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Measurements of TSP, PM_{10}, $PM_{2.5}$, BC, and PM chemical composition from an urban residential location in Nepal

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ABSTRACT

Particulate matter ($PM_{2.5}$, PM_{10}, and TSP), black carbon (BC), and PM constituent components were measured at a location in an urban residential neighborhood of Kathmandu Valley, Nepal. $PM_{2.5}$, TSP, and BC were measured during winter, and $PM_{2.5}$, PM_{10}, and their chemical composition was measured during the summer monsoon periods in 2014. Both indoor and outdoor measurements were collected during the winter season. There was a distinct seasonal profile of $PM_{2.5}$ concentration, with the 24-hour mean larger during winter (76 ± 18 μg/m^3) than monsoon (21 ± 8 μg/m^3). This site was located approximately 15 m above the street, but was still influenced by roadway combustion sources indicated by large BC concentration (15 ± 3 μgC/m^3 during winter) and large rush hour PM and BC peaks. Two rush hour peaks of PM and BC were observed during morning and evening. Indoor and outdoor $PM_{2.5}$ and BC concentration measured during winter were similar suggesting the heavy influence of outdoor traffic activities on such indoor environments. Mean 24-hour TSP during winter and PM_{10} during monsoon was 109 μg/m^3 and 34 μg/m^3, respectively. $PM_{2.5}$ accounted for 75% of TSP during winter and 61% of PM_{10} during monsoon indicating a high degree of influence by $PM_{2.5}$ sources. Sulfate, ammonium, and calcium were the dominant components of water-soluble ions, and silica, iron, aluminum, and barium were the major elements in both PM_{10} and $PM_{2.5}$ during monsoon indicating the most important emission sources as traffic emissions and road/soil/construction-related dust.

1. Introduction

Particulate pollution is one of the major environmental burdens for the residents of the Kathmandu Valley, and a number of studies have confirmed that the region is subjected to highly degraded air quality (ICIMOD, 2007; Aryal et al., 2008, 2009; Gurung and Bell, 2012; Chen et al., 2015; Kim et al., 2015; Shakya et al., 2016). The Nepal government has promulgated standards of 230, 120, and 40 μg/m^3 for 24-hour average of TSP (total suspended particles), PM_{10} (particles with aerodynamic diameter smaller than 10 μm), and $PM_{2.5}$ (particles with aerodynamic diameter smaller than 2.5 μm), respectively (National Ambient Air Quality Standards, 2012). Despite these standards, there was no continuous regulatory monitoring of air quality in Kathmandu Valley during our sampling period, and the information is lacking on the concentrations of these three fractions of PM sizes from the Kathmandu Valley. This study collected simultaneous measurements of different size fractions of PM, and the contribution of $PM_{2.5}$ to PM_{10} and TSP at an urban location in Kathmandu Valley.

Like many developing nations in South Asia, Nepal has a high rate of population growth and a concurrently rapid increase in vehicle registration in Kathmandu, the capital of Nepal. This is thought to lead to increases in air pollution in the valley (ICIMOD, 2007), as vehicle emissions are one of the main sources of particulate matter in the Kathmandu Valley (Shakya et al., 2010; Putero et al., 2015; Kim et al., 2015). Rapid and unplanned urbanization, Kathmandu-centric development, operation of old and poorly maintained vehicles, poor traffic management, and biomass/refuse burning also likely contribute to degraded air quality in the valley (Kim et al., 2015). Restriction of air flow from the valley by the

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surrounding mountains also exacerbates air pollution, and thus 3.5 million valley residents are exposed to high levels of air pollution with a serious threat to their well-being (ICIMOD, 2007). Our recent work (Shakya et al., 2016) found a significant reduction in lung function in a cohort of Kathmandu Valley traffic police that were exposed to high levels of $PM_{2.5}$ and BC.

$PM_{2.5}$ and PM_{10} daily-integrated concentrations have been shown in the range of 100s of $\mu g/m^3$ across a few recent studies in Nepal (Aryal et al., 2009; Gurung and Bell, 2012; Shakya et al., 2016). By using passive sampling techniques, Kiros et al. (2016) recently demonstrated higher air pollution in urban locations compared to suburban and rural locations in Kathmandu Valley. Aryal et al. (2008) found that annual mean PM_{10} was larger by a factor of 1.3–4.8 at busy traffic roads compared to rural, urban and suburban areas in the valley during 2003–2007. To our knowledge, no studies have investigated residential exposure levels of different PM fractions and its chemical constituents, and black carbon (BC) near busy roadways in Kathmandu Valley.

Residents living near major roads in the Kathmandu Valley are continuously exposed to enhanced levels of particulate matter and other air pollutants, and such exposures are likely to have adverse health effects on residents who live in these locations. Because we spend much of our day at indoor locations (Brasche and Bischof, 2005), monitoring of air pollution at roadside residences is essential to better assess the health effects of such pollutants as well as to design air pollution management strategies in urban areas. In a location like Kathmandu Valley, the lack of comprehensive monitoring data also makes it difficult to assess if the regional air quality in the valley complies with the national ambient air quality standards, which further occludes our observation of residential aerosol climatology.

The objectives of this study are to investigate seasonality of air pollution characteristics at a residence near busy roads and to analyze the influence of traffic- and dust-related emission sources in PM levels. In this study, we monitored the levels of three different fractions of PM: $PM_{2.5}$, PM_{10}, and TSP, and BC from an outdoor environment at a residential location in 2014. Measurement of $PM_{2.5}$ from an indoor environment, and the chemical composition of $PM_{2.5}$ and PM_{10} size fractions was also made.

2. Methods

Particulate matter and BC samples were collected at an urban residence located approximately 15 m above a major roadway in Pulchowk, Lalitpur. Lalitpur is one of the three districts (the other two are Kathmandu and Bhaktapur) of Kathmandu Valley.

Monitoring was conducted from an open and uncovered balcony located on the fifth floor of a six-story building in the Pulchowk neighborhood. A conductive inlet tube was used to draw the samples from outdoor into the sampling equipment that was housed inside the room. The balcony was oriented towards the west, and the building was located along one of the major public-transport routes linking Kathmandu with the Lalitpur district. Buildings in this area are typically 4-6 stories high, with commercial uses (commercial, restaurants, light industrial services) at the ground level, and residential units on the upper floors. Most roads are paved with asphalt in this neighborhood. There is a four lane road where this house is located.

Sampling was conducted during the late winter/early spring (February 16 to March 25, 2014) and during the monsoon (August 13 – September 1, 2014) periods. The sampling period was selected to investigate the seasonality of PM pollution in this area. In this manuscript, the late winter and early spring sampling is referred as the 'winter' period. A nephelometer (pDR-1500, Thermo Fisher Scientific, Massachusetts, USA) and a microaethalometer (AE51, Aeth Labs, California, USA) were used for monitoring particulate matter and BC, respectively. The nephelometer uses optimized light scattering for measuring real-time particulate concentrations while the microaethalometer uses the rate of change in absorption of transmitted light at 880 nm to measure black carbon concentrations. Detection limit and precision of pDR-1500 are 1 $\mu g/m^3$ and 0.2%, respectively. Manufacturer-provided impactors or cyclones were used for measuring PM of different size fractions, such as TSP, PM_{10} and $PM_{2.5}$. Semicontinuous measurements of PM and BC were collected at an interval of 5-minutes. The pDR-1500 back filters were exchanged and archived for later chemical analysis every 24 h, and the microaethelometer filter strip was changed every 12–36 h depending on sampling loading and reported filter attenuation of the instrument. Both instruments give error codes when problems are detected (e.g. a filter is exhausted, when flow is low, and for other errors). Such erroneous data were removed from further analysis. Each of the cyclones (for pDR and microaethalometer) were cleaned with deionized water before deployment at each phase of the study. The pDR was zeroed at the beginning of each phase of the study, and they used the manufacturer factory calibration. A summary of measurements is given in Table S1.

2.1. Winter sampling

During the winter sampling, simultaneous levels of TSP, $PM_{2.5}$, and BC were monitored at the outdoor from the balcony of the fifth floor of the residential house. $PM_{2.5}$ (March 13–21, 2014) and BC (February 16 – March 21, 2014) were also measured concurrently at the indoor of the room attached to the balcony at the same floor. Chemical composition were not analyzed from winter samples.

2.2. Monsoon sampling

During the monsoon period, outdoor $PM_{2.5}$ and PM_{10} were monitored from the balcony. There was no measurement of BC, TSP, or indoor levels during the monsoon.

Chemical composition (elements and water-soluble inorganic ions) were analyzed from $PM_{2.5}$ and PM_{10}. Two pDR-1500 samplers were operated at the same time to collect two size fractions of ambient aerosol. A 37 mm polytetrafluoroethylene filter was inserted into each pDR-1500 to collect the particulate matter during monsoon, and sampled aerosol at a low flow rate (<2 L per minute). Two filters were collected daily, and samplers operated for two weeks concurrently for both $PM_{2.5}$ and PM_{10} size fractions (total number of samples = 29; Table S1). Used filters were stored inside petri dishes and shipped to the laboratory at the University of Massachusetts, Amherst for determination of elements and ions.

2.2.1. Elements and ion analysis

The filters were analyzed for trace elements by using energy dispersive X-ray fluorescence (XRF) spectroscopy (Quant'x, Thermo Fisher Scientific, Massachusetts, USA). The XRF was calibrated by using thin film single element standards for each of the measured elements. In each batch of 10 filter analyses, a mixed external standard was also analyzed. The precision of five mixed standard elements run during the analysis were within 0.35–3.1% (as relative standard deviation). The detection limits ranged from 0.1 to 20 ng/cm^2 for the elements (Table 2) except for sodium (Na) and tin (Sn) whose detection limits range from 39 to 75 ng/cm^2. The method follows the US EPA Compendium Method (IO3.3) for inorganic metal speciation.

After XRF analysis, each filter sample was extracted by adding 5 μL of ethanol and 25 ml of high purity water. The solution was sonicated for 2 h and was subsequently analyzed for water-soluble ions by using ion chromatography (ICS1100 with autosampler,

Thermo Fisher Scientific, Massachusetts, USA). Post-hoc chemical composition analysis of $PM_{2.5}$ and PM_{10} was conducted on only the filters collected during monsoon sampling period. No winter aerosol chemical composition was available from this location though semi continuous data were collected.

2.2.2. Meteorology

Archived meteorology data (The Weather Underground, http://www.wunderground.com) was collected from Tribhuvan International Airport, Kathmandu, a site about 7 km from our sampling location. The mean temperature during the winter and monsoon sampling periods was 14 °C and 22 °C, respectively. Relative humidity during winter and monsoon was 64% and 77%, respectively, and there was a total precipitation of 5.7 cm and 35.8 cm during winter and monsoon, respectively.

3. Results and discussion

3.1. TSP, PM_{10}, $PM_{2.5}$, and BC levels

Measurements of $PM_{2.5}$ and TSP from an outdoor environment, and $PM_{2.5}$ from an indoor environment at a roadside residential location in Pulchowk are presented in Fig. 1. $PM_{2.5}$ frequently exceeded 40 μg/m^3 for 5-minute averages in winter 2014 for both outdoor (90% of total measurements) and indoor (79% of total measurements). The 5-minute $PM_{2.5}$ levels exceeded 40 μg/m^3 only 9% of time during monsoon. Recorded hourly average concentrations of TSP were as high as 250 μg/m^3 and as high as 160 μg/m^3 for $PM_{2.5}$ (Fig. 2). One exception to these levels occurs on March 19, 2014, which is discussed later (Section 3.4). $PM_{2.5}$ and PM_{10} measured from this outdoor environment at the same location were mostly below 50 μg/m^3 during the monsoon season in August. In winter, when higher concentrations of PM were recorded, daily average concentration for $PM_{2.5}$ from indoor and outdoor environments, and TSP from outdoor environment was 67 ± 19, 76 ± 18 and 109 ± 39 μg/m^3, respectively.

Not surprisingly, 24-hour $PM_{2.5}$ mean concentration was highly variable depending on the seasons. On average, 24-hour $PM_{2.5}$ concentrations were larger during the winter (76 ± 18 μg/m^3) than during the monsoon (21 ± 8 μg/m^3). An independent *t*-test showed there was a significant difference between $PM_{2.5}$ measurements during the two seasons. Substantial daily rainfall occurred during the monsoon (total precipitation of 35.8 cm) while rain events (>1 cm each) were observed just twice over the winter sampling and previous studies have reported reduced concentrations of PM_{10} (Aryal et al., 2008; Majumder et al., 2012) and $PM_{2.5}$ (Aryal et al., 2009) during the monsoon compared to non-monsoon seasons in the Kathmandu Valley. Such a decrease in PM concentrations due to rainfall has been observed in several studies (Glavas et al., 2008; Hieu and Lee, 2010; Lee and Hieu, 2011), and previous studies have reported reduced concentrations of PM_{10} (Aryal et al., 2008; Majumder et al., 2012) and $PM_{2.5}$ (Aryal et al., 2009) during the monsoon compared to non-monsoon seasons in the Kathmandu Valley.

Study-averaged TSP measured during this study was 109 ± 39 μg/m^3, which was sharply lower compared to work by others in a different urban location in the valley (Table 1; Chen et al., 2015). During the monsoon, daily average PM_{10} and $PM_{2.5}$ were measured as 34 ± 10 and 21 ± 8 μg/m^3, respectively. $PM_{2.5}$ found in this study is comparable to previous studies conducted in urban locations of Kathmandu Valley (Table 1). PM_{10} values in this study were lower than previous studies at urban locations, though most other studies rely on wintertime measurements, and our work was under monsoonal conditions.

BC measured during the winter in this study (15 ± 11 μgC/m^3) was higher than the previous studies from other locations in Kathmandu Valley, such as an urban site in Paknajole (11.6 ± 10.7 μgC/m^3; Putero et al., 2015), and a suburban site located about 1 km away from our site (8.4 ± 5.1 μgC/m^3; Sharma et al., 2012). Shakya et al. (2010) measured elemental carbon (EC) concentration of 4.5 ± 1.2 μgC/m^3 during winter at another urban site in Kathmandu. The observed high BC concentrations compared to previous studies might be because of the close proximity to direct traffic on busy roads. Measurements on previous studies were taken from rooftops, while in this study, the samples were collected from the balcony facing the roads of busy traffic. This suggests that our sampling site, despite the elevation, was still heavily influenced by primary emissions from vehicular activity.

$PM_{2.5}$ contributes about 75 ± 6% of outdoor TSP mass in winter, and it contributes about 61 ± 12% PM_{10} mass in monsoon. The high $PM_{2.5}$/TSP and $PM_{2.5}/PM_{10}$ ratios suggest a strong influence from combustion sources, such as traffic exhaust near roadways or from secondary sources of aerosols. The $PM_{2.5}/PM_{10}$ ratio from this roadside residence was consistent with previous observations of $PM_{2.5}/PM_{10}$ ratio (0.53–0.80) from roadside locations in Paris (Ruellan and Cachier, 2001) and traffic sampling site in Taipei,

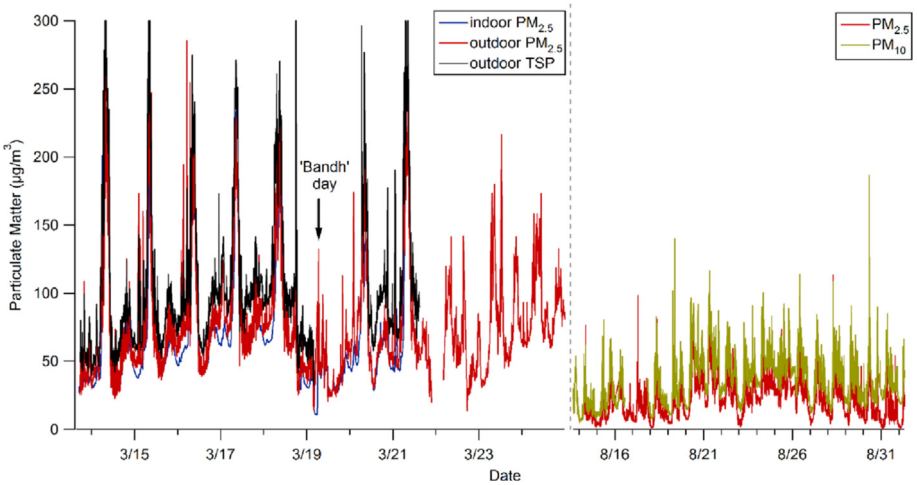

Fig. 1. 5-minute average measurements of TSP and $PM_{2.5}$ during winter season, and PM_{10} and $PM_{2.5}$ during monsoon season at a residential location in Pulchowk during 2014. Indoor $PM_{2.5}$ and outdoor TSP measurements were discontinued after 3/22/2014. There was a rain event (2.7 cm) on 3/22/2014.

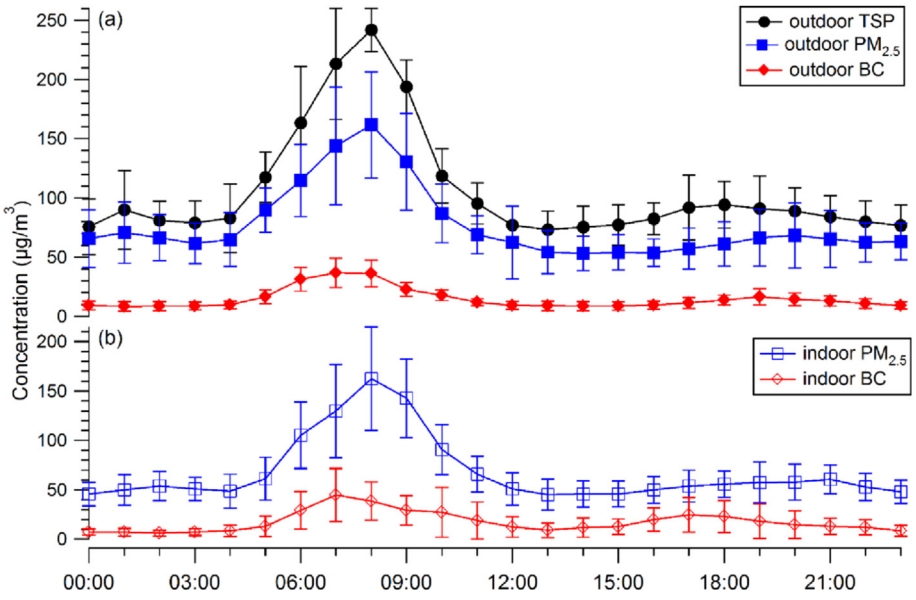

Fig. 2. Hourly variation of TSP, $PM_{2.5}$ and BC from (a) outdoor and (b) indoor environment during winter season in 2014.

Table 1
Recent ambient PM measurements in Kathmandu Valley.

Location	Site types	Duration	PM size fraction	Concentration (μg/m^3)	References
Pulchowk, Lalitpur	Roadside	2/16/2014–3/25/2014	TSP	109 ± 39	This study
	Residential	2/16/2014–3/25/2014	$PM_{2.5}$	76 ± 18	
		8/13/2014–9/1/2014	PM_{10}	34 ± 10	
		8/13/2014–9/1/2014	$PM_{2.5}$	21 ± 8	
Bode, Bhaktapur	Urban	4/2013–3/2014	TSP	199 ± 124	Chen et al., 2015
Paknajole, Kathmandu	Urban	2/1/2013–1/31/2014	PM_{10}	169 ± 113	Putero et al., 2015
		2/1/2013–4/30/2013	$PM_{2.5}$	195 ± 83	
Bode, Bhaktapur	Urban	12/2012–02/2013	PM_{10}	128	Kim et al., 2015
Ten sites	Mixed	02/2008–01/2009	PM_{10}	454–891	Majumder et al., 2012
Thamel, Kathmandu	Urban	2006–2007	$PM_{2.5}$	30–90	Aryal et al., 2009
Godavari, Lalitpur	Rural	1/1/2006–12/31/2006	$PM_{2.5}$	26 ± 19	Stone et al., 2010
Six sites	Mixed	03/2003–02/2006	PM_{10}	50–230	Aryal et al., 2008
Nagarkot, Bhaktapur	Rural	12/1998–10/2000	PM_{10}	11–81	Carrico et al., 2003
			$PM_{2.5}$	8–59	

Taiwan (Li and Lin, 2002). The $PM_{2.5}/PM_{10}$ ratio of 0.77 ± 0.19 was also recorded at another location at high elevation showing the dominance of $PM_{2.5}$ in Kathmandu Valley (Carrico et al., 2003). A $PM_{2.5}/PM_{10}$ ratio larger than 0.60 was recorded in Lalitpur and Kathmandu in 2003–2004 suggesting the influence from vehicle emissions (ICIMOD, 2007), and that this ratio has apparently increased over time. Though $PM_{2.5}$ fraction is compared against TSP during winter and against PM_{10} during monsoon, it seems to suggest a decrease in the amount of fine fraction aerosol during the monsoon, or possibly a relative increase in larger particle concentration. Putero et al. (2015) found the highest PM_1/PM_{10} ratio (0.39) during pre-monsoon season compared to other seasons at an urban location in Kathmandu Valley. Their overall PM_1/PM_{10} ratio (0.29 ± 0.10) was about half the $PM_{2.5}/PM_{10}$ ratio observed in this study. This suggests that our location has less influence from dust sources compared to their site.

3.2. Hourly variation of TSP, PM_{10}, $PM_{2.5}$, and BC

There was a strong diurnal variability for TSP, $PM_{2.5}$, and BC with a large spike in concentration occurring around 7–9 AM and a small peak occurring around 5–7 PM during the winter (Fig. 2). Such rush-hour peaks were observed for both PM and BC levels. An exception to this association occurs during nighttime, when PM increases slightly, but BC appears to remain generally constant throughout the night. These nighttime peaks in $PM_{2.5}$ were absent during the monsoon (Fig. 2). Nighttime PM growth during winter might be attributed to either non-combustion sources or enhanced coagulation and condensation processes of traffic exhaust particles coinciding with decreased mixing layer height, lower temperature and higher relative humidity (Jamriska et al., 2008; Perez et al., 2010). Diurnal variability also exists during the measurements from monsoon; the appearance of a second rush-hour PM peak is more pronounced in monsoon season compared to winter season (Figs. 2 and 3).

When comparison of the hourly average of $PM_{2.5}$ for weekdays (Sunday - Friday) and weekends (i.e. Saturday) was made at this location, there was an overall decrease in $PM_{2.5}$ concentration during the weekends compared to weekdays (Fig. S1), which is likely to be caused by a decrease in road traffic on the weekends. This was true for both winter and monsoon seasons, but the decrease for daily average of $PM_{2.5}$ on weekends during monsoon (21%) was larger than during winter (13%). It should be noted that Saturday is the only day considered as a weekend in Nepal, where Sunday is typically a routine work day. Though the concentrations were smaller on Saturday compared to other days of the week, the

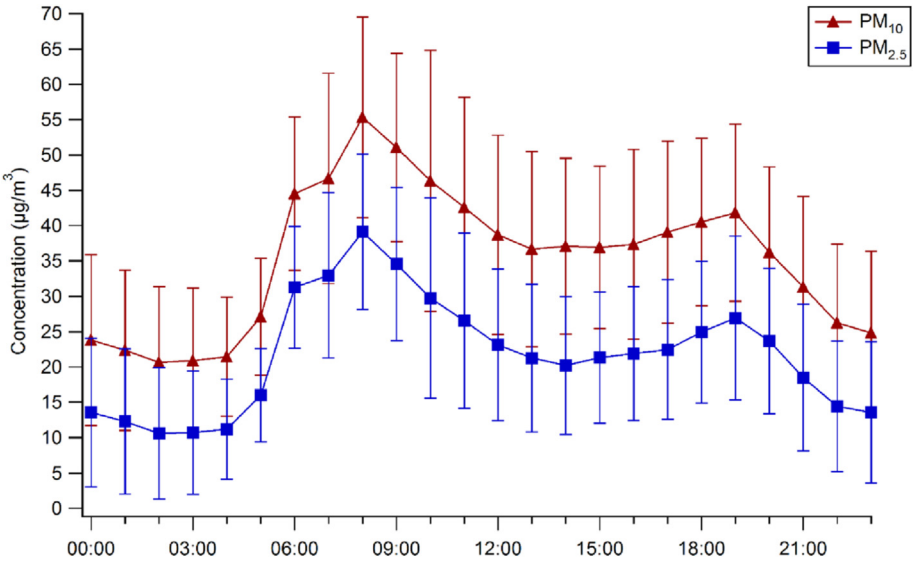

Fig. 3. Hourly variation of $PM_{2.5}$ and PM_{10} during monsoon season in 2014.

hourly trend was similar during both weekdays and weekends. Rush hour peaks of $PM_{2.5}$ were still similar on both weekdays and weekends. But during the monsoon, the peak appeared an hour earlier on weekends compared to weekdays, and the decrease in $PM_{2.5}$ after the morning rush-hour peak was greater during the weekends compared to weekdays. The road at Pulchowk is one of the major routes for public transportation connecting Kathmandu with Lalitpur district, and these public vehicles operate seven days per week.

Hourly average plots show $PM_{2.5}$, BC, and TSP during winter, and $PM_{2.5}$ and PM_{10} during monsoon follow similar trends (Figs. 2 and 3 and Fig. S2). This suggests that PM concentration aligns with combustion-related sources, such as traffic exhaust. Morning rush hour peak profiles also appear similar for $PM_{2.5}$, BC, and TSP. Maxima of $PM_{2.5}$/TSP ratio occurs during nighttime and noon, while the minima of $PM_{2.5}$/TSP ratio coincides with rush hour periods (6–9 am and 4–6 pm). However, no such trend was observed for $PM_{2.5}/PM_{10}$ ratios during monsoon. In contrast to $PM_{2.5}$/TSP ratio trends during winter, $PM_{2.5}/PM_{10}$ ratio increases during rush hours and decreases during the nighttime (Fig. S2). Low $PM_{2.5}$/TSP ratios during rush hours in winter corresponds to increased road dust resuspension related to traffic activities. Low $PM_{2.5}/PM_{10}$ ratio during traffic hours that were attributed to traffic emissions and turbulence, have been observed at an urban site in Barcelona, Spain (Querol et al., 2001). Overall $PM_{2.5}$/TSP ratio during the winter was larger compared to $PM_{2.5}/PM_{10}$ ratio during monsoon. Besides traffic sources, garbage burning near roadsides during winter also likely contributed to the $PM_{2.5}$ burden. The seasonal differences also might be due to the increased contribution of non-traffic exhaust such as road and soil dust, and the preferential loss of $PM_{2.5}$ in rain, where most of these precursors are lost in rainfall, but most PM_{10} is crustal and are quickly resuspended after rain events during the monsoon.

Based on hourly averages, BC level was 1.2 times larger on weekdays than on the weekends. The largest difference occurred in the morning hours when BC concentration was 1.8 times larger on weekdays than on the weekends. Overall hourly average $PM_{2.5}$/BC (5.8 ± 1.3) and TSP/BC (7.8 ± 1.3) ratios in weekdays were similar to $PM_{2.5}$/BC (5.8 ± 1.7) and TSP/BC (8.0 ± 1.6) ratios during weekends in winter. This suggests that though this site is heavily influenced by local combustion sources such as vehicle exhaust, the site could also be influenced by regional air pollution.

3.3. Indoor-outdoor $PM_{2.5}$ and BC associations

The Indoor $PM_{2.5}$ trend at this modern house was very similar to outdoor $PM_{2.5}$ (Fig. 1). The ratio of outdoor to indoor daily average $PM_{2.5}$ concentrations was 1.1 ± 0.1. Indoor sampling was collected from a windowed bedroom. A paired *t*-test indicates a significant difference ($p < 0.05$) between indoor and outdoor $PM_{2.5}$ concentrations. The kitchen was located on a different floor. This building was constructed within the past 10 years, and included modern casement windows and other modern engineering designs. There was no air conditioning in this room. There was one window and one door for access to the balcony in the room. People living in this room were non-smokers. During scheduled power outages, power inverters were used as an alternate source for electricity; no diesel- or gasoline-generators were used. It is likely that this building reflects some of the better quality housing stock available in Kathmandu.

Though hourly $PM_{2.5}$ and BC generally tracked one another, relative differences (normalized by $PM_{2.5}$) between indoor and outdoor $PM_{2.5}$ concentrations were smallest during rush hours and largest during the nighttime (Fig. 2). Indoor $PM_{2.5}$ observed at nighttime (7 PM–5 AM) was 27% lower than corresponding outdoor $PM_{2.5}$, and indoor daytime $PM_{2.5}$ (6 AM–6 PM) was just 8% lower than corresponding outdoor $PM_{2.5}$. There was a stronger correlation between daily outdoor and indoor $PM_{2.5}$ ($R^2 = 0.89$) compared to BC ($R^2 = 0.70$). The mean ratio of 24-hour BC to $PM_{2.5}$ in outdoor was 0.19 ± 0.06, and this was indistinguishable to that in indoor (0.19 ± 0.05) environment. Indoor and outdoor $PM_{2.5}$ levels were highly correlated ($R^2 = 0.95$) with each other during winter (not plotted). Night time samples of $PM_{2.5}$ and BC are less likely to be influenced by local emissions indicated by a small peak in BC. They also appear to diverge from one another suggesting that the source profile overnight shifts from one dominated by local traffic sources to one heavily influenced by long range transport where there is an underlying BC signal which does not change, but a secondary aerosol formation process continues over time in the evening. While a collapsing boundary layer is known to increase aerosol concentration near the surface (Han et al., 2009), one would expect this effect to influence both $PM_{2.5}$ and BC concentrations

equally. Large differences during the nighttime may also be caused by less turbulent air at night during winter in Kathmandu Valley (Aryal et al., 2008), which leads to less mixing with the outdoor air. The sampled location was unlikely to be affected by typical indoor activities (cooking, woodsmoke, etc,) though indoor dust resuspension was more likely to be true during daytime. Particle resuspension during the daytime contributed to indoor PM (>1 μm) in the study at Fresno, California in 1999 (Vette et al., 2001). Despite this, a clear divergence is observed in the data set, suggesting a changing pattern of aerosol sources at this location.

As one might expect, there is a large increase in TSP observed during the morning rush hour event, which is much smaller during the evening rush hour. Despite this, the TSP/$PM_{2.5}$ ratios during rush hours declined in both time periods to approximately 0.65, whereas non-rush hour ratios were 0.80–0.90% (Fig. S2). This indicates the likelihood of a significant coarse mode fraction attributing to rush hours at this location.

The measurements were taken from only one location in the Valley, and this house was newly built, and the windows and doors present in this house might not be representative of the residential houses in the Valley, and the results suggest the heavy influence of outdoor sources on the indoor environment at this location. Vulnerable populations such as homebound individuals (elderly and young children) are thus likely to be exposed to high levels of pollution attributed to local sources, even in an indoor environment. This analysis is based on just one set of observations in a modern home. Thus, additional sampling in different residences are needed to understand the influence of roadside air pollutants in the neighborhoods in Kathmandu Valley.

3.4. Case study: 'Bandh' day

On March 19, 2014, a political uprising, commonly known as a 'Bandh', occurred in Kathmandu Valley. Bandhs are an act of civil disobedience that is called by organizations, political parties, or groups to protest government policies or decisions; they often are called on very short notice. During these events, protestors ask for a near complete shutdown of the entire city during the day closing offices, schools, shops, and roadways to vehicles. Compliance with this request can sometimes be enforced with the threat of violence, and thus most of the city is subdued during these events.

Measurements during the Bandh did not have distinct rush hour peaks for PM and BC, in contrast to other days, and the concentrations were much lower (Fig. 4 and Fig. S3). In previous measurements, PM_{10} was reduced by 30% on a 'Bandh' day on April 23, 2003 compared to the previous day in the Valley (ICIMOD, 2007). In our study, morning rush hour periods (6–10 am) were characterized by a 44% reduction in $PM_{2.5}$ and 32% reduction in BC compared to the average from the entire sampling campaign. 24-hour means of $PM_{2.5}$ and TSP levels were reduced by a factor of two ($PM_{2.5}$: 45 μg/m^3 and TSP: 59 μg/m^3) on a Bandh day compared to the previous day ($PM_{2.5}$: 97 μg/m^3 and TSP: 117 μg/m^3) and the next day ($PM_{2.5}$: 75 μg/m^3 and TSP: 109 μg/m^3) (Fig. 1). This is much a larger reduction than that observed for $PM_{2.5}$ decrease observed during weekends (discussed in section 3.2). There was only a small difference in rush hour levels of $PM_{2.5}$ between weekdays and weekends (Fig. S1).

BC levels also decreased on the Bandh day with 24-hour mean of 8 μgC/m^3 compared to 16 μgC/m^3 and 15 μgC/m^3 on the previous and the next day, respectively. During the early evening, the Bandh relaxed and few vehicles began to operate and commercial shops started to open. The data appeared to track this profile, with $PM_{2.5}$ and BC concentrations indicating an early-morning spike, followed by declining concentrations of PM and BC, and then starting to increase after 5 pm (Fig. 4). Though the overall air quality improved in the Valley during the Bandh day, some locations might see the reverse. Protestors sometimes burn tires on the roads for a blockade causing nearby residents to breathe harmful levels of air pollutants (Shakya et al., 2008).

3.5. Chemical composition of $PM_{2.5}$ and PM_{10} during monsoon season

Among the water-soluble inorganic ions, calcium ion (Ca^{2+}), sulfate (SO_4^{2-}), and ammonium (NH_4^+) were the major ions in both PM_{10} and $PM_{2.5}$ (Table 2). Calcium was the most abundant ion in both PM_{10} and $PM_{2.5}$. Resuspension of dust from roads, construction activities, soil dust, and traffic sources likely contributed to such high calcium concentrations. The three ions, NH_4^+, SO_4^{2-}, and nitrate (NO_3^-) that are formed from secondary aerosol formation processes, account for about half of the eleven water-soluble ions by mass. These three ions were less abundant in PM_{10} (19%) than in $PM_{2.5}$ (28%). SO_4^{2-}, NH_4^+, and potassium ion (K^+) concentrations (normalized by PM mass) were slightly larger in $PM_{2.5}$ than in PM_{10}.

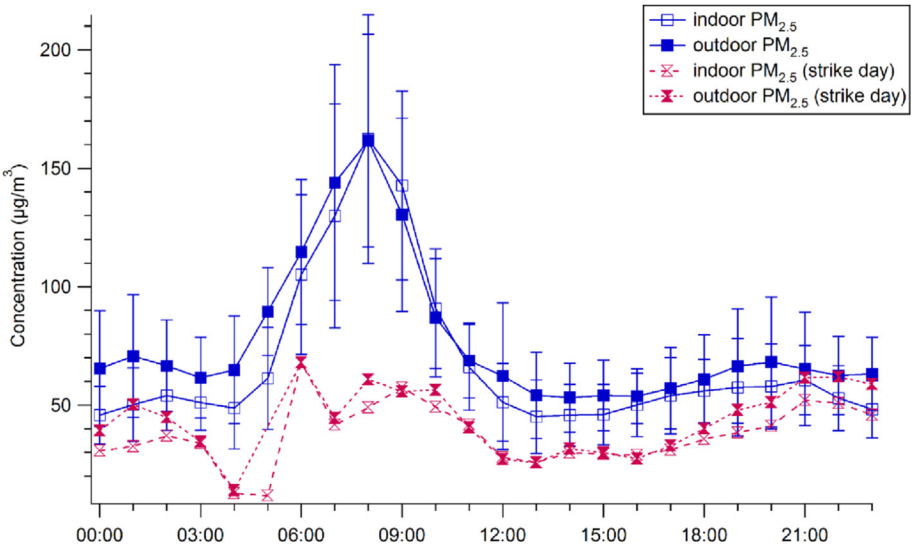

Fig. 4. Hourly variation of $PM_{2.5}$ and BC during all days of the sampling period and on a 'Bandh' day during winter season in 2014.

Table 2
Summary of chemical composition of PM_{10} and $PM_{2.5}$ during monsoon season at Pulchowk, Kathmandu Valley. Concentrations (mean ± one standard deviation) are given in μg/m^3.

Chemical species	PM_{10}	$PM_{2.5}$
PM concentration	35.12 ± 17.21	21.87 ± 14.09
Water-soluble ions		
F^-	0.01 ± 0.01	
Cl^-	0.82 ± 1.20	0.58 ± 0.48
NO_2^-	0.08 ± 0.08	0.02 ± 0.02
NO_3^-	0.17 ± 0.11	0.09 ± 0.06
PO_4^{2-}	0.12 ± 0.14	0.04 ± 0.05
SO_4^{2-}	3.95 ± 2.42	3.66 ± 2.07
Na^+	0.66 ± 1.03	0.28 ± 0.45
NH_4^+	2.49 ± 1.47	2.31 ± 1.30
K^+	0.25 ± 0.14	0.21 ± 0.11
Mg^+	0.09 ± 0.06	0.04 ± 0.03
Ca^{2+}	4.95 ± 0.96	3.86 ± 0.86
Elements		
Na	0.26 ± 0.26	0.23 ± 0.22
Mg	0.22 ± 0.17	0.13 ± 0.11
Al	1.15 ± 0.70	0.55 ± 0.57
Si	2.99 ± 2.09	1.09 ± 0.78
S	1.04 ± 0.63	0.98 ± 0.48
K	0.64 ± 0.36	0.32 ± 0.13
Ca	0.98 ± 0.63	0.38 ± 0.24
Sc	0.08 ± 0.05	0.03 ± 0.02
Ti	0.11 ± 0.07	0.04 ± 0.03
Cr	0.02 ± 0.02	0.01 ± 0.01
Mn	0.02 ± 0.02	0.01 ± 0.01
Fe	1.12 ± 0.72	0.45 ± 0.29
Ni	0.03 ± 0.01	0.02
Cu	0.01 ± 0.01	0.01
Zn	0.13 ± 0.08	0.13 ± 0.05
Ag	0.01 ± 0.02	0.01 ± 0.01
Cd	0.02 ± 0.02	0.01 ± 0.01
Sn	0.02 ± 0.03	0.02 ± 0.02
Ba	0.48 ± 0.47	0.29 ± 0.44

The occurrence of NH_4^+ and K^+ in mainly fine mode aerosol have been reported from the study at mountain site in China (Li et al., 2011). The neutralization ratio $[NH_4^+]/(2[SO_4^{2-}]+[NO_3^-])$ was above unity for both $PM_{2.5}$ and PM_{10} indicating ammonium levels were sufficient to neutralize the aerosol acidity during the monsoon period. This is in contrast to a previous study on bulk aerosol from a different location in Kathmandu, which showed that ammonium was not sufficient to neutralize aerosol acidity during the wintertime (Shakya et al., 2010). These differences suggest that either there were changes in aerosol acidity in different seasons or that there were changes in aerosol sources across these two locations. Both locations are expected to be heavily influenced by traffic emissions. There was no weekday-weekend pattern of water-soluble ions for the sampling period. These ion levels varied from day to day during the week except for Ca^{2+}, whose value remained the same throughout the week. Such a consistent signature of Ca^{2+} could also indicate a prevalent source of road/soil dust and the influence of construction activities in the region.

Elemental potassium (K) and K^+ were moderately well correlated with each other ($R^2 > 0.50$), but the K^+/K ratio was smaller than unity, suggesting the abundance of potassium-containing molecules in these samples (i.e. not ionic potassium). The relatively low content of water-soluble K^+ suggests a small contribution from biomass burning (Watson et al., 2001). The mean K^+/K ratio was smaller in PM_{10} (0.36) than in $PM_{2.5}$ ($K^+/K = 0.59$), showing any likely K arising from dust being present in PM_{10}, and K arising from biomass burning in $PM_{2.5}$. There was an excellent correlation between sulfur (S) measured by XRF spectroscopy and sulfate-sulfur measured by IC for both $PM_{2.5}$ ($R^2 = 0.92$, $p < 0.01$; slope = 0.72) and PM_{10} ($R^2 = 0.85$, $p < 0.01$; slope = 0.85). There was no correlation between calcium measurements by the two techniques, likely caused by the large influence of insoluble calcium species, probably arising from crustal materials which may be only partially soluble.

Sulfate and NH_4^+ occur mainly on fine aerosol mode (~70% by mass) in these data. Nitrate concentrations were larger in PM_{10} than in $PM_{2.5}$ suggesting its presence in the coarse mode. Nitrate in PM_{10} was also strongly correlated with ions such as magnesium ion (Mg^{2+}) and Ca^{2+} ($R^2 > 0.55$). This suggests that NO_3^- may be associated with crustal materials. Work by Rengarajan and colleagues (Rengarajan et al., 2011) has shown a large fraction of $Ca(NO_3)_2$ and/or $Mg(NO_3)_2$ arising from crustal materials in western India, which may partially explain these findings.

Silica (Si), aluminum (Al), iron (Fe), S, calcium (Ca), K, barium (Ba), Na, magnesium (Mg), and zinc (Zn) were the measured elements with the highest concentrations in both PM fractions (Table 2 and Fig. 5). 19 elements presented in Table 2 contributed about 26% of PM_{10} mass and 21% of $PM_{2.5}$ mass. Three elements such as Al, Si, and Fe were the most dominant crustal elements exceeding 1 μg/m^3 in PM_{10}, while only one element, Si, exceeded in $PM_{2.5}$. The elements such as Si, Al, Fe, Ca, scandium (Sc), and titanium (Ti) concentrations in PM_{10} fractions were two times larger than those in $PM_{2.5}$ fractions. In both size fractions, Si had the largest concentration among the analyzed elements. Most of these (Si, Al, Fe, and Ca) are crustal elements that are concentrated in the PM_{10} fraction. This suggests that the sources from the resuspension of road dust, tire wear, and particles from construction materials (Kreider et al., 2010) are the important PM sources in this location. Computation of an enrichment factor for Si, Fe, and Ca by using Al as the reference element and a generic upper continental crust (Taylor and McLennan, 1995) shows larger values (>10) in the $PM_{2.5}$ fraction compared to that in the PM_{10} fraction (<10). An exception was Si, which had smaller values (<10) in both $PM_{2.5}$ and PM_{10} fractions. Enrichment factors suggest that the main influence was from anthropogenic origin on $PM_{2.5}$ and crustal origin on PM_{10} fraction (Cong et al., 2010).

Tracers of oil burning, vanadium (<5 ng/m^3) and nickel (<25 ng/m^3) were found in much smaller concentrations, suggesting that fuel oil (used for home heating) is not an important aerosol source in this area. Elements such as chromium (Cr), copper (Cu) and Zn were lower in both $PM_{2.5}$ and PM_{10} on Saturday compared to other days of the week.

Water-soluble inorganic ions and elements accounted for about half of the PM_{10} (47%) and $PM_{2.5}$ (49%) mass. Carbonaceous aerosol and the respective mass of the compounds related to the elements might account for the difference in PM mass. Stone et al. (2010) found about 40% contribution of carbonaceous aerosol to $PM_{2.5}$ at a rural location in Godavari, Lalitpur in 2006. Shakya et al. (2010) found that about 62% of total speciated aerosol (carbonaceous and water-soluble ions) was composed of total carbon (elemental and organic carbon) at an urban location in Kathmandu during winter 2007–2008.

Soil dust concentrations in $PM_{2.5}$ and PM_{10} were computed by using the following equation (Eldred et al., 1987):

$$\text{Soil dust} = 1.15 \times (1.98 \times \text{Al} + 2.14 \times \text{Si} + 1.2 \times \text{K}^* + 1.4 \times \text{Ca} + 1.67 \times \text{Ti} + 1.36 \times \text{Fe}^*)$$

where, Fe* and K* values were used for their respective natural concentrations after considering their enrichment factors (Marcazzan et al., 2002).

Based on this exercise, soil dust accounted for 33% of measured chemical composition of $PM_{2.5}$ and 42% of that of PM_{10} during the monsoon. This shows that soil dust could be the second most important PM source after vehicular exhaust in Kathmandu Valley.

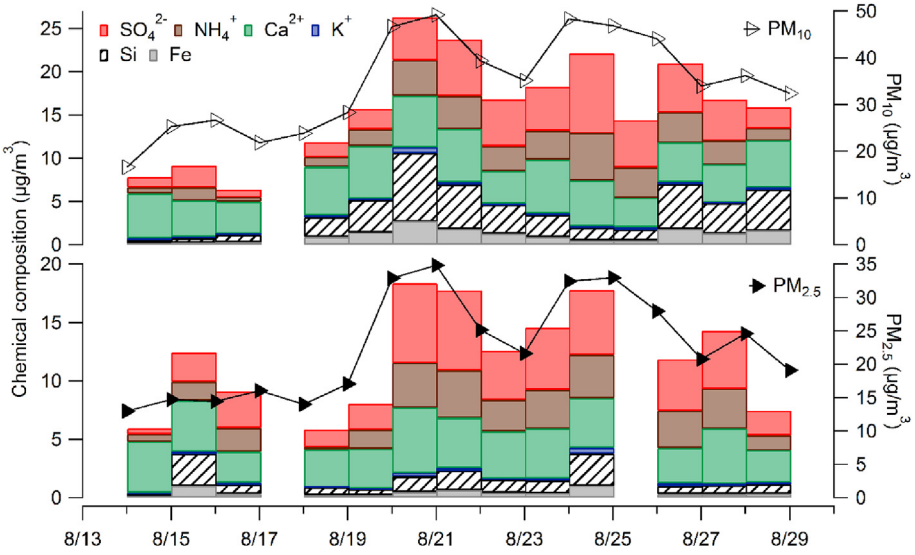

Fig. 5. Chemical composition of major species of $PM_{2.5}$ and PM_{10} during monsoon season in 2014.

Kim et al. (2015) found a large contribution of crustal components (31.7%) to PM_{10} at another urban location in Kathmandu Valley, a finding that is consistent with our results.

3.6. Limitations

A limitation of this study is the comparatively short duration of the sampling timeframe. For example, the seasonal variation (for winter and monsoon seasons) could be assessed only for $PM_{2.5}$. Indoor and outdoor BC concentrations, indoor $PM_{2.5}$ concentrations, and outdoor TSP concentrations were measured during only the winter. Outdoor PM_{10} concentrations and chemical composition of PM_{10} and $PM_{2.5}$ were measured only during monsoon season. Indoor sampling performed in this study may not be representative of local residences in Kathmandu Valley because this house was newly built and had modern engineering designs. This house also used only power inverters as an alternative electricity source.

4. Conclusions

Measurements of PM size fractions in an urban residential location in Pulchowk indicated elevated concentrations during the winter compared to the monsoon season. The results indicate the role of meteorology in alleviating $PM_{2.5}$ pollution in Kathmandu Valley. Frequent rainfall help to reduce overall $PM_{2.5}$ concentrations in the monsoon season and an increased mixed layer height during monsoon compared to winter helped to reduce ground level $PM_{2.5}$ concentrations during nighttime in monsoon. The determination of elements and ions showed that secondary inorganic ions (NH_4^+, SO_4^{2-}, and NO_3^-) accounted more for $PM_{2.5}$ mass (28%) compared to PM_{10} mass (19%) while the opposite was true for crustal elements such as Al, Si, Ca, and Fe. Vehicle emission and resuspended dust were the most important PM sources at this location. There was a large contribution of $PM_{2.5}$ to TSP and PM_{10} suggesting the heavy influence of combustion sources.

A surprisingly high fraction of fine particle intrusion was observed in indoor measurements of $PM_{2.5}$ compared to collocated outdoor measurements. This is in light of the measurement location, which was characterized by a location well above a nearby roadway in a home, which was recently built with modern windows and insulation techniques. This would suggest that indoor levels of PM are likely to be reasonably reflected by nearby outdoor monitors. Further, this is likely to be true for many other homes because domiciles in Kathmandu are not usually built to such modern standards, meaning that infiltration rates will be similar.

Regular monitoring of ambient air pollution and indoor air pollution is important to assess local air quality, and to identify the air pollution emission sources in Kathmandu valley. Average daily concentrations of the sampling campaign exceeded Nepal National Ambient Air Quality Standard for only $PM_{2.5}$ (i.e. 40 $\mu g/m^3$) during winter (76 ± 18 $\mu g/m^3$), and this alone indicates that more work in this region is warranted. Specifically, air pollution exposures of residents living close to busy roads, and concomitant health effects attributed to this exposure, need further study. These results clearly showed that residents living many stories above roadways are still exposed to high levels of particulate pollution, and monitoring during a civil strike also confirms that vehicular emissions are the major source of particulate pollution exposure in these homes.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.apr.2017.05.002.

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