J. Earth Syst. Sci. (2019) 128:88
https://doi.org/10.1007/s12040-019-1118-5

$PM_{2.5}$, PM_{10} and surface ozone over Lumbini Protected Zone, Nepal, during monsoon season of 2012

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MS received 7 July 2017; revised 24 August 2018; accepted 16 October 2018; published online 27 March 2019

Physical characterisation of $PM_{2.5}$, PM_{10} and surface ozone measured during the period from 17 July to 21 August 2012 at four strategic locations in and around the Lumbini Protected Zone, Nepal, is done to assess air quality of the region and understand qualitatively source mechanisms of these pollutants. The measurement locations are Panditarama Lumbini International Vipassana Meditation Centre, Parsahawa, Bhairahawa and Tilaurakot, representing monastic, industrial, urban and control areas, respectively. The overall average concentration of $PM_{2.5}$ at these locations is $\sim 19 \pm 12$, 35 ± 13, 35 ± 11 and 25 ± 6 $\mu g/m^3$ and of PM_{10} is $\sim 25 \pm 11$, 103 ± 41, 58 ± 15 and 32 ± 7 $\mu g/m^3$, respectively. $PM_{2.5}$ never crosses the safe limit of the National Ambient Air Quality Standards of Nepal (NNAAQS) in the monastic and control areas but either crosses the NNAAQS occasionally or remains in its vicinity at the other two locations. The PM_{10} concentration frequently exceeds the safe limit in the industrial area but not in the other remaining areas. The analysis indicates the dominance of the impact of local sources and boundary layer thickness on the atmospheric loadings of the particulate matter. The daily average mixing ratio of surface ozone remains normally low at all the four observational sites although the mixing ratio of ozone at Panditarama Lumbini International Vipassana Meditation Centre is much lower than the NNAAQS but higher than that observed at Tilaurakot.

Keywords. Lumbini; global heritage; air quality assessment; particulate matter; surface ozone.

1. Introduction

The air quality of a region refers to the state of the air within that region and surroundings, which, by virtue of its state, influences human health, plants, animals and natural resources of the region. Good air quality pertains to air which is clean, clear and free from gaseous and particulate pollutants. It is required for preserving the exquisite balance of life on Earth and preserving heritage areas (with monuments thereon) and ecology (Brimblecombe 2003; Ivaskova *et al.* 2015; Turo *et al.* 2016). In contrast, poor air quality can adversely affect human health, plants and animals and degrade the environment (Mohnen *et al.* 1993; Ghio and Devlin 2001; Shindell *et al.* 2012; Stocker *et al.* 2013; Janssen *et al.* 2013; Wu *et al.* 2013; Burney and Ramanathan 2014; Forouzanfar *et al.* 2015; Fuzzi *et al.* 2015; Kelly and Fussell 2015) which in turn may complicate small- to large-scale interactions of various atmospheric parameters, namely physical, chemical, optical and meteorological, leading eventually to climate change and its deleterious repercussions (Colvile *et al.* 2001;

Orru *et al.* 2017). Sources of pollutants can be natural as well as anthropogenic. Major natural sources include volcanic eruption and windblown dust, whereas anthropogenic sources include fossil fuel and biofuel combustion, biomass burning, auto-vehicles, industries, thermal power plants, landfills, etc.

Lumbini, the birthplace of the Lord Buddha, is a world heritage place situated in the Rupandehi district in the southwestern plains of Nepal. Being a world heritage location, the area of Lumbini has got global importance. The world community has taken various steps to conserve the whole area along with its monuments and other features of historical and archaeological significance thereon. The main reason behind the conservation of this area is that once the heritage is destroyed, it cannot be reconstructed in a certain period of time. On losing the originality of this heritage, the history of Buddhism, the peace and the civilisation may be lost and can never be restored in its totality.

Despite the global importance of this heritage region, industrial activity in some areas in and around Lumbini Protected Zone (LPZ) has drastically increased. A report published by the International Union for Conservation of Nature (IUCN; Nepal 2012) describes that the problems caused by industrial expansion within the zone need to be addressed to preserve the sanctity of Lumbini, promote sustainable development of the surrounding communities and conserve the biodiversity of the region. These reports indicate that massive emission of pollutants from the industries in and around the LPZ has now become a cause of concern to this area and its ecosystem.

In view of the above, the World Health Organization (WHO) and the Indian Institute of Tropical Meteorology, Pune, India, collaborated to carry out a pilot study on the air quality of the LPZ in Nepal. As such, measurements of the particulate matter ($PM_{2.5}$ and PM_{10}) and surface ozone were made at four different environmental sites in the LPZ. These sites are Panditarama Lumbini International Vipassana Meditation Centre, Parsahawa, Bhairahawa and Tilaurakot, representing monastic, industrial, urban and control areas, respectively. This research work is an effort to assess the air quality of the above four measurement locations in the LPZ, understand qualitatively the sources of these pollutants and investigate whether there is significant impact of industrial activity on air quality. It should be mentioned that only one research publication on the air quality of Lumbini is available in the literature (Rupakheti *et al.* 2017) which presents air quality data (PM, BC, CO, O_3) measured at Lumbini International Research Institute (LIRI), a Buddhist library in Lumbini, during the pre-monsoon season of 2013.

2. Observational sites, measurement and methodology

Four measurement sites, namely, Panditarama Lumbini International Vipassana Meditation Centre or simply PMC (27°28′34″N; 83°16′28″E; ∼100 amsl), Parsahawa (27°30′16″N; 83°22′22″E; ∼107 amsl), Bhairahawa (27°30′43″N; 83°26′43″E; ∼107 amsl) and Tilaurakot (27°34′47″N; 83°4′47″E; ∼107 amsl), were selected under the collaborative programme, which represent, respectively, monastic zone, industrial area, urban area and control area in the LPZ. The map showing the location of the observational sites along with superimposed ECMWF ERA-Interim reanalysis 10-m wind for 00:00 and 12:00 UTC is shown in figure 1. It may

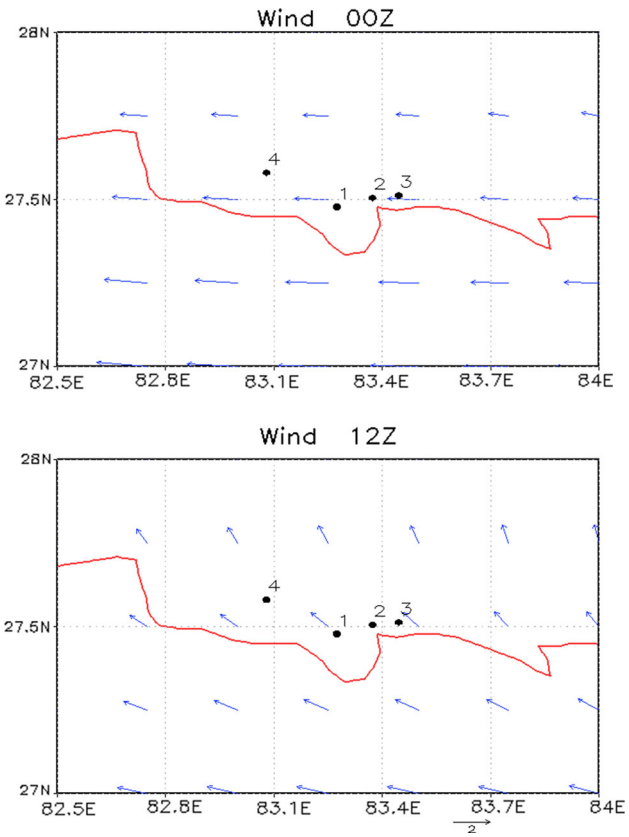

Figure 1. Map showing measurement locations (1: PMC; 2: Parsahawa; 3: Bhairahawa; and 4: Tilaurakot) in the LPZ, Nepal, and superimposed ECMWF ERA-Interim 10-m wind at 00 Z and 12 Z.

Table 1. *Radial distance between pair of measurement locations.*

Sl. no.	Pair of measurement location	Radial distance (km)
1	PMC–Parsahawa	09.29
2	PMC–Bhairahawa	16.42
3	PMC–Tilaurakot	24.63
4	Parsahawa–Bhairahawa	07.15
5	Parsahawa–Tilaurakot	32.57
6	Bhiarahawa–Tilaurakot	38.70

be seen in this figure that morning wind is easterly and afternoon wind is southeasterly.

The observational site, PMC, in the monastic zone has natural greenery with long shady trees and bushes. Tourists from all over the world visit the area all the year round, especially between September and November. The observable major source of pollution in the area is burning of fossil fuel/biofuel mostly for domestic cooking in the forenoon hours of the day. Emission from vehicular activity on the nearby roads is low due to low traffic density. However, the impact of gases emitted from the vehicles on the local atmospheric gas phase chemistry cannot be completely ruled out.

The industrial site at Parsahawa (Gonaha) is the terrace of a residential building which is nearly 12 km by road to the east of the PMC. The area mostly contains cement factories, brick kilns, steel industry, food industry, etc. The vehicular activity on nearly 50-m distant road is high.

The measurement site at Bhairahawa is also the terrace of a residential building which is about 18 km by road to the east of the PMC. It is an urban area where movement of light to heavy vehicles on the paved and unpaved/unrepaired roads is very high. Here, the pollutants are originated mostly due to the vehicular movement. Also, urbanisation
and small-scale industries may be the source of pollution in Bhairahawa.

The measurement site in Tilaurakot is an open ground in front of the office of the Lumbini Development Trust (LDT). It is nearly 29 km by road to the northwest of the PMC. This site is considered as a control site or reference site due to its location being free from major anthropogenic activity of pollutants' emission. Radial distance between different measurement locations is given in table 1.

The present air quality assessment is based on measurements of $PM_{2.5}$, PM_{10} and surface ozone at four different environmental locations in the LPZ during a period from 17 July to 21 August 2012. It may be noted that the India Meteorological Department has categorised the months from June to September as monsoon season. Thus, the above 2 months fall in the monsoon season. Rainfall activity in these 2 months is normally found to be at its peak. Details of the measurement periods at individual sites are given in table 2. This table shows two measurement periods at PMC and Parsahawa sites. This is because the aerosol analyser could not be operated for sufficient number of hours at these locations during earlier observational period due to some technical problem in the casing of the analyser. Later, the same was compensated by running the equipment on some additional days at both the sites. Another point to be noted is that the period of observations shown in figure 1 differs slightly from what is given in table 2. This is because this table shows the whole campaign period, but figure 1 shows the actual period when the equipment remained operational at the respective sites.

Measurement of mass size distribution (MSD) of particulate matter was made with the help of online GRIMM Portable Aerosol Spectrometer (model: 1.108; make: GRIMM, Germany).

Fundamentally, the spectrometer measures number size distribution. But, the number size distribution is converted by the GRIMM's internal algorithm into MSD with the help of C-factor which depends on the particle density, particle shape and refractive index of the particle. The C-factor is calculated on the basis of dust load on the gravimetric filter (polytetrafluoroethylene [PTFE] filter) located before the internal pump of the spectrometer, and the calculated dust weight is shown by the instrument. It is a location-specific non-dimensional factor. The calculated C-factor becomes the input into the dust monitor, and all the dust mass concentration values as well as their averages are multiplied by this factor. It is mentioned that C-factor calculated for PMC, Parsahawa, Bhairahawa and Tilaurakot is 1.05, 1.2, 1.08 and 1.04, respectively. The average density considered in the calibration of the spectrometer is 1.68 g/cm^3. The instrument measures number/MSD of suspended particulate matter in 16 size bins with the smallest size bin of 0.23–0.3 μm and the largest size bin of >20 μm. The reproducibility of the spectrometer over the whole measuring range is ±3%. The uncertainty in the determination of the total aerosol mass by GRIMM 1.108 may be within 10–20% (Nicolas *et al.* 2009).

Table 2. *Details of observational sites and periods during field campaign in the LPZ from 17 July to 21 August 2012.*

Sl. no.	Measurement site (hagl)	Station height (amsl)	Starting date	Ending date
1	Panditarama Meditation Centre, Lumbini (∼1.5 m; monastic zone)	∼100 m	18 July 2012 19 August 2012	22 July 2012 20 August 2012
2	Gonaha-7, Parsahawa (∼4 m; industrial area)	∼107 m	23 July 2012 16 August 2012	30 July 2012 18 August 2012
3	Siddhartha nagerpalika-6, Gallamandi, Banklony, Bhairahawa (∼7 m; urban area)	∼107 m	31 July 2012	7 August 2012
4	Kapilbastu Nagerpalika-12, Shivgadh, Tilaurakot (∼1 m; control area)	∼107 m	8 August 2012	15 August 2012

Note: hagl stands for height in metre above ground level and amsl stands for height in metre above mean sea level.

The instrument is sent to GRIMM Company normally once a year for its calibration or whenever it has to be used in a field campaign programme. Estimation of the masses of $PM_{2.5}$ and PM_{10} has been made with the measurement of MSD.

The mixing ratio of surface ozone was monitored by an online US-EPA approved ozone analyser (model: O342M; Environment S.A., France). The analytical technique of the analyser is based on drawing ambient air with the help of a suction pump. The air is passed through a filter and then analysed at 253.7 nm wavelength (Nishanth *et al.* 2011). The lowest detection limit of the analyser is 0.4 ppbv with minimum response time of 20 s. The analyser needs zero and span calibration which is normally done once in a month with the help of built-in ozonator. The linearity of the analyser is ±1% with noise 0.2 ppb, zero drift less than 1 ppb/week and span drift less than 1%/week.

More details of both the analysers and their measurement techniques can be obtained from their manuals which are available on the websites of the respective companies (http://www.wmo-gaw-wcc-aerosol-physics.org/files/opc-grimm-model--1.108-and-1.109.pdf and http://www.lisa.u-pec.fr/~formenti/Tools/Manuals/O342MA_10.06.pdf). It is mentioned that measurement interval for both the instruments was kept at 15 min.

Data on both the particulate matter and the surface ozone were passed through various quality checks (e.g., stray readings of the analysers were first removed and then Gaussian filter was used for quality control) before starting mathematical analyses.

Data on meteorological parameters like rainfall (RF), wind speed (W) and boundary layer thickness (BL) used in this study are compiled from ECMWF (ERA-Interim) with horizontal resolution of $0.125^\circ \times 0.125^\circ$ (corresponding to $\sim 12 \times 12$ km^2). Details on the data may be obtained from http://www.ecmwf.int/en/research/climate-reanalysis/era-interim. Five days' air mass backward trajectory analysis at a height of 500 m above the observational sites has been done using National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory Hybrid Single Particle Lagrangian Integrated Model (HYSPLIT; http://www.arl.noaa.gov/ready.html) to understand the transportation of pollutants.

3. Results and discussion

3.1 *Time series of the concentration of particulate matters*

Variation of daily mean concentration of $PM_{2.5}$, PM_{10}, RF and W at PMC, Parsahawa, Bhairahawa and Tilaurakot during a period from 19 July to 20 August 2012 is shown in figure 2. Vertical bars shown at individual mean values in the plot indicate standard deviation (σ) about the corresponding means.

This figure reveals that the concentration of particulate matter at PMC and Tilaurakot is much less than the NNAAQS (for $PM_{2.5}$ is 40 $\mu g/m^3$ and for PM_{10} is 120 $\mu g/m^3$, averaged on 24-h duration). The overall average, minimum and maximum concentrations of $PM_{2.5}$ at PMC are $\sim 19 \pm 12$, 9 and 37 $\mu g/m^3$ and at Tilaurakot are $\sim 25 \pm 6$, 16 and 32 $\mu g/m^3$, respectively. Similarly, the overall average, minimum and maximum concentrations of PM_{10} at PMC are $\sim 25 \pm 11$, 12 and 41 $\mu g/m^3$ and at Tilaurakot are $\sim 32 \pm 7$, 22 and 43 $\mu g/m^3$, respectively. The average value and range of variation of $PM_{2.5}$ and PM_{10} at both

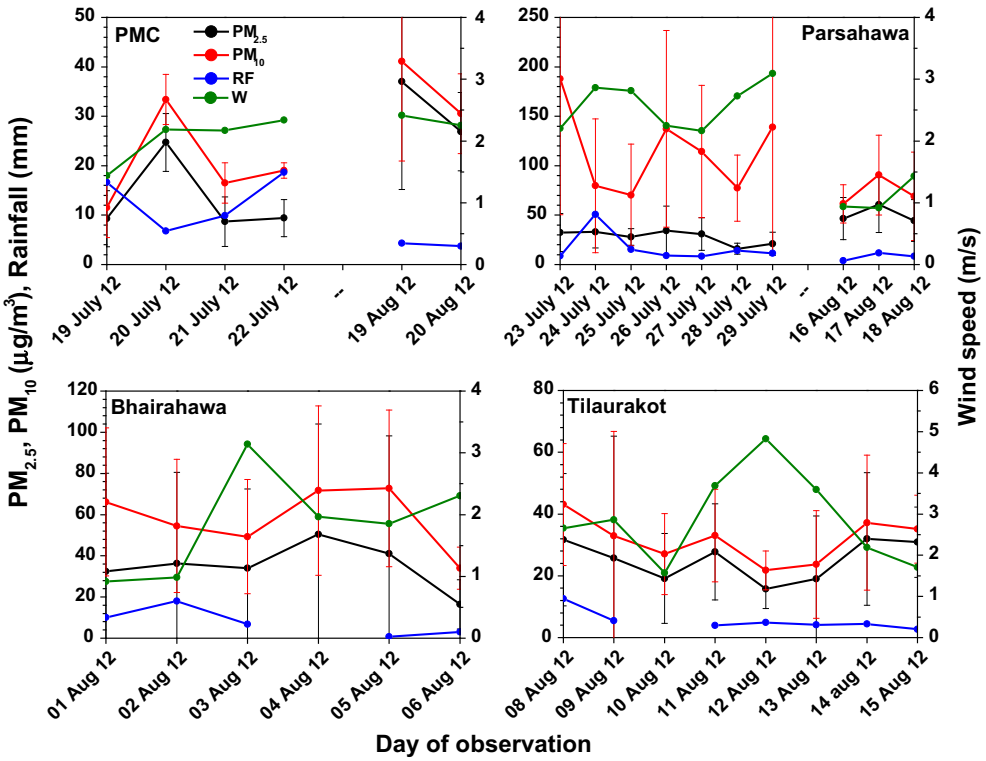

Figure 2. Daily mean concentration of particulate matter, ECMWF ERA-Interim reanalysis rainfall (RF) and wind speed (W) at Panditarama Lumbini International Vipassana Meditation Centre (19–22 July and 19–20 August 2012), Parsahawa (23–29 July and 16–18 August 2012), Bhairahawa (1–6 August 2012) and Tilaurakot (8–15 August 2012) in the Lumbini Protected Zone, Nepal.

the locations indicate that fractional contribution of the fine particles to the total aerosol mass is much higher than that of the coarse particles. It is found to be about 76% at PMC and 79% at Tilaurakot. The result indicates that the sources of particulate matter are weak in these areas, and they mostly generate fine particles. As mentioned above, the main sources of the observed particulate matter in these localities are burning of fossil fuel and biofuel mostly for domestic cooking. Vehicle-driven emission of coarse particles from soil dust and of fine particles from fuel combustion may be less due to less number of vehicles on the nearby roads. Figure 2 also reveals that the rainfall washes out particulate matter effectively at PMC to reduce its concentration, but the wind does not show any remarkable association with this variation. The correlation coefficient between $PM_{2.5}$ and RF at PMC is -0.84 (significant at better than 95%) and between PM_{10} and RF is -0.81 (significant at better than 95%). However, wind appears to influence particulate matter concentration at Tilaurakot, but the correlation coefficient between these two parameters is insignificant. Consequently, there is possibility of the dominance of other factors influencing the concentration of particulate matter at Tilaurakot. There might have been considerable reduction in the coarse particle concentration due to their interception by plants along with stimulated gravity settling of heavier particles caused by abundantly available leafy trees.

Trajectory plots for all the four observational sites are shown in figure 3. It may be seen in this figure that there were only two occasions when winds at PMC originated over the sea, otherwise landlocked. That is, wind over PMC is expected to be more of dry nature during the observation period. The winds reaching the Tilaurakot site are mostly originating over the sea. Thus, the fraction of marine components is expected to be more in the particulate matter at Tilaurakot than that at PMC. The prevalence of marine wind over Tilaurakot might have stimulated gravity settling, particularly of coarse particles, through moistening the aerosol. Consequently, the fraction of coarse particle is more at PMC (24%) than that at Tilaurakot (21%).

Rupakheti *et al.* (2017) have reported that average concentrations of $PM_{2.5}$ and PM_{10} measured

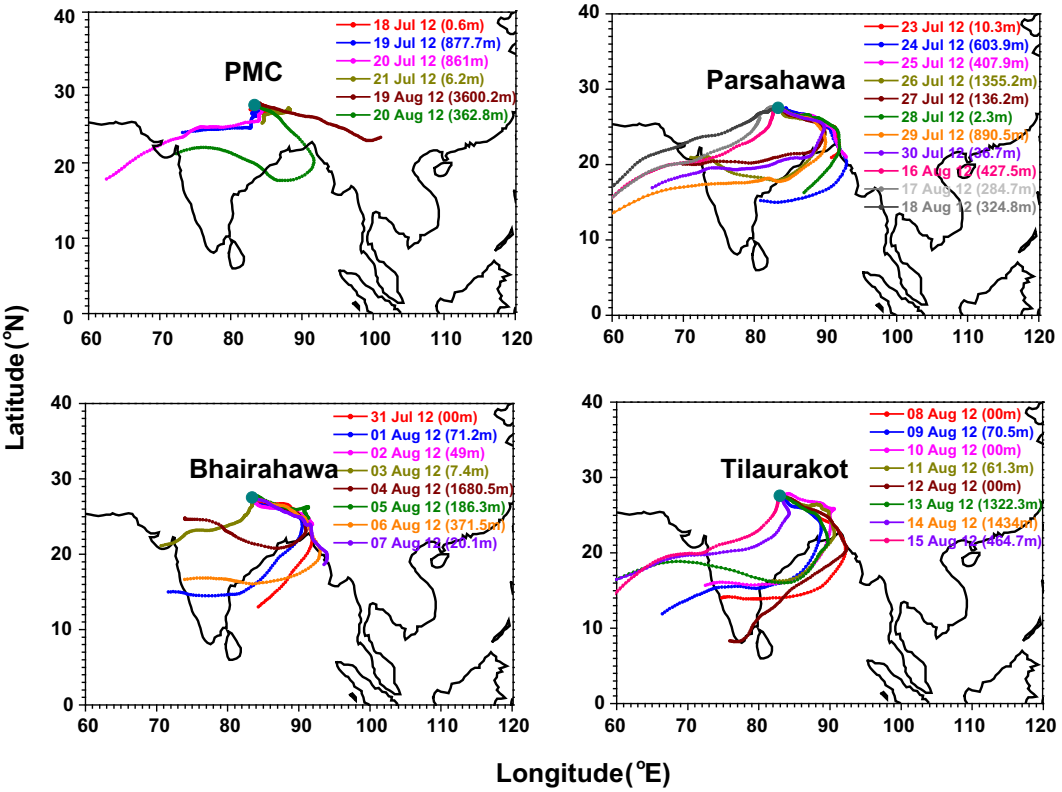

Figure 3. Five days' air mass backward trajectory ending at an altitude of 500 m above ground level at PMC, Parsahawa, Bhairahawa and Tilaurakot during the period of observations at individual locations (note: figures in the bracket beside dates denote initial altitude of the air parcels above ground level).

at the premise of LIRI (∼1.5 km to the north–northeast of PMC) during April–June 2013 (a major period of pre-monsoon season) are ∼53 ± 35 μg/m^3 (range: ∼6−272 μg/m^3) and ∼129 ± 92 μg/m^3 (range: ∼11−604 μg/m^3), respectively. That is, the concentrations of $PM_{2.5}$ and PM_{10} during pre-monsoon season of 2013 are, respectively, ∼2.8 and ∼5.1 times more than the concentration of corresponding parameters obtained at PMC during monsoon season of 2012. They have also reported that coarse mode fraction was ∼60%, which is very high as compared to our result of ∼24%. Higher contribution of coarse aerosols during pre-monsoon season was attributed to wind-blown soil dust from nearby agricultural fields and construction sites.

Daily average concentration of $PM_{2.5}$ and PM_{10} at Bhairahawa varies from 16 to 50 μg/m^3 and from 34 to 73 μg/m^3 with the overall average concentration of 35±11 and 58±15 μg/m^3, respectively. The concentration of $PM_{2.5}$ exceeds the NNAAQS during about 33% of the observational period and remains towards higher concentration level during 50% of the period. This implies that there are strong sources of fine particles. However, the concentration of PM_{10} remains nearly always below the NNAAQS, but above 60 μg/m^3 during 50% of the observational period. Such a high concentration level for quite an appreciable period of time during monsoon season, when effective precipitation scavenging mechanism normally remains active, is a matter of concern, and there is need to adopt some control measures for reducing emissions of the particulate matter in this area. The result indicates that there may be dominance of gas-to-particle conversion mechanism which might have resulted by the presence of gases emitted mostly from the vehicles and the other burning activities. The coarse particles (PM_{10}–$PM_{2.5}$) are generated by vehicle-driven road side dust, wind-blown dust and construction activity. Backward trajectory shown in figure 3 for Bhairahawa indicates that marine component from the Bay of Bengal might have also contributed considerably through transportation.

In general, the trend of variation of $PM_{2.5}$ is same as that of PM_{10} over the period of observation at PMC, Tilaurakot and Bhairahawa. Such a

variation trend implies that the sources (anthropogenic or natural) of $PM_{2.5}$ and PM_{10} in these areas are either same or, if different, originate nearly simultaneously in the same area, and these sources are not much away from the observational sites. Correlation analysis of particulate matter concentration at Bhairahawa with RF and W indicates that the correlation coefficient is insignificant, implying that W and RF are not the dominant factors influencing the concentration of particulate matter at Bhairahawa.

At Parsahawa, a large variation in the daily mean concentration of PM_{10} is observed. The minimum and the maximum concentrations are ∼61 and 188 $\mu g/m^3$, respectively, with the overall average concentration of $\sim 103 \pm 41$ $\mu g/m^3$. The variation trend indicates that the sources of PM_{10} may be in the vicinity of the measurement site. Whenever the wind blows from the source region towards the measurement site, the concentration is highly increased. Otherwise, it remains generally below the NNAAQS. The concentration at Parsahawa is found to exceed the NNAAQS during 30% of the measurement period.

Daily mean concentration of $PM_{2.5}$ is always below the NNAAQS during the first measurement period (23–29 July 2012) at Parsahawa and varies from ∼16 to 34 $\mu g/m^3$. On the contrary, it is always above the NNAAQS during the second measurement period (16–18 August 2012) and varies from ∼45 to 61 $\mu g/m^3$. Actually, the winds during 23–29 July 2012 originated mostly over the Bay of Bengal and travelled as southeasterly through humid zones of north India (figure 3). Consequently, these winds contained high moisture content on reaching Parsahawa. High moisture content in the air and occasional rain induced, respectively, gravity settling and wet removal of aerosol resulting in the low concentration of $PM_{2.5}$ at Parsahawa during this period. However, the winds during 16–18 August 2012 at Parsahawa originated over the Arabian Sea traversed the dry zone path as southwesterly (figure 3) and thus contained low moisture content on reaching there. The prevalence of dry air and occurrence of appreciable dry period at Parsahawa caused building up of fine particles in its boundary layer through direct emission from the nearby industries and vehicles as well as through gas-to-particle conversion resulting in higher concentration of $PM_{2.5}$ during this period. The role of RF in influencing particulate matter concentration at Parsahawa can be understood by the value of correlation coefficient between $PM_{2.5}$ and RF. It is found to be −0.89 which is highly significant (at better than 99%). The overall average concentration of $PM_{2.5}$ is nearly same ($\sim 35 \pm 13$ $\mu g/m^3$) as that at Bhairahawa but much higher than those at PMC and Tilaurakot.

Higher concentration levels of the particulate matter at Bhairahawa and Parsahawa for quite an appreciable period of time are bad not only for these areas but also for other areas of the LPZ because the prevailing easterly to southeasterly winds of the monsoon regime may advect these particulate matter to other clean areas of the LPZ and pollute them.

3.2 *Diurnal variation of the concentration of particulate matter*

Figure 4 shows the diurnal variation of the concentration of particulate matter observed at PMC, Parsahawa, Bhairahawa and Tilaurakot. The diurnal variation of boundary layer thickness is also shown in the plots to understand its influence on the variability of the particulate matter concentration. It is noticed from this figure that the concentration at PMC becomes maximum (∼47 $\mu g/m^3$ for $PM_{2.5}$ and ∼55 $\mu g/m^3$ for PM_{10}) at 21:00 h. The minimum concentrations of $PM_{2.5}$ and PM_{10} are 12 and 19 $\mu g/m^3$, respectively, which occur at 16:00 and 11:00 h, respectively. After 16:00 h, their concentrations rise slowly up to 19:00 h and then faster (∼13 $\mu g/m^3$/h for $PM_{2.5}$ and ∼13.5 $\mu g/m^3$/h for PM_{10}) till the maximum is attained. The maximum concentration at 21:00 h is the result of combined effect of increased vehicular emission after 16:00 h and decreases in the boundary layer thickness. The minimum concentration of $PM_{2.5}$ at 16:00 h and of PM_{10} at 11:00 h is attributed mainly to lower vehicular emission and increases in the boundary layer thickness. A small peak at 12:00 h is caused mainly by very local emission mostly from domestic cooking.

During late-night hours, anthropogenic activity and associated emission stops, and the aerosol removal mechanism, mainly dry deposition, becomes effective. Although the decrease in the boundary layer thickness with the passage of the night tends to initially sustain the concentration level of the particulate matter, the removal mechanism dominates it later, and the concentration of particulate matter continuously goes on decreasing during night. For this reason, the rate of decrease in the concentration of particulate matter from the highest value is slow in the beginning and fast

(~7.5 μg/m^3/h for $PM_{2.5}$ and ~9 μg/m^3/h for PM_{10}) later.

On sunrise at around 07:00 LT, the nocturnal boundary layer breaks and the boundary layer starts developing. This causes further reduction in the concentration of particulate matter. However, morning fumigation, occurring due to the breakup of the nocturnal inversion (Hibberd and Luhar 1996), causes vertical mixing of the pollutants. This phenomenon partially counterbalances reduction in the concentration and delays the time of occurrence of the minimum concentration level in the morning.

Figure 1 shows morning (00 UTC) and evening (12 UTC) wind patterns over the region under study. This figure reveals that the morning wind over the region is easterly, and as the day progresses, it turns to be southeasterly. Thus, the morning wind from Parsahawa and Bhairahawa (industrial and urban regions, respectively) may also be contributing to the particulate matter loading at PMC causing delay in attaining minimum value in the morning.

Diurnal variation of particulate matter at Parsahawa shows minimum concentration of $PM_{2.5}$ at 13:00 h and of PM_{10} at 12:00 h. It is found to be ~15 and 53 μg/m^3, respectively. The maximum concentration of both $PM_{2.5}$ and PM_{10} occurs at 23:00 h which is ~59 and 162 μg/m^3, respectively. On average, the concentration of $PM_{2.5}$ and PM_{10} during night (~43 and 121 μg/m^3, respectively) is found to be more than that during day (~28 and 90 μg/m^3, respectively). This is mainly due to higher boundary layer thickness during day and lower during night, irrespective of the impact of anthropogenic sources. A slight increase in the concentration of $PM_{2.5}$ after 06:00 h is due to domestic cooking utilising mostly biofuel and coal. Morning fumigation may also be contributing to the aerosol loading.

The average concentration of $PM_{2.5}$ and PM_{10} at Bhairahawa is lower (~11 and 32 μg/m^3, respectively) during 09:00–18:00 h and higher (~45 and 70 μg/m^3, respectively) during 19:00–08:00 h. The dominant factor responsible for such a diurnal variation is the boundary layer thickness which is higher during the day and lower during the night (figure 4). The concentration of particulate matter is found to increase after 04:00 h till 06:00 h in the morning. This is mainly due to domestic activity and vehicular movement during morning hours in the urban area of Bhairahawa. The morning

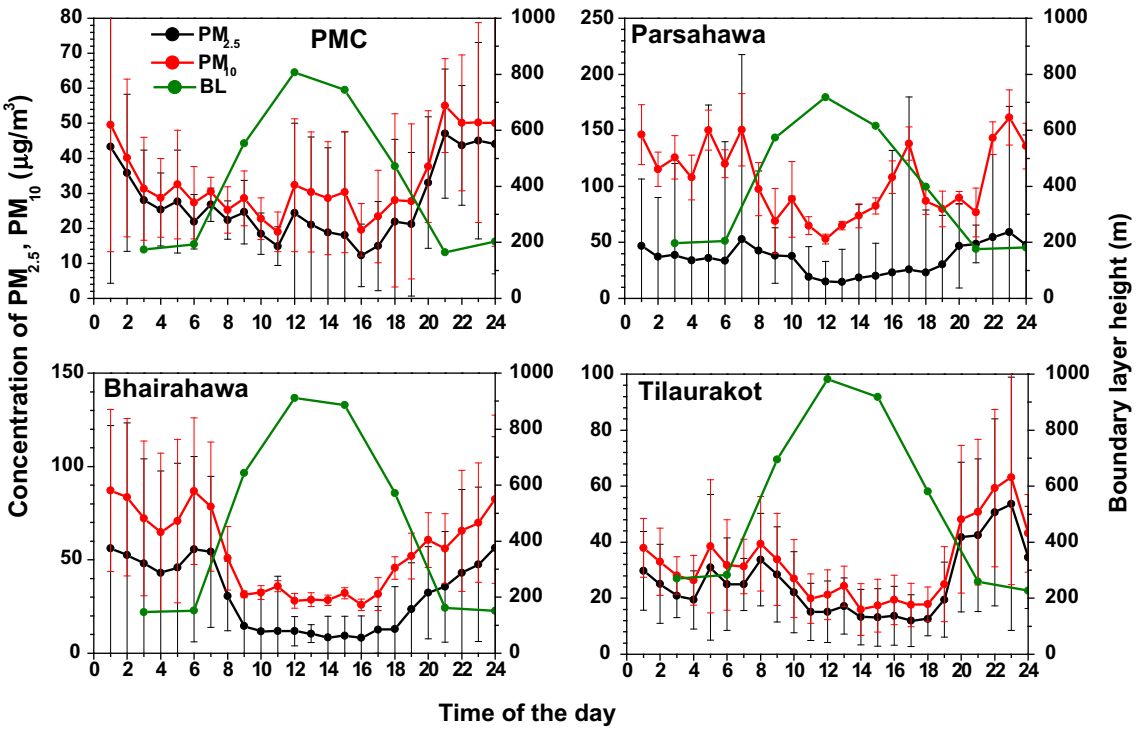

Figure 4. Diurnal variation of the concentration of particulate matter and ECMWF ERA-Interim reanalysis boundary layer thickness (BL) at Panditarama Lumbini International Vipassana Meditation Centre, Parsahawa, Bhairahawa and Tilaurakot in the Lumbini Protected Zone, Nepal.

fumigation occurring due to the breakup of the nocturnal inversion may also be adding to this trend of increase.

As seen at the other three locations, diurnal variation of particulate matter at Tilaurakot also shows daytime minimum and nighttime maximum. The maximum concentration of $PM_{2.5}$ and PM_{10} ($\sim$54 $\mu g/m^3$ and 63 $\mu g/m^3$, respectively) occurs at 23:00 h, whereas the minimum is broad occurring during 11:00–18:00 h with an average concentration of $\sim$14 $\mu g/m^3$ for $PM_{2.5}$ and 19 $\mu g/m^3$ for PM_{10}. The average rate of increase of particulate pollutants during 18:00–23:00 h is 8.2 $\mu g/m^3/h$ for $PM_{2.5}$ and 9 $\mu g/m^3/h$ for PM_{10} which is very high. This indicates the impact of very local anthropogenic activity which may be biofuel/coal burning for domestic cooking and biomass burning to generate smoke for saving the domestic cattle from mosquito bite in the night. Also, lowering of inversion layer contributes to increase in the concentration during this period. It may be noted that evening wind is southeasterly (figure 1). This implies that a little contribution of particulate matter is made by transportation from Parsahawa and Bhairahawa, as the location of Tilaurakot is to the northwest of Parsahawa and Bhairahawa. Daytime minimum concentration is attributed to increase in the boundary layer thickness with occasional washout process.

3.3 *Time series of the mixing ratio of surface ozone*

A day-to-day variation in the average mixing ratio of surface ozone, RF and W at PMC, Parsahawa, Bhairahawa and Tilaurakot is shown in figure 5. It is found from this figure that the mixing ratio at these locations varies from $\sim$13 to 27, from $\sim$11 to 20, from $\sim$9 to 16 and from $\sim$11 to 18 ppb, respectively. The coefficients of variation are 0.3, 0.2, 0.2 and 0.2. This indicates that the variability at all the individual locations is low with maximum at PMC. The overall average mixing ratio of ozone at these locations is $\sim 16 \pm 6$, 14 ± 4, 12 ± 5 and 14 ± 6 ppb, respectively. Eight hourly average mixing ratio of ozone is found to be always far below the NNAAQS (80 ppb, averaged on 8-h duration) during the period of observation. Thus, the quality of air with regard to ozone is very good. Such a low mixing ratio of ozone can occur only in those areas where there is less availability of ozone precursors like hydrocarbon (HC) and oxides of nitrogen (NO_x).

The average mixing ratio of surface ozone measured during pre-monsoon season of 2013 at LIRI was $\sim 47 \pm 20$ ppb (Rupakheti *et al.* 2017), which is $\sim$2.7 times more than that obtained during monsoon season of 2012 at PMC. This indicates that the production mechanism of ozone was more effective during pre-monsoon season than that during monsoon season.

Variation in the W is nearly out of phase of the variation in the ozone mixing ratio at PMC ($r = -0.94$, highly significant) and Parsahawa ($r = -0.73$, significant at better than 95%) indicating thereby dilution in the ozone mixing ratio at these locations due to increase in the vertical mixing caused by turbulence. At the remaining two locations, the relationship of W with the ozone mixing ratio is insignificant implying dominance of some other factors over the W.

It may be seen in figure 5 that the whole observational period witnessed RF with daily average varying from 3.7 to 18.7 mm at PMC, from 2.3 to 50.6 mm at Parsahawa, from 0.7 to 18.0 mm at Bhairahawa and from 2.7 to 12.6 mm at Tilaurakot. The sky remained mostly cloudy in the absence of RF. As such, humidity remained high in the boundary layer, and incoming solar radiation got reduced. Also, NO_2 mixing ratio could have been reduced in the boundary layer through aqueous phase chemical conversion of NO_2 into nitric acid and its subsequent removal by precipitation. Consequently, the photochemical production of surface ozone was reduced. This has been another reason for low level of surface ozone over the Lumbini region.

The above paragraph reveals that there was abundance of water vapour in the boundary layer during the whole observational period. This provided conducive environment for the removal of surface ozone through chemical scavenging (Ali *et al.* 2009). It may be noted that the process of chemical scavenging starts with photolysis of ozone molecule by solar radiation at wavelength (λ) $\leq$ 320 nm giving rise to electronically excited singlet oxygen O($'$D). A small fraction of the produced singlet oxygen reacts with water vapour to yield hydroxyl radical (OH) (Buckley and Birks 1995). The available ozone molecules are scavenged with the OH radical to result into hydroperoxy radical (HO_2) and oxygen molecules. HO_2 further reacts with ozone molecule to yield OH radical and oxygen molecules. The reaction mechanisms involved are as given below:

$$O_3 + h\nu \rightarrow O('D) + O_2, \quad \lambda \leq 320 \text{ nm}, \quad (1)$$

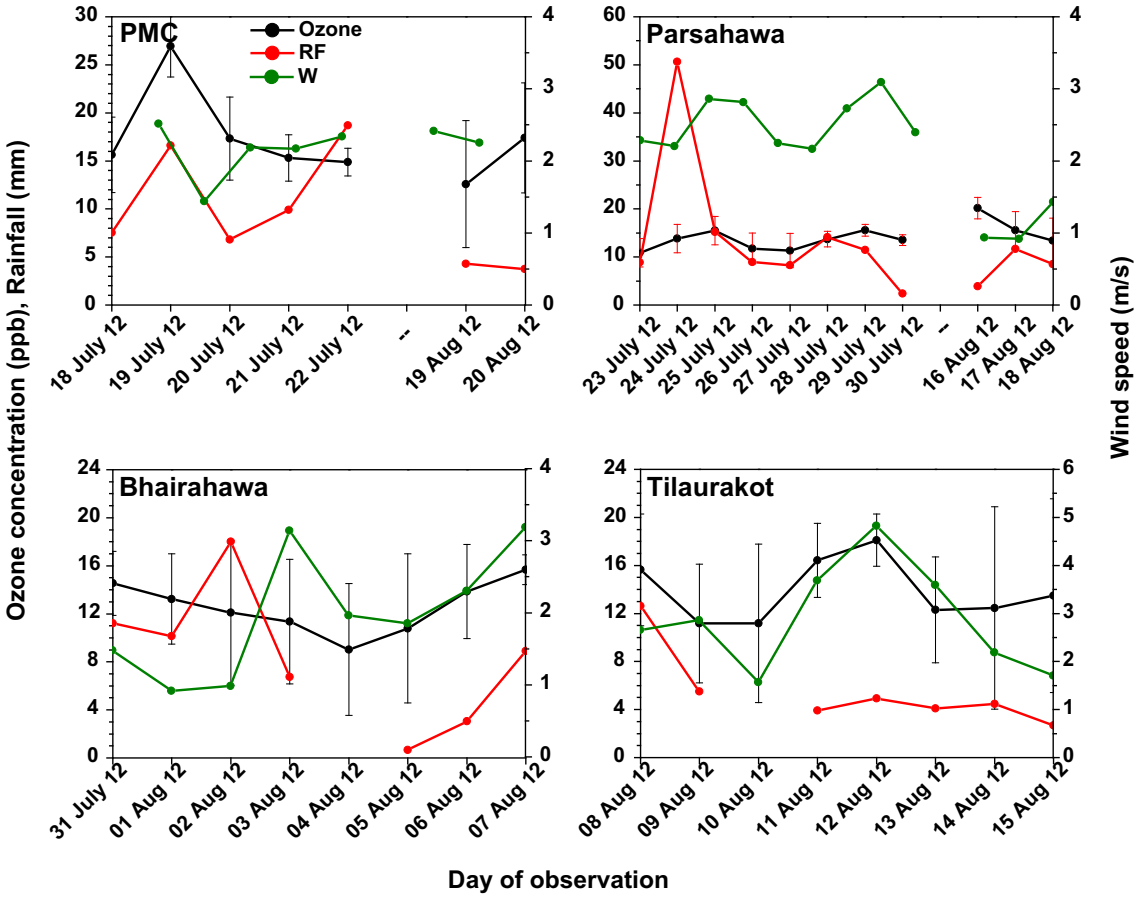

Figure 5. Daily mean mixing ratio of surface ozone, ECMWF ERA-Interim reanalysis rainfall (RF) and wind speed (W) at Panditarama Lumbini International Vipassana Meditation Centre (18–22 July and 19–20 August 2012), Parsahawa (23–30 July and 16–18 August 2012), Bhairahawa (31 July–7 August 2012) and Tilaurakot (8–15 August 2012) in the Lumbini Protected Zone, Nepal.

$$H_2O + O('D) \rightarrow 2OH, \quad (2)$$

$$O_3 + OH \rightarrow HO_2 + O_2, \quad (3)$$

$$HO_2 + O_3 \rightarrow OH + 2O_2. \quad (4)$$

Thus, chemical scavenging has also been responsible for low level of surface ozone over the Lumbini region. However, the reduction in the surface ozone by this mechanism might not have been significant due to the occurrence of cloudy sky or rain for most of the observational period.

3.4 *Diurnal variation of the mixing ratio of surface ozone*

Diurnal variation of surface ozone observed at PMC, Parsahawa, Bhairahawa and Tilaurakot is shown in figure 6. This figure reveals that the mixing ratio of ozone at all the locations rises from low value in the morning to peak value during midday and then decreases to low value somewhere in the night. The peak mixing ratio attained at these locations is 22, 16, 17 and 20 ppb, respectively occurring during 12:00–15:00 h. The variation matches well with the diurnal variation of boundary layer thickness which is known to have in-phase relation with the incoming solar radiation. The correlation coefficient between surface ozone mixing ratio and boundary layer thickness at PMC, Parsahawa, Bhairahawa and Tilaurakot is 0.93, 0.91, 0.96 and 0.97, respectively (statistically significant at better than 99%). The result implies that photochemistry is the main source mechanism of ozone in these areas.

The mixing ratio of surface ozone increases from the lower values in the morning to the peak values in the afternoon at the rate of 0.8 ppb/h at PMC, 0.4 ppb/h at Parsahawa, 0.8 ppb/h at Bhairahawa and 0.7 ppb/h at Tilaurakot. It

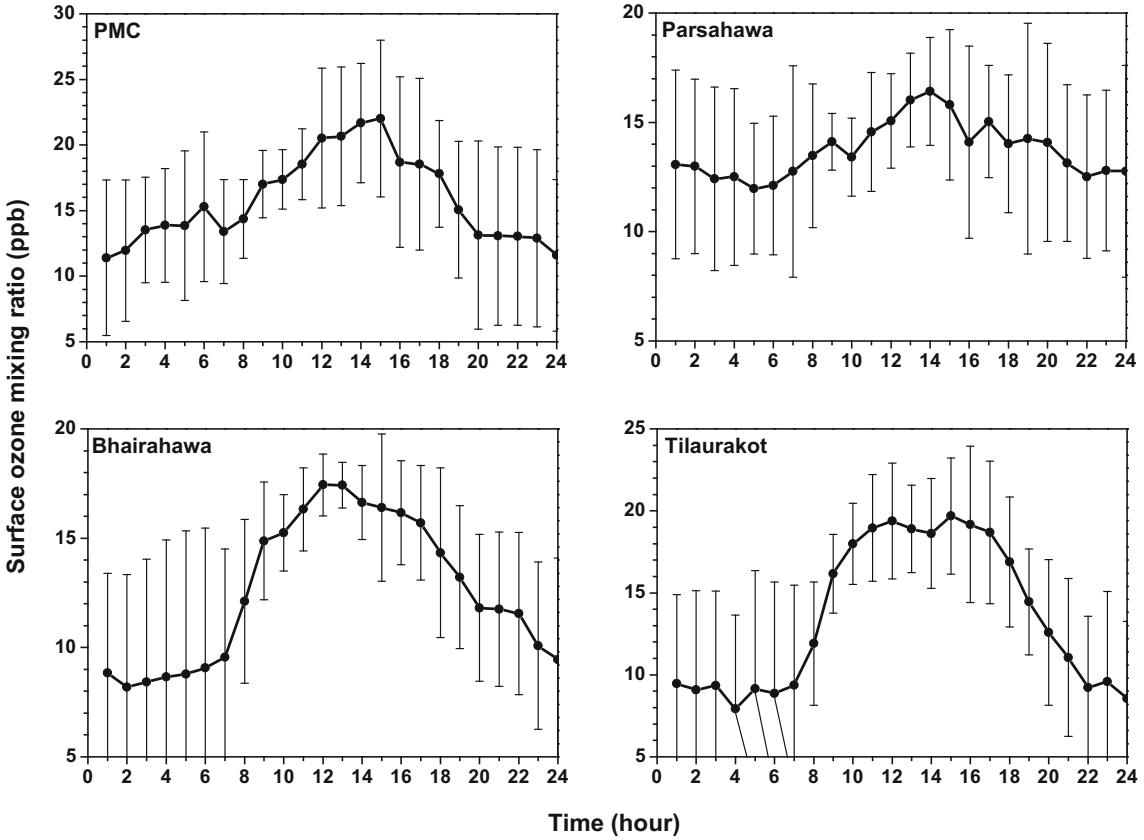

Figure 6. Diurnal variation of the mixing ratio of surface ozone at Panditarama Lumbini International Vipassana Meditation Centre, Parsahawa, Bhairahawa and Tilaurakot in the Lumbini Protected Zone, Nepal.

decreases from the peak values in the midday to the lower values in the night at the rate of 1.2, 0.4, 0.9 and 1.2 ppb/h. The result implies that the photochemistry is more active at PMC, Bhairahawa and Tilaurakot observational sites but less active at Parsahawa. The photochemistry at Parsahawa is less active due to less availability of incoming solar radiation caused by attenuation of the same either by clouds and rain of the monsoon season or by high aerosol loadings particularly in the coarse size range produced and transported mainly from the nearby industries during dry weather condition.

3.5 *Comparison of pollutants' levels*

Daily and diurnal variation of the concentration of particulate matter and mixing ratio of surface ozone at PMC, Parsahawa, Bhairahawa and Tilaurakot indicate that PMC and Tilaurakot fall in unpolluted areas. Tilaurakot comes in the cleanest area with respect to ozone. Consequently, Tilaurakot may be safely taken as a control or reference site for comparing pollution levels of the other areas in the LPZ. For comparison, the average percentage deviation of the concentration of pollutants at PMC, Parsahawa and Bhairahawa with respect to that at Tilaurakot is estimated. It is found that the coarse particles at Parsahawa and Bhairahawa have a very large positive deviation (∼223% and 83%, respectively). Fine particles at these locations also have an appreciable positive deviation (∼38% and 39%). However, the fine and the coarse particles at PMC show a negative deviation. This clearly indicates that Parsahawa may be treated as a dominant source region for coarse particles in the LPZ. Bhairahawa also acts as a potential source region for the coarse particles. Both these areas may be treated as a source region for fine particles also. The area of PMC is very clean with regard to particulate pollutants.

The percentage positive deviation in ozone mixing ratio is maximum at PMC (∼24%). This implies that the area of PMC intercepts elevated level of ozone in comparison to those at other locations in the LPZ, though remaining at a level far below the safe limit.

4. Conclusions

A pilot study is carried out on the physical characterisation of $PM_{2.5}$, PM_{10} and surface ozone measured at four different environmental locations in the LPZ, Nepal, during the period from 17 July to 21 August 2012. The following broad conclusions are drawn based on the study.

Air quality with regard to particulate matter is good at Panditarama Meditation Centre (monastic zone) and Tilaurakot (control area) but nearly bad at Bhairahawa (urban area) and Parsahawa (industrial area). The major sources of particulate emissions are local, but there is possibility of advective transport of particulate matter from Bhairahawa and Parsahawa areas to the nearby areas of the LPZ by prevailing easterly to southeasterly winds of the monsoon regime. Hence, control measures need to be taken to mitigate it before it adversely affects the clean areas of the LPZ. Boundary layer thickness plays a dominant role in the diurnal distribution of the particulate matter.

Comparison of the present analysis with that reported in the literature indicates that pollution level during the monsoon season is much lower than that during the pre-monsoon season.

There is no ozone pollution in the LPZ during the study period. The surface ozone level at PMC is high in comparison to that at the control site but is within the safe limit. Tilaurakot can be safely taken as a control site for comparison of the pollution status of the other areas of the LPZ.

Acknowledgements

The authors are grateful to the Director, IITM, Pune, for his kind support and guidance for carrying out this work. The authors are also grateful to Mr Terrence Thompson, Sr Advisor, Environmental Health, WHO, Nepal, for funding; Dr Sudan Panthi, WHO, Nepal, for his suggestion and guidance during the measurements of air quality parameters in the LPZ; and Mr Rajesh Kohar, technical support person appointed for this campaign by WHO, Nepal. The meteorological data obtained from ECMWF-ERA Interim are also acknowledged. Special thanks go to Guruji Bhikkhu Vivekananda and Guruma Sayalay Bhaddamanika of the PMC and authorities of the Lumbini Development Trust, Nepal, who provided all possible support to this programme.

References

Ali K, Beig G, Chate D M, Momin G A, Sahu S K and Safai P D 2009 Sink mechanism for significantly low level of ozone over the Arabian Sea during monsoon; *J. Geophys. Res.* **114** D17306, https://doi.org/10.1029/2008JD011256.

Brimblecombe P 2003 *The Effects of Air Pollution on the Built Environment*; Imperial College Press, London.

Buckley P T and Birks J W 1995 Evaluation of visible-light photolysis of ozone water-cluster molecules as source of atmospheric hydroxyl radical and hydrogen peroxide; *Atmos. Environ.* **29** 2409–2415, https://doi.org/10.1016/1352-2310(95)91997-7.

Burney J and Ramanathan V 2014 Recent climate and air pollution impacts on Indian agriculture; *Proc. Nat. Acad. Sci. USA* **111** 16319–16324, https://doi.org/10.1073/pnas.1317275111.

Colvile R N, Hutchinson E J, Mindell J S and Warren R F 2001 The transport sector as a source of air pollution; *Atmos. Environ.* **35** 1537–1565.

Forouzanfar M H, Alexander L, Anderson H R, Bachman V F, Biryukov S, Brauer M, Burnett R, Casey D, Coates M M and Cohen A 2015 Global, regional, and national comparative risk assessment of 79 behavioural, environmental and occupational, and metabolic risks or clusters of risks in 188 countries, 1990–2013: A systematic analysis for the global burden of disease study 2013; *Lancet* **386** 2287–2323, https://doi.org/10.1016/S0140-6736(15)00128-2.

Fuzzi S, Baltensperger U, Carslaw K, Decesari S, Denier Van Der Gon H, Facchini M C, Fowler D, Koren I, Langford B, Lohmann U and Nemitz E 2015 Particulate matter, air quality and climate: Lessons learned and future needs; *Atmos. Chem. Phys.* **15** 8217–8299.

Ghio A J and Devlin R B 2001 Inflammatory lung injury after bronchial instillation of air pollution particles; *Am. J. Respir. Crit. Care Med.* **164** 704–708.

Hibberd M F and Luhar A K 1996 A laboratory study and improved PDF model of fumigation into a growing convective boundary layer; *Atmos. Environ.* **30(21)** 3633–3649.

IUCN (International Union for Conservation of Nature) Nepal 2012 Environmental impact assessment of industrial development around Lumbini, the birthplace of the Lord Buddha, World Heritage Property, Report prepared for UNESCO, Kathmandu, Nepal, 8p.

Ivaskova M, Kotes P and Bradman M 2015 Air pollution as important factor in construction materials deterioration in Slovak Republic; *Proc. Eng.* **108** 131–138.

Janssen N A H, Hoek G, Simic-Lawson M, Fischer P, Bree L, Brink H, Keuken M, Atkinson R W, Anderson H R, Brunkreef B and Cassee F R 2013 Black carbon as an additional indicator of the adverse health effects of airborne particles compared with PM_{10} and $PM_{2.5}$; *Environ. Health Perspect.* **119** 1691–1699.

Kelly J F and Fussell J C 2015 Air pollution and public health: Emerging hazards and improved understanding of risk; *Environ. Geochem. Health* **37** 631–649.

Mohnen V A, Goldstein W and Wang W C 1993 Tropospheric ozone and climate change; *Air Waste* **43(10)** 1332–1334, https://doi.org/10.1080/1073161x.1993.10467207.

Nicolas J, Yubero E, Galindo N, Gimenez J, Castaner R, Carratala A, Crespo J and Pastor C 2009 Characterization of events by aerosol mass size distribution; *J. Environ. Monit.* **11** 394–399.

Nishanth T, Ojha N, Satheesh Kumar M K and Naja M 2011 Influence of solar eclipse of 15 January 2010 on surface ozone; *Atmos. Environ.* **45** 1752–1758.

Orru H, Ebi K L and Forsberg B 2017 The interplay of climate and air pollution on health; *Curr. Environ. Health Report* **4** 504–513, https://doi.org/10.1007/s40572-017-0168-6.

Rupakheti D, Adhikary B, Praveen P S, Rupakheti M, Kang S, Mahata K S, Naja M, Zhang Q, Panday A K and Lawrence M G 2017 Pre-monsoon air quality over Lumbini, a world heritage site along the Himalayan foothills; *Atmos. Chem. Phys.* **17** 11041–11063, https://doi.org/10.5194/acp-17-11041-2017.

Shindell D, Kuylenstierna J C, Vignati E, van Dingenen R, Amann M, Klimont Z, Anenberg S C, Muller N, Janssens-Maenhout G and Raes F 2012 Simultaneously mitigating near-term climate change and improving human health and food security; *Science* **335** 183–189.

Stocker T F, Qin D, Plattner G-K, Tignor M, Allen S K, Boschung J, Nauels A, Xia Y, Bex V and Midgley P M 2013 IPCC: Climate change 2013: The physical science basis; In: *Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change,* Cambridge University Press, Cambridge, UK and New York, NY, USA, 1535p.

Turo F D, Proietti C, Screpanti A, Fornasier M F, Cionni I, Favero G and De Marco A 2016 Impacts of air pollution on cultural heritage corrosion at European level: What has been achieved and what are the future scenarios; *Environ. Pollut.* **218** 586–594.

Wu S, Deng F, Wang X, Wei H, Shima M, Huang J, Lv H, Hao Y, Zheng C, Qin Y, Lu X and Guo X 2013 Association of lung function in a panel of young health adults with various chemical components of ambient fine particulate air pollution in Beijing, China; *Atmos. Environ.* **77** 873–884.

Corresponding editor: Suresh Babu