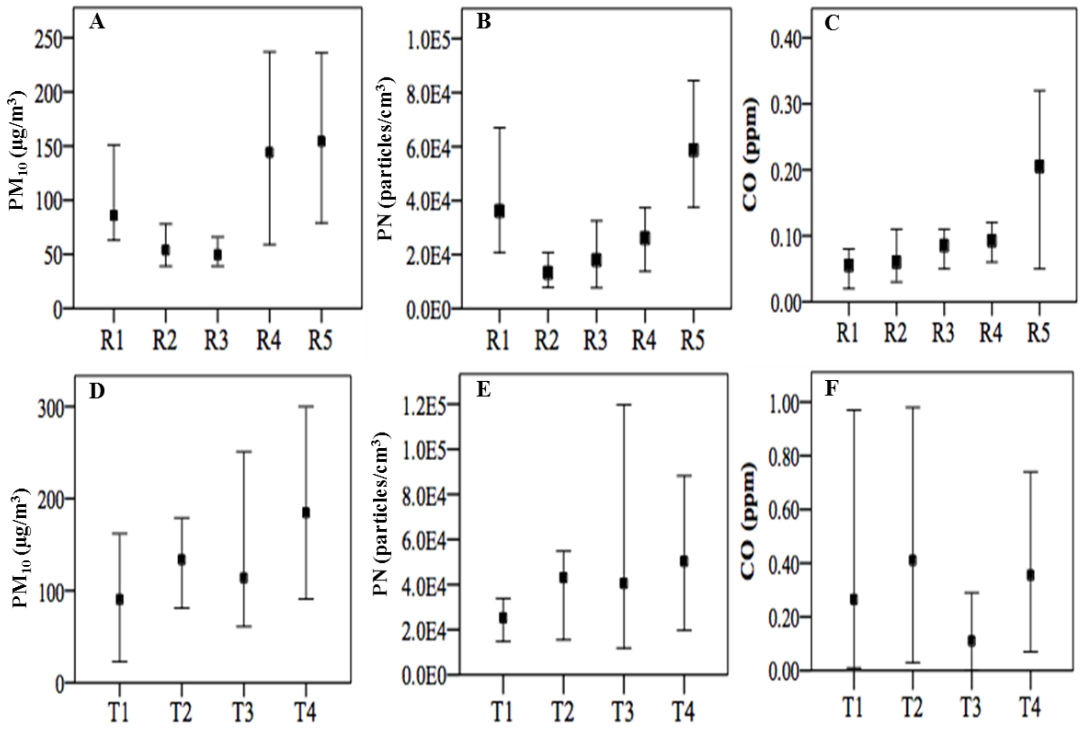

Figure 5.3: Variations of mean pollutant concentrations. A, B and C are on-road concentrations for different road segments (R1 – R5) of the EWH. D, E and F are on-road concentrations while driving through district towns connected by the EWH (T1 – T4).
Whiskers present minimum and maximum mean concentrations during the campaign, while shaded square present overall mean concentrations.

5.3.3 Comparison with other on-road studies

In Table 5.2, a summary of previous mobile studies, most of which were conducted on major roadways in large cities in both developed and developing countries, are compared with the current study. No studies for similar environments (such as roads spanning into rural and mountain areas) or for comparable traffic volumes were available for comparison. However, it should be noted that there has been a rapid increase in the number of on-road studies around the world, including tunnels and roadside investigations, which are not directly comparable with this work. While PN concentrations have been the primary focus of many studies, differences existed in terms of the particle size ranges that were measured (based on the different models of Condensation Particle Counters (CPC) and Scanning Mobility Particle

Sizers (SMPS) used). The instrument used for this study (NanoTracer) has a narrower range compared with the CPC and SMPS instruments used by other studies. This narrow range, together with the correction factor that was computed by running the NT side by side with the CPC 3787, may have resulted in an underestimation of our total PN concentrations. Given the remoteness of the study site and the challenging environments in Bhutan, it was not feasible for us to use CPCs and SMPSs. Notwithstanding the differences, PN concentrations for R1 – R5, as well as the four district town roads (T1 – T4) were significantly lower than studies summarized in Table 5.2. However, daytime PN concentration (1.5×10^4 particles/cm^3) reported for a rural road in Zurich (Bukowiecki et al., 2002) was comparable with the mean PN concentrations obtained for R2 and R3 (1.31×10^4 and 1.87×10^4 particles/cm^3). However, the type of rural environment and road distance included for mobile sampling were not mentioned in the paper, while the current study involved a range of geographic conditions and a distance of 68 – 193 km, depending on the road segment. Further, Morawska et al. (2008) conducted a meta-analyses of 71 studies that quantified PN concentrations in different environments, including on-road and tunnel environments. The reported mean concentrations were highest for on-road and tunnel environments, being 7.15×10^4 and 1.77×10^5 particles/cm^3, respectively. The mean PN concentrations obtained for the EWH, including the four district town roads, were up to five times lower than the mean concentrations reported for on-road studies, but in general, they were comparable with other microenvironments, such as street canyon and roadside levels.

For CO, although two studies (Westerdahl et al., 2005, Westerdahl et al., 2009) used a similar instrument (TSI Q-Trak) to one used in this study, concentrations (median and mean) were at least an order of magnitude lower for the present site. We

have not come across any study reporting on-road PM_{10} levels using a mobile sampling method. Nevertheless, concentrations obtained for most of the road segments were higher than the World Health Organization (WHO) short-term guideline (24 hours) of 50 $\mu g/m^3$ (WHO, 2006).

Our results for predominantly rural roads in Bhutan exhibited high concentrations at times, especially for PM_{10} and PN. This is not surprising since higher pollutant concentrations are expected on roadways, given their proximity to vehicle emissions (Knibbs et al., 2009). Therefore, commuters, as well as road maintenance workers and roadside residents along the EWH, are potentially exposed to elevated concentrations of pollutants. Future studies should focus on investigating the dispersion patterns of roadway air pollutants and defining the health impacts of long-term exposure in Bhutan and other developing countries, given that land development in these areas favours the location of houses in close proximity to major roads.

5.3.4 Estimated source contributions to on-road concentrations

Table 5.3 shows the mean concentrations for the characterized sources and the extent of their contributions (γ) to on-road pollution levels for R1, R2, R3 and R4. It can be seen from Table 5.3 that for R1, the highest PM_{10}, PN and CO were all contributed by road works. The highest PM_{10} for R2 was contributed by roadside combustion, while PN and CO were contributed by vehicles. For R3, the highest PM_{10} and CO were contributed by vehicles and PN by road works. Unpaved roads made the highest PM_{10} contribution for R4, while the highest PN was contributed by vehicles and CO by roadside combustion. While no specific source trend was observed for the road segments, the highest overall PM_{10} contribution was made by unpaved roads, and PN and CO by vehicles. This was expected, since exhaust emissions are the principal source for PN and CO (Fruin et al., 2008, Morawska et al., 2008), while coarse particle

emissions can be a dominant source on unpaved roads (Kam et al., 2012). These findings highlight the diversity of sources that contribute to on-road pollution in Bhutan, and that there is substantial variability in the dominant source between locations.

As explained earlier, source characterization and their contributions could not be assessed for R5. However, as traffic density was highest along this stretch of the road, and road conditions were better than for other road segments (no major road works and relatively few settlements), it was expected that traffic contribution were the dominant source for R5. Also, since we relied on visual observations for source characterization, our calculation of peak concentrations may not be accurate in terms of timing and the interception of simultaneous sources. Since the study was conducted in a low resource country and in very challenging environments, the option of using more sophisticated methods and instrumentation was limited. Further, we did not test the contribution from the research vehicle's own exhaust, however several studies that relied on mobile platforms have reported this to be negligible, both during travelling as well as when the research vehicles were stopped (Weijers et al., 2004, Wang et al., 2009, Apte et al., 2011).

Table 5.2: Summary of related previous on-road studies (PM_{10}: $\mu g/m^3$, PN: particles/cm^3, CO: ppm)

Study	Pollutants	Instrumentation	Time average	Particle size range	On-road concentration
This study East-West Highway, Bhutan; 570 km; Normal driving; 6 measurements between November 2012 and January 2013; Successful data from > 2900 km	PM_{10} (mean)	DustTrak, TSI	5 seconds		49 – 148 [a] 90 – 184 [b]
	PN (mean)	NanoTracer, Philips, Aerasense	5 seconds	10 – 300 nm	$1.31 \times 10^4 – 5.74 \times 10^4$ [a] $2.53 \times 10^4 – 5.03 \times 10^4$ [b]
	CO (mean)	IAQ-CALC, model 7545, TSI	5 seconds		0.06 – 0.19 [a] 0.11– 0.36 [b]
(Bukowiecki et al., 2002) Route covering downtown, suburban & rural areas in Zurich, Switzerland; Measurements between February 2001-May 2002	PN (mean)	TSI SMPS, DMA 3071	3 minutes	7 – 310 nm	7.8×10^4 [c] 1.5×10^4 [d]
Kittelson et al. (2004) Minnesota Highways, USA; 5 days sampling in November 2000; 800 km distance covered	PN (mean)	TSI 3025A CPC		3 – 1000 nm	4.03×10^5
		TSI 3071 SMPS		8 – 300 nm	1.03×10^5
Westerdahl et al. (2005) Los Angeles Freeways, USA (110N, 10E, 710S), ~120 km; Normal driving; 4 day measurements in February & April 2003	PN (median)	TSI portable CPC, model 3007	10 seconds	10 – 1000 nm	4.70×10^4 -1.90×10^5
	CO (median)	TSI Q-Trak Plus monitor, model 8554			1.9 – 2.7
Westerdahl et al. (2009) 4th Ring Road & Balding Expressway, Beijing; 26.6 km; 2 weeks measurement in summer 2007	PN (median)	FMPS, TSI, model 3091	1 seconds	6 – 560 nm	8.17×10^4 [e] 7.82×10^4 [f]
	CO (median)	Q-Trak, TSI, model 7655	10 seconds		4.0 [e] 3.7 [f]
Apte et al. (2011)* 19.5 km mixed route in New Delhi, India; Normal driving; 31 days, 62 trips, 160 measurements in February-May 2010 (early spring through mid-summer)	PN (mean)	TSI portable CPC, model 3007		10 – 1000 nm	2.90×10^5

[a]: Mean range for the 5 road segments (R1 – R5) of the East-West Highway. [b]: Mean range for 4 district town roads (T1 – T4). [c]: Urban daytime for clear weather period for November 2001. [d]: Rural daytime for clear weather period for November 2001. [e]: On-road no traffic control daytime. [f]: On-road traffic control daytime. *: In-vehicle concentration for an auto-rickshaw, a semi enclosed three-wheeled vehicle that offers little protection from exhaust plumes and therefore, reflects on-road levels.

Table 5.3: Peak concentrations from on-road and roadside sources and their contributions to on-road pollution levels (PM_{10}: $\mu g/m^3$, PN: particles/cm^3, CO: ppm)

Road Segment	Pollutants	Vehicles	γ1	Road works	γ2	*Unpaved roads	γ3	Roadside combustion	γ4
R1	PM_{10}	268	3.0	468	5.9	304	3.5	352	4.2
	PN	6.96×10^4	2.0	9.66×10^4	3.2	-	-	7.10×10^4	2.1
	CO	0.59	12.9	0.65	14.3	-	-	0.32	6.5
R2	PM_{10}	430	9.0	358	7.4	274	5.4	476	10.1
	PN	1.57×10^5	13.9	5.01×10^4	3.8	-	-	5.28×10^4	4.0
	CO	1.03	23.2	1.36	31.0	-	-	0.54	11.7
R3	PM_{10}	305	6.4	258	5.3	162	2.9	279	5.8
	PN	9.95×10^4	8.5	1.68×10^5	15.1	-	-	8.47×10^4	7.1
	CO	0.91	14.2	0.41	5.8	-	-	0.14	1.3
R4	PM_{10}	433	4.1	312	2.7	888	9.4	444	4.2
	PN	9.58×10^4	3.3	3.72×10^4	0.7	-	-	5.35×10^4	1.4
	CO	0.48	11.8	0.26	5.9	-	-	1.52	39.5
Overall	PM_{10}	359	5.1	349	4.9	407	5.9	387.75	5.5
	PN	1.05×10^5	5.4	8.80×10^4	4.3	-	-	6.55×10^4	3.0
	CO	0.75	15.4	0.67	13.7	-	-	0.63	12.8

γ1: ratio of mean peak concentration from vehicles to mean on-road background concentration.
γ2: ratio of mean peak concentration from road works to mean on-road background concentration.
γ3: ratio of mean peak concentration from unpaved roads to mean on-road background concentration.
γ4: ratio of mean peak concentration from roadside combustion to mean on-road background concentration.
*: PN and CO were not characterized from unpaved roads since they are tracers for combustion activities (Morawska et al., 2008).

5.4 Conclusions

We quantified on-road air quality using a vehicle-based sampling method along the 570 km EWH in Bhutan, making it one of the longest roads to be investigated so far. This major roadway is an economic lifeline for a significant proportion of Bhutan's population and transverses diverse geographical features, as well as both rural and urban settlements. This means that there was a diversity of pollution sources over its length, as opposed to the dominant influence of vehicle

emissions on urban roads.

Apart from vehicle emissions, other dominant pollution sources on the EWH were road works, unpaved stretches of road and both vehicle and non-vehicle roadside combustion. The on-road pollution levels varied according to location, with the highest concentrations (PM_{10}, PN and CO) for R5, the final 70 km segment of the EHW, where traffic density was also the highest. In terms of source contributions, although no specific overall trend was observed for any of the road segments, the highest PM_{10} contribution was made by unpaved roads, and PN and CO by vehicles. One definite conclusion that we can draw from these findings is that a diversity of sources contribute to on-road pollution in Bhutan, and that there is substantial variability in the dominant source between locations. For the four district towns connected by the EWH, the concentrations were highest in T2 and T4, most likely reflecting the positive association between pollution levels and the relative size of the towns and urban activities.

Compared with other studies conducted on metropolitan city roads around the world, we found typically lower pollution levels for all of the pollutants investigated. However, short-term levels were relatively high, with PM_{10} levels exceeding the WHO short-term 24-hours guideline for most of the road segments. These findings highlight that road commuters, and people living and working near roads in Bhutan, can be exposed to elevated levels of pollutants. Any associated public health risks can be exacerbated by the tendency of people to settle along the roads in Bhutan, combined with ever-increasing traffic volumes. In the recent years, there have been numerous studies in the Himalayan region focussing on household air pollution, in relation to biomass fuel usage and traditional cookstoves. However, this is the first study we are aware of, to investigate pollution levels relevant to population exposure

on and near popular mountain roads. Future studies with a focus on the dispersion patterns of roadway pollutants and the assessment of health impacts from on- and near-road exposure would be of significance.

5.5 Acknowledgements

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Supplementary Information (SI)

Mobile assessment of on-road air pollution and its sources along the East-West Highway in Bhutan

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Table S5.1: Summary statistics of half hourly traffic density for different road segments of the EWH.

Road segments	Date	Minimum	Maximum	Mean	SD	Total
R1	20-Nov-12	0	18	8	5	82
	30-Nov-12	2	17	7	4	59
	16-Dec-12	5	19	13	5	128
	23-Dec-12	6	16	11	4	67
	01-Jan-13	1	19	7	5	67
	08-Jan-13	4	11	8	3	47
R2	21-Nov-12	0	11	5	3	81
	30-Nov-12	0	27	6	7	101
	16-Dec-12	0	9	5	3	67
	23-Dec-12	2	11	6	3	88
	01-Jan-13	2	12	5	3	61
	08-Jan-13	0	22	5	5	85
R3	22-Nov-12	0	26	12	9	82
	29-Nov-12	2	17	9	6	47
	17-Dec-12	2	16	10	5	57
	22-Dec-12	2	13	9	5	35
	02-Jan-13	6	11	8	2	47
	07-Jan-13	1	4	3	2	11
R4	22-Nov-12	11	81	26	23	283
	29-Nov-12	0	41	16	12	148
	17-Dec-12	1	76	20	22	225
	22-Dec-12	6	31	17	9	155
	02-Jan-13	3	44	18	14	180
	07-Jan-13	0	50	14	15	151
R5	22-Nov-12	8	44	28	15	111
	29-Nov-12	24	77	43	20	256
	17-Dec-12	19	83	63	30	251
	22-Dec-12	17	69	40	18	240
	02-Jan-13	6	45	33	18	131
	07-Jan-13	18	67	52	19	258

SD: Standard Deviation

Table S5.2: Summary statistics of PM_{10}, PN and CO for different roads segments (R1 – R5) of the EWH.

(A)	R1				
	Mean	**SD**	**25th %**	**Median**	**75th %**
20-Nov-12					
PM_{10}	63	236	7	11	28
PN	6.70×10^4	1.35×10^5	1.46×10^4	2.91×10^4	6.32×10^4
16-Dec-12					
PM_{10}	78	70	60	67	75
PN	2.84×10^4	4.98×10^4	1.39×10^4	1.79×10^4	2.58×10^4
CO	0.08	0.41	0.00	0.00	0.00
23-Dec-13					
PM_{10}	63	129	31	42	62
CO	0.04	0.22	0.00	0.00	0.10
01-Jan-13					
PM_{10}	74	130	37	46	63
PN	2.82×10^4	4.69×10^4	1.06×10^4	1.47×10^4	2.68×10^4
CO	0.08	0.31	0.00	0.00	0.00
08-Jan-13					
PM_{10}	151	303	101	118	126
PN	2.08×10^4	3.64×10^4	1.13×10^4	1.50×10^4	2.01×10^4
CO	0.02	0.17	0.00	0.00	0.00

(B)	R2				
	Mean	**SD**	**25th %**	**Median**	**75th %**
30-Nov-12					
PN	2.08×10^4	5.64×10^4	5.37×10^3	7.83×10^3	1.27×10^4
16-Dec-12					
PM_{10}	53	127	26	43	58
PN	1.40×10^4	2.96×10^4	4.95×10^3	7.61×10^3	1.22×10^4
CO	0.03	0.23	0.00	0.00	0.00
23-Dec-13					
PM_{10}	39	162	8	19	38
CO	0.06	0.15	0.00	0.00	0.10
01-Jan-13					
PM_{10}	45	210	13	25	34
PN	7.81×10^3	1.67×10^4	3.02×10^3	4.93×10^3	8.51×10^3
CO	0.11	0.67	0.00	0.00	0.00
08-Jan-13					
PM_{10}	78	142	52	61	77
PN	1.02×10^4	2.48×10^4	4.79×10^3	6.49×10^3	9.69×10^3
CO	0.04	0.25	0.00	0.00	0.00

(C)	R3				
	Mean	**SD**	**25th %**	**Median**	**75th %**
22-Nov-12					
PN	3.25×10^4	6.21×10^4	1.15×10^4	2.00×10^4	3.59×10^4
17-Dec-12					
PM_{10}	39	65	18	28	37
PN	1.99×10^4	5.02×10^4	5.35×10^3	7.50×10^3	1.36×10^4
CO	0.08	1.20	0.00	0.00	0.00
22-Dec-13					
PM_{10}	47	20	36	41	48
PN	1.28×10^4	1.62×10^4	6.21×10^3	8.00×10^3	1.21×10^4
CO	0.05	0.40	0.00	0.00	0.00
02-Jan-13					
PM_{10}	46	153	15	20	29
PN	1.67×10^4	4.61×10^4	5.29×10^3	7.98×10^3	1.33×10^4
CO	0.10	0.84	0.00	0.00	0.00
07-Jan-13					
PM_{10}	66	33	40	58	87
PN	7.69×10^3	6.31×10^3	4.74×10^3	5.94×10^3	9.02×10^3
CO	0.11	0.18	0.00	0.00	0.10

(D)	R4				
	Mean	**SD**	**25th %**	**Median**	**75th %**
29-Nov-12					
PN	3.74×10^4	9.55×10^4	8.81×10^3	1.18×10^4	2.40×10^4
17-Dec-12					
PM_{10}	237	859	33	42	104
PN	2.98×10^4	6.29×10^4	7.59×10^3	1.10×10^4	2.67×10^4
CO	0.12	0.73	0.00	0.00	0.10
22-Dec-13					
PM_{10}	131	281	43	82	123
PN	3.08×10^4	6.50×10^4	7.77×10^3	1.60×10^4	2.85×10^4
CO	0.09	0.45	0.00	0.00	0.00
02-Jan-13					
PM_{10}	59	243	19	22	31
PN	1.38×10^4	2.91×10^4	5.90×10^3	7.54×10^3	1.35×10^4
CO	0.10	0.40	0.00	0.00	0.00
07-Jan-13					
PM_{10}	149	335	73	93	121
PN	1.85×10^4	4.10×10^4	6.62×10^3	8.91×10^3	1.37×10^4
CO	0.06	0.44	0.00	0.00	0.00

(E)	R5				
	Mean	**SD**	**25th %**	**Median**	**75th %**
17-Dec-12					
PM_{10}	117	295	35	54	94
PN	5.45×10^4	7.07×10^4	1.37×10^4	2.97×10^4	6.80×10^4
CO	0.15	0.35	0.00	0.00	0.10
22-Dec-13					
PM_{10}	185	213	97	125	207
PN	8.45×10^4	1.50×10^5	2.20×10^4	3.68×10^4	7.18×10^4
CO	0.32	0.94	0.10	0.10	0.20
02-Jan-13					
PM_{10}	79	146	26	35	63
PN	3.76×10^4	6.74×10^4	8.32×10^3	1.47×10^4	3.37×10^4
CO	0.05	0.30	0.00	0.00	0.00
07-Jan-13					
PM_{10}	236	689	92	125	174
PN	5.83×10^4	1.11×10^5	1.46×10^4	2.62×10^4	5.10×10^4
CO	0.30	0.97	0.00	0.00	0.20

Table S5.3: Summary statistics of PM_{10}, PN and CO concentrations for district towns (T1 – T4) connected by the EWH.

(A)	PM_{10} (µg/m^3)				
Towns	**Date**	**Minimum**	**Maximum**	**Mean**	**SD**
T1	20-Nov-12	5	1438	23	58
	16-Dec-12	50	313	85	49
	23-Dec-12	16	351	52	54
	1-Jan-13	27	928	162	158
	8-Jan-13	59	483	130	79
T2	16-Dec-12	38	557	160	109
	22-Dec-12	36	662	179	149
	1-Jan-13	17	806	114	114
	7-Jan-13	21	449	81	49
T3	17-Dec-12	30	175	65	36
	22-Dec-12	41	231	61	27
	2-Jan-13	18	610	78	111
	7-Jan-13	111	1972	251	256
T4	17-Dec-12	41	929	91	86
	22-Dec-12	188	4134	300	369
	2-Jan-13	27	4184	113	331
	7-Jan-13	121	2152	234	242

(B)		PN (particles/cm^3)			
Towns	**Date**	**Minimum**	**Maximum**	**Mean**	**SD**
T1	20-Nov-12	4.86×10^3	1.35×10^5	3.03×10^4	2.97×10^4
	16-Dec-12	8.42×10^3	1.64×10^5	2.21×10^4	2.13×10^4
	1-Jan-13	5.11×10^3	2.11×10^5	3.39×10^4	3.39×10^4
	8-Jan-13	5.87×10^3	9.59×10^4	1.48×10^4	1.19×10^4
T2	22-Nov-12	7.28×10^3	4.94×10^5	5.32×10^4	8.15×10^4
	16-Dec-12	4.44×10^3	2.40×10^5	5.48×10^4	4.51×10^4
	22-Dec-12	2.94×10^3	2.47×10^5	5.31×10^4	5.34×10^4
	1-Jan-13	1.58×10^3	3.60×10^5	3.85×10^4	5.11×10^4
	7-Jan-13	5.26×10^2	6.90×10^4	1.55×10^4	1.34×10^4
T3	22-Nov-12	5.56×10^4	1.00×10^6	1.20×10^5	1.28×10^5
	29-Nov-12	6.56×10^3	2.78×10^5	4.59×10^4	5.16×10^4
	17-Dec-12	5.83×10^3	1.41×10^5	2.30×10^4	2.73×10^4
	22-Dec-12	6.39×10^3	3.14×10^4	1.20×10^4	5.12×10^3
	2-Jan-13	5.03×10^3	5.94×10^4	1.18×10^4	8.76×10^3
	7-Jan-13	1.16×10^4	1.49×10^5	3.05×10^4	2.36×10^4
T4	17-Dec-12	5.71×10^3	1.00×10^6	3.93×10^4	6.43×10^4
	22-Dec-12	2.84×10^4	1.00×10^6	8.82×10^4	1.59×10^5
	2-Jan-13	2.89×10^3	5.64×10^5	1.97×10^4	4.61×10^4
	7-Jan-13	1.03×10^4	9.86×10^5	5.38×10^4	9.91×10^4

(C)		**CO (ppm)**			
Towns	**Date**	**Minimum**	**Maximum**	**Mean**	**SD**
T1	16-Dec-12	0.00	0.60	0.02	0.08
	23-Dec-12	0.00	0.90	0.06	0.14
	1-Jan-13	0.00	5.40	0.97	1.24
	8-Jan-13	0.00	0.10	0.01	0.03
T2	16-Dec-12	0.00	4.70	0.47	0.81
	22-Dec-12	0.00	4.70	0.98	1.24
	1-Jan-13	0.00	3.50	0.16	0.48
	7-Jan-13	0.00	0.80	0.03	0.08
T3	17-Dec-12	0.00	0.50	0.08	0.10
	22-Dec-12	0.00	0.10	0.00	0.02
	2-Jan-13	0.00	1.70	0.29	0.55
	7-Jan-13	0.00	1.10	0.07	0.20
T4	17-Dec-12	0.00	1.60	0.07	0.22
	22-Dec-12	0.10	15.20	0.54	1.79
	2-Jan-13	0.00	3.30	0.07	0.33
	7-Jan-13	0.00	9.60	0.74	1.05

Figure S5.1: Sections of the East-West Highway at different locations.

Figure S5.2: Pollution sources along the East-West Highway.

A: Smoke from the roadside settlement from cooking and heating activities using firewood. B: Roadside fire lit by workers. C: Incense burning at one of the mountain passes near the Buddhist stupa. D: Road resurfacing work. E: Road widening work. F: Traffic movement along the unpaved road following stopover at one of the roadwork sites.

Impact of traffic regulation at the roadwork site

During the campaign there was a major road widening work at R4, spanning a kilometer (site location, 439 – 440 km from the eastern side). At the worksite traffic was regulated and allowed only at specific times to expedite the work. The road towards the capital from the worksite was narrow and downhill until the bridge located at 467 km (from the eastern side). That stretch of road also had a quarry site and a large fleet of trucks transporting stone-chips to the two hydropower plants which were under construction downstream. As such the road between the worksite and the bridge was heavily riddled with potholes and unpaved sections. On 17 December 2012, the research vehicle arrived at the road widening site at 14:12 o'clock when road was closed for traffic. When the road was cleared, the research vehicle had to travel in a convoy with other stranded vehicles. There was thick cloud of dusts all along the road until the bridge (total distance 27 km), beyond which the traffic gradually spread out. As a result the highest PM_{10} concentration during the campaign was obtained during this event (SI Figure S5.3). The mean PM_{10} concentration for the same stretch of R4 was 679 ± 1531µg/m^3 on 17/12/2012, which was over four times higher than the trip average of 166 ± 367 µg/m^3 from other days (with mean ranging from 56 ± 136 to 239 ± 467 µg/m^3, respectively), while mean CO concentration (0.21 ± 0.40 ppm) was over two times higher than the trip average from other days (0.09 $^{\pm}$ 0.65 ppm). However, there was no large variation of mean PN concentration ($5.52 \times 10^4 \pm 6.72 \times 10^4$ particles/cm^3), which was comparable with the mean trip average concentration ($4.05 \times 10^4 \pm 8.18 \times 10^4$) from other days. In fact, the mean PN concentration was similar on 22/12/2012 and higher on 29/12/2012 than on 17/12/2012 for the same stretch of R4. This was because PN readily undergo coagulation sink when particle mass concentrations are high (Apte et al., 2011). This

phenomenon has been empirically proven by Mönkkönen et al. (2004). They found a linear relationship between PN and PM_{10} concentrations of up to 300 µg/m^3. Beyond this concentration PN decreased with increasing mass concentrations due to effective coagulation sink provided by particle mass.

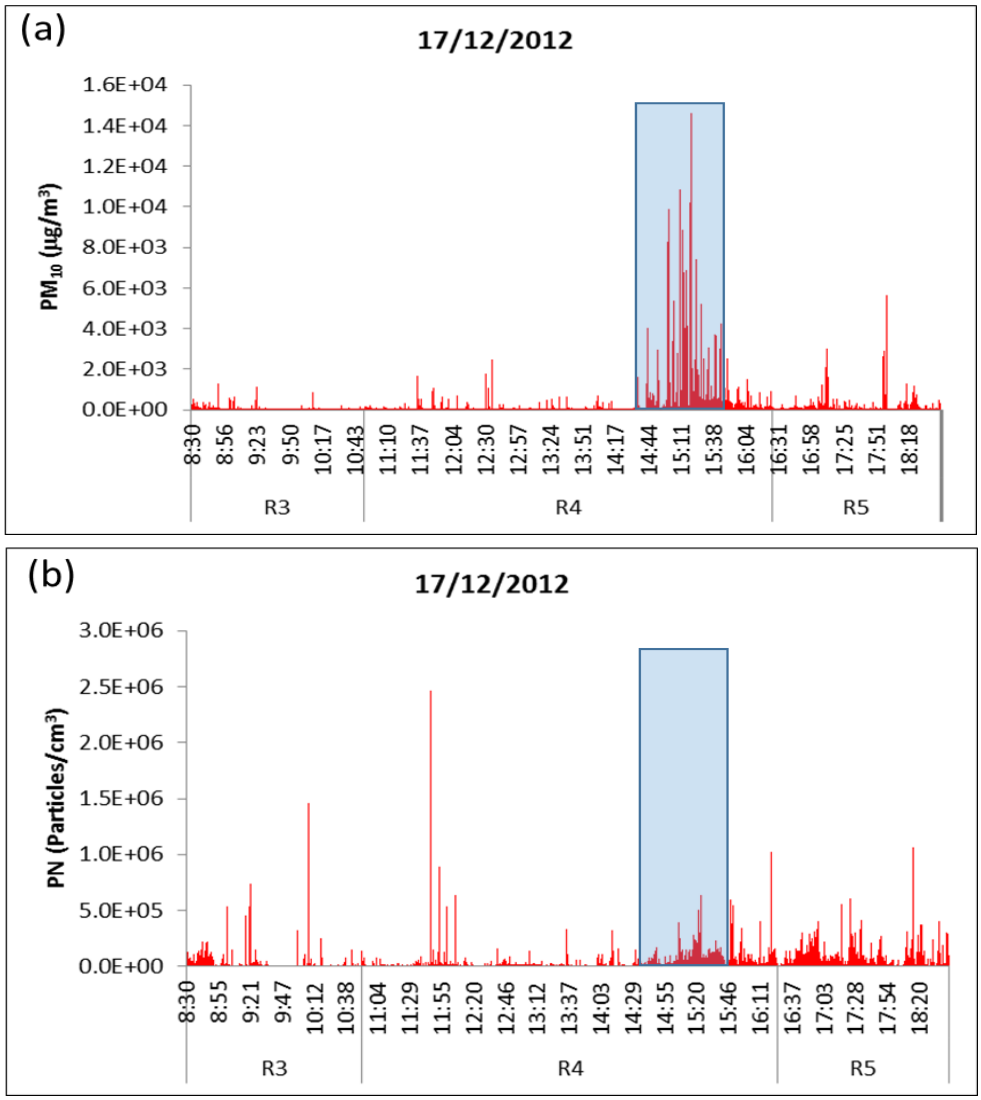

Figure S5.3: Time-series pollutant concentrations of PM_{10} (a) and PN (b) for R3 – R4 on 17/12/2012.

Shaded areas present the pollution levels between 439 and 467 km stretch of R4, when the research vehicle travelled in a convoy with other stranded vehicles following road closure at the worksite.

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Chapter 6: Tackling the Health Burden from Household Air Pollution (HAP): Development and Implementation of New WHO Guidelines

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Statement of Contribution

Statement of Contribution

The authors listed below have certified* that:

- they meet the criteria for authorship in that they have participated in the conception, execution, or interpretation, of at least that part of the publication in their field of expertise;
- they take public responsibility for their part of the publication, except for the responsible author who accepts overall responsibility for the publication;
- there are no other authors of the publication according to these criteria;
- potential conflicts of interest have been disclosed to (a) granting bodies, (b) the editor or publisher of journals or other publications, and (c) the head of the responsible academic unit, and
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In the case of this chapter: *Tackling the Health Burden from Household Air Pollution (HAP): Development and Implementation of New WHO Guidelines*

Contributor	Statement of contribution*
Tenzin Wangchuk QUT Verified Signature Date: 24/08/2015	Contributed to manuscript writing, a section on household air pollution problems in Bhutan.
Nigel Bruce*	Developed the structure and reviewed the manuscript.
Carlos Dora*	Led the project and reviewed the manuscript.
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Abstract

Household air pollution (HAP), arising mainly from the combustion of solid and other polluting fuels, is responsible for a very substantial public health burden, most recently estimated as causing 3.5 million premature deaths in 2010. These patterns of household fuel use have also important negative impacts on safety, prospects for poverty reduction and the environment, including climate change. Building on previous air quality guidelines, the WHO is developing new guidelines focused on household fuel combustion, covering cooking, heating and lighting, and although global, the key focus is low and middle income countries reflecting the distribution of disease burden. As discussed in this paper, currently in development, the guidelines will include reviews of a wide range of evidence including fuel use in homes, emissions from stoves and lighting, household air pollution and exposure levels experienced by populations, health risks, impacts of interventions on HAP and exposure, and also key factors influencing sustainable and equitable adoption of improved stoves and cleaner fuels. GRADE, the standard method used for guidelines evidence review may not be well suited to the variety and nature of evidence required for this project, and a modified approach is being developed and tested. Work on the guidelines is being carried out in close collaboration with the UN Foundation Global Alliance on Clean cookstoves, allowing alignment with specific tools including recently developed international voluntary standards for stoves, and the development of country action plans. Following publication, WHO plans to work closely with a number of countries to learn from implementation efforts, in order to further strengthen support and guidance. A case study on the situation and policy actions to date in Bhutan provide an illustration of the challenges and opportunities involved,

and the timely importance of the new guidelines and associated research, evaluation and policy development agendas.

Keywords

Household air pollution, Air quality guidelines, Fuel combustion, Exposure, Health risk, Bhutan.

6.1 Introduction

Well into the second decade of the 21st Century, almost 3 billion people (around 6 –700 million households) still rely on solid fuels (wood, animal dung, crop wastes, charcoal and coal) for their everyday cooking and heating needs. Combustion of household fuels is responsible for a very substantial global burden of disease, estimated at almost 2 million premature deaths for 2004 (WHO 2009). The most recent estimate of disease burden published by IHME for the year 2010 reported 3.5 million premature deaths, the main reason for the apparent increase being the addition of cardiovascular disease as an outcome (Lim et al., 2012). The incomplete combustion of solid fuels in traditional open fires and stoves leads to levels of household air pollution (HAP) which far exceed WHO air quality guidelines (WHO 2005), causing a wide range of respiratory diseases, adverse pregnancy outcomes and cancer affecting people across the whole life course. This burden occurs mainly in developing countries where solid fuels are used extensively for cooking, and also for heating (Figure 6.1).

Although the use of solid fuels for cooking has declined globally from around 60% of homes in 1980 to 41% in 2010, the world's growing population means that the number of people affected, some 2.8 billion, is the almost the same now as it was 30 years ago (Bonjour et al., 2012, WHO 2012). At current rates of change, the total number of people using solid fuels is expected to increase (IEA, 2011).

This is not only a health risk in developing countries, solid fuels are also used extensively for heating in more developed countries where poorer quality stoves, inadequate ventilation and maintenance contribute a smaller but largely unknown health burden (Noonan et al., 2007). Other household fuels used widely in both developing and developed countries, including kerosene (paraffin) and gas for both heating and lighting, also emit health damaging pollutants with exposure levels depending on the condition of the device and adequacy of ventilation (Lam et al., 2012).

For the 1.3 billion people with no access to electricity (and many more with intermittent supply) (IEA, 2011), lighting is most commonly obtained from simple kerosene lamps, which are an important source of HAP and an important risk for burns and fires (Lam et al., 2012). Children are also at risk of poisoning from drinking kerosene which is commonly purchased and stored in soft drink and water bottles (Yach, 1994). The majority of households without electricity are the same homes as those habitually using solid fuels for cooking. They are among the world's poorest people, who face multiple other threats to health, including from unsafe water and sanitation, infectious diseases, and inadequate access to health services.

Until recently, progress with the implementation of policies and interventions for improving access to clean and safe household energy has been very slow at best in most affected countries. The launch in 2010 of the UN Foundation's Global Alliance for Clean Cookstoves (http://www.cleanccokstoves.org), and the ambitious UN-led initiative on Sustainable Energy for All (http://www.sustainableenergyforall.org) are providing a renewed momentum to address this issue.

The new WHO indoor air quality guidelines on household fuel combustion will make an important contribution to these efforts through providing the scientific

basis for selecting household energy interventions which can substantially improve the health of users, and ensuring that these benefits are realised as quickly and equitably as possible. Building on existing WHO ambient and indoor air quality guidelines (WHO 2005; WHO 2009; WHO 2010), this new volume will be global in scope but with a particular focus on developing countries, in recognition of the distribution of disease burden. All main applications of energy in the home will be addressed, principally cooking, heating, lighting. The guidelines work will be conducted in two phases. Phase I will see the development and publication of the Guidelines volume, with initial guidance on implementation. Phase II will be directed at supporting countries in implementation, evaluating the progress made, and developing further technical support based on this experience.

The aim of this paper is to describe the development of these new guidelines, the rationale for the areas of evidence reviewed, methods for assessing the strength of evidence available for making recommendations, and plans for supporting and evaluating country implementation. In order to illustrate the circumstances and policy challenges involved in developing countries seeking to address this issue, a case study from one country, Bhutan, where the authors of this paper investigate HAP at the moment, is reported. The new WHO household air pollutions guidelines were featured at Healthy Buildings 2012.

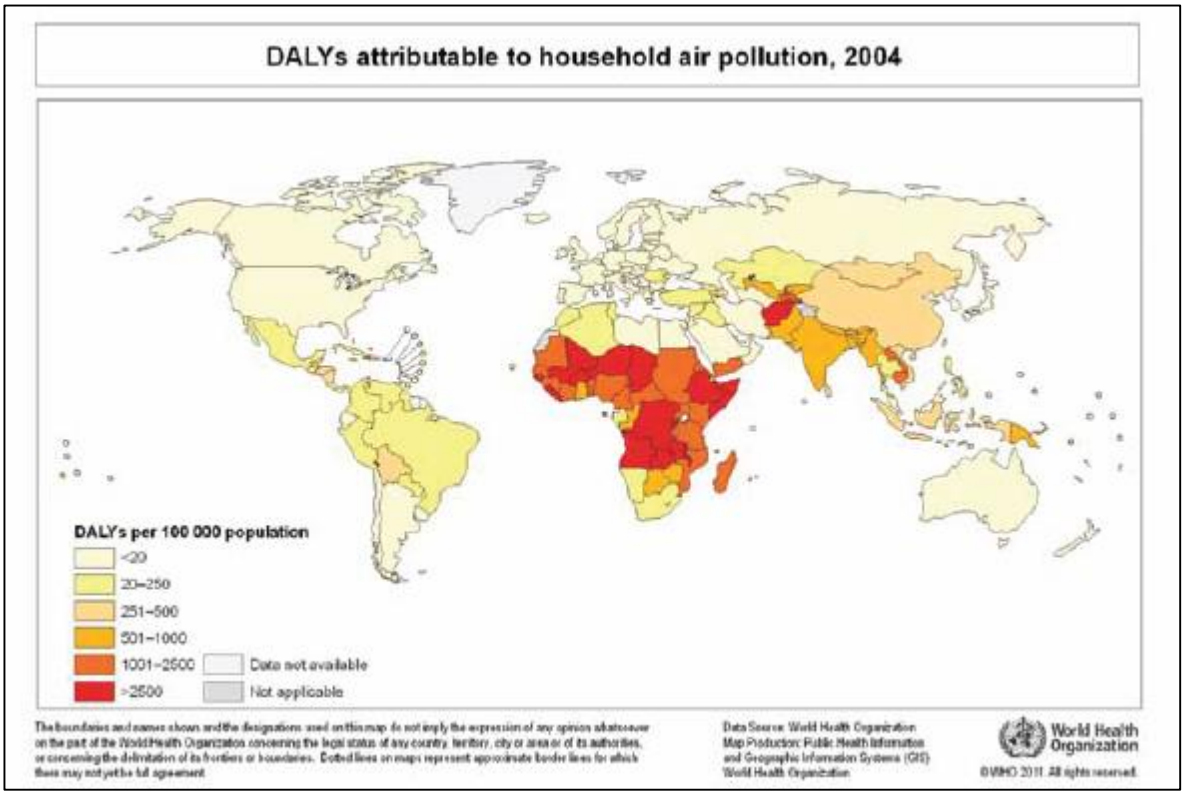

Figure 6.1: Burden of disease, expressed as DALYs (disability-adjusted life years), attributable to household air pollution from cooking with solid fuels, for the year 2004.
Source: WHO Global Health Observatory (2012).

6.2 Evolution of the new IAQG

These new Guidelines are the third in a series of WHO indoor air quality guidelines products. Following publication of the WHO ambient (outdoor) AQG Global Update in 2005 (WHO 2005), work began to develop indoor guidelines with three components: (i) Selected pollutants (WHO 2010), (ii) Dampness and Mould (WHO 2009), and (iii) Household Fuel Combustion, (this project).

The 2005 Global update included particulate matter (PM_{10} and $PM_{2.5}$), and introduced for the first time the concept of 'intermediate targets', set at three levels (IT1 – 3) (Table 6.1). The purpose of these was to allow a staged approach towards meeting the actual guideline levels in settings where current levels are very high, and rapid compliance with the guideline would be seen as impractical. This is an

important concept and strategy that is being carried through to the new Guidelines. The 2005 update also included a Chapter on household air pollution (Chapter 9), with a focus on the situation and challenges for developing countries. This discussed proposals that, alongside the conventional guideline levels of pollutants, technology and fuel-base guidelines would also be required. Thus, in the absence of actual measurements of HAP and exposure (which will not be available on a population basis), proxies based on survey information on fuels, technologies and household characteristics will need to be used. It was also argued that a limited number of ad hoc studies measuring HAP and exposure will still be required, to 'calibrate' such survey-based information to absolute concentrations.

The volume on Specific Pollutants reviewed the evidence for nine pollutants (see Box 1), six of which are combustion related. Particulate matter was not included in this volume, as the available evidence and guideline levels already covered in the 2005 Update were stated to apply equally to the indoor environment. This volume also included a new 24-hr guideline for carbon monoxide of 7 mg/m^3, in response to growing evidence of health impacts from chronic exposure, in addition to the already well recognised toxic effects from more acute exposures (Table 6.1).

Box 1: Selected pollutants (WHO 2010)

*1. Benzene**
*2. Carbon monoxide**
*3. Formaldehyde**
*4. Napthalene**
*5. Nitrogen dioxide**
*6. Poly aromatic hydrocarbons (PAH)**
7. Radon
8. Trichlorethylene
9. Tetrachlorethylene

** Combustion related*

Table 6.1: Guidelines for $PM_{2.5}$ from the WHO Global Update (2005) and Carbon monoxide from Selected Pollutants (2010)

Pollutant	**Guideline or target**	**Exposure period**	**Level**
$PM_{2.5}$ ($\mu g/m^3$)	Guideline	Annual average	10
	IT-3		15
	IT-2		25
	IT-1		35
Carbon monoxide (mg/m^3)	Guideline	8-hour	10
	Guideline	24-hour	7
Source: WHO			

6.3 Phase I: Development and publication of new guidelines for household fuel combustion

The core of the new Guidelines is a set of systematic evidence reviews that will form the basis of recommendations. These are complemented by description of an emissions model, based on work designed to relate emission rates to predicted HAP concentrations in a standard box model (Johnson et al., 2011). The reviews cover a wide range of topics, from background material on household fuel use globally, through levels of emissions, household pollution and exposure to health impacts and the effectiveness of the different types of stoves and fuels available as interventions. In recognition of the challenges for adoption of interventions in low-income settings, the guidelines also include a review of enabling and limiting factors for adoption and sustained use of improved stoves and clean fuels (see Box 2).

The evidence on which these reviews draw is very diverse, including nationally representative surveys, laboratory based testing of stoves, quasi-experimental field studies of the impacts of stoves on HAP and exposure, and a range of epidemiological studies of health risks that include cross sectional studies, analytic observational studies, and a very few randomised trials. Bringing this varied body of evidence together presents a considerable challenge, and not least for applying the

standard methods recommended by WHO for assessing the strength and quality of evidence used for making policy recommendations.

This methodology, known as GRADE (Grading of Recommendations Assessment, Development and Evaluation), was originally developed for the type of evidence available to medical interventions, and mainly RCTs (See GRADE working group http://www.gradeworkinggroup.org/index.htm). In summary, GRADE is used to categorise bodies of evidence used for making recommendations into four levels, namely high, moderate, low and very low (Balshem et al., 2011). Randomised trials enter the process as high, while all observational designs (including quasi-experimental) enter as low. This initial assessment can be downgraded according to five criteria, and upgraded according to three others (Table 6.2).

Using this methodology, bodies of evidence that are dominated by observational evidence, as is common in environmental health due to the practical and ethical constraints on conducting randomised trials, tend to result in GRADE assessments which are low or very low.

Recent years have seen growing awareness of the limitations of this approach for application to the evidence use for policy on complex interventions common in public and environmental health work, although some argue that GRADE can work effectively for such evidence (Durrheim and Reingold, 2010; Schunemann et al., 2010; Rehfuess et al., 2011).

Building on this debate and the undoubted value of GRADE, work is underway to apply a modified version in the new guidelines. The new approach recognises that the majority of evidence available contributes to a set of steps in a causal chain (from intervention to emissions, to household concentrations, to exposure and finally to health outcomes), and that few studies provide evidence on

the overall impacts on health of introducing a cleaner household energy intervention. Each of these steps (or combinations) for which there is a body of evidence can be assessed separately, prior to reviewing the coherence of all available evidence in the context of the causal chain model.

The approach to assessing evidence makes an important distinction between (i) the question of whether the association is causal, for which Bradford-Hill viewpoints are used, and (ii) the strength of evidence for the intervention effect size, for which the modified GRADE is being used. Thus, although there are common aspects to these two perspectives, it is possible to have good evidence that an association between HAP exposure and a specific disease outcome is causal (and by implication that reducing exposure will reduce the risk of that disease), but rather weaker evidence as to the precise size of the effect of an intervention to reduce exposure. The modified version of GRADE being applied to these guidelines will give more weight to quasi-experimental study designs, and also take into account two other factors. These are (i) consistency of effect across varied settings and study designs, and (ii) analogous evidence such as where effects on an outcome consistent with dose are also seen with other sources of combustion pollution including ambient air pollution, second-hand smoking and active smoking (Rehfuess et al., 2011).

Decisions on how strong a recommendation should be informed are not only by the strength of the available evidence, but also by other factors, including interventions costs, values and preferences, the balance of benefits and harms, and economic assessments of costs and benefits, (See guidance from WHO GRC: http://www.who.int/kms/guidelines_review_committee/en/index.html). In the case of the new IAQG, these can have an important bearing on recommendations, and all are subject of reviews and discussion (Box 2).

The draft evidence review chapters were discussed in detail at a Guidelines Development Group (GDG) meeting, held in New Delhi, India, between 24 – 26 April 2012, where provisional recommendations were drafted. The meeting was hosted by the Indian Council of Medical Research and attended by around 30 international experts and ten Indian and international observers. Following external peer review and WHO clearance, the guidelines are expected to be published during 2013.

During the development of the guidelines, WHO is working closely with the UN Foundation's Global Alliance initiative on international standards for cookstoves. Established thus far as voluntary standards through an 'international workshop agreement' overseen by the International Organisation on Standardisation (ISO), these set tiers of performance across four dimensions (fuel efficiency, total emissions, indoor emissions safety), (See http://www.iso.org/iso/catalogue_detail?csnumber=61975). The health impacts, based on levels of emissions, relate to existing WHO air quality guidelines for $PM_{2.5}$ and carbon monoxide, and future refinement of the levels used for the tiers will be informed by the evidence compiled in the new WHO Guidelines.

Box 2: Topics for evidence reviews

- *Fuels and technologies used for cooking, heating, lighting and other uses*
- *Emissions from a representative range of fuel/technology options*
- *Modelling emissions to meet WHO air quality guidelines*
- *Air pollution and exposure levels experienced by households*
- *Health risks from HAP exposure from solid, liquid and gaseous fuels.*
- *Risks of burns and poisoning from household fuels*
- *Impacts of interventions on HAP, exposure and health in practice*
- *Key enabling and limiting factors for sustainable adoption at scale*
- *Intervention costs, financing options (including climate finance) and economic evaluation*
- *Emergency and humanitarian settings*

Table 6.2: GRADE table for assessing the quality of bodies of evidence for making recommendations

Study design	**Initial score**	**Reduce score for these factors**	**Increase score for these factors**	**Quality of evidence**
Randomised trials	High	Risk of bias: -1 (serious) -2 (very serious)	Large effect: +1 (Large) +2 (Very large)	High (++++)
		Inconsistency: -1 (serious) -2 (very serious)	Dose response: +1 (Evidence of a gradient)	Moderate (+++)
Observational studies	Low	Indirectness: -1 (serious) -2 (very serious)	All plausible residual confounding: +1 (would reduce a demonstrated effect) +1 (would suggest a spurious effect if not effect was observed)	Low (++) Very Low (+)
		Imprecision: -1 (serious) -2 (very serious		
		Publication bias: -1 Likely -2 Very likely		
Source: (Balshem et al., 2011).				

6.4 Phase II: Implementation and evaluation

Following publication of the guidelines, WHO will provide technical support to all countries to assist with implementation of the guideline recommendations. The WHO recognises, however, that, particularly for low income countries, there are a number of significant challenges in bringing about a rapid transition to clean, safe and efficient household energy for all segments of society, including:

- The people worst affected by HAP exposure and burns from traditional household fuel use are also the poorest. They are viewed by investors and markets as having low purchasing power and representing high risk, and hence in current market conditions cannot easily access improved energy technologies and fuels.
- Unlike the wider (urban) environment, the home cannot easily be subjected to regulation, so that mitigation must look to a wider range of policy instruments, including education, market development, and innovations in finance and other measures to strengthen both supply and demand for cleaner solid fuel technologies and clean fuels (Bruce et al., 2006).
- Regulation has a place however, including around product standards referred to above, and in respect of pollution of the outdoor environment to which household fuel combustion makes a surprisingly large contribution (some16% globally, substantially more in some regions) (Lim et al., 2012), and policy will need to take advantage of these opportunities.
- Those responsible for implementing mitigation policies do not have to hand routinely available data on air pollution in homes. In order to monitor progress, it will be necessary, in addition to some targeted household air pollution assessment, to develop more sophisticated survey instruments that

will provide the necessary data on stove type and use, ventilation and other factors, which can be linked to information in the WHO Guidelines (and new evidence as this becomes available) on how these relate to actual levels of household air pollution and health risk. By way of example for this last point, current routinely used survey instruments, e.g. for the DHS, do not distinguish between a traditional solid fuel stoves and advanced combustion devices with, for example, fans which have been shown to have much superior performance at least in laboratory tests (Jetter et al., 2012). As a consequence, all solid fuel homes are currently allocated the same (high) level of risk. New sets of questions are required to provide more nuanced assessments of all energy uses in the home (*e.g.* cooking, heating and lighting), and will complement existing and new field studies reporting actual levels of HAP and exposure achieved, and evidence on the related levels of health risk as is being compiled in the new WHO guidelines.

An initial 3-year implementation and evaluation phase is proposed, during which WHO plans to work closely with a number of countries, while developing guidance and tools that can be used more widely. The specific objectives of Phase II will be to (a) provide technical support in preparing an Action Plan and (b) evaluate experience within the countries in developing and starting to implement policy in order to revise and update implementation guidance. Action plans, which are being developed in collaboration with the UN Foundation Alliance, will include tools for situation assessment, methods for evaluation of alternative interventions prior to implementation, opportunities for regulation and application of the evolving ISO standards, awareness raising and market development, financing options, and strategy for monitoring and evaluation. Updating will involve incorporation of new evidence

relevant to the guidelines, in particular on field-base testing and evaluation of the performance, acceptability and adoption of cleaner technologies and fuels, and associated programmatic experience.

6.5 How will implementation work in practice in developing countries: Example of Bhutan, a potential candidate?

Bhutan is a small eastern Himalayan nation, landlocked and bordered by India and China. Some 70% of Bhutan's population of 634,982 live in rural areas (RGoB, 2006) and the rural population primarily consists of subsistence farmers. Around 70% of Bhutan's total land area is under forest cover (RGoB, 2010). People live in traditional houses made of wood, stone and mud, and cooking takes place either in kitchen attached to the outer wall of the house, or in enclosed separate kitchens.

The government of Bhutan has plans to electrify all rural houses by 2013. A record maintained with the Bhutan Electricity Authority showed that as of December 2010, a total of 68,590 rural households of the total 88,642 have been electrified (Personal Communication). However, except for lighting where electricity has become the primary source, multiple fuels are still used for cooking in rural areas. While rice is generally cooked in an electric rice cooker, curry for example, is still usually cooked over traditional stoves using wood. In general, wood is used for all the intensive cooking purposes as well as for heating. Thus the primary energy source for most Bhutanese households remains biomass, which includes firewood, woodchips and animal dung (DoE, 2009). Figure 6.2 shows the percentage of households relying on different energy sources for lighting and cooking in Bhutan and (DoE, 2005).

It is important to note that the electrified rural households were found to consume only about 25% less fuel wood than un-electrified houses, with per capita wood consumption of 10.4 and 7.8 tonnes per annum respectively for rural un-

electrified and electrified houses (DoE, 2005). This is because many household activities (in addition to cooking food) such as preparation of animal fodder, brewing, and distilling local liquor, which are energy and time intensive, require fuel wood. There is a significant difference in energy usage patterns between urban and rural areas and while urban households mostly rely on electricity and petroleum products, rural areas consume significant amount of wood, the latter accounting for 96% of the total fuel wood consumption (DoE, 2005). Bhutan consumed a total of about 725,000 tonnes of firewood in 2005, accounting for 57.7% of the total primary energy supply mix. Per capita wise this has been rated as the highest consumption per capita in the world (DoE, 2005).

Cooking using biomass is normally done only on traditional mud stoves (Figure 6.3), and even in the houses which are electrified, this type of stove is still used for cooking some dishes (*e.g.* curry), or just to lower the costs of cooking since wood is widely available and free. Without a chimney (which is the case for a large fraction of households), all of the emissions are released directly into the indoor environment. The thermal efficiency of traditional stoves was found to range from 8 – 18% (DoE, 2005).

During winter months the use of metal stoves, where wood is burnt for space heating, locally known as *bukhari* (Figure 6.4) is becoming increasingly common in both urban and rural areas. Although these stoves have chimneys, during initial lighting, the door of the combustion chamber is left open to allow sufficient air for wood to catch fire. Usually, a small amount of kerosene is used for initial lighting of the wood. This normally generates a lot of smoke, which disperses within the indoor space. Also, strong winds outside can force smoke back through the chimney into the indoor environment. It should be added that the presence of a chimney does not

completely resolve the HAP problem: while it certainly improves indoor air quality in houses where biomass combustion takes place, it leads to outdoor air pollution, and hence penetration of the combustion products back to the indoor environment. The higher the density of the houses, which is the case for larger villages or towns, the greater this problem becomes.

An effort to reduce household air pollution and pressure on forests by promoting 'improved' stoves was initiated by a number of organisations in the early 1980s. The National Women's Association of Bhutan was one of the first organizations to disseminate this program with financial and technical assistance from UNICEF and the government. In the early 1990s, this program became a national project. An estimated 14,000 to 15,000 improved stoves were installed, all free of cost in different parts of the country (Palit and Garud 2010). The latest initiative of improved stove dissemination in some of the rural areas is being carried out by a local NGO called the Tarayana Foundation in collaboration with the ALSTOM Foundation (Pindirica, 2011).

A number of qualitative reviews have been carried out to assess the benefits of the new stoves following dissemination. In two of the projects funded by the UNDP, users reported that wood consumption was reduced by about 50%, and fuel collection time lowered significantly (Ugyen and Giri 2004). Further, this report also mentions perceived health benefits reported by the people with less smoke exposure. However, several factors have limited sustained use of improved stoves. Over time, stoves became non-functional and people eventually switched back to traditional stoves. The major drawback perceived by the users was that stoves had limited heat radiation capacity for space heating and prolonged cooking hours (Ugyen and Giri, 2004; Palit and Garud, 2010). Thus, users ended up modifying the pothole dimensions in order to

increase the heat output, causing structural damage to the stoves. Further, chimneys were simply removed as some users perceived fire hazards and also because traditionally rural people use smoke to dry chilli and grain in the kitchen.

6.5.1 What are the research questions when conducting research on HAP in a developing country like Bhutan affected by household fuel combustion, and how, in the context of this, would the new WHO guidelines be implemented?

A team of academics from QUT, Brisbane, UK, Poland and Bhutan (including all the authors of this paper), aims to address this very important topic and to identify the pollution levels, their sources, drivers, the trends of personal exposure and establish effectiveness of selected interventions. Based on the knowledge built by studies conducted in many developing countries, including Lao PDR lead by one of us (LM) (Mengersen et al., 2011a, Mengersen et al., 2011b, Morawska et al., 2011) which established the existence of high exposures and related health effects, but not necessarily how to address it, we considered that following specific questions need to be addressed in a quantitative way:

1. What is the contribution of household fuel combustion to household air pollution (HAP) as well as ambient air pollution in rural Bhutan and what are its drivers?
2. How effective is control of indoor exposure to products of household combustion with exhausting them outdoors as a sole mitigation?
3. How does personal exposure vary between different age/gender groups and what drives it?
4. Can a set of appropriate interventions, shown to have low emissions in laboratory testing, be identified and their community acceptability, performance and affordability demonstrated?

5. What are the policy, technical and capacity issues that need to be addressed in implementing the new WHO Guidelines on Household Fuel combustion, and specifically in seeking to attain Guideline levels for $PM_{2.5}$ and CO?

This knowledge will allow for recommendations to combat HAP in Bhutan and contribute to global experience on the application of the new WHO Guidelines for household fuel combustion.

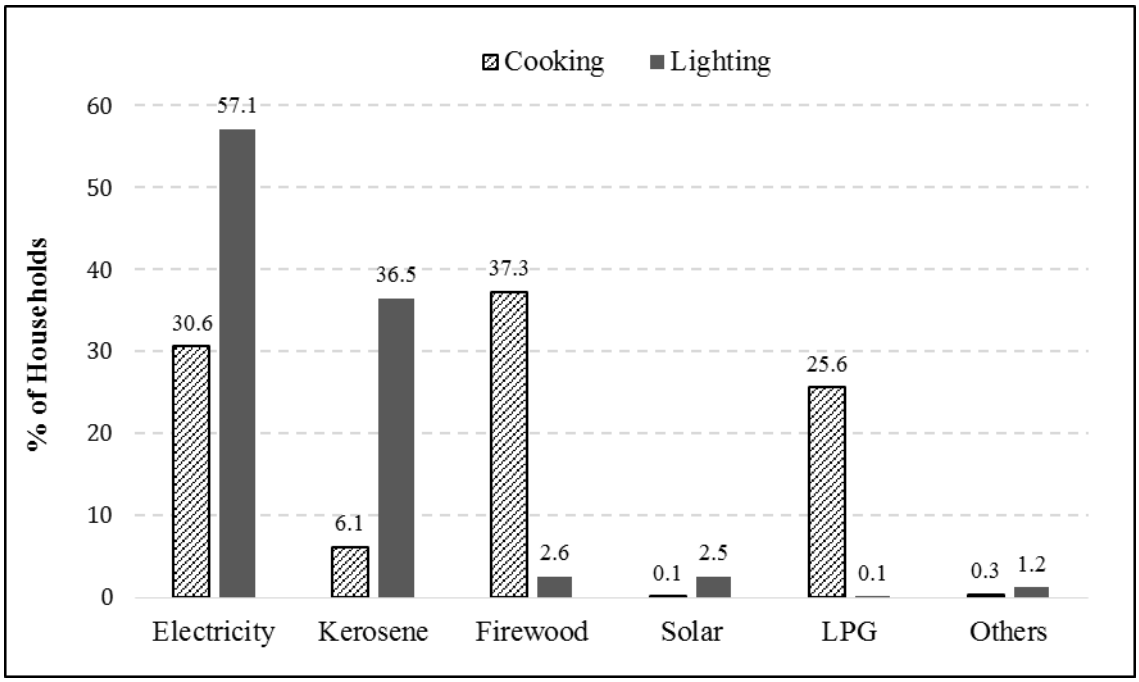

Figure 6.2: Primary energy sources for cooking and lighting in Bhutan.
Source (RGoB, 2006).

Figure 6.3: A traditional mud stove. The stove consists of one to two openings through which fuel and ash is removed, and two or three potholes, with raised lumps where pots can rest.

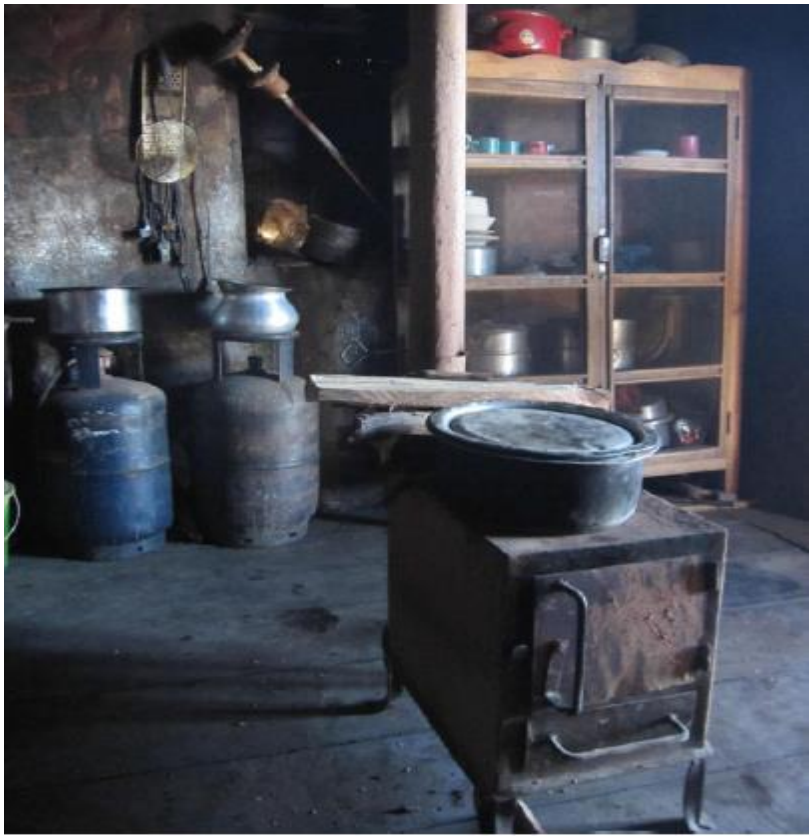

Figure 6.4: A bukhari used for space heating, made of metal and with a chimney. It can also be seen an evident fuel stacking taking place in this house.

6.6 Relevance of the WHO guidelines on household fuel combustion to developed countries?

Solid fuels, particularly wood are used in many countries of the developed world as a means of heating or creating a 'warm' ambiance, including for example US, Canada, (*e.g.* Wilton et al., 2012; Cavanagh et al., 2012; Longley et al., 2012; Noonan et al., 2011), as well as Australia and New Zealand. Cities like Launceston (Luhar et al., 2000) in Tasmania, or Christchurch and other cities in New Zealand are notoriously blanketed by smoke from indoor wood combustion during cold winter months when low wind speeds and temperature inversion worsen the situation; with ambient PM_{10} level frequently exceeding the national standards (which is 50 $\mu g/m^3$ as a 24-hour mean; the WHO guidelines in addition specify annual average of 20 $\mu g/m^3$). While measures have been taken to address the situation by in particular replacement the old inefficient stoves with more modern, lower emission devices (O'Connell et al., 2010), there is still much to be done to reach acceptable ambient air quality. There is much less quantitative information on indoor air quality and the effect the heaters have on it, and as stated by Longley et al., (2012): Studies on the impact of wood burner replacement interventions are rare, especially in terms of impact on indoor air quality and exposure, or by Wilton et al., (2012): …less is understood about the impact of these burners on indoor air quality. However, Noonan et al., (2011) have reported time trends in household and outdoor air pollution associated with replacement of woodstoves in Libby, Montana.

There is no doubt that the types of the wood-burners used in developed countries, particularly those modern and more efficient, generate far less household air pollution than for example the stoves used in Bhutan and depicted in Figures 6.3 and 6.4, and therefore interventions to improve the situation will be different. It is

likely that in developing countries the interventions will initially enable to achieve one of the intermediate targets (WHO 2005), although, for all populations, it remains to be seen whether solid fuel combustion in homes is compatible with the WHO guideline levels for $PM_{2.5}$ or whether only clean fuels can achieve this. The new WHO Guidelines on Household Fuel Combustion will be an important tool for decision-makers in setting new regulations and policy, and will focus the attention of regulatory as well as the research communities on the need for comprehensive characterisation of air quality affected by solid fuel combustion as well as of human exposure.

6.7 Conclusions

Household fuel combustion remains a major public health priority well into 21st Century, as a result of air pollution, safety, and impacts on development and climate. This is primarily an issue for developing countries, but it also presents risks for developed countries. The new WHO Guidelines will provide evidence-based information on stove/fuel performance and associated health risks for decision-making in this challenging policy area, and in helping to ensure equitable access to clean and safe household energy technologies and services. To support this, guidance on implementation, based on evidence of key enabling factors and barriers for adoption and sustained use, will also be included. Following publication of the guidelines during 2013, WHO plans to work closely with a number of countries and international partners to support and evaluate implementation, and use this experience to update guidance as countries and stakeholders get to grips with the challenges and opportunities.

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Chapter 7: Conclusion

7.1 Introduction

There is a large body of epidemiological studies linking adverse health outcomes (both short and long-term) with exposure to particles and gaseous pollutants in various microenvironments. The dominant microenvironments in terms of time spent by people are homes, schools and transportation microenvironments, particularly on roads. Therefore, air quality in these environments needs to be characterized since people are potentially exposed to high concentrations of multiple pollutants.

In general, developing countries experience severe air quality problems. Household air pollution is the major risk factor since large populations rely on solid fuels for domestic energy requirements. Residential environments therefore present extremely high concentrations of combustion products, resulting in intense exposure when people are indoors. Due to cultural and social practices, exposure is disproportionately higher for women and infants compared to other population sub-groups.

Research has also shown that the effects of air pollution are more pronounced for children than adults. The importance of education in modern society means that more children are attending schools even in developing countries. A healthy environment is crucial for learning and for the development of children. However, studies have shown that school environments present substantial air quality problems due to the presence of multiple sources in both indoor and outdoor environments.

Pollution levels along major roadways have been the focus of previous investigations due to typically high traffic emissions on roadways, leading to high exposure for road commuters and roadside residents. It is difficult to capture temporal and spatial distributions of pollution levels on roadways using conventional fixed site

monitors. Because of this problem studies have relied on mobile platforms, which can be conveniently used on roadways during real-world driving. Mobile methods have proven to be extremely robust for a wide range of air quality characterization objectives along roadways.

There are several limitations with the current literature for developing countries. Particle mass has been the main focus of air quality characterization studies in residential environments, with hardly any study reporting PN emissions from biomass stoves. A handful of studies that characterized air quality in schools and transportation microenvironments were done in major urban settings. But a significant percentage of the population live in rural areas, where air quality is affected by both local sources and long-range transport of urban and regional pollutants. There is a definite need to extend the focus of air quality investigations to rural areas in order to assess the health risks of exposure to pollutants.

The overall aim of this project was to characterize and quantify air pollution in various indoor and outdoor microenvironments in rural Bhutan. Investigation in Bhutan was felt to be necessary since there was no empirical information that could be used to support policy formulations for effective management of air pollution problems.

The first part of the project involved characterization of pollutant concentrations and emission rates during actual cooking and heating using biomass stoves in rural homes. The second part of the project characterized air quality in a school outdoor environment and evaluated the factors driving their concentrations. The third part of the project characterized air quality and primary sources for the 570 km East-West Highway.

Given the remoteness of the study location, portable real-time instruments were used for sampling: DustTraks for particle mass (PM_{10} and $PM_{2.5}$), NanoTracer for PN, and Q-Trak for CO, CO_2, temperature and relative humidity. NO_2, VOCs and carbonyls were sampled passively using Radiello dosimeters.

7.2 Principal significance of the findings

Execution of this project in a low resource country and remote location in the Himalayas presented considerable challenges. The logistical problems in getting the instrumentation from Australia, in-field maintenance and fixing of malfunctioning instruments, getting replacements, and frequent power outages and road inaccessibility to study sites have been major setbacks for the project. Despite presenting immense challenges, the project was successful and the outcomes highlighted significant air quality problems in rural Bhutan. This is in contrast to a commonly held perception among many Bhutanese that rural areas enjoy pristine air quality. The specific findings and conclusions from the research are discussed in the following sections, and schematically presented in Figure 7.1, while Table 7.1 presents an overall comparison of this study with similar studies from other developing countries.

The first part of the project presented in Chapter 4, characterized pollutant concentrations ($PM_{2.5}$, PN and CO) and emission rates from different biomass stoves during real-world operations. Investigations were done in four village houses (H1 – H4) focussing on common stove types and cooking activities. Three stage measurements, before, during, and after the activity were done in all the houses. Measurements in H1 were conducted during heating using a metal chimney stove, in H2 when cooking rice using a traditional mud stove, in H3 during fodder preparation

using a stone tripod stove, and in H4 during liquor distillation using a traditional mud stove.

The results showed extremely high concentrations of pollutants when stoves were operated, which were significantly higher than background concentrations. Further, it was found that one hour of decay after the cooking activity had ceased did not lower the concentration to background level. This was because technically the fire was not put out since unburnt wood with amber was buried in the ash to sustain the fire for the subsequent cooking activity, which extended the source emission time. Investigation also showed that indoor concentrations were influenced by neighborhood activities, where smoke from neighboring houses easily penetrated to the houses under investigation.

For PN, peak concentrations when stoves were operating exceeded 1×10^6 particles/cm^3 which was beyond the upper resolution of the instrument, resulting in a breakdown of the instrument. Therefore, open wood fire sources in residences in rural Bhutan present an extreme environment for current real-time instrumentation, indicating a need for a sample dilution system for such studies.

Activity wise, liquor distillation in H4 resulted in the highest pollutant concentrations, since this activity is generally more time and fuel intensive compared to other cooking activities. The estimated mean emission rates were also highest during liquor distillation compared to other activities. However, during startup lighting of stoves, the metal chimney stove (used for heating) presented the highest emission rates. This was due to use of kerosene and woodchips to start the fire, which emitted thick smoke, compared to the use of softer wood only for the startup lighting of other stoves. The significant findings of this study were that stove emission rates were several orders of magnitude higher than the WHO recommended emission rate

targets for household stoves, and emission rates reported for gas and electric stoves in the literature. Both pollutant concentrations and emission were more influenced by the type of activities than the type of stoves used. These evidences clearly suggest a need for systematic intervention for reduction of HAP in rural Bhutan. While a shift in energy sources towards cleaner fuels would offer the greatest benefit, there are logistical and economic challenges for rural population in Bhutan. The costs associated with clean fuels and limited accessibility to LPG and kerosene will compel people to rely on wood, which is plentiful and can be collected free. Dissemination of improved cookstoves with proper education and adaptions focused on cultural, social and behavioral patterns would significantly reduce the levels of exposure and its health burden.

Since classroom air quality is largely influenced by outdoor sources, the second part of the project (presented in Chapter 5), investigated outdoor air quality in a school environment and also evaluated the factors driving pollutant concentrations. Measurements for multiple pollutants (PM_{10}, $PM_{2.5}$, CO, NO_2, VOC, and carbonyls) were conducted for 16 weeks, spanning wet and dry seasons.

Analysis of daily time-series concentrations showed distinct peaks, coinciding with the school's morning cleaning time. A more pronounced peak of PM_{10} than $PM_{2.5}$ or PN, indicted greater impact of re-suspension of coarse particles from the school ground during cleaning time. Daily mean concentrations exceeded the WHO 24-hours guidelines on 12% and 18% of the measurement days for PM_{10} and $PM_{2.5}$, respectively. The majority of the exceedance occurred during dry season days. Overall, dry season presented higher pollutant concentrations than wet season for majority of the pollutants. The seasonally segregated mean concentrations of both PM_{10} and $PM_{2.5}$ presented higher values than the WHO annual guidelines for both wet

and dry seasons. Mean CO concentrations were below the detection limit of the instrument throughout the study period. Only wet season results were available for PN due to instrument malfunction, and they presented a comparable mean concentration with the worldwide ambient clean background level.

The highest pollutant concentrations were found on four days during which open wood fire cooking was done in the school ground, when the school was conducting a sports competition and a prayer ceremony. On those four days children were actively engaged in outdoor activities, which potentially exposed them to high concentrations of biomass combustion products. Since these events were observed each year, making alternative cooking plans or a shift towards clean energy source would minimise the health risks that children were subjected to.

Apart from sources within the school, it was found that pollutant concentrations were associated with combustion activities in the community surrounding the school. This study was timely since the school and sounding locations have been earmarked for township development. Therefore, this study will provide a useful comparison for changes in air quality due to urban activities in the future.

The third part of the project presented in Chapter 6, characterized on-road air quality and contributions from the principal sources on the East-West Highway in Bhutan. The road under investigation, 570 km in length is an economic lifeline for all the eastern districts and some of the districts in the central and western Bhutan. The road is characterized by the presence of a number of combustion and non-combustion sources.

A total of six measurements of $PM_{10,}$ PN and CO were conducted using vehicle based mobile methods for the entire length of the road. For the purposes of the study, the entire road length has been divided into five segments (R1 – R5),

considering the road length between two district towns as a single road segment. The contributions from different on-road and proximate sources were assessed based on interception of sources by the research vehicle, and by accounting for time and location of the sources. Additionally, the first ever traffic survey for Bhutan was conducted for the EWH during the campaign.

The results showed the highest traffic density for R5 compared to other road segments, which is the final segment of the EWH, connecting the capital city to other districts. Light passenger vehicles constituted 75% of the traffic volume followed by heavy vehicles 14%, medium vehicles 8% and two-wheeler with 2%. Besides vehicles, the dominant sources were road works, unpaved roads, and roadside combustion activities. While pollutant concentrations varied widely between the road segments, the highest concentrations were observed for R5, for all the pollutants investigated. Peak concentrations of PM_{10} were always observed for a section of R4 (distance of 27 km) throughout the campaign. The road in question had major road works being undertaken at the time of the campaign and the entire 27 km distance was unpaved due to ongoing works. Between the districts towns, T2 and T4 presented the highest concentrations compared to T1 and T3. This points to the impact of the relative size of towns and associated urban activities. In terms of source contributions, substantial variability in the dominant source was observed between different road segments. However, the overall highest contributions were made by the unpaved road for PM_{10} and vehicles for PN and CO. The findings of this study highlighted high exposure levels for road commuters, roadside residents and those working along the EWH.

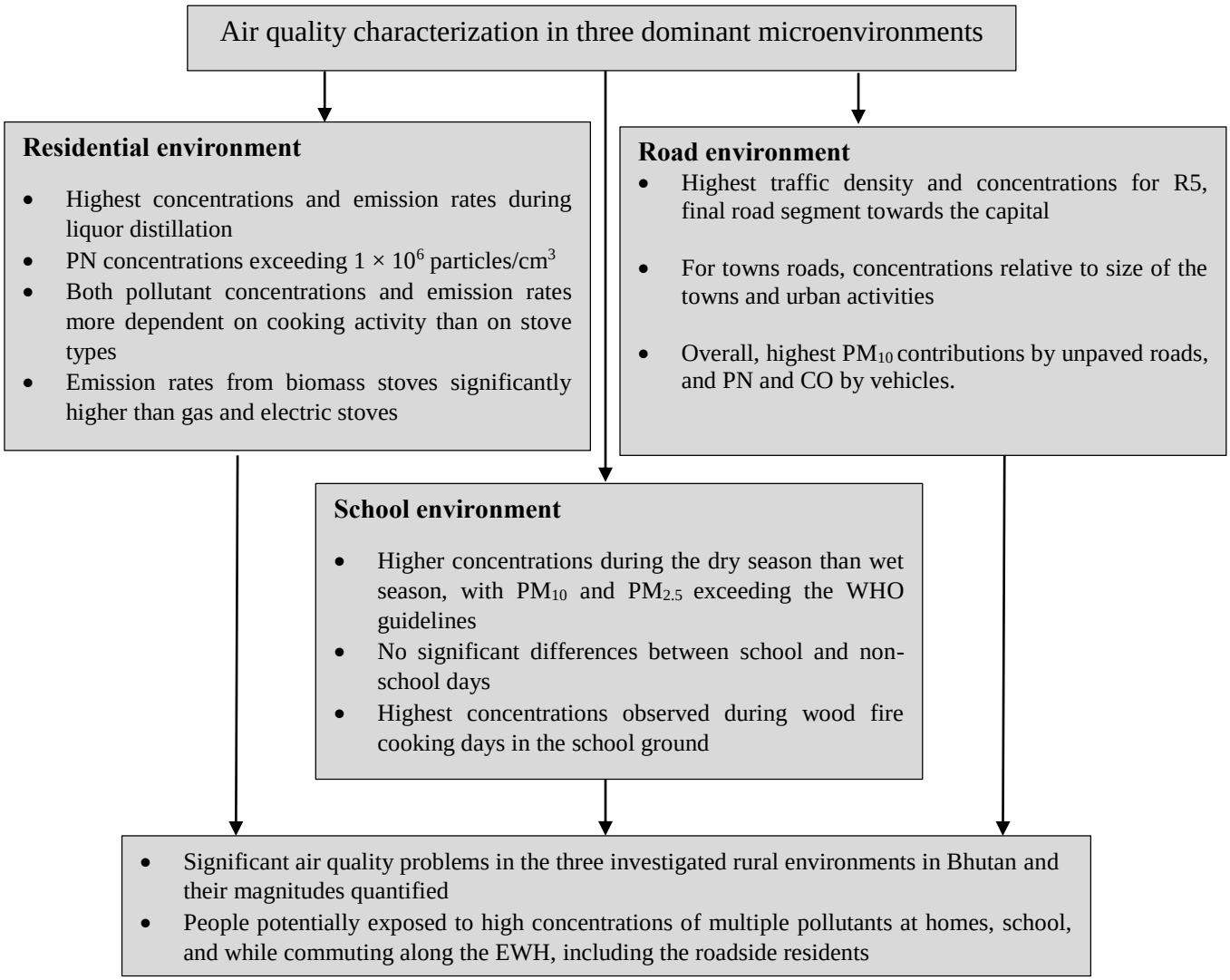

Figure 7.1: Schematic presentation of summary of the main findings

Table 7.1: Comparison of findings of this study with similar studies in developing countries**

Study	Microenvironment	Rural/Urban	Pollutant	Instrumentation	Fuel	Mean concentration	Sampling details
This Study, Kanglung, Bhutan	Residential	Rural	$PM_{2.5}$ PN CO	DustTrak, TSI. NanoTracer, Philips, Aerasense. Q-Trak, TSI.	Wood	1510 – 2550 µg/m^3 2.03 × 10^3 part/cm^3 3.56 – 14.01 ppm	Cooking time, 34 minutes to 2 hours, and 5 hours for heating
(Ellegard, 1996), Maputo, Mozambique	Residential	Sub-urban	PM<7.1µm	Gil-Air SC Pump, Gilian Instrument Corp.	Wood	1200 µg/m^3	Average cooking time: 1.5 hours
(Lodhi and Zain-al-Abdin, 1999), Malaysia	Residential	Rural	SPM CO	Respirable aerosol mass monitor. SPE carbon monoxide detector.	Wood	300 µg/m^3 2.9 ppm	Cooking hours
(Naeher et al., 2000), Xela, Guatemala	Residential	Rural	$PM_{2.5}$ CO	SKC pump, MIE cyclone. Draeger CO electrochemical sensor	Wood	5310 µg/m^3 22.9 ppm	Cooking time
(Balakrishnan et al., 2002), Tamil Nadu, India	Residential	Rural	PM_4	Battery-operated, constant-flow pumps supplied by SKC Inc.	Wood	847 µg/m^3	Cooking time
This study, Kanglung, Bhutan	School outdoor	Rural	PM_{10} $PM_{2.5}$ PN CO NO_2 Formaldehyde	DustTrak, TSI. DustTrak, TSI. NanoTracer, Philips, Aerasense. Q-Trak, TSI. Radiello passive dosimeter.		27^a, 36^b µg/m^3 13^a, 29^b µg/m^3 2.56 × 10^{3a} part/cm^3 >DL 0.07^a, 1.73^b µg/m^3 2.37^a, 2.41^b µg/m^3	24 hours for particles and CO, 16 weeks of monitoring 7 days for NO_2 and formaldehyde
(Lee and Chang, 2000), Hong Kong	School indoor	Urban	PM_{10} NO_2 Formaldehyde	Dust Trak, TSI. Thermo Electron (Model 42) Chemiluminescence NOx Analyser. SKC formaldehyde monitoring kit.		21 – 617 µg/m^3 31 – 67 µg/m^3 > DL - 27 µg/m^3	Continuous monitoring Off-school hours 24 hours

Table 7.1: Continued

Study	Microenvironment	Rural/Urban	Pollutants	Instrumentation	Fuel	Mean concentration	Sampling details
(Zhao et al., 2008), Taiyuan, Shanxi Province, China	School outdoor	Urban	NO_2 Formaldehyde	Diffusion samplers, IVL Swedish Environmental Research Institute Ltd. SKC UME, (Eighty Four, PA,USA).		52.3 µg/m^3 5.8 µg/m^3	7 days
(Goyal and Khare, 2009), Delhi, India	School outdoor	Urban	PM_{10} $PM_{2.5}$	GRIMM make environmental dust monitor, Model-107		531.1[c], 162.4[e] 244.7[c], 37.7[e]	6 hour average of yearlong monitoring
(Chithra and Shiva Nagendra, 2012), Chennai, India	School indoor	Urban	PM_{10} $PM_{2.5}$ CO	GRIMM environmental dust monitor Model 1.107. Indoor Air Quality Meter, TSI.		149[c], 95[d] µg/m^3 61[c], 32[d] µg/m^3 0.10[c], 0.11[d] ppm	Daily average, 60 days measurements
This study, East-West Highway, 570 km, Bhutan	On-road, mobile measurements	Rural	PM_{10} PN CO	DustTrak, TSI. NanoTracer, Philips, Aerasense. Q-Trak, TSI.		49 – 148 µg/m^3 1.31×10^4 – 5.74×10^4 part/cm^3 0.06 – 0.19 ppm	6 measurements for the entire road length
(Westerdahl et al., 2009), 4th Ring Road & Balding Express Way, 26.6 km, Beijing	On-road, mobile measurements	Urban	PN	FMPS, TSI, Model 3019		8.17×10^4 part/cm^3 (Median)	2 weeks measurements
(Apte et al., 2011), 19.5 km mixed route, New Delhi, India	*On-road in-vehicle, mobile measurements	Urban	PN	TSI portable CPC, Model 3007		2.90×10^5 part/cm^3	31 days, 62 trips, 160 measurements

DL: Detection Limit, [a]: Wet season, [b]: Dry season, [c]: Winter, [d]: Summer, [e]: Non-winter, *Measurements were done inside an auto-rickshaw, a semi enclosed three-wheeled vehicle, where in-vehicle concentrations actually reflects on-road levels.

**: Notwithstanding the differences in instrumentation, measurement protocols and quality control criteria used, high concentrations of particulate matter for the investigated microenvironments in the study are consistent with similar studies from other developing countries.

In summary, this research focused on characterization and quantification of multiple pollutants in three dominant microenvironments (homes, school outdoor and on-road) in terms of time spent by people. The research was focused on the setting of Bhutan since no quantitative air quality studies have been conducted there till date. Due to the lack of standard reference instruments in Bhutan, on-site calibration of the instruments used in the study could not be conducted. Thus all calibrations were done at the International Laboratory for Air Quality and Health (ILAQH), Queensland University of Technology, Brisbane, Australia. Notwithstanding this limitation, the overall findings highlighted significant air quality problems in the microenvironments investigated originating from a diversity of sources, indicating that people were potentially exposed to high concentrations of multiple pollutants. The outcomes will be of significance not only for supporting policy formulation in Bhutan, but also in other developing countries, particularly for Himalayan nations given the shared geographical zones and common cultural and social practices in the entire Himalayan region. The current Bhutan Air Quality Standards for particulate matter (see Table 2.1) are set at much higher values than what has been adopted in many developing countries. In light of the findings of this study, a revision of current values is recommended.

7.3 Directions for future research

While quantification and characterization of pollution levels in various microenvironments are important, it is exposure to pollutants that drives health impacts. The future studies should focus on time-activity patterns and magnitude of exposure, especially for women and children who spend the majority of their time indoors and close to a fire source. Likewise, children's exposure in the school environment is not known from the current study, warranting the attention of future

studies. Similarly, exposure assessments for road commuters and for people living and working along roadsides will be of significance. Additionally, specific health effects of exposure in various microenvironments would shed light on the actual health burden faced by Bhutan in relation to air quality problems.

The current study in village homes observed several potential social, cultural and behavioral patterns of occupants that could potentially elevate exposure to household combustion products. Future study focussing on the role of traditional household practices as influential parameters of exposure, and measures suggesting minimization of exposure to HAP will be of significance.

Due to the limited number of instruments the current study could not conduct simultaneous indoor and outdoor measurements. In relation to household fuel combustion, since it also contributes to outdoor pollution, the immediate outdoor surrounding can experience high pollution levels. Because people spend time outdoors, outdoor air quality in villages should be characterized. Simultaneous indoor and outdoor measurements will be critical to quantify the magnitude of exposure in indoor and outdoor environments. Likewise, since the current project focused only on the school outdoor environment, the contribution from indoor sources were not known. While many studies have characterized simultaneous indoor-outdoor air quality in schools, these findings cannot be adapted to Bhutanese environment given differences in structure types, building materials, furniture, ventilation systems, and indoor activities conducted.

The current road study focused only on on-road pollution levels using a mobile method. Due to limited instrumentation, the study could not conduct any ambient and fixed site monitoring along the EWH. Future on-road studies should consider fixed site monitoring to account for the temporal variations along roadways.

Finally, given complicated terrain and geography in Bhutan, in-situ air quality measurements combined with meteorology measurements and modeling work could provide better information about the impacts and relationships between sources and receptors.

7.4 References

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