2017/07/03

Dear Editor,

We would like to express our sincere thanks for serving as the editor for our work.

We would like to inform you that we have revised the manuscript significantly based on the comments by the two reviewers. New interpretation and analysis along with some new figures have been done to answer the questions raised by the reviewers. The “Introduction” section has been shortened based on the suggestions from one of the reviewer. At the same time, the purpose behind incorporating the model in our study has also been defined clearly.

We, at this stage, strongly believe that the quality of the manuscript has improved and look forward to hearing positive result from your side.

Sincerely Yours,

Dipesh Rupakheti and Prof. Shichang Kang, on behalf of all coauthors

Pre-monsoon air quality over Lumbini, a world heritage site along the Himalayan foothills

by D. Rupakheti et al.

Review of Rupakheti et al. (Report #1, Anonymous Referee #4)

The authors present PM, BC, CO and O_3 concentrations measured at Lumbini during April-June 2013 and explained meteorology, pollutant concentrations by conducting WRF-Stem model simulation. They also estimated the regional contributions of CO and aerosol composition to local air quality based only on the model simulation results. This reviewer full agree that the presented observational data set in this study are unique and very useful to understating the level of air pollution in the study area. However, in this revised manuscript, there are several important issues on model simulation and scientific discussion. Therefore, this revised manuscript cannot be accepted in its current form. Before publishing in ACP, several points should be clarified.

We would like to thank the reviewer for his/her constructive comments and suggestions. It seems to us that the reviewer provided suggestions based on the ACPD version (date: 17th June, 2016) of the manuscript which makes it difficult to address all of the concerns. For example, based on the earlier reviewers' comments/suggestions, model based aerosol chemical composition has already been removed. However, we have tried our best to accommodate all of the suggestions to the possible extent. Please find the reviewer's comments in black and our replies in blue. The changes in the revised manuscript are colored in red.

Specific comments and suggestions are below:

L68, Fig 1: Information given in Figs 1, 2 and 5 are overlapped. This reviewer recommends to merge Fig 1 and Fig 5. That is, plot both monthly mean AOD and winds for separately in April, May and June. Those figures will give more direct insight on aerosol distribution and regional-scale circulation during the intensive measurement period. Fig 2 is not necessary in main body text, so please move it to the supplement.

As suggested by the reviewer, Figure1 and Figure 5 have been merged which is the new Figure 1 in the revised version of the manuscript. Likewise, Figure 2 has been moved to the supplementary information section as Figure S1.

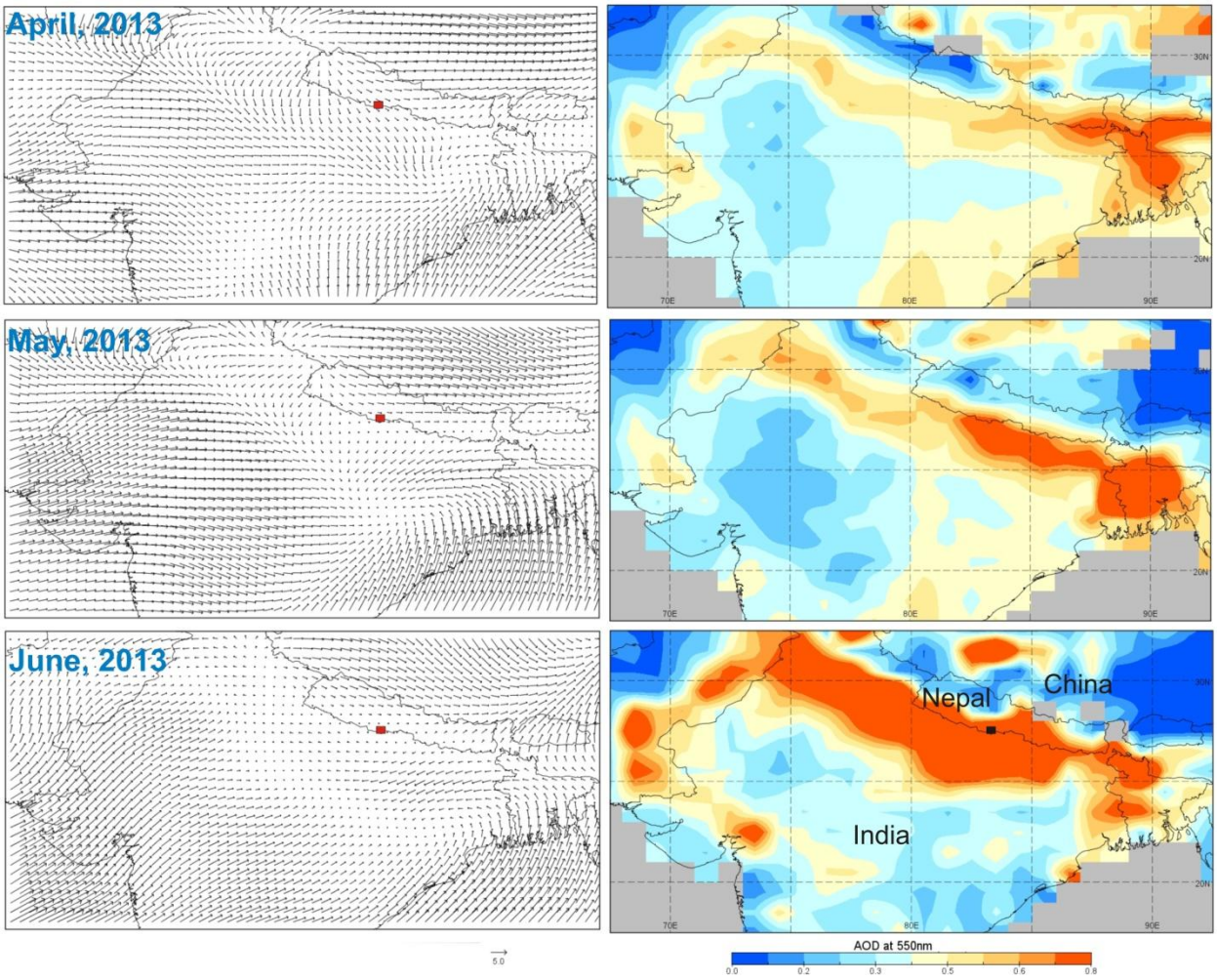

Figure 1. Monthly synoptic wind (at 1000 hPa) for April, May and June 2013, based on NCEP/NCAR reanalysis data where the orientations of arrows refer to wind direction and the length of arrows represents the magnitude of wind (m/s). Red square box in the figure (left) represents the location of Lumbini. Figures on the right column represent monthly aerosol optical depth acquired with the MODIS instrument aboard TERRA satellite. High aerosol loading can be seen over the entire Ingo-Gangetic Plains (IGP). Light gray color used in the figure represents the absence of data.

L68, Fig 1: Which version of MODIS TERRA data have used? Why the authors are not used MODIS Aqua data?

Version 6 (Level 3) of the MODIS TERRA AOD data has been used in this study. We looked at both Aqua and Terra images, which are as follows. They are not significantly different. The difference between the AOD values from two satellites could be due to the true diurnal signal or the retrieval error (Wang et al., 2010).

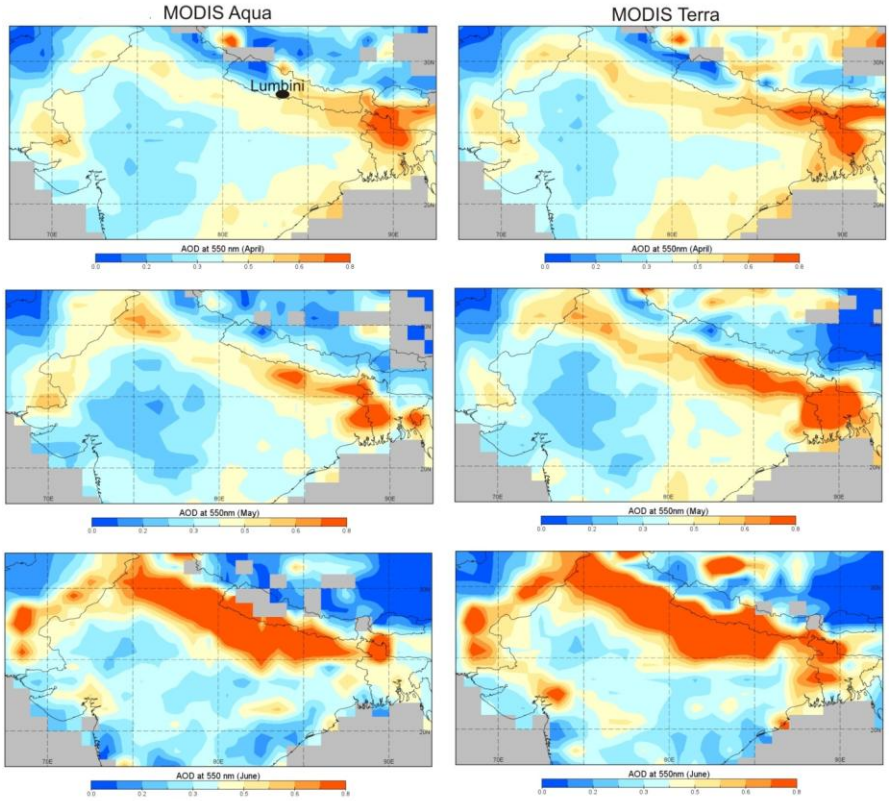

Figure: Monthly average MODIS Aqua and Terra AOD over the South Asian region during April-June, 2013. The black dot indicates the location of Lumbini.

Reference:

Wang, L., Wang, Y., Xin, J., Li, Z., & Wang, X. (2010). Assessment and comparison of three years of Terra and Aqua MODIS Aerosol Optical Depth Retrieval (C005) in Chinese terrestrial regions. Atmospheric Research, 97(1), 229-240.

L54-139: This reviewer recommends to reduce the length of INTRODUCTION section with deleting sentences are not closely related the topic of this paper. For example, the authors emphasize many times in the paper that Lumbini is a UNESCO world heritage. This is interested to the authors, but not to all readers. So please minimize the statements on this.

&

L 99 ~ : As the authors mentioned, sulfuric acid is more critically important in historical heritages. Please more carefully and clearly explain why the air pollutants presented in this study is important should added in the INTRODUCTION.

Agreed. The unwanted length of the Introduction section has been reduced along with the minimal use of information on UNESCO world heritage in this section. The deleted sentences are indicated with the strikethrough. In addition, the significance of the monitored species has been mentioned in the Introduction.

L128: remove "Aerosol optical depth – not discussed on the present study". This is not necessary here.

Agreed and has been removed.

L 165: Table 1 - The sampling period should be more clearly clarified. As shown in Fig. 6, all instruments had not properly operated during the study period.

Done. All instruments ran successfully for the entire duration of the campaign except PM instrument. The following text has been inserted in Section 3.2.1.

"The gap in the figure (for PM time series) is due to the power interruption to the instrument."

L169: What's the uncertainty of PM concentration measured with GRIMM EDM164? Especially the quantitative uncertainty of EDM164 for such a high PM concentration level should be discussed, because PM concentration in here is estimated from light scattering measurements.

PM concentrations have been observed highly variable as evident in the standard deviations. For example, variability in PM_1, $PM_{2.5}$ and PM_{10} concentration for the month of April is 67.8 %, 60.2 % and 61.8 % respectively from their mean. Similar values were obtained during the month of

May whereas lower values were obtained for the month of June (PM_1: 43.6%, $PM_{2.5}$: 45.3 % and PM_{10}: 54.1 %). The instrument deployed for PM concentration measurement (GRIMM EDM164) is able to measure the particles mass concentration in the range of 0.1-6000 µg/m^3 with the size ranging between 0.25-32 µm with an accuracy of ±5% over the entire measurement range (GRIMM EDM-164 manual; available at : http://wiki.grimm-aerosol.de/images/3/31/GRIMM_EDM_164_datasheet.pdf). Given the high range of the instrument for PM concentration monitoring as compared to that found on our study site, we strongly believe that the data provided by the instrument is highly trustworthy.

Figures 3 & 6 : There is large differences between observations and model simulations. First, more specific explanation and discussion on why the model results were not well agreed with the observations must be addressed. Why the WRF-STEM simulation cannot well simulate the precipitation events and why there is big difference in RH, WD and WS. Why BC is too underestimated compare to the aethalometer data? This should be made for all parameters. This is very important to convincing the results given in Section 3.3 and Section 3.4, as the authors mentioned in L261-262.

The revised version of the paper (submitted on 10th February, 2017) has already addressed this issue. For your information, various sentences on observation and model comparison (on the concentration part) were already removed as suggested by the previous reviewers. In addition, we have applied some general statistics to understand the relationship between observed and modeled species. Please see our response to another reviewer.

Since this is not only modeling based study, it is beyond the scope of the current paper to do sensitivity analysis with different physics scheme or initial and boundary conditions to improve the meteorological prediction. Besides, comparing one station data point with model grid representing 25x25 km is always difficult. We present the comparison results of model to observation to indicate model performance over Lumbini region, not as model validation. We do not have the local emissions inventory and thus we are using the global EDGAR emission in our model. There are plenty of published papers that have used global EDGAR emissions for regional modeling analysis.

Figure 3: WD should NOT be plotted with solid line, because, for example, WD at 355 and 5 degree is almost the same direction. So make a plot with dots.

Agree. The line graph for the WD has been replaced by the dots.

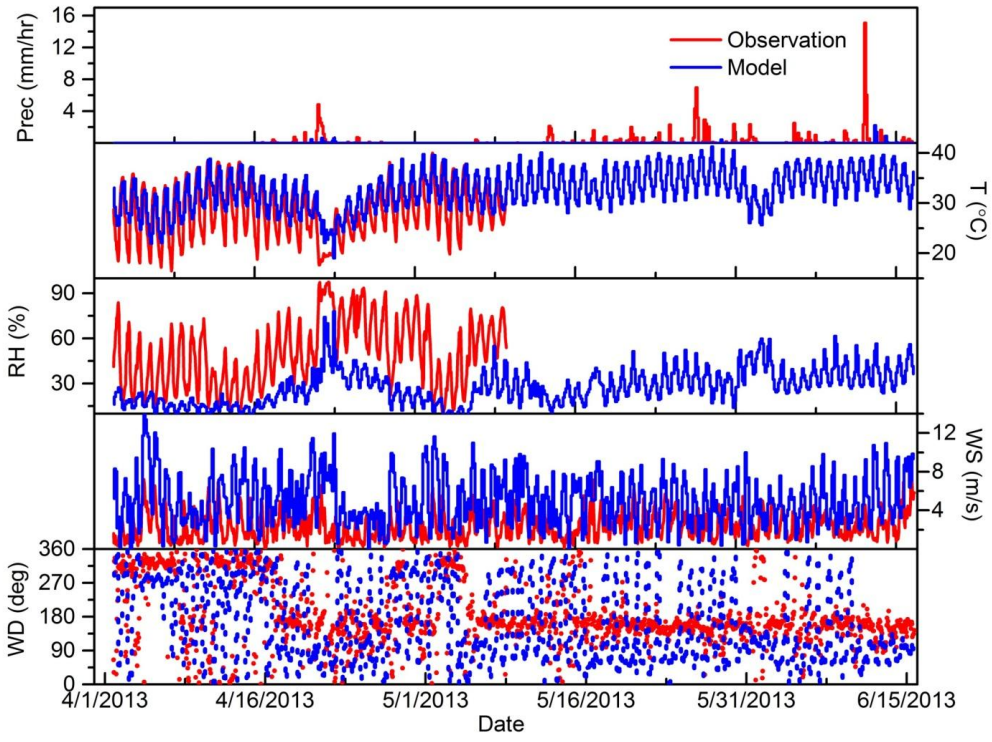

Figure 2. Time series of hourly average observed (red) and model estimated (blue) meteorological parameters at Lumbini, Nepal for the entire measurement period during 1 April to 15 June 2013

L242-243: It is hard to agree to this argument. As shown in Fig. 3, there is large difference between modeled and observed wind speed.

This sentence was already revised during the first revision. We would like to reiterate that the model was able to capture the pattern but not the magnitude; the magnitude by the model is overestimated as compared to the observation which is a common feature of WRF model. Past studies have also proved that the WRF model generally overestimates the WS (Borge et al., 2008; Mohan & Bhati, 2011; Hu et al., 2013; Gunwani & Mohan, 2017).

References

Borge, R., Alexandrov, V., Del Vas, J. J., Lumbreras, J., & Rodríguez, E. (2008). A comprehensive sensitivity analysis of the WRF model for air quality applications over the Iberian Peninsula. Atmospheric Environment, 42(37), 8560-8574.

Mohan, M., & Bhati, S. (2011). Analysis of WRF model performance over subtropical region of Delhi, India. Advances in Meteorology, 2011.

Hu, X. M., Klein, P. M., & Xue, M. (2013). Evaluation of the updated YSU planetary boundary layer scheme within WRF for wind resource and air quality assessments. Journal of Geophysical Research: Atmospheres, 118(18).

Gunwani, P., & Mohan, M. (2017). Sensitivity of WRF model estimates to various PBL parameterizations in different climatic zones over India. Atmospheric Research, 194, 43-65.

L258-259: This reviewer also cannot agree to this conclusive sentence. Apparently, the observed RH is two times higher than the modeled one. There is no evidence that RH by model is how well captured the regional variation. Is there a reference data to back this up?

RH values are highly underestimated by the model, however as previously mentioned as in the case of temperature (Section 3.1); the model does not show significant changes in RH during the measurement campaign when the observations stopped working.

L265- Figure 5: How about the winds at 850 hPa or 700 hPa pressure level?

New figures on the winds at 850 hPa have been plotted as follows. Please see the revised text for the discussion.

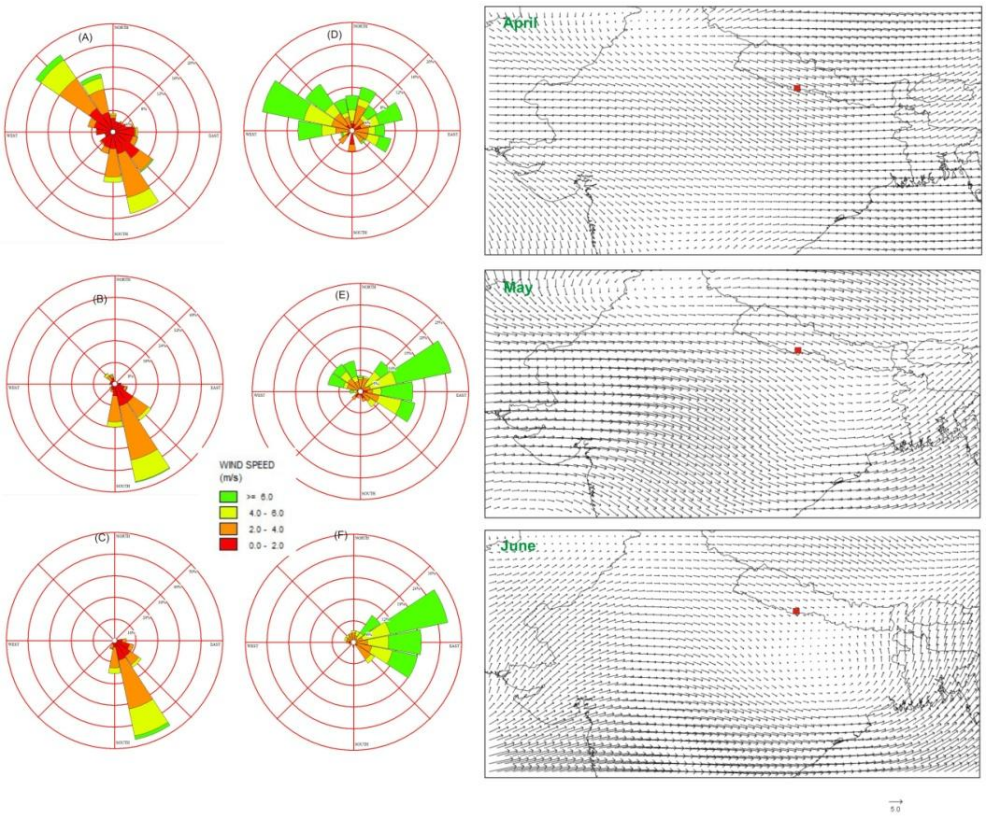

Figure S3: Wind rose of wind speed and wind direction obtained from the observation (A, B, C) and from the model (D, E, F) for the months of April, May and June 2013 respectively. The right panel shows the synoptic scale wind (850 hPa) during three months of the campaign.

L266-267: Here, what "calm winds" means? This discussion in Figure 6 is conflict the winds discussed in Figures 3 and 4. Please clarify.

"Calm winds" has been replaced by weak winds. The wind direction in Figure 3 (now Figure 2) has been replaced with the dot plot whereas Figure 4 deals with the monthly wind rose plot. However, no weather parameters have been plotted in Figure 6 as the reviewer has indicated. So, we are unable to address this comment.

Figure 6: As commented above, more explanations are needed why there is a large discrepancy between modeled and observed values. Without clarifying this, the results given in the next sections (sections 3.3.2. and 3.4) are not truly reliable.

As previous reviewers have also indicated this, the model output results and associated text have already been revised with unrelated text removed.

L384-385: This reviewer understands the PBL height observation was not available during the measurement period. However, the modeled PBL height has also large uncertainty and not believes it. The authors cited several previous works, but need to add some information on the PBL height, not general seasonal characteristics.

As suggested by the reviewer, we have added following sentences in the PBL description section:

The daily average PBL height obtained from the model is compared with published values (*Wan et al., 2017*) which indicate that the value is captured by our model during initial measurement period and overestimated in the months of mid May onwards. The monthly average diurnal variation also showed that the boundary layer height was maximum during 15:00 local time which coincides with the period of lowest concentration of the pollutants.

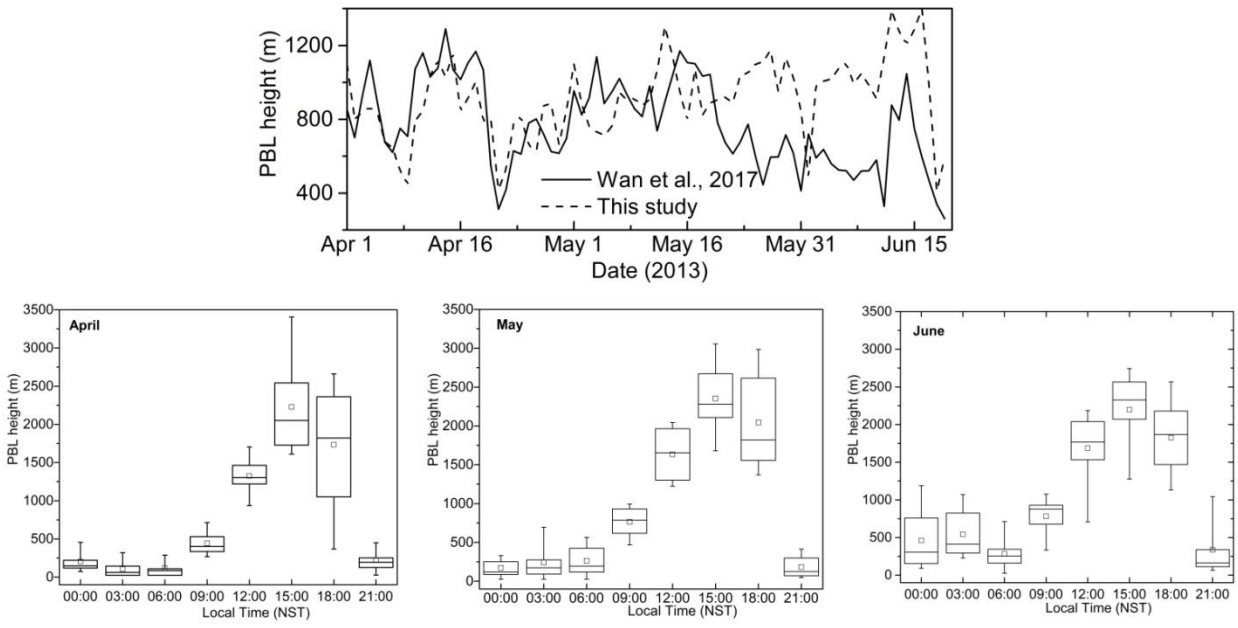

Figure 6. Daily time series of PBL height obtained from the model and reported values over Lumbini (obtained from Wan et al., 2017). The lower panel shows the monthly average diurnal variation of the PBL height. The square mark in each box represents the mean PBL height,

bottom and top of the box represents 25th and 75th percentile, top and bottom of the whisker represents 90th and 10th percentile respectively.

Reference

*Wan, X., Kang, S., Li, Q., Rupakheti, D., Zhang, Q., Guo, J., . . . Cong, Z. (2017). Organic molecular tracers in the atmospheric aerosols from Lumbini, Nepal, in the northern Indo-Gangetic Plain: Influence of biomass burning. Atmos. Chem. Phys. Discuss., 2017, 1-40. doi:10.5194/acp-2016-1176 (**Accepted for ACP**).*

L407-408: The authors mentioned 'Global Monthly Fire Location Products' were used. However, daily data were used in Figure 9. Please clarify this.

Thanks for pointing out the mistake. The daily data on forest fire were obtained from the FIRMS platform of NASA Earthdata. Correction has been done.

L416: Clearly present how much higher? This is very vague sentence.

We have revised the sentence as:

High AAE values (~ 1.6) during these two events are also an indication of presence of BC of biomass burning origin.

L421: Quality of Figure 10 is very bad. It's hard to read.

We replaced it with high-resolution figure which is now Figure 8. The new figure is given below.

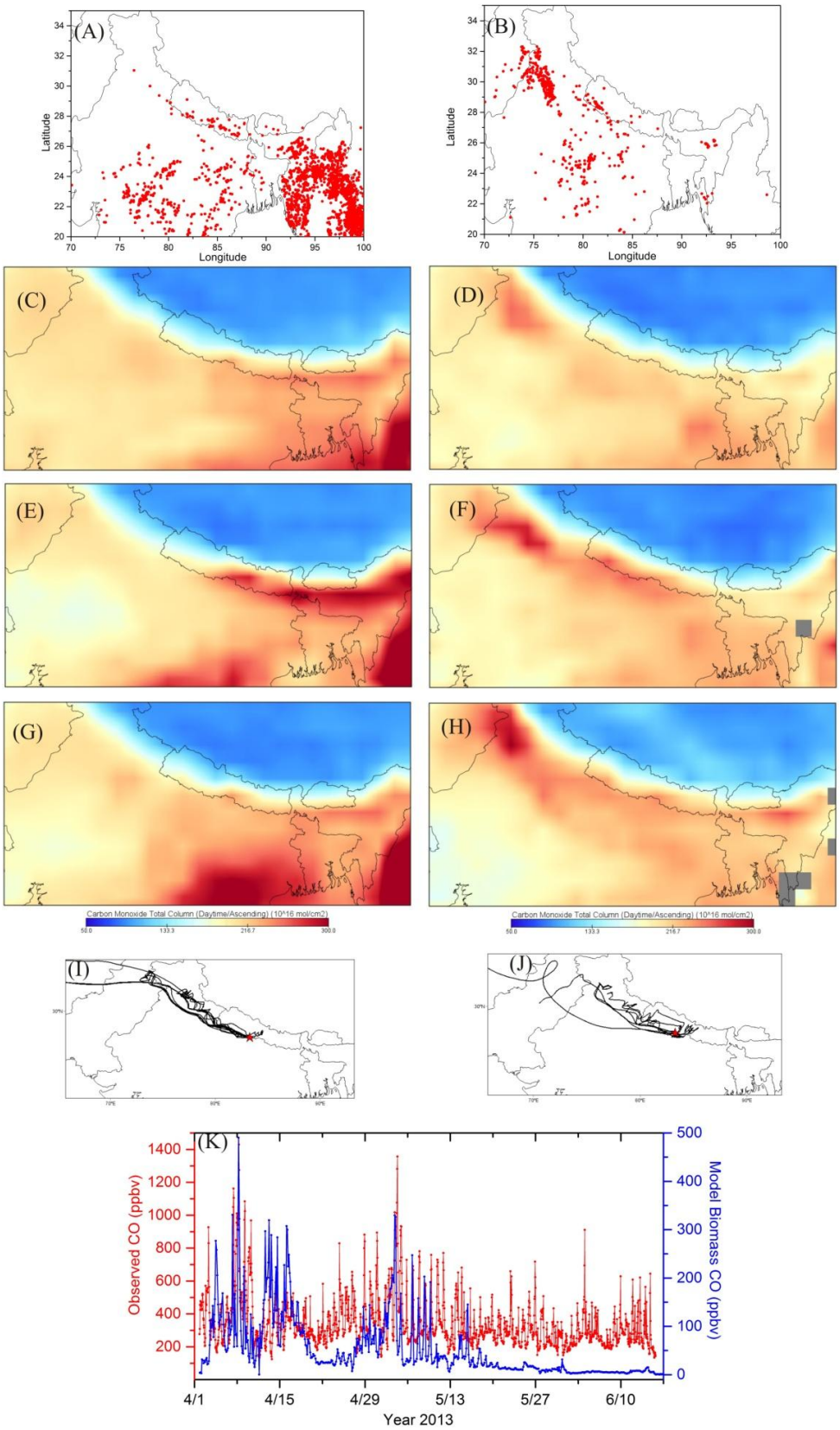
(A)
(B)
Latitude
Longitude
(C)
(D)
(E)
(F)
(G)
(H)
Carbon Monoxide Total Column (Daytime/Ascending) (10^16 mol/cm2)
50.0
133.3
216.7
300.0
(I)
(J)
(K)
Observed CO (ppbv)
Model Biomass CO (ppbv)
Year 2013
4/1
4/15
4/29
5/13
5/27
6/10

Figure 8. Active fire hotspots in the region acquired with the MODIS instrument on Aqua satellite during (A) Event-I (7-9 April) and (B) Event-II (3-4 May). CO emissions, acquired with AIRS satellite, in the region two days before (3-5 April), during (7-9 April) and two days after (10-12 April) Event-I are shown in panels (C), (E) and (G), respectively while panels (D), (F) and (H) show CO emissions two days before (1-2 May), during (3-4 May) and two days after (5-6 May) the Event-II. Panels (I) and (J) represent the 6-hr interval HYSPLIT back trajectories during Event I and II, respectively. Location of the Lumbini site is indicated by the red star in the panel (I and J). Observed CO versus Model open burning CO illustrating the contribution of forest fires during peak CO loading is shown in panel (K).

Figure 10: How the authors get the modelled biomass CO concentration? Generally the modelled biomass CO concentration is not capture the observed CO concentration. What is the major reason that the author gusse? This reviewer cannot agree to the sentence given in Li435-436.

STEM model can tag the CO emissions originating from biomass burning separately. Comparison of the model to observed CO is done by adding the biomass CO and anthropogenic CO together. This is a standard modeling technique employed by other air quality models as well. Since anthropogenic emissions do not change significantly in a weekly time scale and the temporal variability of the modeled biomass CO matches the temporal variability in the observation, it is inferred that the peak CO events are due to biomass burning. Meteorology could have played a role but we do not see a huge difference in modeled meteorology during the campaign period and observations are not available throughout the campaign.

Figure 10 (I) and (J): Instead of wind roses, regional-scale wind patterns will be more helpful to understand the transport in the interested region.

Thank you for pointing this. From the ground-based observation, we see the local winds coming from the south (for Event-I) whereas the HYSPLIT air mass and synoptic wind both showed that the air mass passed over the fire events in NW IGP. We have replaced the wind rose with the HYSPLIT back trajectories and corrected the text accordingly in section 3.3.1. In addition, the figures for regional-scale wind pattern (during these two events) have been provided in Figure S8 (supplementary materials).

L441-450 and Figure 11: The two ozone peaks were possible contributed by local pollution, induced by NO2, but also by the transport from the fire plume. However, satellite NO2 data shown in Figure 11 is not direct evidence of the effects of fires on high ozone concentration. Clarify this.

It is likely that the local pollution as well as regional pollution (transported from NW IGP region, as indicated by synoptic wind in Fig S8) contributed to the ozone peak. However, we are not able to quantify the individual contributions, even with the model simulation because ozone was not simulated in this experiment. These statements have been added to the text in Section 3.3.1.

L455 and Figure 12: It should be provided how the authors calculated the contributions from different countries? Can you provide PSCF results to back this up? In addition, have the authors estimated other pollutants (i.e., PM2.5 and PM10) for their contribution like CO?

The source regions are identified using CO as a tracer as this is the standard techniques employed by other air quality models (Liu et al., 2003; Price et al., 2003; Pfister et al., 2005; Chen et al., 2009). CO emissions can be tagged according to the country of origin or any given area in the model and subsequently calculate the resulting concentration. The result shown in Figure 10 (current revised version) is done using this methodology. PSCF results are beyond the scope of the current manuscript. We haven't estimated the source regions for other pollutants like PM.

References

Chen, D., Wang, Y., McElroy, M. B., He, K., Yantosca, R. M., & Sager, P. L. (2009). Regional CO pollution and export in China simulated by the high-resolution nested-grid GEOS-Chem model. Atmospheric Chemistry and Physics, 9(11), 3825-3839.

Price, H. U., Jaffe, D. A., Doskey, P. V., McKendry, I., & Anderson, T. L. (2003). Vertical profiles of O3, aerosols, CO and NMHCs in the northeast Pacific during the TRACE-P and ACE-Asia experiments. Journal of Geophysical Research: Atmospheres, 108(D20).

Pfister, G., Hess, P. G., Emmons, L. K., Lamarque, J. F., Wiedinmyer, C., Edwards, D. P., ... & Sachse, G. W. (2005). Quantifying CO emissions from the 2004 Alaskan wildfires using MOPITT CO data. Geophysical Research Letters, 32(11).

Liu, H., Jacob, D. J., Bey, I., Yantosca, R. M., Duncan, B. N., & Sachse, G. W. (2003). Transport pathways for Asian pollution outflow over the Pacific: Interannual and seasonal variations. Journal of Geophysical Research: Atmospheres, 108(D20).

Figure 14: The authors only showed wavelength dependency of normalized light-absorption coefficients for two time periods. First question is why the authors do not show all times? The authors can present with time (x-axis), wavelength (y-axis) and normalized light-absorption coefficient with different color. This figure will be more helpful to understand the difference of light-absorption coefficient in around 380 nm wavelength. Second, the difference at 380 nm in current Figure 14 is statistically significant? And how many data points were used? Last question for Figure 12 is that data during the fire periods discussed section 3.2 were included or excluded here?

The time period selected for Figure 14 (now Figure 11) refer to the periods when the BC concentration was highest and lowest as inferred from the diurnal variation of the BC (Figure 5). Our interest was to study the inclination of the curve during biomass burning dominated period (highest peak in the morning) and fossil fuel dominated period (during afternoon since there is the absence of cooking activities) in Lumbini. We chose those two periods of the day in this study. A new figure has been drawn (shown below) to understand the time series of the normalized light absorption which clearly indicates the highest values of the light absorption at the lowest wavelength. However, to our understanding, this figure possesses difficulties for comparison with reported normalized light absorption values (from literature) which leads us to retain the original figure. But we have included the normalized curve for both of the events which clearly indicated the inclination towards the biomass burning curve. The difference at 380 nm in Figure 11 is statistically insignificant at $p<0.05$. The number of data points used for the cooking and non-cooking periods is 58 for each (excluding the data during two events). Figure 12 does not demonstrate the influence of forest fire, thus we are unable to provide our response to the later part of the query.

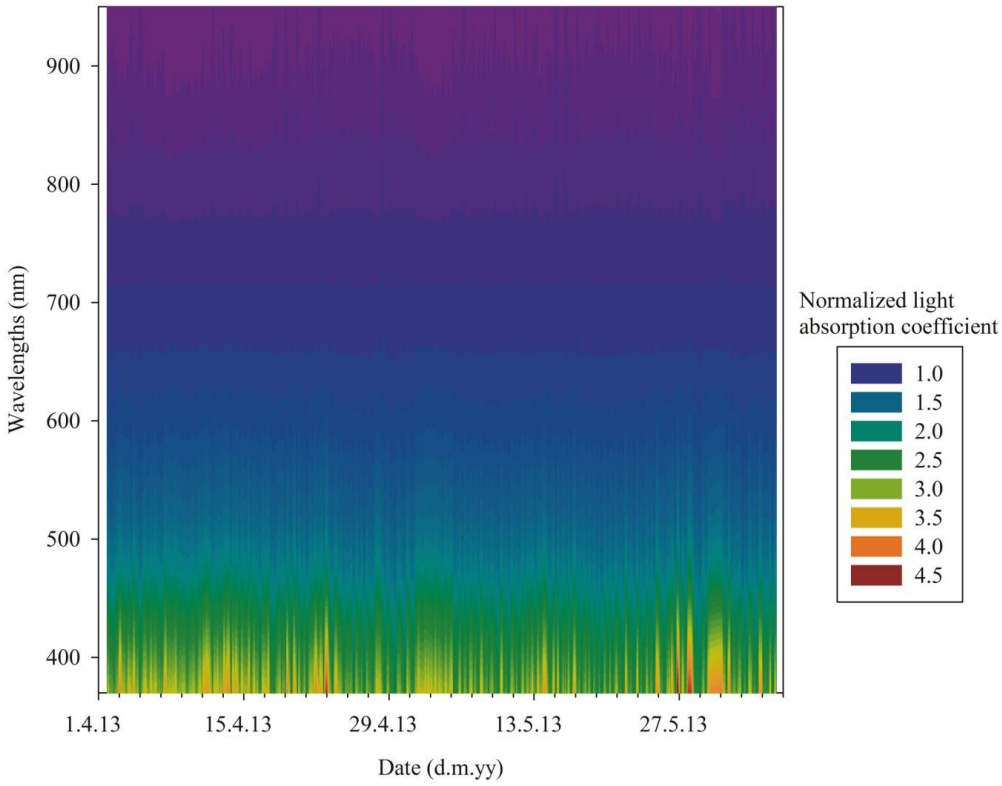

Figure: Time series of the normalized light absorption coefficient (normalized at 700nm) observed at Lumbini during the pre-monsoon of 2013.

Review of Rupakheti et al. (Report #2, Anonymous Referee #3)

Comments

Overall comment: The authors have improved the MS significantly, especially the monitoring data analysis and interpretation. The major uncertainty however still remains with the modeling part and it was, as pointed out by the authors, mainly due to the emission input data.

We would like to thank the reviewer for his/her comments/suggestions on our work. Please find the reviewer's comments in black and our replies in blue. The changes in the revised manuscript are colored in red.

Major comments:

It is not clear how authors extracted/projected the emission provided by EDGAR-HTAP_v2 for the simulation period and how the emissions were segregated temporally (hourly, daily etc.) for the simulation.

Emissions provided by the EDGAR-HTAP_v2 were re-gridded using four point interpolation technique available in the STEM model emissions preprocessor. The STEM model has been used extensively to study air pollution in Asia. The model has parameterized diurnal emission profile built in the emission preprocessor. Biomass emissions vary on a daily basis as per the burning event detected by FINN model while other anthropogenic emissions are constant over the year without seasonality.

One of the reasons of the discrepancy between the modeled and monitored levels is the point-based monitoring as compared to model produced grid average values. However, the model significantly underestimated all species, especially PM. Even CO levels were not reasonably produced as shown in Figure 6, the two events were not reproduced that well (as stated in line 509) as the modeling results appear to be fluctuating continuously during the period.

Why modeled PM levels are not presented in Figure 6? It is suggested that authors include scatter plots to show the relationship between the monitoring and modeling results for each species in Figure 6. Due to the uncertainty in the model output, all the results and discussion based on the model results may be questionable, i.e. those discussed in Section 3.3.2 (line 524).

The PM composition and quantity discussion are removed in the current version of the manuscript based on the suggestions provided by the previous reviewers. To show improvement in emission inventory and model development, PM comparison statistics between model and observations are shown in Table 3.

Section 2.3: provide the reasons why this particular model was selected, i.e. if it performs better than other models for the region etc., and how the emission input data was prepared for the modeling period.

STEM model has been used extensively to study air pollution in Asia and other parts of the world since its creation in 1987. The model PI, Professor Gregory R. Carmichael has more than 25000 paper citations (Google Scholar Search) mostly based on STEM model studies. We believe that all models have strengths and weaknesses. The authors chose this model because of familiarity with this model.

Emissions provided by the EDGAR-HTAP_v2 were projected using the four point interpolation technique available in the STEM model emissions preprocessor. The model has parameterized diurnal emission profile built in the emission preprocessor. Biomass emissions can vary on a daily basis as per the burning event detected by FINN model while other anthropogenic emissions are constant over the year without seasonality.

Section 3.1: WRF overestimated temperature and wind speed, underestimated precipitation and RH. Common statistical measures should be used to assess the model performance. For wind direction, because of the circular scale of the measurements (near 0 and near 360 degrees are almost the same) the interpretation of the time series should be made with caution or should be avoided. The comparison should be made for different wind sectors or simply by comparing the modelled windroses with the observed windroses to be presented along in Figure 4.

Agree. Based on the suggestion of the reviewer, we calculated correlation, Root Mean Square Error (RMSE) and Mean Absolute Difference (MAD) for the observed and modeled meteorological parameters. The correlation (r) for wind direction, wind speed, temperature and relative humidity were found to be 0.18, 0.22, 0.87 and 0.71 (all values at $P<0.001$) respectively.

Similarly, Root Mean Square Error (RMSE) and Mean Absolute Difference (MAD) were also calculated for the meteorological parameters. RMSE (MAD) values were found as 121.16 (105.67), 3.55 (3.10), 31.66 (27.24), 3.94 (3.28) for wind direction, wind speed, relative humidity and temperature respectively. The values obtained in our study are comparable with those from Delhi during summer as reported in Mohan & Bhati (2011) using the WRF model.

Considering the circular scale of the wind direction measurement, we have replaced the line plot previously used for the time series of the wind speed by the dots. This suggestion was provided by another reviewer as well. Now, based on the suggestion by this reviewer, we have plotted the wind rose diagram for the model based values and presented along with the measurement based values as shown in the Figure S3 below. However, please note that comparing wind direction from a point source measurement to a model grid is always difficult. Besides, comparing surface wind direction is more challenging than at higher altitudes where more synoptic winds prevail. In the absence of vertical wind direction observations, we show the surface wind comparison just to indicate model performance, not as model validation. In addition, we also show NCEP/NCAR reanalysis plots in the figure to illustrate the difficulty in comparing wind direction for air pollution transport.

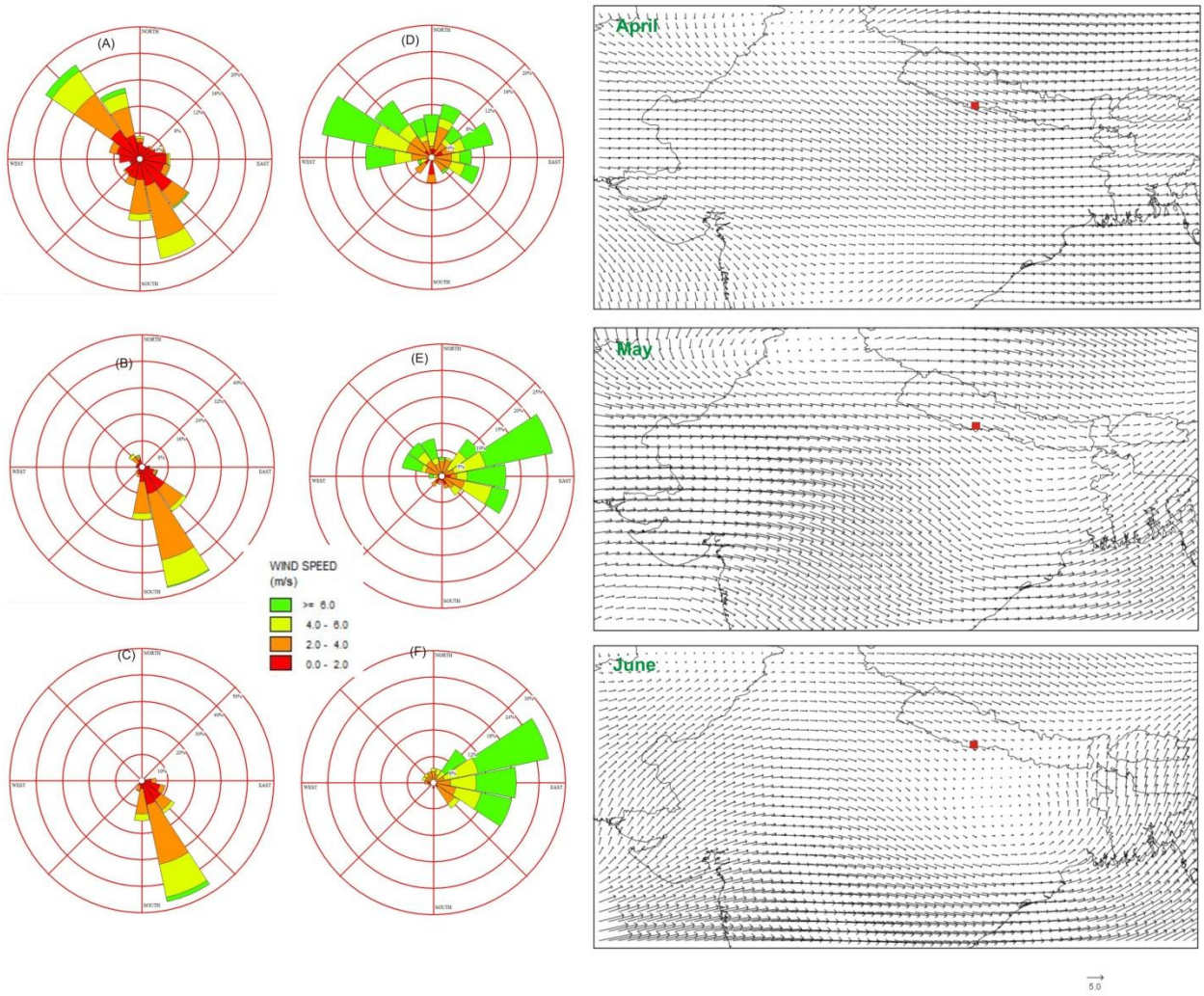

Figure S3: Wind rose of wind speed and wind direction obtained from the observation (A, B, C) and from the model (D, E, F) for the months of April, May and June 2013 respectively. The right panel shows the synoptic scale wind (850 hPa) during three months of the campaign.

Reference

Mohan, M., & Bhati, S. (2011). Analysis of WRF model performance over subtropical region of Delhi, India. Advances in Meteorology, 2011.

Suggestion: Since the purpose of using WRF-STEM model was "...to understand pollution source region as well as the contribution of open biomass burning to air quality in Lumbini..." as stated in lines 126-127, it is suggested that authors can use alternative ways of the data analysis

to achieve the same aims. For example, analysis of wind field (such as Figure 5) or HYSPLIT trajectories and source locations (hotspots, urban etc.) to show the potential of regional transport of the biomass smoke to the site.

Thank you for the suggestions. Another reviewer has suggested to move the synoptic scale wind to Figure 1 (along with the AOD) which we have agreed and revised the Figure 1 (please see our response to another reviewer). We have included the HYSPLIT (in Figure 8) to understand the possible source region for pollutants during two events. However, we wish to note that using back trajectories to identify source regions are also uncertain as identified by Jaffee et al. (1999).

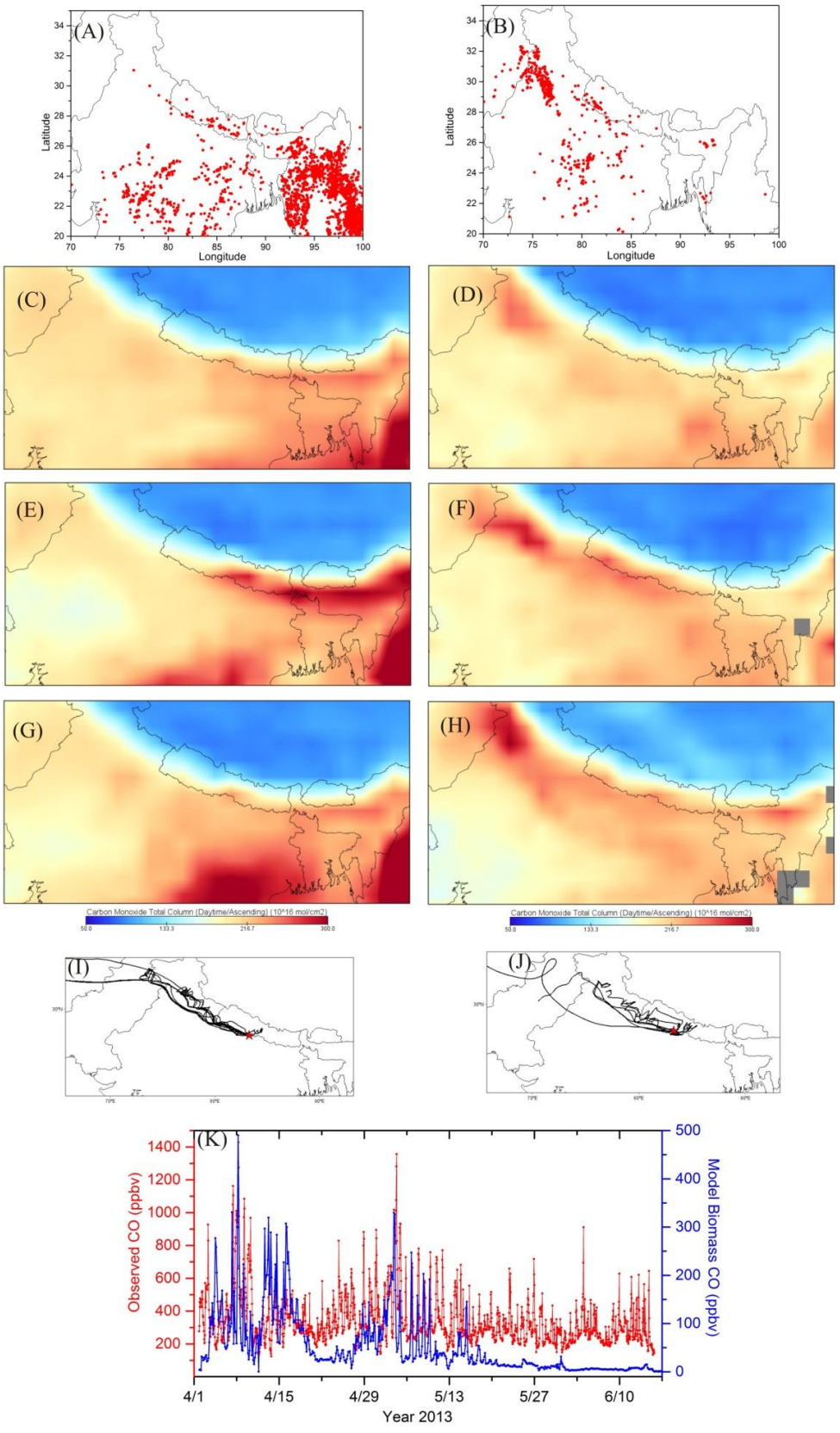
(A)
(B)
Latitude
Longitude
(C)
(D)
(E)
(F)
(G)
(H)
Carbon Monoxide Total Column (Daytime/Ascending) (10^16 mol/cm2)
50.0
133.3
216.7
300.0
(I)
(J)
(K)
Observed CO (ppbv)
Model Biomass CO (ppbv)
Year 2013
4/1
4/15
4/29
5/13
5/27
6/10

Figure 8. Active fire hotspots in the region acquired with the MODIS instrument on Aqua satellite during (A) Event-I (7-9 April) and (B) Event-II (3-4 May). CO emissions, acquired with AIRS satellite, in the region two days before (3-5 April), during (7-9 April) and two days after (10-12 April) Event-I are shown in panels (C), (E) and (G), respectively while panels (D), (F) and (H) show CO emissions two days before (1-2 May), during (3-4 May) and two days after (5-6 May) the Event-II. Panels (I) and (J) represent the 6-hr interval HYSPLIT back trajectories during Event I and II, respectively. Location of the Lumbini site is indicated by the red star in the panel (I and J). Observed CO versus Model open burning CO illustrating the contribution of forest fires during peak CO loading is shown in panel (K).

Reference

Jaffe, D., Anderson, T., Covert, D., Kotchenruther, R., Trost, B., Danielson, J., ... & Harris, J. (1999). Transport of Asian air pollution to North America. Geophysical Research Letters, 26(6), 711-714.

Minor comments:

Line 180: remove word “dust” because not only dust particles but all the particles

Done.

Line 387: the air pollution levels may not be the right/only criteria to classify an area into semiurban or rural etc. Please rephrase.

Done. The new sentence now reads as:

In addition, average BC and CO concentrations in Lumbini were found falling in between concentrations observed at rural sites (up to 6 times higher) and cities in the region (see Table 2), indicating that Lumbini, in a way, can still be considered as semi-urban location.

Line 449: too long a sentence. Please improve the written language.

Done. We have rephrased the sentence as:

Increase in CO concentrations in the evening hours might be due to transport of CO from source regions upwind of Lumbini which along with the local emissions get trapped under reduced Planetary Boundary Layer (PBL) heights.

Line 617: the content above shows important influence from open burning but the discussion in this section seems to be biased toward residential cooking.

The main aim of this section is to understand the influence of biomass burning on the air quality in Lumbini. Main source of biomass burning in the study area is residential cooking (as shown in Figure 11 and 12 in the current revised version). Due to this fact, we believe that the explanation represents the biomass burning which in a way stands for the residential cooking.

Pre-monsoon air quality over Lumbini, a world heritage site along the Himalayan foothills

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Abstract

Lumbini, in southern Nepal, is a UNESCO world heritage site of universal value as the birthplace of Buddha. Poor air quality in Lumbini and surrounding regions is a great concern for public health as well as for preservation, protection and promotion of Buddhist heritage and culture. We present here results from measurements of ambient concentrations of key air pollutants (PM, BC, CO, O_3) in Lumbini, first of its kind for Lumbini, conducted during an intensive measurement period of three months (April-June 2013) in the pre-monsoon season. The measurements were carried out as a part of the international air pollution measurement campaign; SusKat-ABC (Sustainable Atmosphere for the Kathmandu Valley - Atmospheric Brown Clouds). The main objective of this work was to understand and document the level of air pollution, diurnal characteristics and the influence of open biomass burning on air quality in Lumbini. The hourly average concentrations during the entire measurement campaign ranged as follows: BC: 0.3 - 30.0 µg m^{-3}, PM_1: 3.6-197.6 µg m^{-3}, $PM_{2.5}$: 6.1 - 272.2 µg m^{-3}, PM_{10}: 10.5 - 604.0 µg m^{-3}, O_3: 1.0 - 118.1 ppbv, and CO: 125.0 - 1430.0 ppbv. These levels are comparable to other very heavily polluted sites in South Asia. Higher fraction of coarse mode PM was found as compared to other nearby sites in the IGP region. ΔBC/ΔCO ratio obtained in Lumbini indicated considerable contributions of emissions from both domestic and transportation sectors. The 24-h average $PM_{2.5}$ and PM_{10} concentrations exceeded the WHO guideline very frequently (94% and 85% of the sampled period, respectively), which implies significant health risks for the residents and visitors in the region. These air pollutants exhibited clear diurnal cycles with high values in the morning and evening. During the study period, the worst air pollution episodes were mainly due to agro-residue burning and regional forest fires combined with meteorological conditions conducive of pollution transport to Lumbini. Fossil fuel combustion also contributed significantly, accounting for more than half of the ambient BC concentration according to aerosol spectral light absorption coefficients obtained in Lumbini. WRF-STEM, a regional chemical transport model, was used to simulate the meteorology and the concentrations of pollutants to understand the pollutant transport pathways. The model ~~was able to reproduce the temporal variation in the pollutant concentrations well; however,~~ estimated values were ~ 1.5 to 5 times lower than the observed concentrations for CO and PM_{10} respectively. Model simulated regionally tagged CO tracers showed that the majority of CO came from the upwind region of

Ganges Valley. Model needs significant improvement in simulating aerosols in the region. Given the high pollution level, there is a clear and urgent need for setting up a network of long-term air quality monitoring stations in the greater Lumbini region.

1. **Introduction**

The Indo-Gangetic plain (IGP) stretches over 2000 km encompassing a vast area of land in northern South Asia: the eastern parts of Pakistan, most of northern and eastern India, southern part of Nepal, and almost all of Bangladesh. The Himalayan mountains and their foothills stretch along the northern edge of IGP. The IGP region is among the most fertile and most intensely farmed region of the world. It is a heavily populated region with about 900 million residents or 12% of the world's population. Four megacities - Lahore, Delhi, Kolkata, and Dhaka are located in the IGP region, with dozens more cities with populations exceeding one million. The region has witnessed impressive economic growth in recent decades but unfortunately it has also become one of the most polluted, and an air pollution 'hot spot' of local, regional and global concern (Ramanathan et al., 2007). Main factors contributing to air pollution in the IGP and surrounding regions include emissions from vehicles, thermal power plants, industries, biomass and fossil fuel used in cooking and heating activities, agricultural activities, crop residue burning and forest fires. Air pollution gets transported long distances away from emission sources and across national borders. As a result, the IGP and adjacent regions get shrouded with a dramatic annual buildup of regional scale plumes of air pollutants, known as Atmospheric Brown Clouds (ABC), during the long and dry winter and pre-monsoon seasons each year (Ramanathan and Carmichael, 2008). Figure 1 shows monthly synoptic wind and mean aerosol optical depth (AOD) during April-June, 2013 over South Asia. ~~acquired from the MODIS instrument onboard TERRA satellite over South Asia for a period of December 2012-June 2013.~~ Very high aerosol optical depth along the entire stretch of IGP reflects severity of air pollution over large areas in the region.

Poor air quality continues to pose significant threat to human health in the region. In a new study of global burden of disease released recently, Forouzanfar et al. (2015) estimated that in 2013 around 1.7 million people died prematurely in Pakistan, India, Nepal, and Bangladesh as a result of air pollution exposure, nearly 30% of global total premature deaths due to air pollution. Air

pollution also affects precipitation (e.g. South Asian monsoon), agricultural productivity, ecosystems, tourism, climate, and broadly socio-economic and national development goals of the countries in the region (Burney and Ramanathan, 2014;Shindell, 2011;Ramanathan and Carmichael, 2008). It has also been linked to intensification of cold wave and winter fog in the IGP region over recent decades (Lawrence and Lelieveld, 2010 and references therein;Safai et al., 2009;Ganguly et al., 2006). Besides high levels of aerosol loading as shown in Fig. 1, Indo-Gangetic plains also have very high levels of ground level ozone or tropospheric ozone (O_3) (e.g., Ramanathan and Carmichael (2008)). It which is a toxic pollutant to plant and human health, and a major greenhouse gas (IPCC, 2013;Shindell, 2011;Mohnen et al., 1993). South Asia, in particular IGP region, has been projected to be the most ozone polluted region in world by 2030 (Stevenson et al., 2006). Majority of crop loss in different parts of the world results from effects of ozone on crop health and productivity (Shindell, 2011). Burney and Ramanathan (2014) also reported a significant loss in wheat and rice yields in India from 1980 to 2010 due to direct effects of black carbon (BC) and ozone (O_3). BC and O_3 are two key short-lived climate pollutants (SLCP). Similarly, species like fine particles and carbon monoxide (CO) are potent to health damages by posing impacts upon the respiratory and cardiovascular system and even also to the climate system (Singh et al., 2017 and references therein). Because of the IGP's close proximity to the Himalaya-Tibetan plateau region, this once relatively clean region, is now subjected to increasing air pollution transported from regions such as the IGP, which can exert additional risks to sensitive ecosystems in the mountain region (e.g., (Lüthi et al., 2015;Marinoni et al., 2013;Duchi et al., 2011). However, air pollution transport pathways to Himalayas are still not yet fully understood.

Monuments and buildings made with stones are vulnerable to air pollution damage (Brimblecombe, 2003;Gauri and Holdren, 1981). Sulfur dioxide, which forms sulfuric acid upon reaction with water, is the most harmful substance for the monuments as it can corrode and damage them (Baedecker et al., 1992;Gauri and Holdren, 1981). Indo-Gangetic plains are rich in archeological, cultural and historical sites and monuments and many of them are inscribed as UNESCO World Heritage Site. For example, among many other such sites in IGP are the Archaeological Ruins at Moenjodaro (Pakistan), Taj Mahal in Agra and Mahabodhi Temple Complex in Bodh Gaya (India), Lumbini (Nepal), and ruins of the Buddhist Vihara at Paharpur

~~(Bangladesh) (World Heritage List; UNESCO, website: http://whc.unesco.org/en/list). The Taj Mahal is one of the seven wonders of the modern world and India's greatest landmark. At the end of the last century, the government of India realized the growing problem of air pollution damage to Taj Mahal and started a program to save the monument.~~ A recent study has reported that deposition of light absorbing aerosol particles (black carbon, brown carbon) and dust is responsible for the discoloration of Taj Mahal, a world famous monument in India (Bergin et al., 2015). Lumbini, located near the northern edge of the central Ingo-Gangetic plain, is famous as the birthplace of the Lord Buddha ~~. Lumbini is~~ and thus a UNESCO world heritage site of outstanding universal value to humanity.~~, inscribed in the UNESCO list since 1997. The site, with valuable archaeological remains of the Buddhist Viharas (monasteries) and Stupas (memorial shrines), as well as modern temples and monasteries, is a center of attraction and visited by hundreds of thousands of pilgrims, scientists, scholars, yogis, and tourists every year~~. Since the study area is renowned due to its historical and archaeological significance, Lumbini is getting the worldwide attention also for poor air quality in the region. There was no regular air quality monitoring in Lumbini at the time of our measurement campaign.

Through this study, we want to understand the level of air pollution, its diurnal characteristic, and the influence of open biomass burning on air quality in Lumbini. We carried out continuous measurements of ambient concentrations of key air pollutants (particulate matter, black carbon, carbon monoxide, ozone) and meteorological parameters during an intensive measurement period of three months (April-June) in the year 2013. These are the first reported pollutant measurements for Lumbini. A regional chemical transport model called Sulfur Transport and dEposition Model (STEM) was used to simulate the variations of meteorological parameters and air pollutants during the observation period ~~to understand pollution source region as well as the contribution of open biomass burning to air quality in Lumbini~~ to examine the extent to which a state-of-the-art, widely-used air quality model is able to simulate the data, as an indication for where there are still gaps in our knowledge and what further measurements and emissions dataset developments are needed. Model simulated regionally tagged CO tracers were used to identify emission source regions impacting pollutant concentration observed at Lumbini. Satellite data has also been used to understand the high pollution events during the monitoring period. These measurements were carried out as a part of the SusKat-ABC international air pollution

measurement campaign (*M. Rupakheti, manuscript in preparation for ACPD*) jointly led by the International Centre for Integrated Mountain Development (ICIMOD), Kathmandu, Nepal and Institute for Advanced Sustainability Studies (IASS), Potsdam, Germany.

2. Experimental set up

2.1 Sampling site

The Lumbini measurement site (27º29.387′ N, 83º16.745′ E, elevation: ~100 m above sea level) is located at the premise of the Lumbini International Research Institute (LIRI), a Buddhist library in Lumbini. Lumbini lies in the Nepal's southern lowland plain or *Terai* region, termed as "bread basket of Nepal" due to the availability of very fertile land suitable for crop production, which forms the northern edge of the Indo-Gangetic Plains (IGP). About 25 km north of Lumbini the foothills begin while the main peaks of the Himalayas are 140 km to the north. The remaining three sides are surrounded by flat plain land of Nepal and India. The site is only about 8 km from the Nepal-India border in the south. A three storied 10 m tall water tower was used as the platform for the automatic weather station (AWS) whereas remaining instruments were placed inside a room near the base of the tower. Figure S1 shows the location of Lumbini, the Kenzo Tange Master Plan area of the Lumbini development project, and the sampling tower. An uninterrupted power back up was set up in order to assure the regular power supply even during hours with scheduled power cuts during the monitoring period. The nearby premises of the monitoring site consist of the LIRI main office and staff quarters. Further away is a museum, a local bus park for the visitors to Lumbini, the office of the Lumbini Development Trust, monasteries, and thinly forested area with grassland within the master plan area. Outside of the master plan area lie vast area of agricultural fields, village pockets, and several brick kilns and cement industries. A local road (black topped), that cuts through the master plan area, lies about 200 m north of the sampling site and experiences intermittent passing of vehicles. According to the Ministry of Culture, Tourism and Civil Aviation of Nepal over 130 thousand tourists (excluding Nepalese and Indian citizens) visited the Lumbini area in 2014 (http://tourism.gov.np/en).

2.2 Monitoring Instruments

The summary of instruments deployed in Lumbini is presented in Table 1. All data were collected in Nepal Standard Time (NST) which is GMT +05:45 hour. PM_1, $PM_{2.5}$ and PM_{10} mass concentrations were monitored continuously with GRIMM EDM164 (GRIMM Aerosol Technik, Germany) which uses the light scattering at 655 nm to derive mass concentrations. Similarly, aerosol light absorptions at 7 wavelengths (370, 470, 520, 590, 660, 880, 950 nm) were measured continuously with an Aethalometer (Model AE-42, Magee Scientific, USA), averaging and reporting data every 5 min. It was operated at a flow rate of 5 l min^{-1}. No cut-off was applied for inlet; hence the reported concentration of BC is total suspended BC particles. As described by the manufacturer, ambient BC concentration is derived from light absorption at 880 nm using a specific mass absorption cross section. To obtain BC concentration in Lumbini, we used a specific mass absorption cross-section value of 8 m^2 g^{-1} for the 880 nm channel. A similar value has been previously used for BC measurement in the Indo-Gangetic plain (Praveen et al., 2012). To remove the filter loading effect, we used correction method suggested by Schmid et al. (2006) which was also used by Praveen et al. (2012) for BC measurements at a rural site in the Indo-Gangetic plain. Surface ozone (O_3) concentration was measured continuously with an ozone analyzer (Model 49*i*, Thermo Scientific, USA) which utilizes UV (254 nm wavelength) photometric technology to measure ozone concentration in ambient air. CO analyzer (Model 48*i*, Thermo Scientific, USA) was used to monitor ambient CO concentration which is based on the principle that CO absorbs infrared radiation at the wavelength of 4.6 microns. The ambient air was drawn through 6-micron pore size SAVILLEX 47 mm filter at the inlet in order to remove the ~~dust~~ particles before sending air into the CO and O_3 analyzers using a Teflon tube. The filters were replaced every 7-10 days depending on particle loading, based on manual inspection. CO instrument was set to auto-zero at a regular interval of 6 hours. Local meteorological parameters (temperature, relative humidity, wind speed, wind direction, precipitation, and global solar radiation) were monitored with an automatic weather station (AWS) (Campbell Scientific, Loughborough, UK), recording data every minute.

2.3 Regional chemical transport model

Aerosol and trace gas distributions were simulated using a regional chemical transport model. Sulfur Transport and dEposition Model (STEM), a 3D eulerian model that has been used

extensively in the past to characterize air pollutants in South Asian region was used to ~~interpret~~ understand observations at Lumbini (Kulkarni et al., 2015;Adhikary et al., 2007). The Weather Research and Forecasting (WRF) model (Skamarock et al., 2008) version 3.5.1 was used to generate the required meteorological variables necessary for simulating pollutant transport in STEM. The model domain was centered at 24.94º N latitude and 82.55º E longitude covering a region from 3.390º N to 43.308º N latitude and 34.880º E to 130.223º E longitude. The model has 425×200 horizontal grid cells with grid resolution of 25×25 km and 41 vertical layers with top of the model set at 50 mbar. The WRF model was run from November 1, 2012 to June 30, 2013. However, for this study, modeled data only from April to June 2013 have been used. The WRF model was initialized with FNL data available from NCAR/UCAR site (http://rda.ucar.edu/datasets/ds083.2/).

The tracer version of the STEM model provides mass concentration of sulfate, BC (hydrophilic and hydrophobic), Organic carbon (OC), sea salt (fine and coarse mode), dust (fine $PM_{2.5}$ and PM_{10}), CO (biomass and anthropogenic) and region tagged CO tracers. STEM model domain size, resolution and projection are those of the WRF model. Details about tracer version of the STEM model is outlined elsewhere (Kulkarni et al., 2015;Adhikary et al., 2007). Anthropogenic emission of various pollutants (CH_4, CO, SO_2, NO_x, NMVOC, NH_3, PM_{10}, $PM_{2.5}$, BC and OC) used in this analysis were taken from the EDGAR-HTAP_v2 (http://edgar.jrc.ec.europa.eu/htap_v2/index.php?SECURE=123). Open biomass burning emissions on a daily basis during the simulated period were taken from data obtained from the FINN model (Wiedinmyer et al., 2011). Both these emissions were re-gridded to the STEM model domain using four point interpolation techniques available in the STEM model emissions preprocessor. As with the WRF model, the STEM model was run from November 2, 2012 to June 30, 2013 however, data presented here are only during the intensive field campaign period.

3. Results and discussions

3.1 Meteorology

Hourly average time series of various meteorological parameters like precipitation in mm hr^{-1} (Prec), temperature in ºC (T), relative humidity in % (RH), wind speed in m s^{-1} (WS) and

direction in degree (WD) during the monitoring period are shown in Figure 2. Meteorological parameters were obtained with the sensors at the height of ~12 m from the ground. Meteorology results from ~~simulations using~~ WRF model simulations have been used to ~~compare and fill the data gaps~~ indicate if any significantly different air mass was present during the measurement campaign after the meteorological observations malfunctioned. Precipitation data was derived from TRMM satellite (TRMM_3B42_007 at a horizontal resolution of 0.25º) from the Giovanni platform (http://giovanni.gsfc.nasa.gov/giovanni/) as the rain gauge malfuntioned during the sampling period. Precipitation data from TRMM (Figure 2) show that Lumbini was relatively dry in the early portion of the measurement campaign while as the pre-monsoon edged closer to the monsoon onset, the site did experience some rainfall events. This lowered aerosol loading in the later half of the measurement campaign due to washout. Comparison of WRF model outputs with TRMM data shows that the model under-predicts rainfall through out the campaign.

Average observed temperature for the sampling period until the sensor stopped working (on 8th May, 2013, i.e., for 38 days of measurement) was 28.1ºC (minimum: 16.5ºC, maximum: 40ºC). Average temperature from the model, during same period, was 31ºC with values ranging between 19 - 40ºC. As shown in Figure 2, the model captures the synoptic variability of temperature and is mostly within the range of daily values. However, the model has a high bias and does not capture well daily minimum temperature values. In addition, the model does not show any large variation in temperature for the campaign period after the sensors stopped working. This insight will be useful to interpret pollution data later on. For the same period (until the sensor stopped working), the average (observed) RH was ~ 50% (ranging from 10.5 to 97.5%) whereas the model showed the average RH to be ~ 23% with values ranging between 6 to 78%. RH values are highly underestimated by the model, however~~; the synoptic scale variability is adequately captured by the model~~ as previously mentioned, the model does not show significant changes in RH during the measurement campaign after the observations stopped working.

Average observed wind speed during the study period was 2.4 m s^{-1}, with hourly values ranging between 0.03 - 7.4 m s^{-1} whereas from the WRF model average wind speed was found to be 3.2 m s^{-1} (range: 0.06 - 11.1 m s^{-1}). Diurnal variation of observed hourly average wind speed

suggested that wind speeds were lower during nights and mornings while higher wind speed prevailed during day time, with average winds > 3 m s^{-1} up to ~ 3.3 m s^{-1} between 09:00-13:00 local time (Supplementary materials, Figure S2, lower panel). High speed strong winds (> 4 m s^{-1}) were from the NW direction during the month of April which later switched to almost opposite direction, i.e., SE direction from the month of May onwards. ~~Figure 3 shows~~ The monthly wind rose plot using the data from both observation and modeling where the difference in the pattern could be potentially due to the data resolution is shown in Figure S3 ~~(using WRPLOT view from the Lakes Environmental, http://weblakes.com/).~~ Comparing modeled wind direction prediction skills at the surface with one point measurement is not sufficient. However, in the absence of other measurements, we also show the comparison of wind direction as an indication of model performance not as model validation. ~~Since there are no glaringly large biases in the observed surface wind direction, and the lack of measured upper wind data even from nearby region, we use the model to interpret pollutant transport to Lumbini.~~ Discrepancy on model results might have occurred due to various factors inherently uncertain in a weather prediction using a model. Besides, air pollution transport also occurs via elevated layers and is not limited to surface winds. We show NCEP/NCAR reanalysis plots at 850 hPa in Fig. S3 to illustrate the distinctly differing wind direction compared to the surface winds seen from observations as well as NCEP/NCAR reanalysis plot at 1000 hPa shown in Fig. 1. There are no upper wind measurement data nearby Lumbini to show model performance. ~~However~~ Regardless, we believe that air quality model~~ed~~ data is vital for understanding pollutant transport in an area where observation data are non-existent or are incomplete.

~~The monthly mean synoptic wind for the month of April, May and June is presented in Figure 5. NCEP/NCAR reanalysis monthly data of winds at 1000 mbar were used to study the wind pattern. The red dot in the figure indicates the location of Lumbini. NCEP/NCAR data showed the dominance of calm winds over the measurement site. Similar type of wind directions were observed over Kanpur, India, also in the IGP, during the pre-monsoon season (Srivastava et al., 2011).~~

3.2 Air Quality

3.2.1 General overview, PM ratios and influence of meteorology on pollution concentrations

Figure 3 shows hourly averaged time series of observed BC, PM_1, $PM_{2.5}$, PM_{10}, ~~CO and~~ O_3 and CO observed at Lumbini during the study period. Similar temporal behaviour was shown by BC, particulate matter fractions (PM_1, $PM_{2.5}$ and PM_{10}) and CO. The gap in the figure (for PM time series) is due to the power interruption to the instrument. BC concentrations during the measurement period ranged between 0.3-29.9 µg m^{-3} with a mean (±SD) value of 4.9 (±3.8) µg m^{-3}. BC concentrations in Lumbini during pre-monsoon months are lower compared to BC concentrations observed in the Kathmandu Valley because of high number of vehicles plying on the street, brick kilns and other industries in Kathmandu valley (Sharma et al., 2012;Putero et al., 2015). The lowest concentration was observed during a rainy day (21-22 April) whereas the highest concentration was observed during a period of forest fire (detailed in Section ~~3.4~~ 3.3). For the entire measurement period, we found average (of hourly average values) PM_1: 35.8±25.6 µg m^{-3} (minimum-maximum range: 3.6 - 197.6 µg m^{-3}), $PM_{2.5}$: 53.1±35.1 µg m^{-3} (6.1 - 272.2 µg m^{-3}), PM_{10}: 128.9±91.9 µg m^{-3} (10.5-603.9 µg m^{-3}) and coarse-mode ($PM_{10-2.5}$): 75.65±61.67 µg m^{-3} (1.98-331.80 µg m^{-3}). The coarse-mode fraction was ~ 60% of the PM_{10}. The share of coarse-mode aerosol to PM_{10} in Lumbini was higher than that observed in other sites in the IGP; Guwahiti, India (42%) (Tiwari et al., 2017) and Dibrugarh, India (9-16%) (Pathak et al., 2013) both in eastern IGP and Delhi (38%) (Tiwari et al., 2015) in western IGP indicating the higher contribution of coarse aerosols in Lumbini, likely lifted from soils from nearby agricultural fields and construction materials by stronger winds during pre-monsoon season. Similar value of coarse-mode fraction, as in Lumbini, has been reported by Misra et al. (2014) at Kanpur for dust dominated and mixed aerosols events.

The share of BC in PM fractions were found to be ~13% in PM_1, 9% in $PM_{2.5}$ and ~4% in PM_{10} but the correlation coefficients of BC with three PM fractions were found to be 0.89 (PM_1), 0.88 ($PM_{2.5}$) and 0.69 (PM_{10}), indicating the commonality in the sources of these pollutants. The contribution of BC in PM_1 was found to be of ~12% in Kanpur during February-March (Kumar et al., 2016a) similar to Lumbini. Regarding the share of BC in PM_{10}, the share observed in Lumbini (~4%) was similar to that observed over Varanasi (~340 km due south of our site) in central IGP (5%) (Tiwari et al., 2016) and Dibrugarh in eastern IGP (~5%) (Pathak et al., 2013).

Thus our results indicate that despite our station being located at the northern edge of the IGP along the foothills of the Himalayan range, its aerosol characteristics are similar to those found in heavily polluted sites in the central and eastern IGP.

In Lumbini, the average (hourly) share of PM_1 in $PM_{2.5}$, PM_1 in PM_{10} and $PM_{2.5}$ in PM_{10} were found to be ~70%, 34% and 47% respectively. The share of average (sampling period) coarse-mode aerosols to PM_{10} (60%) was found to be higher as compared to that of average fine mode i.e., $PM_{2.5}$ (40%). Regarding other sites in IGP region, $PM_{2.5}/PM_{10}$ ratios were reported to be 56% in Kanpur (Snider et al., 2016), 60% in Varanasi (Kumar et al., 2015), 57% in Guwahiti (Tiwari et al., 2017), 90% in Dribugarh (Pathak et al., 2013) and 62% in Delhi (Tiwari et al., 2015) indicating local differences within IGP as well as suggesting that influence of combustion sources at Lumbini is still lower compared to other locations in Indian section of the IGP. A recent study (Putero et al., 2015) reported the PM_1/PM_{10} during pre-monsoon of 2013 was found to be 0.39 in the Kathmandu Valley of Nepal. Lumbini has significantly lower vehicle emissions and human population than the Kathmandu Valley yet the ratios are similar, indicating the importance of regional combustion sources in Lumbini for finer aerosols (PM_1), and soil-based emissions such as road dust in the Kathmandu Valley. Future studies will need to explore the emission sources around Lumbini in much greater detail. Lower $PM_{2.5}/PM_{10}$ in Lumbini as compared to other regions mentioned earlier could be due to emissions from cement industries located within 15 km distance from the measurement site. Cement factories emit coarse sized particles but we are not able to distinguish in our measurement without having an analysis if certain marker species. Trivedi et al. (2014) reported a ratio of 0.39 (during pre-monsoon) over Delhi, which is lower than the ratio in Lumbini. The lower ratio in Delhi was due to the presence of coarse sized windblown desert dust and suspended soil materials due to strong winds.

The observed 24-hour average particulate matter concentrations ($PM_{2.5}$ and PM_{10}) were found frequently higher than the WHO prescribed guidelines for $PM_{2.5}$ (25 µg m^{-3}) and PM_{10} (50 µg m^{-3}) with PM2.5: exceeding 94% and PM_{10}: 85% of the measurement period of 53 days in Lumbini.

Observed CO concentrations ranged between 124.9-1429.7 ppbv with an average value of 344.1±160.3 ppbv. CO concentration observed in Lumbini is lower than that of Mohali, Western India where the average concentration was 566.7 ppbv during pre-monsoon season due to intense

biomass and agro-residue burning over the region (Sinha et al., 2014). Temporal variation of CO concentrations is similar to that of BC as both of these species are emitted during incomplete combustion of fuel. Moreover, a very strong correlation (r = 0.9) was observed between BC and CO. Past studies have shown that the ratio of BC to CO depends upon multiple factors like site location, combustion characteristics (fuel and technology) at the sources, and type of air mass (Girach et al., 2014;Pan et al., 2011;Zhou et al., 2009). Formation of the soot depends on the carbon to oxygen ratio of fuel whereas CO can also be produced naturally due to the oxidation of VOCs (Girach et al., 2014). Figure 4 shows the comparison of the average ΔBC/ΔCO ratio (0.021) at Lumbini with that obtained from other sites. Please refer to Figure S4 in the supplementary materials for the time series of ΔBC/ΔCO ratio observed in Lumbini. We used the method described by Pan et al. (2011) to calculate the ΔBC/ΔCO values. The ratio was calculated using the equation $(BC-BC_0)/(CO-CO_0)$ assuming the background values (BC_0 or CO_0) as 1.25 percentile of the data. The ΔBC/ΔCO ratio in Lumbini is similar to that obtained at a suburban site, Pantnagar in India (0.017) (Joshi et al., 2016) and in Maldives (0.017) (Dickerson et al., 2002). As compared to Lumbini, the different ΔBC/ΔCO ratio obtained over megacities such as Beijing and Shanghai are due to the higher number of gasoline and diesel vehicles (Zhou et al., 2009). However, the ratio obtained at Lumbini were within the range of emission ratios from diesel used in transport sector (0.0013-0.055), coal (0.0019-0.0572) and biofuels (0.0087-0.0266) for domestic activities (Verma et al., 2010 and references therein). The hourly averaged observed ozone concentration ranged between 1.0 and 118.1 ppbv with a mean value of 46.6±20.3 ppbv during the sampling period. The 8-hr maximum O_3 concentration exceeded WHO guidelines of 100 µg m^{-3} (WHO, 2006) during 88% of the measurement period. Our results clearly indicate that the current pollution levels in Lumbini is of great concern to health of the people living in the region as well as over a million visitors who visit Lumbini, as well as ecosystems, particularly agro-ecosystem, especially in warm and sunny pre-monsoon months.

The relationship of wind speed (WS) with aerosol and gaseous pollutants in Lumbini is shown in Figure S5 (Supplementary information). We were interested in studying the relationship between wind speed and the pollutants since the wind governs the horizontal dilution of the pollutants (Huang et al., 2012) and also likelihood of lifting soil dust. Except ozone, all other pollutants

exhibited negative correlation with wind speed. BC shows negative correlation (r = -0.42) with the wind speed which is similar with other pollutants as well (as can be seen from the figure). Past studies have also reported a similar negative correlation of BC with wind speed over urban and sub-urban areas (Huang et al., 2012;Cao et al., 2009;Ramachandran and Rajesh, 2007;Sharma et al., 2002;Tiwari et al., 2013) indicating that the locally generated BC can accumulate in the atmosphere during lower wind speed conditions (Cao et al., 2009). Tiwari et al. (2013) also reported similar negative correlation (r = -0.45) during the pre-monsoon season over Delhi. On the other hand, secondary pollutants like ozone exhibited a positive relation with the WS (r=0.38) indicating the WS could be one of the potential factors of high ozone in Lumbini. Solar radiation is one of the most important factors for production of ozone in the atmosphere (Naja et al., 2003). The correlation of hourly ozone concentration with solar radiation (not shown here) was found to be 0.41 whereas wind speed during the daytime only (06:00-18:00) showed very weak correlation of 0.02 with ozone, indicating the calm condition as conducive to formation and accumulation of ozone in the region.

Interestingly, the highest concentrations of all measured pollutants were obtained when the wind speed was less than 1 m s^{-1}. In a separate analysis (not shown here), we considered only the WS >1 m s^{-1} and calculated the correlation coefficients to investigate the influence of regional emissions. We found the similar correlation values as previous when all WS values were considered (BC vs WS = -0.41, CO vs WS = -0.42, O_3 vs WS= 0.29, PM_1 vs WS= -0.40, $PM_{2.5}$ vs WS= -0.38, PM_{10} vs WS= -0.33). The correlation of WS (>1 m/s) with concentration of air pollutants elucidates that air pollution over Lumbini is not only of the local origin, it is rather transported from other nearby regions as well.

Past studies near this site have been focused on the cities like Kathmandu (Sharma et al., 2012;Ram et al., 2010;Panday and Prinn, 2009;Putero et al., 2015) and Kanpur (Ram et al., 2010) and agro-residue burning dominated regions of IGP (Rastogi et al., 2016;Sinha et al., 2014;Sarkar et al., 2013) or a remote mountain location in India (Naja et al., 2014). Very high aerosol loading is observed in South Asia during pre-monsoon, mostly over the IGP region (Supplementary materials, Figure S6). As this is the first study over an IGP site located in Nepal, pollution concentrations observed at Lumbini were compared with other sites in the region (Table 2). Different sites located at urban, semi-urban and remote locations were used for

comparison to get a clear comparative picture of the situation at Lumbini amongst other locations in the region. Pre-monsoon seasonal average $PM_{2.5}$ concentration in Lumbini has been found to be lower than the megacity like Delhi (Bisht et al., 2015) and north-western IGP (Sinha et al., 2014), possibly due to higher level of emissions (from traffic and biomass burning, respectively) over those regions. In addition, average BC and CO concentrations in Lumbini were found falling in between concentrations observed at rural sites (up to 6 times higher) and cities in the region (see Table 2), indicating that Lumbini, in a way, can still be considered as semi-urban location. The hourly average O_3 concentration in Lumbini were found to be higher than the cities like Kathmandu (Putero et al., 2015) and Kanpur during pre-monsoon season (Gaur et al., 2014). However from a mesoscale perspective, the hourly average O_3 concentrations were lower at Lumbini as compared to base camp of Mt. Everest region due to the uplift of polluted air masses (Marinoni et al., 2013), stratospheric intrusion (Cristofanelli et al., 2010) and even the regional or long-range transport of the air pollutants (Bonasoni et al., 2010) to the high altitude site.

Regarding the monthly average concentration, the concentrations of all measured pollutants decreased as the pre-monsoon months advanced. The monthly average concentrations of the monitored species are shown in the Figure S7 along with the monthly fire hotspots over the region. Reduction in concentration (except PM) during the month of May (as compared to April) could be attributed to the fewer fire events during May as well as previously discussed washout by rainfall. Two peak pollution episodes were observed during the first half of April and May which is discussed in more detail in the next section.

3.2.2 Observation-model inter-comparison

Chemical transport models provide insight to observed phenomena; however, interpretation has to take into account model performance before arriving at any conclusion. This section describes pollution concentrations simulated by the WRF-STEM model. A comparison of model calculated pollutant concentration along with the minimum and maximum concentrations of various pollutants (with observation) is shown in Table 3. The model based concentrations used here are instantaneous values for every third hour of the day. BC concentrations ranged between 0.4-3.7 $\mu g\ m^{-3}$ with a mean value of 1.8±0.7 $\mu g\ m^{-3}$ for a period of 1st April-15th June 2013. The average model BC concentration was ~2.7 times lower than the observed BC. Regarding PM_1, $PM_{2.5}$ and PM_{10}, the model simulated average concentration was 12.3±5.5 (0.9-41.7) $\mu g\ m^{-3}$, 17.3±6.7 (1.9-

48.3) µg m^{-3} and 25.4±12.9 (2.1-68.8) µg m^{-3}, respectively. The model estimated values were lower by the factor of 3 and 5 respectively than the observed concentrations. The data show that model needs much improvement in its ability to adequately predict observed aerosol characteristics at Lumbini. Since pollutant concentration is a function of emissions, transport and transformation and deposition, improvements in any of these areas would improve the model. However, given observation insights by PM ratios, it seems that improvements are much needed in the emissions of primary aerosols. Current emissions do~~es~~ not account for trash burning, roadside dust and increasingly newer industries, especially emissions from cement factories that have propped up in recent years.

Average observed CO concentration was 255.7±83.5 ppbv, ranging between 72.2-613.1 ppbv, with average model CO ~1.35 times lower than observed. Time series comparison of modeled CO versus observation is shown in Figure 3. Apart from two peak episodes the model does a better job in predicting CO concentration over Lumbini. Previous study using the STEM model over Kathmandu valley showed that the model was able to capture annual BC mean value but completely missed the concentrations during pre-monsoon and post monsoon period (Adhikary et al., 2007). Similar behavior is seen this time for CO where the model misses the peak values but reasonably captures CO concentration after mid-May when no biomass burning events are observed (model to observation ratio improves to 1.16). STEM model CO performance can be significantly improved via better constraining emissions of open biomass burning as discussed in Section 3.3. This activity is beyond the scope of this current paper although the improvements are underway for all these sectors.

3.2.3 Diurnal variations of air pollutants and boundary layer height

In the emission source region, diurnal variations of primary pollutants provide information about the time dependent emission activities (Kumar et al., 2016b). Figure 5 shows the diurnal variation of hourly averaged concentrations of measured pollutants during the sampling period. Primary pollutants like BC, PM and CO showed typical characteristics of an urban environment, i.e., diurnal variation with a morning and an evening peak. However, Lumbini data shows higher concentrations in the evenings compared to morning hours. Elevated concentrations can be linked to morning and evening cooking hours for BC and CO where emission inventory show that residential sector has significant contribution. However, explanation for elevated evening

concentration compared to morning needs further investigation. Increase in the depth of boundary layer, reduction in the traffic density on the roads, absence of open biomass burning during mid-day and increasing wind speed often contribute to the dispersion of pollutants resulting in lower concentration during afternoon. Diurnal variation of wind direction (Supplementary information, Figure S2, upper panel) shows the dominance of wind coming from south (mainly during the month of May and till mid-June). Morning and evening period experienced the winds coming from the southeast direction while the winds were predominantly from southwest direction during late afternoon. Increase in CO concentrations in the evening hours might be due to transport of CO from source regions upwind of Lumbini which along with the local emissions ~~which~~ gets trapped under ~~lower~~ reduced Planetary Boundary Layer (PBL) heights ~~in evening and night time~~. Ozone concentration was lowest in the morning before the sunrise and highest in late afternoon around 15:00 PM after which concentrations started declining, exhibiting a typical characteristic of a polluted urban site. Photo-dissociation of accumulated NO_x reservoirs (like HONO) provides sufficient NO concentration leading to the titration of O_3 resulting in minimum O_3 just before sunrise (Kumar et al., 2016b). The PBL height (in meters (m)) was obtained from the WRF model as observations were not available. ~~Figure 7 shows the monthly diurnal variation of the model derived PBL height.~~ The study period average PBL height over Lumbini was ~ 910 m (ranging between 24.28 and 3807 m observed at 06:00 and 15:00 respectively). The daily average PBL height obtained from the model is compared with published values (Wan et al., 2017) as shown in Figure 6, which indicate that the value is captured by our model during initial measurement period and overestimated in the months of mid May onwards. As the pre-monsoon month advances, PBL height also increased. The monthly average PBL height was 799 m, 956 m and 1014 m respectively during the month of April, May and (1^{st}-15^{th}) June. As presented in the figure, the monthly average diurnal variation also showed that the boundary layer height was maximum during 15:00 local time during each month which coincides with the period of lowest concentration of the pollutants. ~~The fluctuations of modeled PBL height correspond well with the diurnal variation of observed pollutants like BC, CO and PM with the period of lower boundary height experiencing higher pollution concentration.~~

3.3 Influence of forest fires on Lumbini air quality

3.3.1 Identification of forest fire influence over large scale using in-situ observations satellite and model data

Forest fires and agricultural biomass burning (mostly agro-residue burning in large scale) are common over the South Asia and the IGP region during pre-monsoon season. North Indo-Gangetic region is characterized by fires even during the monsoon and post-monsoon season (Kumar et al., 2016b;Putero et al., 2014). These activities influence air quality not only over nearby regions but also get transported towards high elevation pristine environments like Mt. Everest (Putero et al., 2014) and Tibet (Cong et al., 2015a;2015b). So, one of the main objectives of this study was to identify the influence of open burning on Lumbini air quality. Average wind speed during the whole measurement period was 2.4 m s^{-1}. Based on this data, open fire counts within the grid size of 200×200 km centering over Lumbini was used for this analysis assuming that the emissions will take a maximum period of one day to reach our monitoring site. Forest fire counts were obtained from MODIS satellite data product ~~called Global Monthly Fire Location Products- MCD14ML~~ Fire Information for Resource Management System (FIRMS). More on this product has already been described by Putero et al. (2014). Figure 7 shows the daily average ΔBC/ΔCO ratio, aerosol absorption Ångstrom exponent (AAE) which is derived from Aethalometer data and daily open fire count within the specified grid. The green box in the figure is used to show two peak events (presented earlier in Fig. 3) with the elevated BC and CO concentrations observed during the monitoring period. The first peak was observed during 7-9 April and second peak during 3-4 May, 2013. Two pollutants having biomass burning as the potential primary source: BC and CO were taken in consideration. High AAE values (~ 1.6) ~~higher~~ during these two events ~~(~ 1.6)~~ are also an indication of presence of BC of biomass burning origin. The chemical composition of TSP filter samples collected at Lumbini also showed higher concentration of Levoglucosan, a biomass burning tracer in Lumbini during the pre-monsoon season as compared to other seasons of the year (Wan et al., 2017). Wan et al. (2017) also reported that the higher correlation between K^+ with tracers of dust (Ca^{2+} and Mg^{2+}) indicated that dust is the main source of potassium in Lumbini.

Contrary to our expectation, we could not observe any significant influence of forest fire within the specified grid of 200x200 km (or the influence of local forest fire on the air quality over Lumbini was not observed). Therefore, a wider area, covering South and Southeast Asian

regions, was selected for the forest fire count. Figure 8 (A-B) shows the active fire hotspots from MODIS, over the region, during the peak events which shows the first peak could have occurred due to the forest fire over the eastern India region whereas the second peak was influenced by the forest fire over western IGP region. Moreover, in order to strengthen our hypothesis, we have utilized satellite data products for various gaseous pollutants like CO and NO_2 (Atmospheric Infrared Sounder (AIRS) for CO and Ozone Monitoring Instrument (OMI) for NO_2 both obtained from Giovanni platform). Figure 8 (C-H) shows the daytime total column CO before, during and after occurrence of two events (peaks) as stated earlier. Atmospheric Infrared Sounder (AIRS) satellite with daily temporal resolution and 1°×1° spatial resolution have been utilized to understand the CO concentration over the area. CO concentration over Lumbini during both of the peaks confirmed the role of open fires ~~on either sides of~~ over the IGP region for elevated concentration of CO in Lumbini. To further strengthen our finding, the aid of HYSPLIT back trajectories plots ~~of local wind speed and direction~~ was taken. Figure 8 (I-J) represent the 6-hourly back trajectories ~~wind rose plot~~ only for these two events respectively. However, the back trajectories (during both events) indicated that the air mass passed over the fire events in the north western IGP. We note that using back trajectories to identify source regions are also uncertain as identified by Jaffe et al. (1997). ~~Wind rose plots also confirm the wind blowing from those two forest fire regions affected the air quality in Lumbini region~~. Figure 8 (K) shows model biomass CO peak coincident with observed CO. Although the magnitudes are significantly different, the timing of the peaks is well captured by the model. This, we believe, is due to the fact that satellite based open fire detection also has limitation as it does not capture numerous small fires that are prevalent over south Asia which usually burn out before the next satellite overpass. More research is needed to assess the influence of these small fires on regional air quality.

In a separate analysis (not shown here), elevated O_3 concentration during these two events were also observed. Average O_3 concentration before, during and after the events were found to be 46.2±20.3 ppbv, 53.5±31.1 ppbv and 50.3±20.9 ppbv respectively (Event-I) whereas it was found to be 54.8±23.8 ppbv, 56.7±35 ppbv and 55.6±13.4 ppbv respectively (Event-II). Increased ozone concentrations during the high peak events have been analyzed using the satellite NO_2 concentration over the region considering the role of NO_2 as precursor for ozone

552 formation. Daily total column NO_2 were obtained from OMI satellite (data available at the
553 Giovanni platform; http://giovanni.gsfc.nasa.gov/giovanni/) at the spatial resolution of
554 0.25º×0.25º. Figure 9 shows the NO_2 column value before, during and after both events. Even for
555 the NO_2, maximum concentrations were observed during these two special events. It is likely that
556 the local as well as regional pollution (transported from NW IGP region as indicated by synoptic
557 wind in Figure S8) contributed to the ozone peak. However, we are not able to quantify the
558 individual contributions, even with the model simulation, because ozone was not simulated in
559 this experiment.

560 **3.3.2 Identifying regional and local contribution**

561 WRF-STEM model has been used to identify the anthropogenic emission source region
562 influencing the air quality over Lumbini. As previously explained, the model is able to capture
563 the observed CO concentration when intense open burning events were not present. A recent
564 study (Kulkarni et al., 2015) has explored the source region contribution of various pollutants
565 over the Central Asia using similar technique. Figure 10 (A) shows the average contribution
566 from different regions on CO concentration over Lumbini during the whole measurement period.
567 Major share of CO was from the Ganges valley (46%) followed by Nepal region (25%) and rest
568 of Indian region (~17.5%). Contribution from other South Asian countries like Bangladesh and
569 Pakistan were ~ 11% whereas China contributed for ~1% of the CO concentration in Lumbini.
570 Regarding the monthly average contribution, the Ganges Valley and Nepal's contribution were
571 almost equal during the month of April (~34% and ~37% respectively) but increased for the
572 Ganges Valley region during the month of May (~44%) and got reduced for Nepal region
573 (~25%) (Figure S9).
574 Figure 10 (B) is the time series of percentage contribution to total CO concentration during
575 whole measurement period showing different air mass arriving at a 3 hourly intervals. During the
576 whole measurement period, majority of the CO reaching Lumbini were from the Ganges valley
577 (mainly the states of Punjab, Haryana, Uttar Pradesh, Bihar and West Bengal) region with the
578 contribution sometimes reaching up to ~80%. Other India (central, south, east and north) regions
579 also contributed significantly. Bangladesh's contribution in CO loading was seen only after mid-
580 April lasting for only about a week and after the first week of May. The contribution from
581 Bangladesh was sporadic comparing to other regions. Highest contribution from this Bangladesh

region was observed after the first week of June with the arrival of monsoonal air mass. Pakistan also contributed for the CO loading significantly. Others region as mentioned in the figure covered the regions like Afghanistan, Middle east, West Asia, East Asia, Africa and Bhutan. Contributions from these regions were less than 5%. Contribution from China was not evident till the first week of June where a specific air mass arrival shows contribution reaching up to 25% of total CO loading.

A sensitivity analysis was performed for emission uncertainty in the model grid containing Lumbini. Lumbini and surrounding regions in the recent years has seen significant rise in urban activities and industrial activity and related emissions which may not be accurately reflected in the HTAPv2 emissions inventory. A month long simulation was carried out with emissions from Lumbini and the surrounding four grids off and another simulation with Lumbini and surrounding four grid's emissions increased by 5 times the amount from HTAPv2 emissions inventory. The results are shown in Figure 10 (C) as percentage increase or decrease compared to model results using the current HTAPv2 emissions inventory. The black line shows concentration as 100% for the current HTAPv2 emissions inventory. Despite making Lumbini and the surrounding grids emissions zero, model calculation shows pollutant concentration on average is still about 78% of the original value indicating dominance of background and regional sources compared to local source in the model. Increasing emissions 5 times for the Lumbini and surrounding four grids only increases the concentration on average by 151%. Thus uncertainty in emissions are not a local uncertainty for Lumbini rather for the whole region which needs to be better understood for improving model performance against observations at Lumbini.

3.4 Does fossil fuel or biomass influence the Lumbini air?

The aerosol spectral absorption is used to gain insight into nature and potential source of black carbon. This method enables to analyze the contributions of fossil fuel combustion and biomass burning contributions to the observed BC concentration (Kirchstetter et al., 2004). Besides BC, other light absorbing (in the UV region) aerosols are also produced in course of combustion, collectively termed as organic aerosols (often also called brown carbon or BrC) (Andreae and Gelencsér, 2006). Figure 11 shows the comparison of normalized light absorption as function of the wavelength for BC observed at Lumbini during cooking and non-cooking hours and also for the both events. Our results are compared with the published data of Kirchstetter et al. (2004)

612 and that observed over a village center site of Project Surya in the IGP (Praveen et al., 2012)
613 (figure not shown). We discuss light absorption data from two distinct times of the day. The
614 main reason behind using data during 07:00-08:00 h and 16:00-17:00 h is these periods represent
615 highest and lowest ambient concentration (Fig. 5). Also these period represent cooking and non-
616 cooking or high and low vehicular movement hours (Praveen et al., 2012). To understand the
617 influence of biomass and fossil fuel we plotted normalized aerosol absorption at 700 nm
618 wavelength for complete aethalometer measured wavelengths in Fig. 11. Kirchstetter et al.
619 (2004) reported OC absorption efficiency at 700 nm to be zero. Thus we normalized measured
620 absorption spectrum by 700 nm wavelength absorption. Since aethalometer does not provide 700
621 nm wavelength absorption values, we used methodology followed by Praveen et al. (2012). Our
622 results show that the normalized absorption for biomass burning aerosol is ~3 times higher at
623 370 nm compared to that at 700 nm whereas fossil fuel absorption is about 2.6 times higher at
624 the same wavelength. In addition, the curve obtained for the both events are inclined towards the
625 published biomass burning curve. The normalized curve obtained during both cooking and non-
626 cooking period lies in between the standard curve of Kirchstetter et al. (2004). As shown in Fig.
627 11, the curve obtained for the prime cooking time is closer towards the published curve on
628 biomass burning whereas that obtained during the non-cooking time is closer towards the
629 published fossil fuel curve. Similar result was also observed over the Project Surya village in the
630 IGP region (Praveen et al., 2012;Rehman et al., 2011). This clearly indicates there is contribution
631 of both sources: biomass as well as fossil fuel on the observed BC concentration over Lumbini.

632 In order to identify fractional contribution of biomass burning and fossil fuel combustion to
633 observed BC aerosol, we adopted the method described by Sandradewi et al. (2008). Wavelength
634 dependence of aerosol absorption coefficient (b_{abs}) is proportional to $\lambda^{-\alpha}$ where λ is the
635 wavelength and α is the absorption Ångstrom exponent. The α values ranges from 0.9-2.2 for
636 fresh wood smoke aerosol (Day et al., 2006) and between 0.8-1.1 for traffic or diesel soot
637 (references in Sandradewi et al. (2008)). We have taken α value of 1.86 for biomass burning and
638 1.1 for fossil fuel burning as suggested by previous literature (Sandradewi et al., 2008). Figure
639 12 shows diurnal variation of the biomass burning BC. Minimum contribution of biomass
640 burning to total BC concentration was observed during 04:00-06:00 local time (only about 30%
641 of the total BC). As the cooking activities start in morning, the contribution of biomass BC starts

to increase and reaches about 50%. Similar pattern was repeated during evening cooking hours. Only during these two cooking periods, fossil fuel fraction BC was lower. Otherwise it remained significantly higher than biomass burning BC throughout the day. On average, ~40% of BC was from biomass burning whereas remaining 60% was contributed by fossil fuel combustion during our measurement period. Interestingly, this is the opposite of the contributions that were concluded by Lawrence and Lelieveld (2010). Lawrence and Lelieveld (2010) concluded that ~60% BC from biomass versus ~40% fossil fuel, based on a review of numerous previous studies to be likely for the outflow from Southern Asia during the winter monsoon. When we compared observed Ångstrom exponent with Praveen et al. (2012), we noticed that Lumbini values were lower than Project Surya Village center site. This implies Surya village center had higher biomass fraction, also it was observed absorption Ångstrom exponent exceeded 1.86 during cooking hours which indicates 100% biomass contribution. The difference is attributed to the fact that Lumbini sampling site is not a residential site like Surya village which can capture cooking influence efficiently. Further Lumbini sampling site is surrounded by commercial activities such as a local bus park, hotels, office buildings and industries and brick kilns slightly further away. Although the reason for this difference is not clear, it is an indication of the important role of diesel and coal emissions in the Lumbini and upwind regions.

4. **Conclusions**

Our measurements, a first for the Lumbini area, have shown very high air pollution ~~concentration~~ at Lumbini. Black carbon (BC), carbon monoxide (CO), ozone (O_3) and particulate matter (PM_{10}, $PM_{2.5}$ and PM1) were measured during the pre-monsoon of 2013 as a regional site of the *SusKat-ABC campaign*. Average pollutant concentrations during the monitoring period were found to be: BC: 4.9±3.8 µg m^{-3}; CO: 344.1±160.3 ppbv; O_3: 46.6±20.3 ppbv; PM_{10}: 128.8±91.9 µg m^{-3} $PM_{2.5}$: 53.14±35.1 µg m^{-3} and PM_1: 36.6±25.7 µg m^{-3} which is comparable with other urban sites like Kanpur and Delhi in the IGP region. However, our study finds higher fraction of coarse mode PM in Lumbini as compared to other sites in the IGP region. In addition, ΔBC/ΔCO ratio obtained in Lumbini was within the range of emission from both ~~domestic~~ residential and transportation sectors, indicating them as potential key sources of BC and CO, and likely most of PM_1 in Lumbini. The diurnal variation of the pollutants is similar to that of any urban location,

with peaks during morning and evening. However, our results show higher evening concentration compared to morning concentration values and needs further research to explain this behavior. During our measurement period, air quality in Lumbini was influenced by regional forest fires as shown by chemical transport model and satellite data analysis. A regional chemical transport model, WRF-STEM was used to ~~interpret~~ understand observations. Inter-comparison of WRF-STEM model outputs with observations showed that the model underestimated the observed pollutant concentrations by a factor of ~1.5 to 5 but was able to capture the temporal variability. Model uncertainties are attributed mostly to uncertainties in meteorology and regional emissions as shown from sensitivity analysis with local emissions. Region-tagged CO as air-mass tracers are employed in WRF-STEM model to understand the anthropogenic emission source region influencing Lumbini. Our analysis shows that the adjacent regions; mostly the Ganges valley, other parts of India and Nepal accounted for the highest contribution to pollutant concentration in the Lumbini. The normalized light absorption curve clearly indicated the contribution to BC in Lumbini from both sources: biomass as well as fossil fuel. On average, ~40% BC was found to be from the biomass burning and ~60% from fossil fuel burning.

Various improvements and extensions would be possible in future studies. More reliable functioning of the AWS (temperature and RH sensor, rain gauge) would have allowed more in-depth analysis of the relationship between meteorological parameters and pollutants concentration. Continuous measurements of air pollutants throughout the year would allow for annual and seasonal variation study. Improvements in the model are much needed in its ability to simulate observed meteorology. Significant uncertainty lies with regional emissions inventory developed at national and continental scale versus local bottoms up inventory and pollutant emissions from small scale open burning not captured by satellites. There is a clear need for setting up of a continuous air quality monitoring station at Lumbini ~~(UNESCO World Heritage Site)~~ and the surrounding regions for long-term air quality monitoring.

Data availability

The data used for this manuscript can be obtained by sending an email to the corresponding authors and/or to IASS (Maheswar.Rupakheti@iass.potsdam.de) and/or to ICIMOD (arnico.panday@icimod.org). Modeling data can be obtained from B. Adhikary (Bhupesh.adhikary@icimod.org).

Authors' contributions

M.R. and M.L. conceived the Lumbini portion of the SusKat experiment. M.R. and A.K.P. coordinated the Lumbini field campaign. D.R. and K.S.M conducted the field observations at Lumbini. B.A. designed and ran the WRF-STEM model. P.S.P., B.A. and D.R. finalized the manuscript composition. D.R., P.S.P, B.A., M.R. and S.K. conducted the data analysis. D.R. and B.A. prepared the manuscript with inputs from all coauthors.

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993 **Table 1**. Summary of instruments deployed during monitoring in Lumbini

Instrument (Model)	Manufacturer	Parameters	Inlet/sensor height (above ground)	Sampling interval	Sampled period
Environmental Dust monitor (EDM 164)	GRIMM Aerosol Technik, Germany	PM_1, $PM_{2.5}$, PM_{10}	5 m	5 min	04/02-05/10, 06/02-06/13
Aethalometer (AE42)	Magee Scientific, USA	Aerosol light absorption at seven wavelengths, and BC concentration	3 m	5 min	01/04-05/06
CO analyzer (48*i*)	Thermo Scientific, USA	CO concentration	3 m	1 min	01/04-15/06
O_3 analyzer (49*i*)	Thermo Scientific, USA	O_3 concentration	3 m	1 min	01/04-15/06
Automatic Weather Station (AWS)	Campbell Scientific, UK	T, RH, WS, WD, Global Radiation, Precipitation	12 m	1 min	01/04-15/06

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996 **Table 2.** Comparison of $PM_{2.5}$, BC, CO and O_3 concentrations at Lumbini with those at other sites in South Asia

Sites	Characteristics	Measurement period	$PM_{2.5}$ (µg m^{-3})	BC (µg/m^3)	CO (ppbv)	O_3 (ppbv)	References
Lumbini, Nepal	Semi-urban	Pre-monsoon, 2013	53.1±35.1	4.9±3.8	344.1±160.3	46.6±20.3	This study
Kathmandu, Nepal	Urban	Pre-monsoon, 2013	-	14.5±10	-	38.0±25.6	(Putero et al., 2015)
Mt. Everest, Nepal	Remote	Pre-monsoon	-	0.4±0.4	-	61.3±7.7	(Marinoni et al., 2013)
Delhi, India	Urban	Pre-monsoon (night-time)	82.3±50.5	7.70±7.25	1800±890	-	(Bisht et al., 2015)
Kanpur, India	Urban	June 2009-May 2013, April-June	-	2.1±0.9	721±403	27.9±17.8	(Gaur et al., 2014) (Ram et al., 2010)
Mohali, India	Semi-urban	May, 2012	104±80.3	-	566.7±239.2	57.8±25.4	(Sinha et al., 2014)
Mt. Abu, India	Remote	Jan 1993-Dec 2000, pre-monsoon	-	0.7±0.14	131±36	39.9±10.8	(Naja et al., 2003) (Das and Jayaraman, 2011)

997 **Table 3**. Inter-comparison of observed and model simulated hourly average concentrations of air
998 pollutants during the measurement campaign period. Unit: BC and PM in μg/m^3 and CO in ppbv.

Pollutants	Observed (mean and range)	Modeled (mean and range)	Ratio of mean (observed/modeled)
BC	4.9 (0.3-29.9)	1.8 (0.4-3.7)	2.7
PM_1	36.6 (3.6-197.6)	12.3 (0.9-41.7)	3
$PM_{2.5}$	53.1 (6.1-272.2)	17.3 (1.9-48.3)	3
PM_{10}	128.8 (10.5-604.0)	25.4 (2.1-68.8)	5
CO	344.1(124.9-1429.7)	255.7 (72.2-613.1)	1.35

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Figures

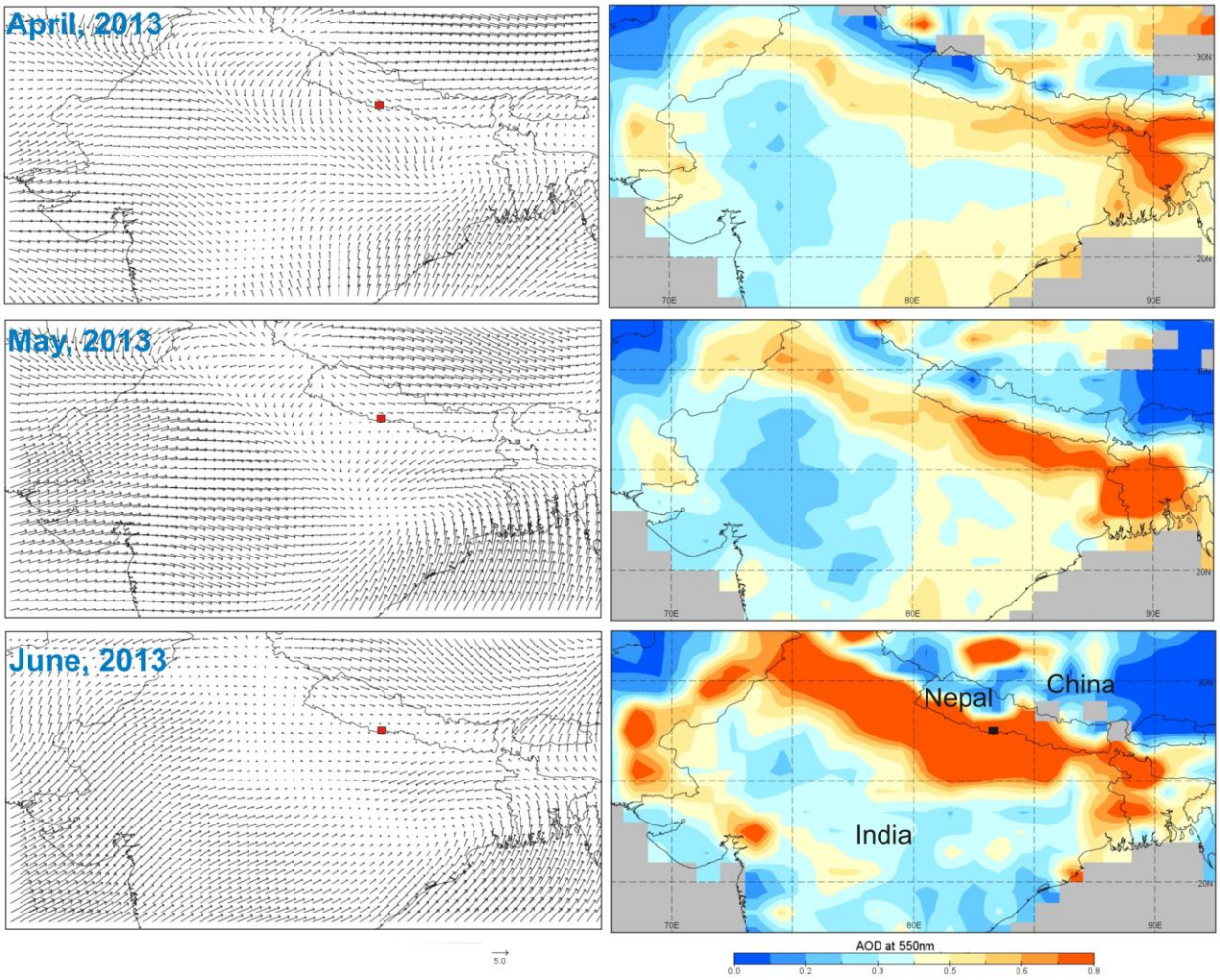


Figure 1. Monthly synoptic wind (at 1000 hPa) for April, May and June 2013, based on NCEP/NCAR reanalysis data where the orientations of arrows refer to wind direction and the length of arrows represents the magnitude of wind (m/s). Red square box in the figure (left) represents the location of Lumbini. Figures on the right side represent monthly aerosol optical depth acquired with the MODIS instrument aboard TERRA satellite. High aerosol loading can be seen over the entire Ingo-Gangetic Plains (IGP). Light gray color used in the figure represents the absence of data.

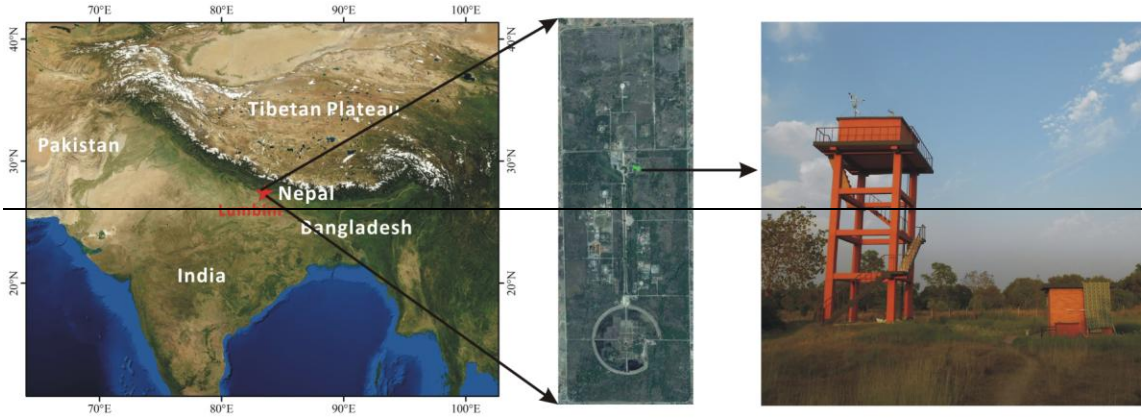


Figure 2. Location of sampling site in Lumbini in southern Nepal (left panel). The middle panel shows the Kenzo Tange Master Plan Area of Lumbini while the right panel shows the sampling tower in the Lumbini Master Plan Area.

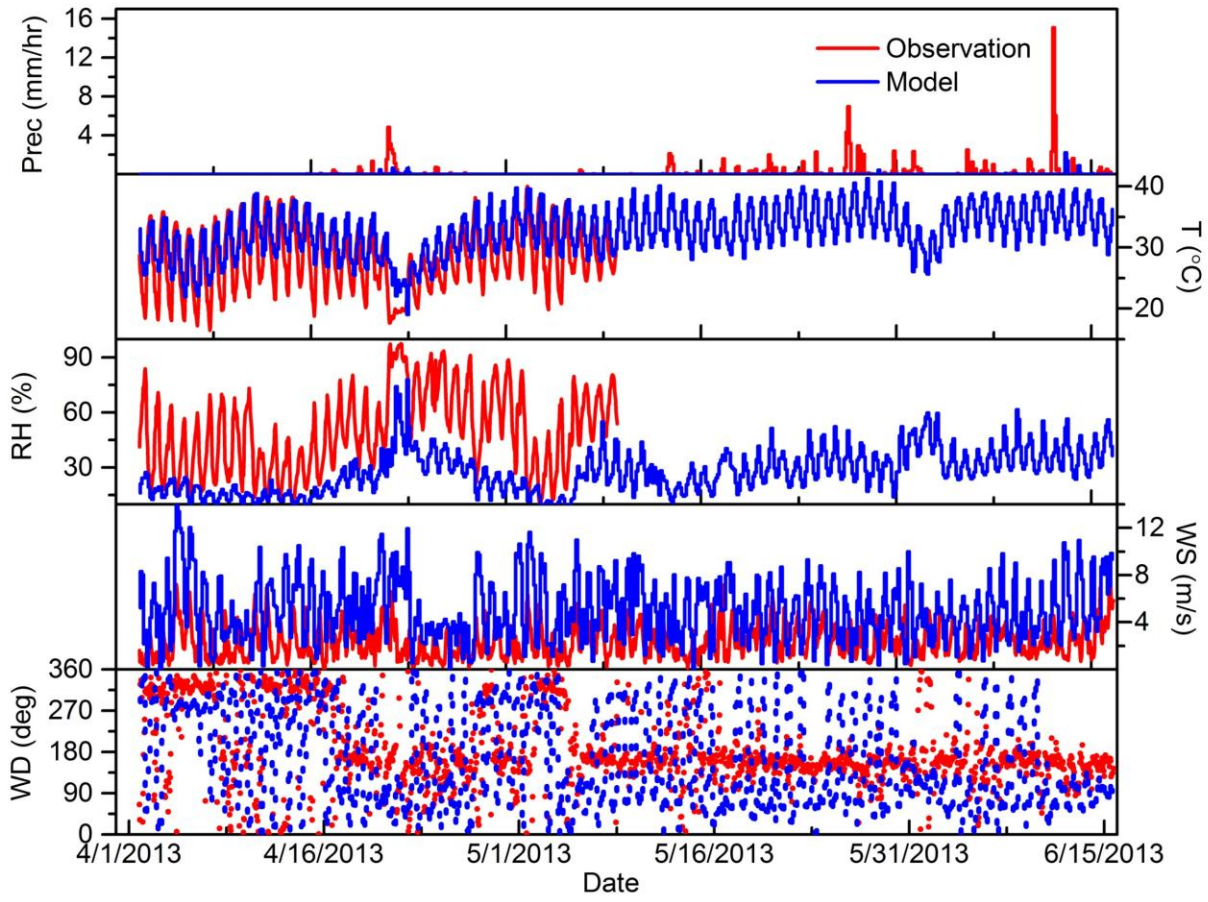
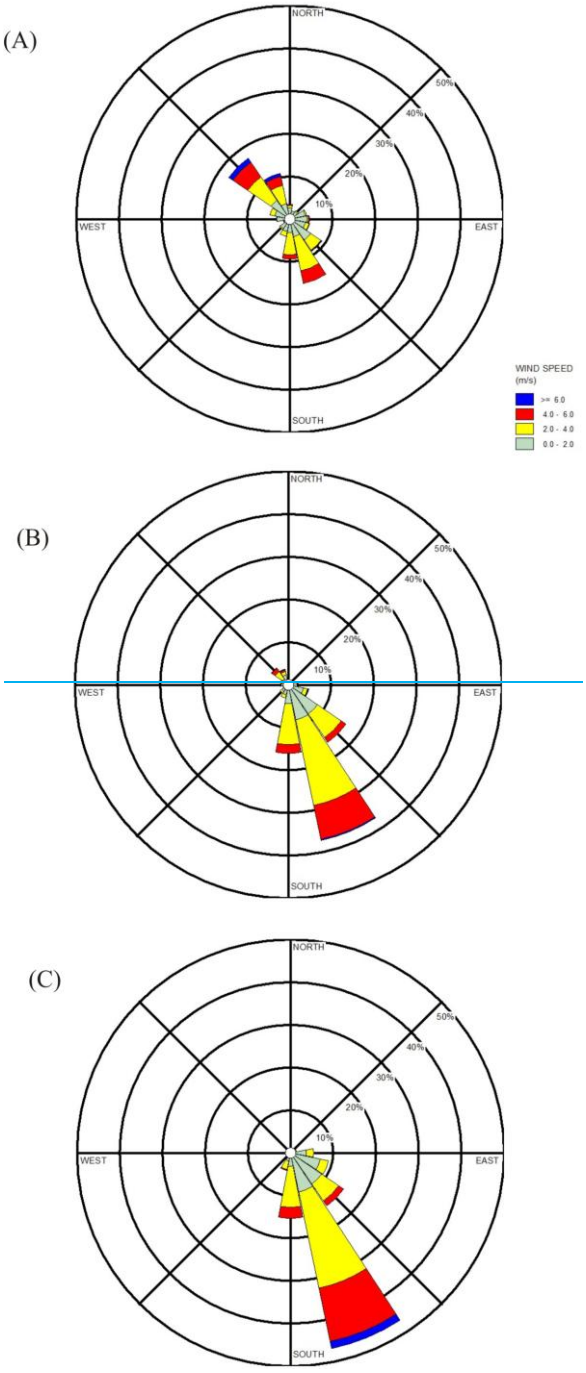


Figure 2. Time series of hourly average observed (red) and model estimated (blue) meteorological parameters at Lumbini, Nepal for the entire sampling period from 1 April to 15 June 2013.



1022

1023 **Figure 4**. Wind rose of wind speed and wind direction observed at Lumbini during the month of

1024 (A) April, (B) May, and (C) (1st-15th) June 2013.

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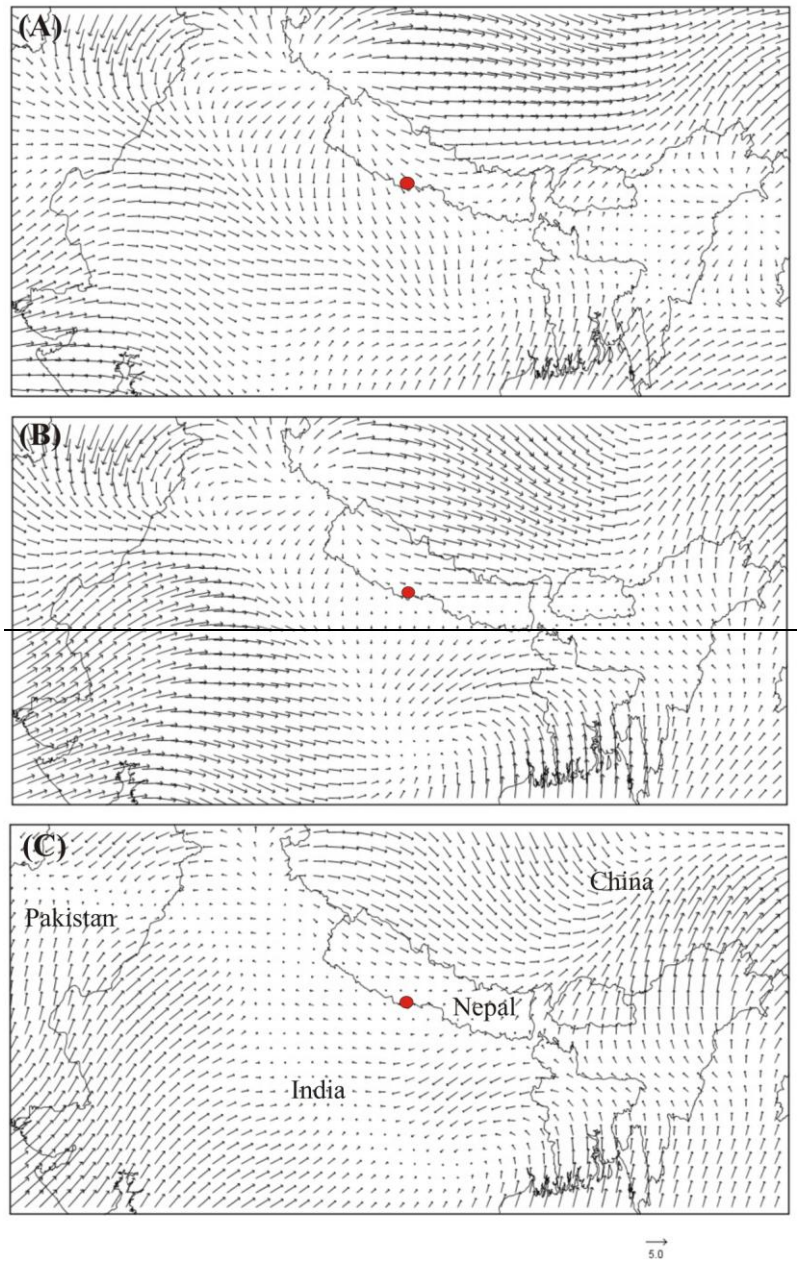

Figure 5. Monthly synoptic surface winds for the month of (A) April, (B) May and (C) June 2013, based on NCEP/NCAR reanalysis data. Orientations of arrows in the figures refer to wind direction whereas the length of arrows represents the magnitude of wind speed (m/s). Red dot in the map represents the location of Lumbini.

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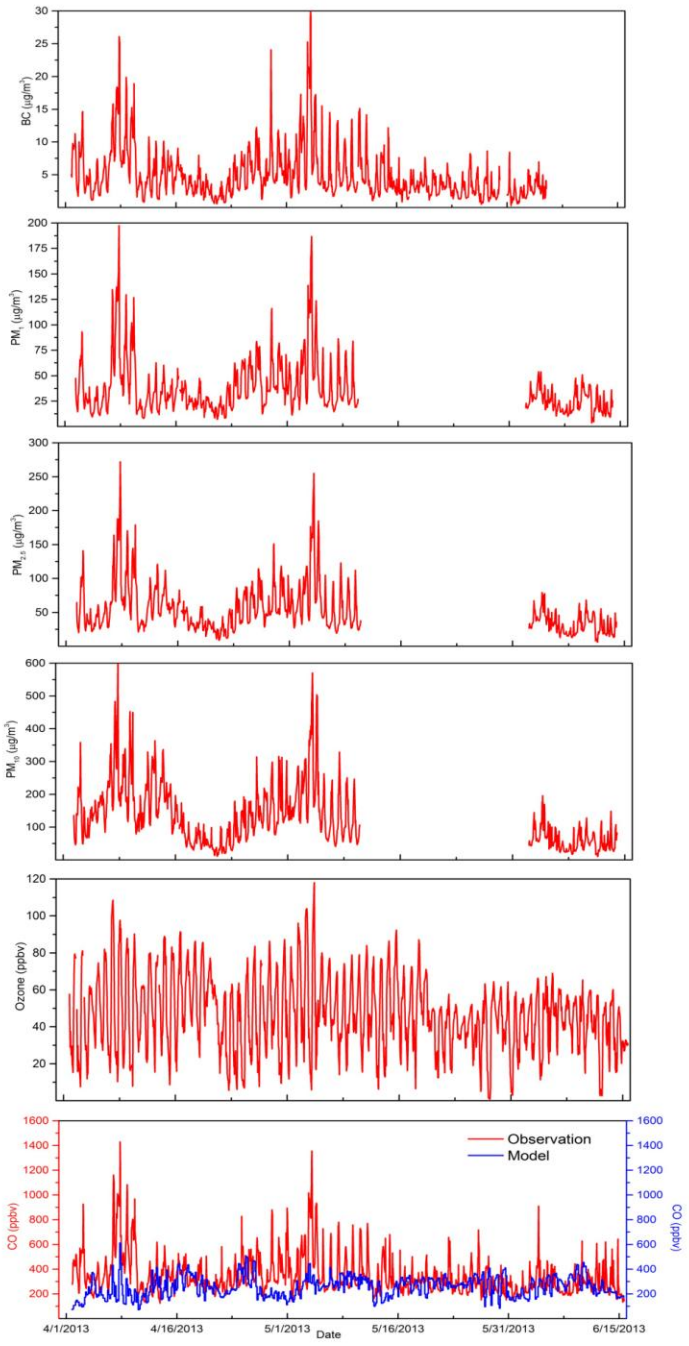


1035

1036 **Figure 3.** Time series of the observed (red line) and model estimated (blue line) hourly average
1037 concentrations of BC, PM_1, $PM_{2.5}$, PM_{10}, O_3 and CO at Lumbini, Nepal for the entire sampling
1038 period from 1 April to 15 June 2013.

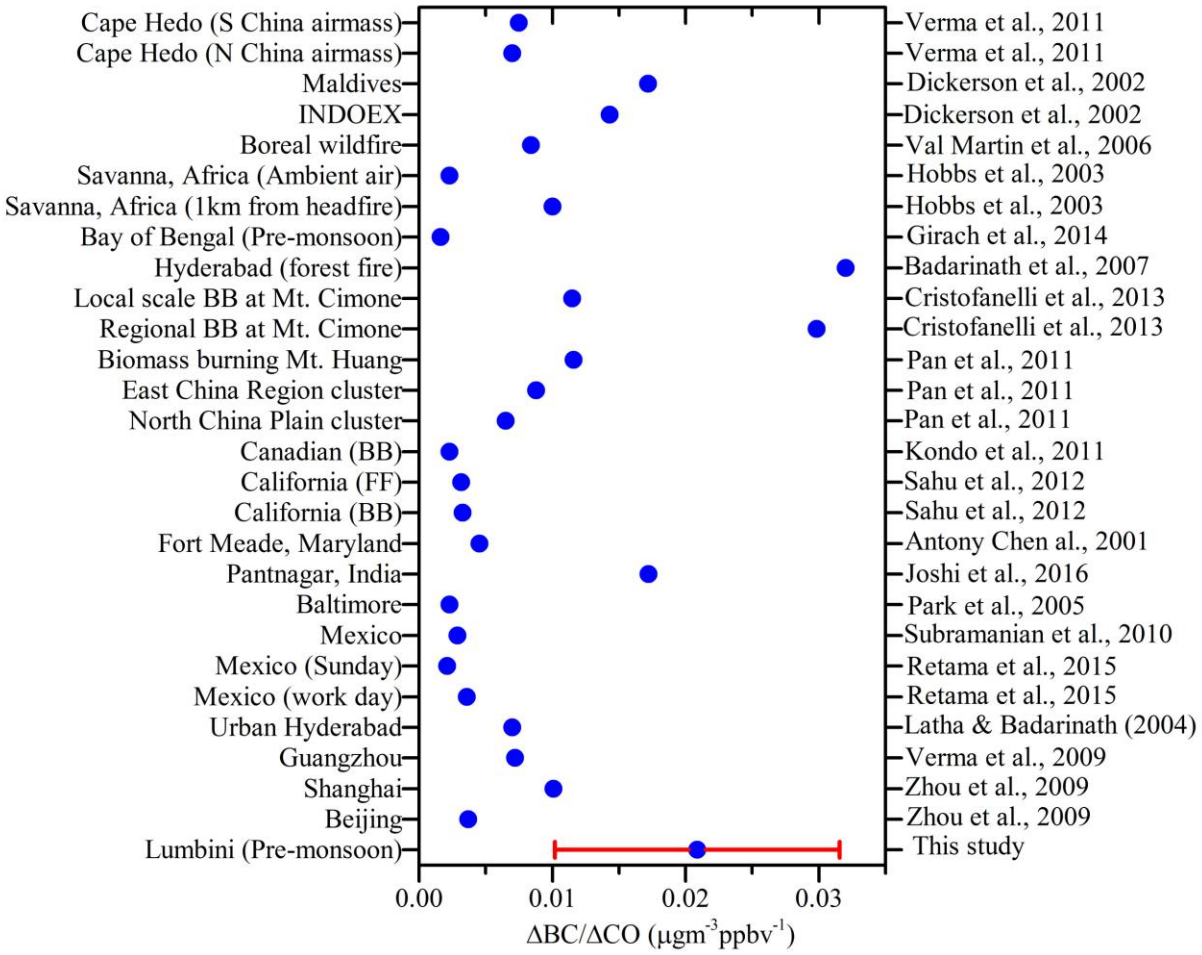


Figure 4. Comparison of BC concentrations to CO concentrations (ΔBC/ΔCO) ratios obtained for Lumbini with other sites. The red horizontal bar represents standard deviation.

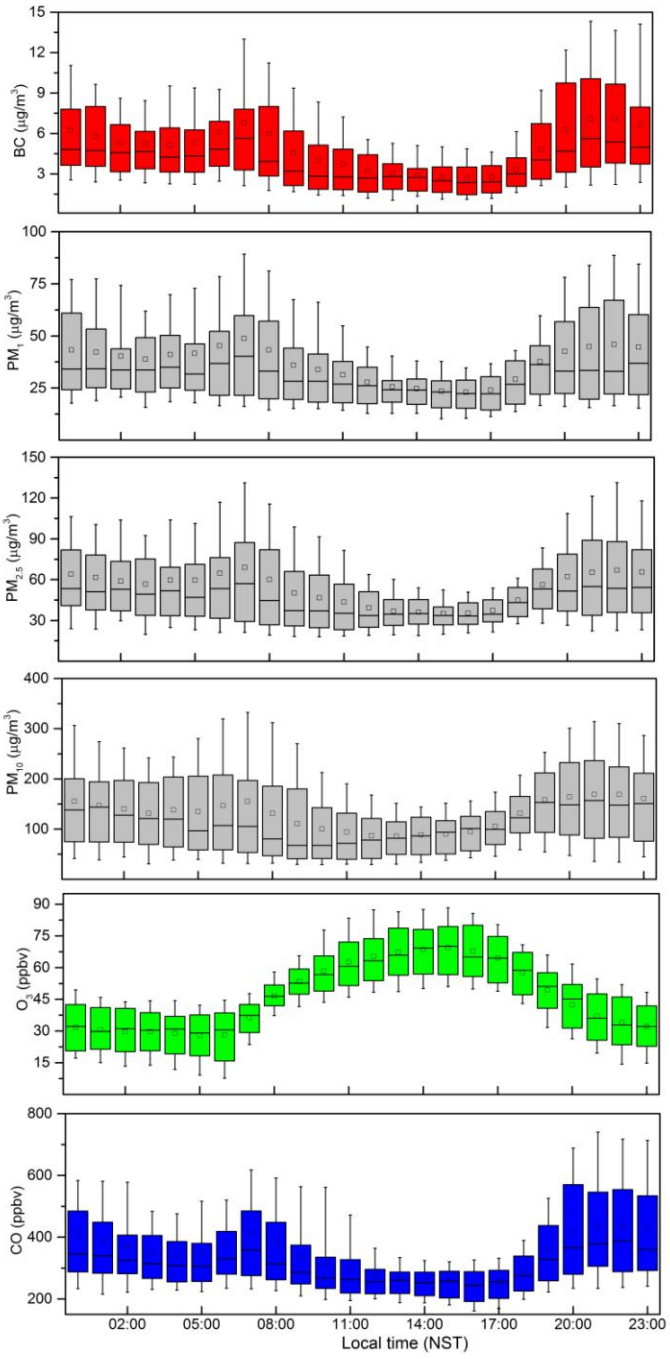


Figure 5. Diurnal variations of hourly average ambient concentrations of BC, PM_1, $PM_{2.5}$, PM_{10}, O_3 and CO at Lumbini during the monitoring period (1 April -15 June 2013). In each box, lower and upper boundary of the box represents 25th and 75th percentile respectively, top and bottom of the whisker represents 90th and 10th percentile respectively, the mid-line represents median, and the square mark represents the mean for each hour.

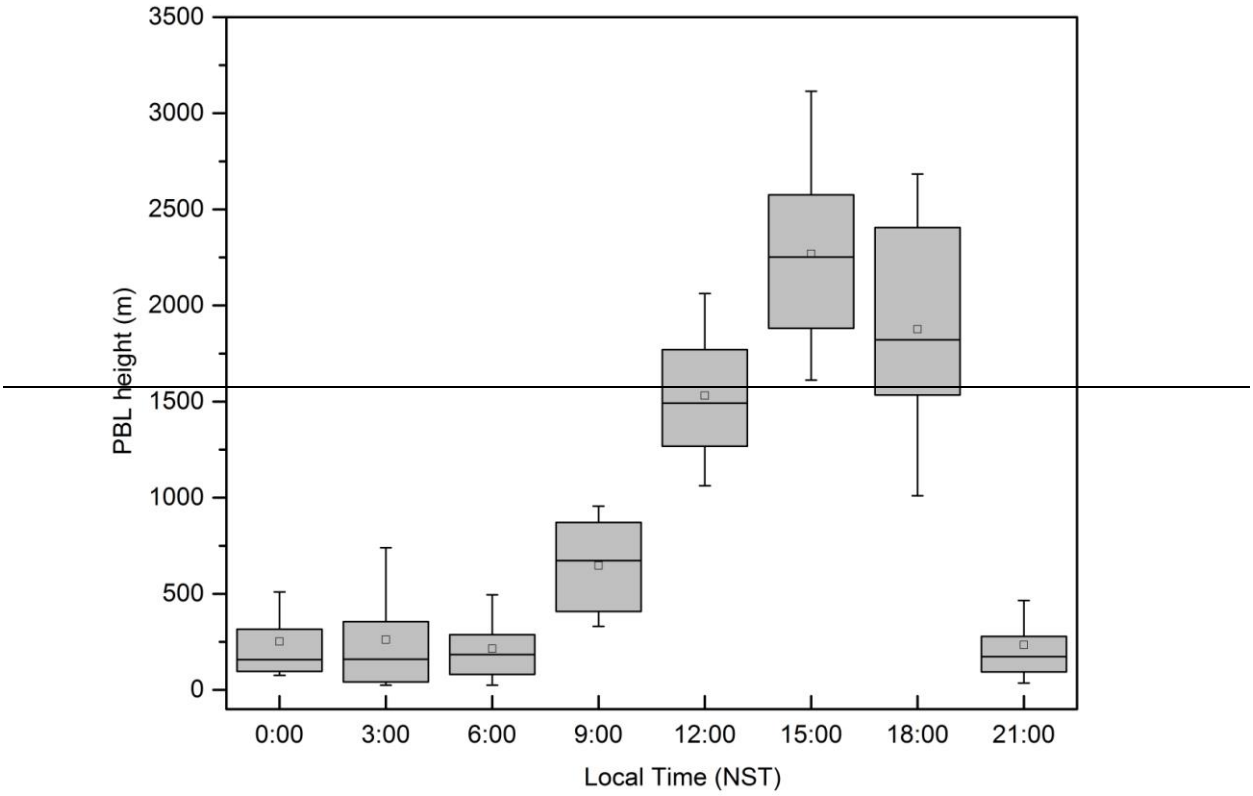
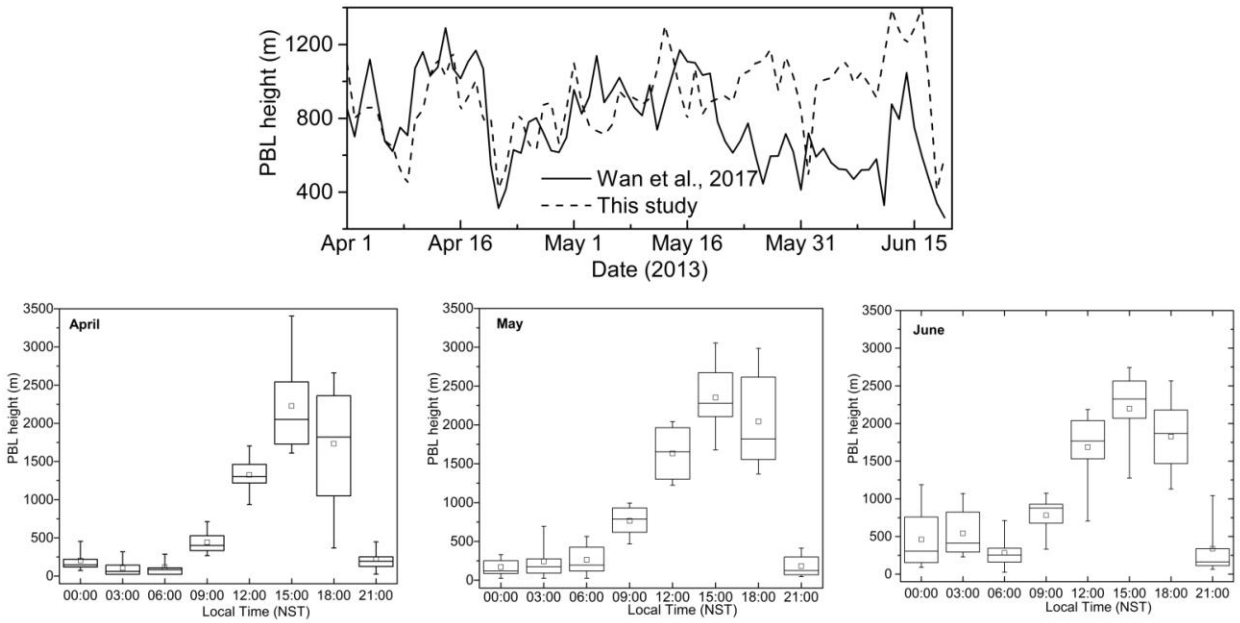


Figure 9. Diurnal variation of the planetary boundary layer (PBL) height at Lumbini obtained for every three hours of each day from the WRF-STEM model for the sampling period. The square mark in each box represents the mean PBL height, bottom and top of the box represents 25th and 75th percentile, top and bottom of the whisker represents 90th and 10th percentile respectively.



1056

1057 Figure 6. Daily time series of PBL height obtained from the model and reported values over
1058 Lumbini (obtained from Xin et al., 2017). The lower panel shows the monthly average diurnal
1059 variation of the PBL height. The square mark in each box represents the mean PBL height,
1060 bottom and top of the box represents 25th and 75th percentile, top and bottom of the whisker
1061 represents 90th and 10th percentile respectively.

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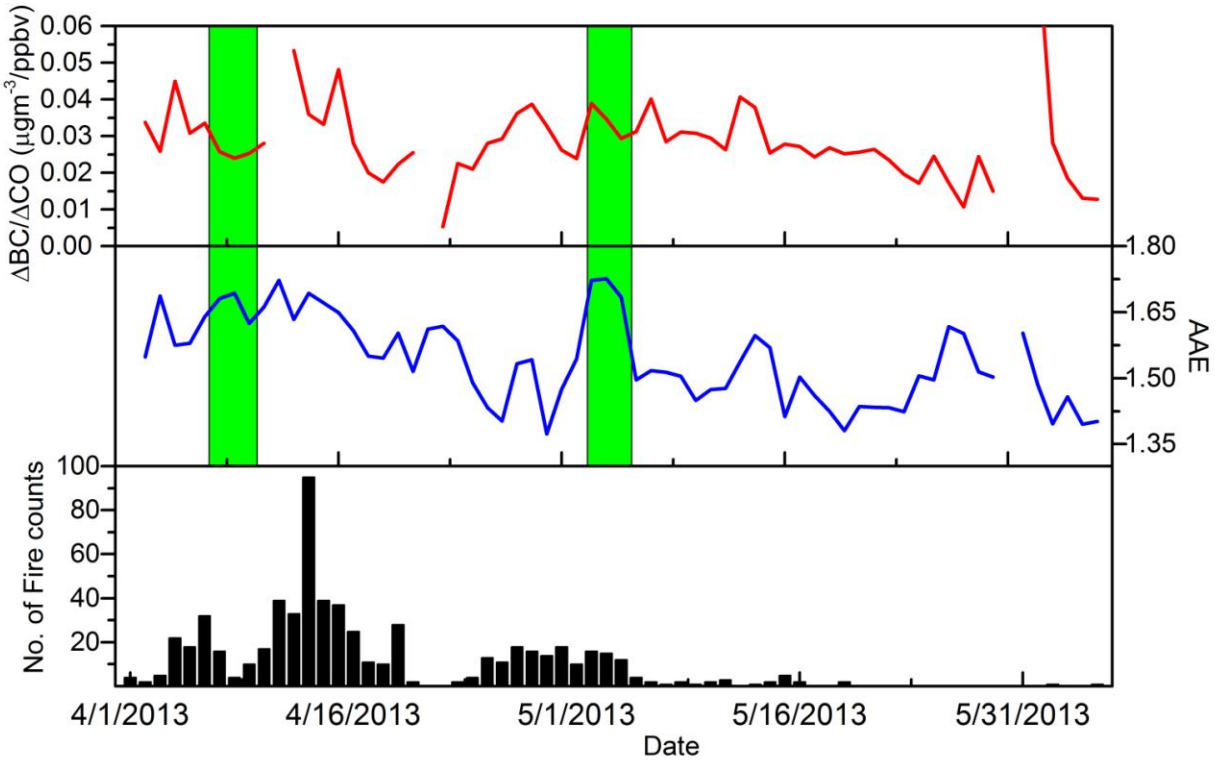
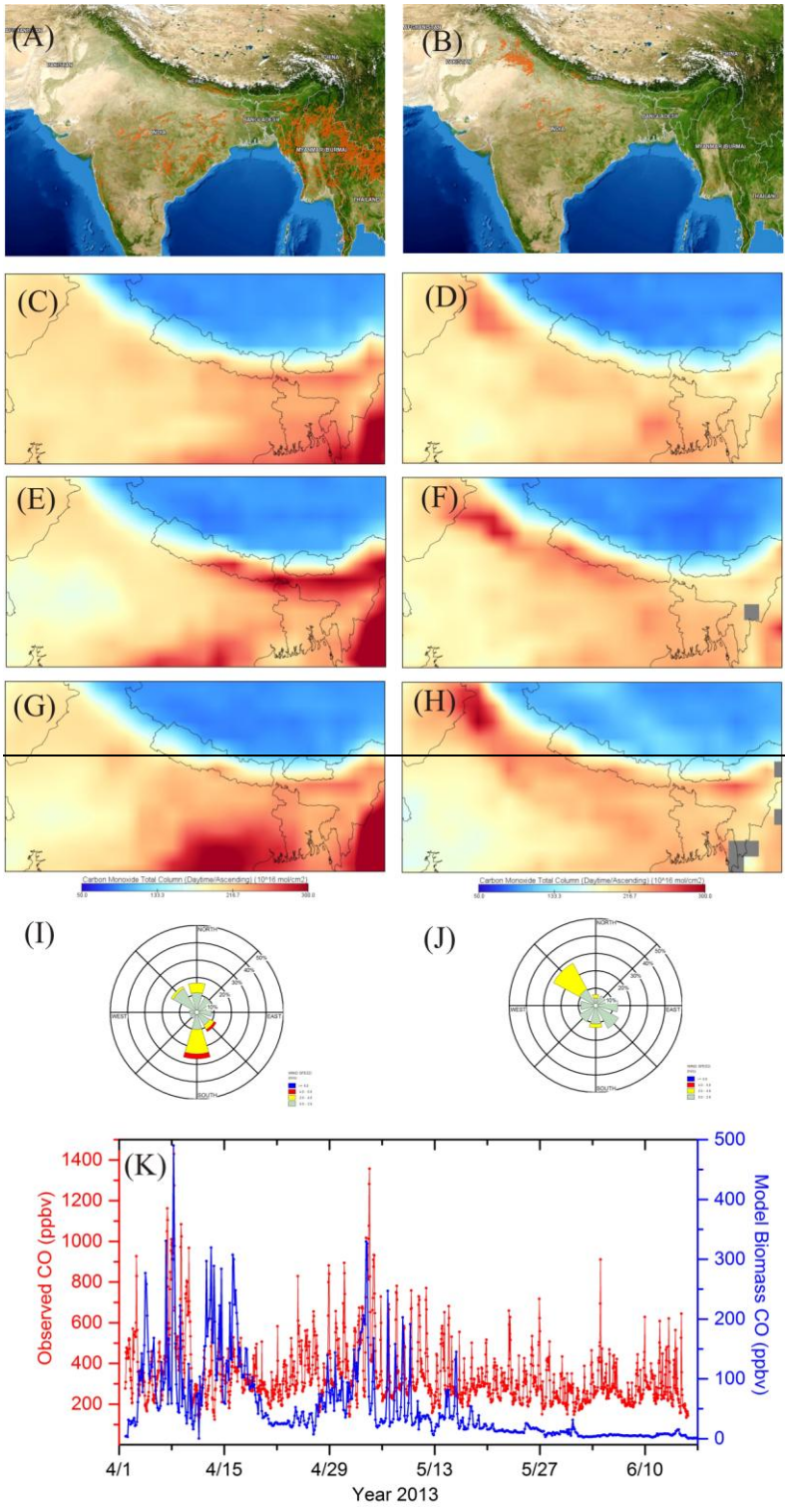


Figure 7. Time series of daily average ΔBC/ΔCO ratio, absorption Ångstrom exponent (AAE), along with fire counts acquired with the MODIS instrument onboard TERRA satellite for a 200×200 km grid centered at Lumbini. Two rectangular green boxes represent time of two episodes with high peaks in CO and BC concentrations as shown in earlier figures.



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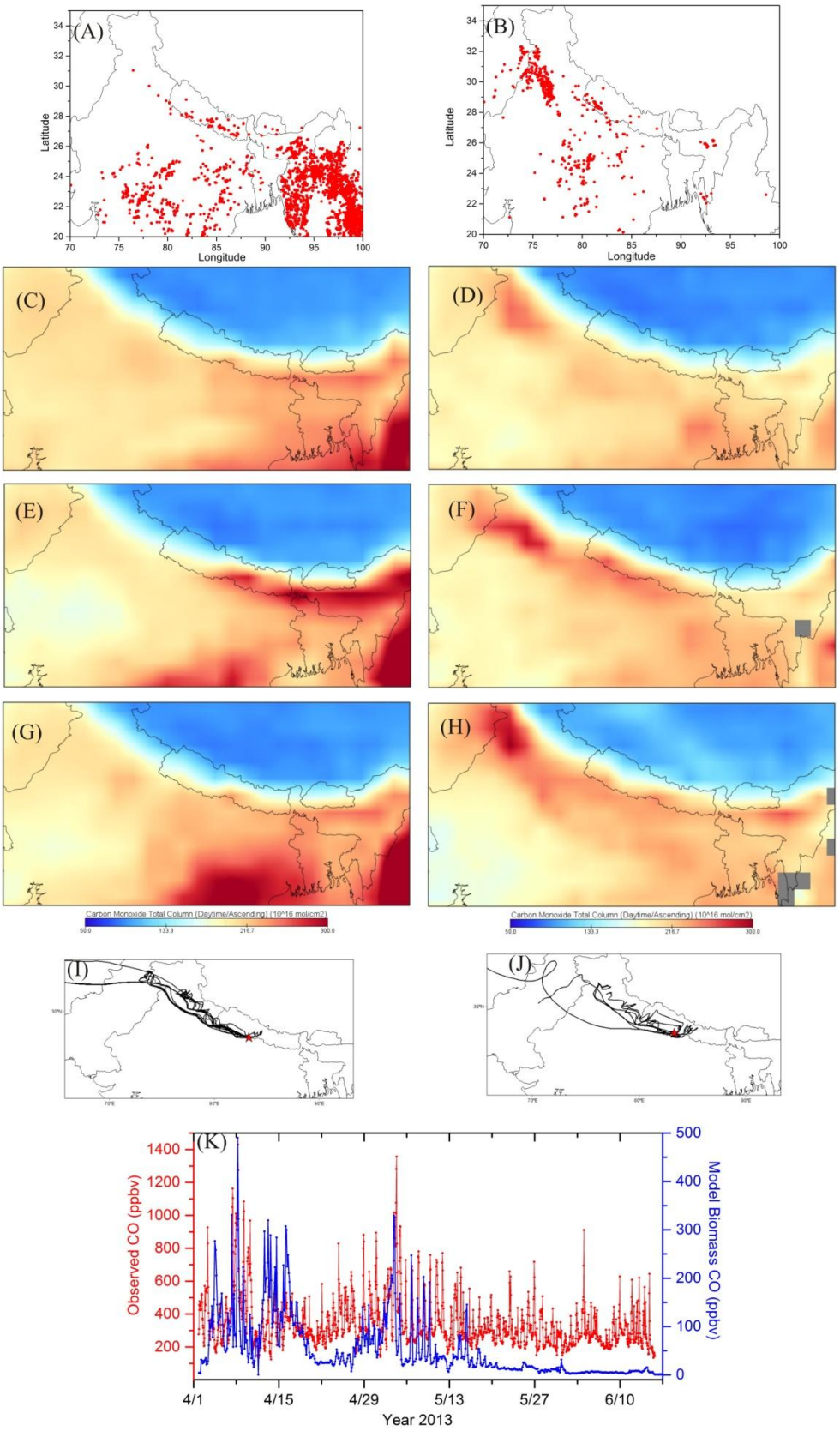
1070 ~~Figure 9. Active fire hotspots in the region acquired with the MODIS instrument on Aqua~~

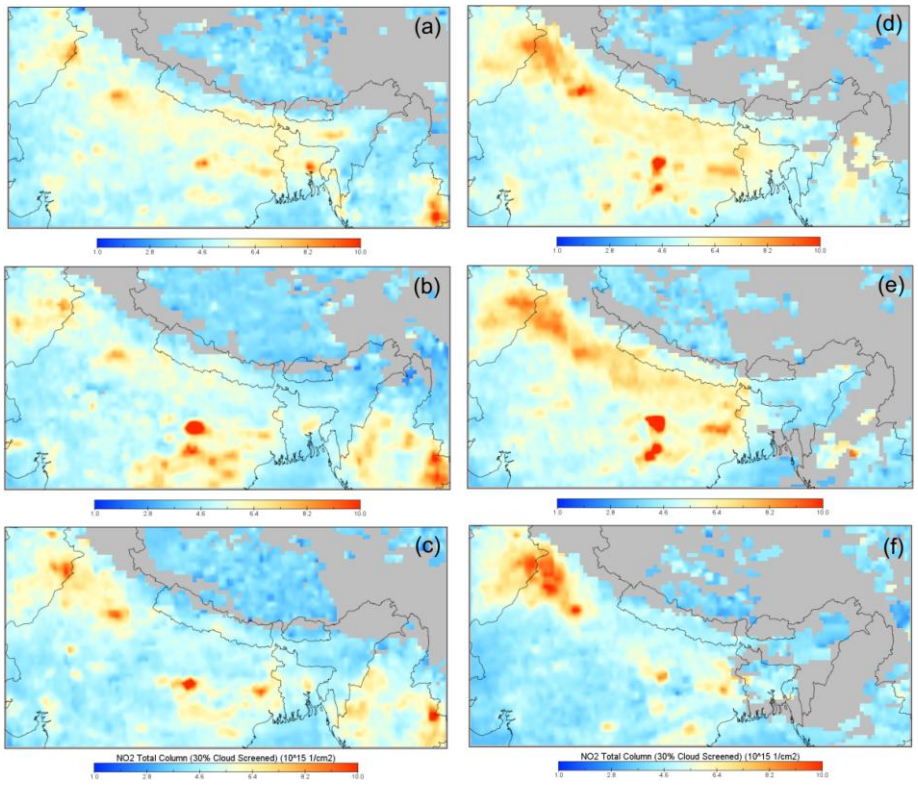
1071 ~~satellite during (A) Event-I (7-9 April) and (B) Event-II (3-4 May). CO emissions, acquired with~~

~~AIRS satellite, in the region two days before (3-5 April), during (7-9 April) and two days after (10-12 April) the Event-I are shown in panels (C), (E) and (G), respectively while panels (D), (F) and (H) show CO emissions two days before (1-2 May), during (3-4 May) and two days after (5-6 May) the Event-II. Panels (I) and (J) represent the 6-hr interval HYSPLIT back trajectories during Event I and II, respectively. Location of the Lumbini site is indicated by the red star in the panel (I and J). Observed CO versus Model open burning CO illustrating the contribution of forest fires during peak CO loading is shown in panel (K).~~


(A)
(B)
Latitude
Longitude
(C)
(D)
(E)
(F)
(G)
(H)
Carbon Monoxide Total Column (Daytime/Ascending) (10^16 mol/cm2)
(I)
(J)
(K)
Observed CO (ppbv)
Model Biomass CO (ppbv)
Year 2013
4/1
4/15
4/29
5/13
5/27
6/10

1079

Figure 8. Active fire hotspots in the region acquired with the MODIS instrument on Aqua satellite during (A) Event-I (7-9 April) and (B) Event-II (3-4 May). CO emissions, acquired with AIRS satellite, in the region two days before (3-5 April), during (7-9 April) and two days after (10-12 April) the Event-I are shown in panels (C), (E) and (G), respectively while panels (D), (F) and (H) show CO emissions two days before (1-2 May), during (3-4 May) and two days after (5-6 May) the Event-II. Panels (I) and (J) represent the 6-hr interval HYSPLIT back trajectories during Event I and II, respectively. Location of the Lumbini site is indicated by the red star in the panel (I and J). Observed CO versus Model open burning CO illustrating the contribution of forest fires during peak CO loading is shown in panel (K).



1090

1091 **Figure 9.** NO_2 total column obtained with OMI satellite over the region (a) before, (b) during,
1092 and (c) after the Event- I. The panels (d), (e), (f) show NO_2 total column before, during and after
1093 the Event- II.

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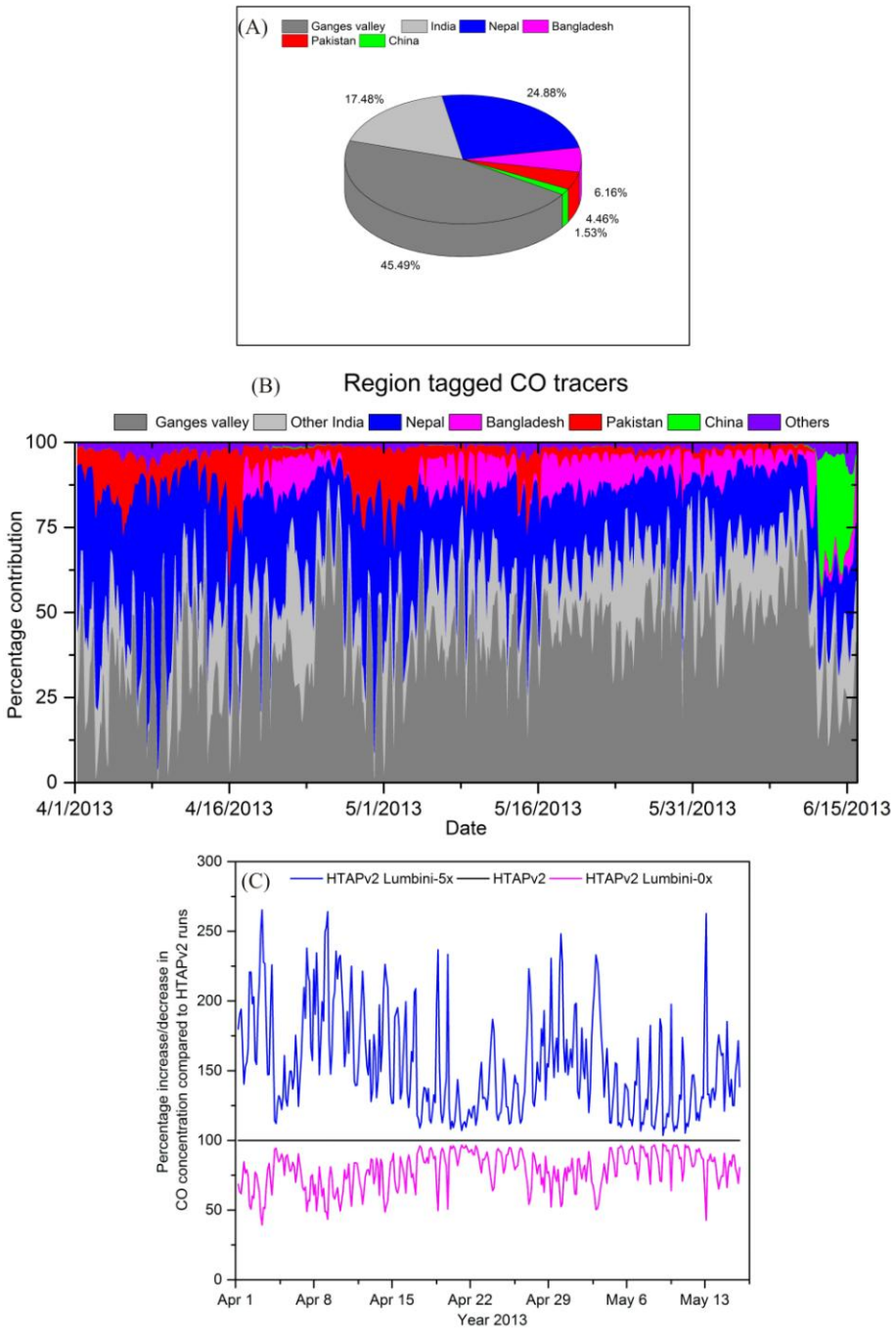
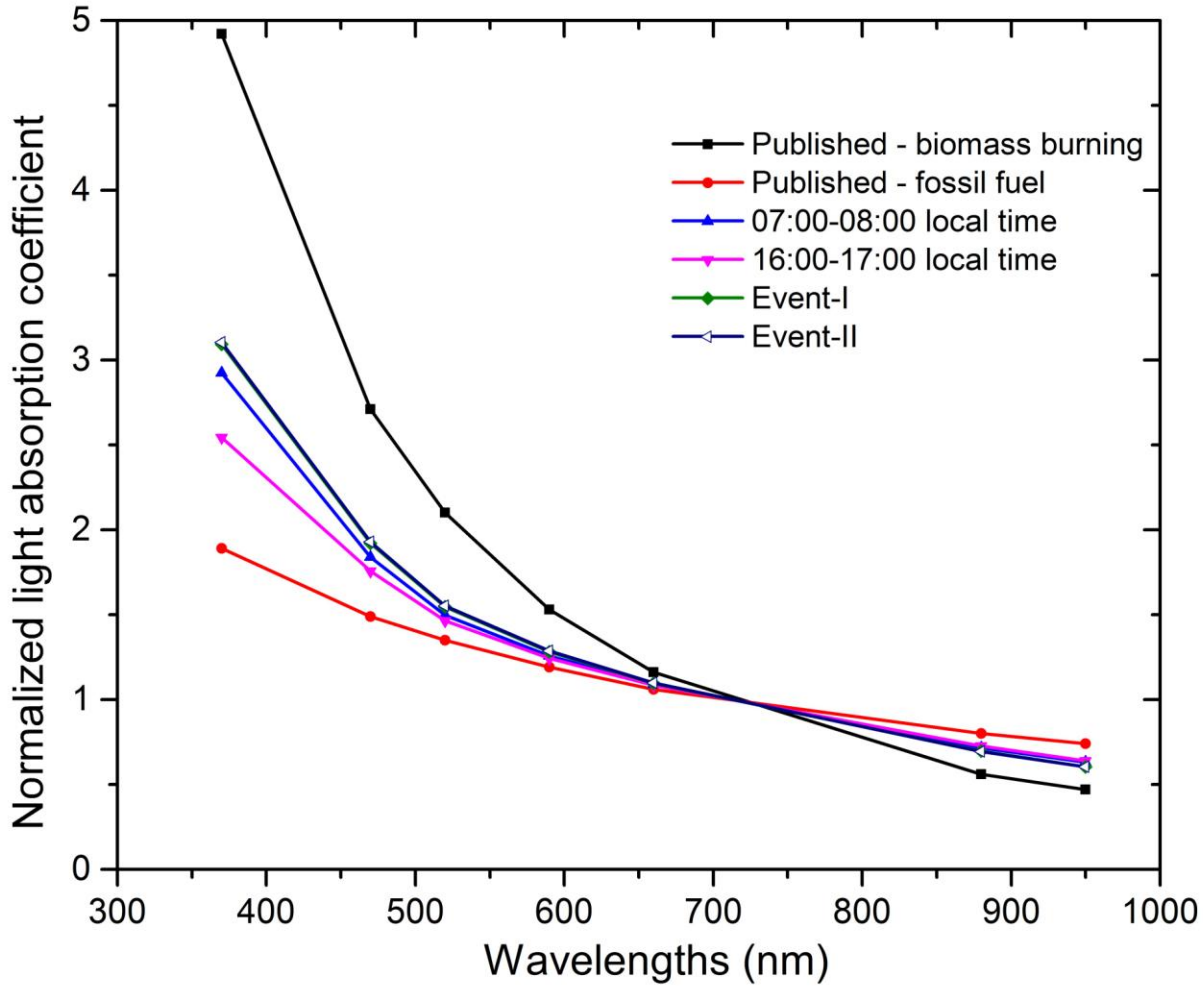


1096

1097 **Figure 10.** (A) WRF-STEM model estimated contributions of various source regions to average
1098 CO concentration in Lumbini for the sampling period, (B) time series of region tagged CO tracer
1099 during the whole measurement period using HTAP emission inventory and (C) Figure showing
1100 percentage increase/decrease in CO concentration with different emissions scenario.

1101



1102

1103 **Figure 11.** Comparison of normalized spectral light absorption coefficients obtained during the
1104 prime cooking (07:00-08:00 local time) and non cooking time (16:00-17:00 LT) at Lumbini with
1105 published data from Kirchstetter et al. (2004).

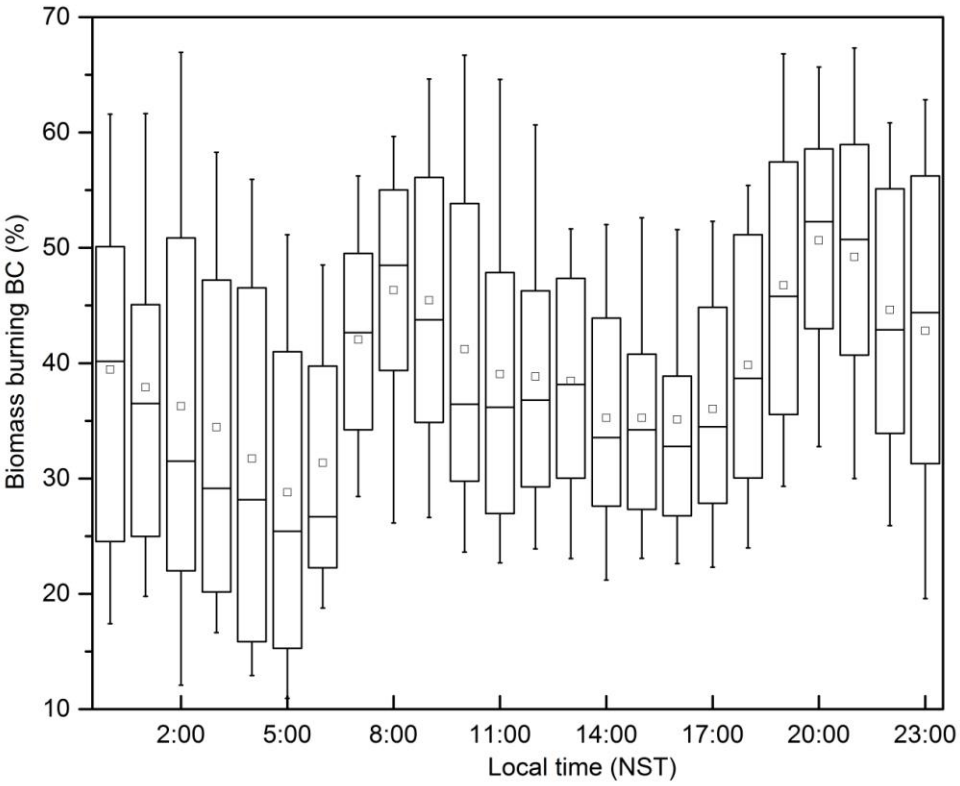


Figure 12. Diurnal variation of the fractional contribution of biomass burning to ambient BC concentration at Lumbini for the measurement period. In each box, lower and upper boundary of the box represent 25th and 75th percentile, respectively, top and bottom of the whisker represents 90th and 10th percentile, respectively. The mid-line in each box represents median while the square mark represents the mean for each hour.