Response to reviewers' comments on Mahata et al. 2018

We would like to thank the anonymous reviewers for their comments and suggestions which, we believe, have supported to improve the quality of the current manuscript. We have tried our best to incorporate both reviewers' comments in the manuscript. In the following responses, the reviewers' original comments are in black, authors' responses in blue and changes in the manuscript in red.

Anonymous Referee #1

General remarks

This paper reports air pollution (ozone and CO) in the Kathmandu area over longer time periods than hitherto available. Air pollution in this region is an important problem and reliable information covering all seasons is an important contribution to research on these issues. And I agree with the authors that the high ozone mixing ratios observed during the pre-monsoon period is of a high concern for human health and ecosystems, in the region. Here I would encourage the authors to go beyond what is presented in the paper and (briefly) discuss possible mitigation options (following the idea of "policy relevant, not policy prescriptive").

However, I have also some reservations about the interpretation of some aspects of the reported data and also some issues with the presentation. I suggest taking these points into account when revising the paper. If this is done in an appropriate way, I suggest that the Editor accepts the paper for publication in ACP.

We would like to thank you for considering that our study is of high importance for the region. We have tried our best to incorporate the suggestions in the revised manuscript.

Comments in detail

One aspect that is only discussed in passing in the paper is the role of stratospheric intrusions as a source of ozone in the upper troposphere in the region (e.g., Wang et al., 2012). Thus, ozone at higher altitudes in the troposphere could be enhanced independent of tropospheric pollution. I suggest that this aspect should be better discussed in the paper.

Thank you for noting the importance of stratospheric intrusions in the troposphere. We have included this fact in the manuscript in lines 524-538.

The diurnal profiles of O_3 mixing ratios (Figure 7) at three sites Bode and Pakanajol in the Valley and Nagarkot, a hilltop site normally above the Kathmandu Valley's boundary layer shows, notably in the morning hours, that the residual layer above the Kathmandu Valley's mixing layer contains a significant amount of ozone. Based on the surface ozone data collected at Paknajol during 2013-14, Putero e al. (2015) concluded that downward mixing of ozone from the residual layer contributes to surface ozone in the Kathmandu Valley in the afternoon hours (11:00-17:00 local time). It is likely that the same source has also contributed to higher ozone mixing ratios at Nagarkot. Such mixing has been observed at other sites as well. Wang et al. (2012) reported that the increase in downward mixing of O_3 from the stratosphere to the middle troposphere (56%) and the lower troposphere (13%) in spring and summer in Beijing. The downward flux was highest in the middle troposphere (75%) in winter. Similarly, Kumar et al. (2010) reported more than 10 ppb of stratospheric contribution to surface ozone at a high altitude site (in Nainital) during January to April. However, there were no significant stratospheric intrusions seen in spring and summer (seen only in winter) at Nepal Climate Observatry - Pyramid (NCO-P) located near the basecamp of Mt. Everest (Putero et al., 2016).

And discuss more about it in lines 547-557.

A study by Putero et al., (2015), based on O_3 mixing ratio measurements at Paknajol in the Kathmandu Valley, as a part of the SusKat-ABC campaign, has reported that the dynamics (both by horizontal and vertical winds) plays a key role in increased O_3 mixing ratios in the afternoon in the Kathmandu Valley. They estimated that the contribution of photochemistry varied as a function of the hour of the day, ranging from 6 to 34 %. Unfortunately, no viable NOx measurements were obtained at any site in the Kathmandu Valley and surrounding mountain ridges during the SusKat-ABC campaign. Speciated VOCs were measured at Bode only for about 2 months but NOx was not available for the same period. Therefore we were not able to discern quantitatively proportional contributions of NOx, VOCs and intrusion (chemistry vs. dynamics) from the free troposphere or lower stratosphere to observed O_3 concentrations at Nagarkot, Bode and other sites in the Valley.

Further, I suggest more comparison of the ozone pollution found at the Kathmandu valley with pollution levels elsewhere in the world (e.g. Huszar et al., 2016). Are the close to zero ozone

values reported here (due to NO titration) also found in other regions of the world? These questions are important for mitigation strategies, because to achieve significant ozone reduction over cities in central Europe, the emission control strategies have to focus on the reduction of VOCs (Huszar et al., 2016).

Thanks for the suggestions. We have compared the level of O_3 observed in this study with the values reported in studies at other sites in different parts of the world. The new texts are in lines 498-503.

Similar patterns of ozone mixing ratios were observed at other sites in northern South Asia. For example, higher O_3 mixing ratios were observed in the afternoon (84 ppb) and lower during the night and early morning hours (10 ppb) at Kullu Valley, a semi-urban site located at 1154 m asl, in the North-western Himalaya in India (Sharma et al. 2012). A similar dip in O_3 value in the dark hours was observed at Ahmedabad, India by Lal et al. (2000).

And in lines 560-565.

However, air quality management plans need to consider carefully the reduction strategies of NMVOCs or NOx while aiming at mitigating the O_3 pollution in the Kathmandu Valley. If the correct strategy (NMVOCs vs. NOx) is not applied, then O_3 mixing ratios could increase, for example, as seen in Huszar et al. (2016) where they report that reducing NMVOCs in urban areas in central Europe leads to O_3 reduction whereas the focus on NOx reduction results in O_3 increase.

I repeat my comment on Fig. 1 from the initial/quick review here: I find the Google Earth figure not appropriate. The yellow pins are strange and the blue letters are difficult to read against the background. I suggest changing to a figure showing the locations of the sites in a map showing the orography clearly.

Thank you for the suggestion to improve the quality of Fig. 1. We have replaced the Google Earth figure by new one which is showing the orography clearly in map.

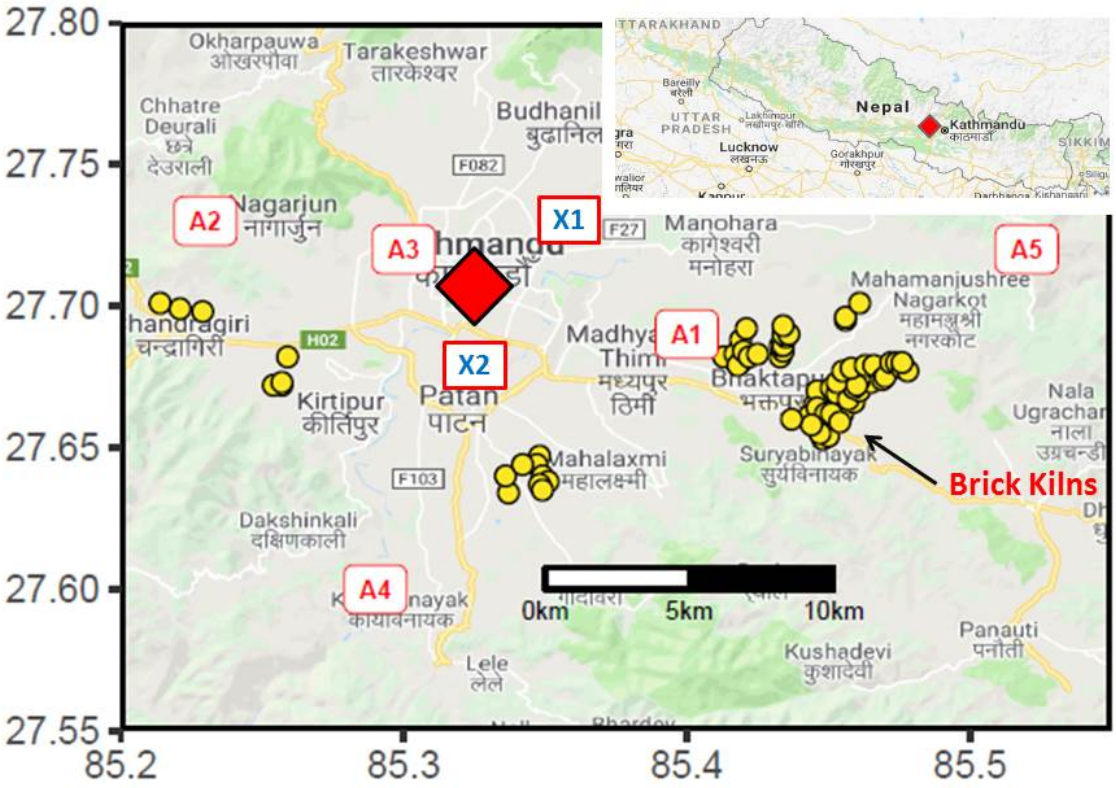

Figure 1. Observation sites in the SusKat-ABC international air pollution campaign during 2013-2014 in the Kathmandu Valley. A1 = Bode, A3 = Paknajol, and A4 = Naikhandi were selected within the valley floor and A2 = Bhimdhunga and A5 = Nagarkot on the mountain ridge. Naikhandi site is also near the Bagmati River outlet. Past study sites, Bouddha (X1) and Pulchowk (X2), which are referred in the manuscript, are also shown in the Figure. Source: Google map.

I also suggest to state the calendar months, not just the seasons. This is done in l. 271, but it should also be stated in the introduction and in the abstract.

It has been included in the introduction and abstract according to the reviewer's suggestion in lines 47-48 and lines 140, 166-167 and 171-172.

The value of for the CO flux at Bode is given to three significant numbers, is this really appropriate? Do you have an error estimate for this number? I think this value is an important result from this study so it deserves some attention.

We would like to thank you for pointing to error estimates in estimates of CO emission fluxes. Also, thank you for considering the CO flux estimate as an important result of our study.

- We have corrected the numbers to show only one significant digit in the abstract, chapter 3.5 and conclusions in the manuscript.
- Uncertainty in the CO flux estimate is introduced by the measurement uncertainty of the instruments, both for the CO mixing ratios and the mixing layer height (MLH). These contribute to the estimated CO emission fluxes varying over a wide range. In order to emphasize this wide variability in the estimated CO emission fluxes, we now show in Figure 8, the mean, interquartiles, and the minimum values of CO flux under our assumption.

Finally, I could very well imagine that the data presented in this paper are of interest to other researchers as well. Therefore I suggest to add a comment on data availability to the paper.

It is now required in the ACP to have a data availability section. The data availability section is included in lines 800-803 in the manuscript.

Data Availability: The observational data collected for this study will be made public through the SusKat website of IASS. They are also available upon direct request sent to maheswar.rupakheti@iass-potsdam.de and khadak.mahata@iass-potsdam.de.

Minor issues

• l. 31: drop ‘on’

Incorporated.

• l. 32: ‘pollutants’

Incorporated.

• l. 37: add altitude for Naikhandi

Incorporated.

• l. 42: State ‘how long’ extended

The campaign was extended until March 2014. This is included in the manuscript in line 43.

• l. 46: state the calendar months, not everybody is familiar with these seasons.

Calendar months are included in lines 47- 48.

• l. 46/47: ‘due to the emissions from brick kiln industries’ How do you know? How much of this is speculation/hypothesis how much is really shown in the paper?

Thank you for your question. Reviewer # 2 has also asked the same question. Our arguments are based on the previous studies. Previous studies carried out at the Bode site during the SusKat-ABC campaign have attributed over a dozen brick kilns located near Bode as strong sources of BC and EC (Kim et al., 2015; Mues et al., 2017), NMVOCs (Sarkar et al, 2016; Sarkar et al., 2017), SO2 (Kiros et al., 2016) and CO (Mahata et al., 2017), and the enhanced concentrations were observed during nighttime and mornings when winds blew from east and southeast bringing emissions from the location of the brick kilns to the observation site. Thus, we have rephrased the sentence (in lines 46-51 in abstract) as follows to better articulate it, and also explained in the main text with reference to other studies.

Seasonally, CO was higher during pre-monsoon season (March-May) and winter (December-February) season than during monsoon season (June-September) and post-monsoon (October-November) season. This is primarily due to the emissions from brick industries, which are only operational during this period (January-April), as well as increased domestic heating during winter, and regional forest fires and agro-residue burning during the pre-monsoon season.

The information is added in the texts in lines 415- 420 as follows:

Previous studies carried out at the Bode site during the SusKat-ABC campaign have attributed over a dozen brick kilns located near Bode as strong sources of BC and EC (Kim et al., 2015; Mues et al., 2017), NMVOCs (Sarkar et al, 2016; Sarkar et al., 2017), SO2 (Kiros et al., 2016) and CO (Mahata et al., 2017), and the enhanced concentrations were observed during nighttime

and mornings when winds blew from east and southeast bringing emissions from the location of the brick kilns to the observation site.

• l.50: in which way did the meteorology play a role?

Thank you for the question. The role of meteorology has been explained in the abstract (lines 54-57) adding sentences as follows:

The wind is calm and easterly in the shallow mixing layer, with a mixing layer height (MLH) of about 250 m, during the night and early morning. The MLH slowly increases after the sunrise and decreases in the afternoon. As a result, the westerly wind becomes active and reduces the mixing ratio during the day time.

• l. 52: 'Some influence' is a bit vague, can you be more specific here?

Our study and a companion study by Bhardwaj et al. (2017) have identified the influence of emissions outside the Kathmandu Valley on the increase in ozone concentrations in the valley. The sentence has been rephrased in lines 57-60 as follows:

Furthermore, there was evidence of an increase in the O_3 mixing ratios in the Kathmandu Valley as a result of emissions in the Indo-Gangetic Plain (IGP) region, particularly emissions from biomass burning, including agro-residue burning.

• l. 54: The value of 4.92 is given to three significant numbers, is this really appropriate? Do you have an error estimate for this number?

Thank you for the suggestion. We agree. Thus, we kept it in round figure as discussed above in general comments. For example, 4.92 is rounded off to 4.9 in line 62.

• l. 63: 'as well as': which effect dominates?

Unfortunately, no viable NOx measurements were obtained at any site in the Kathmandu Valley and surrounding mountain ridge during the SusKat-ABC campaign. Speciated VOCs were measured at Bode only for about 2 months, but NOx data was not available for the same period. Therefore we were not able to discern proportional contributions of NOx, VOCs, and intrusion from free troposphere or lower stratosphere to observed O_3 concentrations at Bode and other sites in and around the Valley. A study by Putero et al., (2015), based on O_3 measurement at Paknajol in the Kathmandu Valley, as a part of the SusKat-ABC campaign, has reported that the photochemistry plays a key role (larger role than the dynamics) in surface O_3 enhancement before noon and, together with the photochemistry, the boundary layer dynamics (both horizontal and vertical winds) also plays a role in increasing the O_3 mixing ratios in the afternoon (11:00-17:00 local time) in the Kathmandu Valley. They estimated that the contribution of photochemistry varied as a function of the hour of the day, ranging from 6 to 34 %. Due to unavailability of data on NOx and VOCs, we did not estimate which effect (chemistry vs. dynamics) is dominant in this case. Thus, we only slightly rephrased the sentence in lines 71-72 as follows:

…. air at the high-altitude site, as also indicated by Putero et al., (2015) for the Paknajol site in the Kathmandu Valley, as well as….

This is further explained in the lines 603- 605.

….as well as entrainment of ozone due to dynamics (both intrusion of ozone rich free tropospheric air into the boundary layer, and regional scale horizontal transport of ozone), as explained in case of Paknajol by Putero et al. (2015).

• l. 65: on the basis of which assessment can you say ‘due to’?

This sentence would require more detailed explanation than is appropriate for the abstract, thus we have deleted this sentence from the abstract.

• l. 80: one further impact of local pollution could also be convective uplift to tropopause altitudes and transport into the extra-topical stratosphere in the monsoon season (e.g. Tissier and Legras, 2016, and references therein).

Thank you for this suggestion, which has been included in the manuscript. Revised sentences are as follows in lines 92-95.

Similarly, pollutants are also uplifted to the tropopause by convective air masses and transported to the extratropical stratosphere during the monsoon season (Tissier and Legras., 2016; Lawrence and Lelieveld, 2010; Fueglistaler et al., 2009; Highwood and Hoskins, 1998).

• l. 93: 2017 → 2018 • l. 97: also toxic outdoors?

Incorporated in lines 113 and 120.

• l. 115: measured → reported measurements

Incorporated in line 138.

• l. 133: for the Kathmandu . . .

Incorporated in line 157.

• l. 167: O3

Incorporated in line 192.

• l. 227: define ‘AWS’

Automatic weather station (AWS) has been defined in line 241.

• l. 286: due to a problem

Incorporated in line 324.

• l. 320, 321: How do you know?

Past studies references have been included in lines 366-368 in the manuscript.

The morning peak at Bode was influenced by nighttime accumulation of CO along with other pollutants from nearby brick kilns (Sarkar et al., 2016; Mahata et al., 2017; Mues et al., 2017) and recirculation of air from above (Panday and Prinn, 2009).

• l. 339: CO mixing ratios

Incorporated in line 391.

• l. 453: stratospheric intrusions are mentioned here but only in passing.

Thank you for the suggestion. We have been inserted few sentences to elaborate it a bit in lines 547-557.

A study by Putero et al., (2015), based on O_3 mixing ratio measurements at Paknajol in the Kathmandu Valley, as a part of the SusKat-ABC campaign, has reported that the dynamics (both by horizontal and vertical winds) plays a key role in increased O_3 mixing ratios in the afternoon in the Kathmandu Valley. Unfortunately, no viable NOx measurements were obtained at any site in the Kathmandu Valley and surrounding mountain ridges during the SusKat-ABC campaign. Speciated VOCs were measured at Bode only for about 2 months but NOx was not available for the same period. Therefore we were not able to discern quantitatively proportional contributions of NOx, VOCs and intrusion (chemistry vs. dynamics) from the free troposphere or lower stratosphere to observed O_3 mixing ratios at Nagarkot, Bode and other sites in the Valley.

• l. 472: make → draw

Incorporated in line 573.

• l. 506: give altitude of Nagarkot here. Also the statement here is a bit vague, can you be more quantitative here (instead of 'but is also').

Thank you for the suggestion. We have rephrased the sentence in lines 608-611 as follows:

The ozone mixing ratios are relatively constant throughout the day at Nagarkot (~1901 m asl), which, being a hilltop site, is largely representative of the lower free tropospheric regional pollution values, however, it is also affected by ozone production from precursors transported from the Kathmandu Valley due to westerly winds during the afternoon hours.

• Mues et al. (2017); citation is missing

Missing citation has been included in the reference.

• l. 546: be specific what is meant with 'this'

This represents mass per unit area. It has been changed in line 657.

• l. 611: change to 'an observation connected to'

Incorporated in line 749.

• l. 617: drop 'the' • l. 622: episode days → episodes

Incorporated in lines 755 and 760.

• l. 632: are these ozone values typical for down-mixing?

As explained earlier, a study by Putero et al., (2015), based on O_3 measurement at Paknajol in the Kathmandu Valley, as a part of the SusKat-ABC campaign, has reported that together with the photochemistry, the boundary layer dynamics (both horizontal and vertical winds) also plays

a role in increasing the O_3 mixing ratios in the afternoon (11:00-17:00 local time) in the Kathmandu Valley. They estimated that the contribution of photochemistry varied as a function of the hour of the day, ranging from 6 to 34 %. A companion study by Bhardwaj et al. (2018) as also indicated role of dynamics in ozone levels in the Kathmandu Valley. The values we have observed are typical for down mixing. We have now cited these two previous studies in lines 769-773.

The diurnal cycle showed evidence of photochemical production, larger scale advection of polluted air masses as well as possible down-mixing of O3 during the daytime, as also observed by Putero et al., (2015) at Paknajol, with the hourly mixing 632 ratio at the polluted site increasing from typically 5-20 ppb in the morning to an early afternoon peak of 60-120 ppb (Putero et al., 2015; Bhardwaj et al., 2018).

• l. 646-650: perhaps two sentences here

Agree. The long sentence has been broken down to two as follows in lines 786-790.

This points out the need for the development of updated comprehensive emission inventory databases for this region. The improved emission inventory is necessary to provide more accurate input data to model simulations to assess air pollution processes and mitigation options for the Kathmandu Valley and the broader surrounding region.

• l. 711: This paper is now accepted

Incorporated in lines 855-858 in the reference section.

• Figs. 5 and 7: can you show error bars in these figures?

Thank you for the suggestions on adding error bars in Figs. 5 and 7. It will add value in the figures. The error bars has included in revised Figures 5 and 7 as suggested.

References

Fueglistaler, S., Dessler, A. E., Dunkerton, T. J., Folkins, I., Fu, Q., and Mote, P. W.: Tropical tropopause layer, Rev. Geophys., 47, RG1004, doi:10.1029/2008RG000267, 2009.

Highwood, E. J. and Hoskins, B. J.: The tropical tropopause, Q. J. Roy. Meteorol. Soc., 124, 1579–1604, doi:10.1002/qj.49712454911, 1998.

Huszar, P., Belda, M., and Halenka, T.: On the long-term impact of emissions from central European cities on regional air quality, Atmos. Chem. Phys., 16, 1331–1352, doi:10.5194/ acp-16-1331-2016, 2016.

Lal, S., Naja, M., and Subbaraya B. H.: Seasonal variations in surface ozone and its precursors over an urban site in India, Atmos. Environ., 34, 2713-2724, doi: 10.1016/S1352-2310(99)00510-5, 2000.

Putero, D., Cristofanelli, P., Sprenger, M., Škerlak, B., Tositti, L., and Bonasoni, P.: STEFLUX, a tool for investigating stratospheric intrusions: application to two WMO/GAW global stations. Atmos. Chem. Phys., 16, 14203–14217, doi:10.5194/acp-16-14203-2016, 2016.

Sharma, P., Kuniyal, J. C., Chand, K., Guleria, R. P., Dhyani, P. P., and Chauhan, C.: Surface ozone concentration and its behavior with aerosols in the northwestern Himalaya, India. Atmos. Environ. 71, 44-53, doi:10.1016/12.042, 2013.

Tissier, A.-S. and Legras, B.: Convective sources of trajectories traversing the tropical tropopause layer, Atmos. Chem. Phys., 16, 3383–3398, doi:10.5194/acp-16-3383-2016, 2016.

Wang, Y., Konopka, P., Liu, Y., Chen, H., Müller, R., Plöger, F., Riese, M., Cai, Z., and Lü, D.: Tropospheric ozone trend over Beijing from 2002–2010: Ozonesonde measurements and modeling analysis, Atmos. Chem. Phys., 12, 8389–8399, doi:10.5194/acp-12-8389-2012, 2012.

Anonymous Referee #2

The manuscript "Observation and analysis of spatio-temporal characteristics of surface ozone and carbon monoxide at multiple sites in the Kathmandu Valley, Nepal" by Mahata and co-authors provides an analysis of CO and O3 measurements carried out at 4 sites in the Kathmandu valley during the course of one year. Due to this good data coverage the analysis allows for a more thorough analysis than previously possible and also provides some valuable CO emission estimates. The paper is well written and organised and after minor corrections suitable for publication in ACP.

We would like to thank you for considering our study valuable, and for providing constructive comments to improve the quality of our analysis. We have tried our best to address your comments and suggestions in the revised manuscript.

Minor comments

L46 and elsewhere: Here a strong statement is made about the significant contribution of brick kilns to the observed CO concentrations. However, there is little actual proof of this shown in the manuscript. This could be improved by indicating the location of the kilns in relation to the measurement locations and a more thorough analysis/description of the nighttime wind pattern. Both of which would allow for a more creditable source attribution. Since there were also other atmospheric tracers measured at Bode, couldn't one of them (e.g. SO2 also be used to support the kiln contribution?

We would like to thank you for the suggestion. We have included the following evidence to more concretely attribute the influence of brick kiln on CO mixing ratios.

- As you suggested, we have revised the Figure 1 by marking the locations of brick kilns near the sampling site.
- Other studies conducted at the Bode site during the SusKat-ABC campaign attributed nearby brick kilns as strong sources of BC and EC (Kim et al., 2015; Mues et al., 2017), NMVOCs (Sarkar et al, 2016; Sarkar et al., 2017), SO2 (Kiros et al., 2016) and CO (Mahata et al., 2017), and the enhanced concentrations were during nighttime and mornings when winds blew from east and southeast bringing emissions from the location of the brick kilns to the observation site.

The revised text reads in lines 415-420 as follows:

Previous studies carried out at the Bode site during the SusKat-ABC campaign have attributed over a dozen brick kilns located near Bode as strong sources of BC and EC (Kim et al., 2015; Mues et al., 2017), NMVOCs (Sarkar et al, 2016; Sarkar et al., 2017), SO2 (Kiros et al., 2016) and CO (Mahata et al., 2017), and the enhanced concentrations were observed during nighttime and mornings when winds blew from east and southeast bringing emissions from the location of the brick kilns to the observation site.

L50, 51: Please mention in which way meteorology played a key role.

The role of meteorology has been included in lines 54-57(see also response to comment L50 by the reviewer 1). The new text reads as follows:

The wind is calm and easterly in the shallow mixing layer, with a mixing layer height (MLH) of about 250 m, during the night and early morning. The MLH slowly increases after the sunrise and decreases in the afternoon. As a result, the westerly wind becomes active and reduces the mixing ratio during the day time.

L72: Please split this number into casualties due to indoor and outdoor pollution. The first number seems to be the more important one in the light of your study.

Thank you for the suggestion. The impact on premature death due to outdoor and indoor air pollution has been included in lines 83-85.

The latest WHO report shows that the indoor and outdoor air pollution are each responsible for about 4 million premature deaths every year (http://www.who.int/airpollution/en/).

L83ff: Please also mention the special topographical and meteorological conditions (poor ventilation) that characterize the basin and further deteriorate air quality.

We have included the following line and also provided a reference on ventilation in lines 100-105.

In Kathmandu topography also plays a major role: the bowl-shaped Kathmandu Valley is surrounded by tall mountains and only a handful of passes. Topography is a key factor in governing local circulations, where low MLH (typically in the range 250 m to 1,500 m) and calm winds, have been observed particularly during nights and mornings. This in turn results in poor ventilation (Mues et al., 2017). Overall, this is conducive to trapping air pollutants and the deterioration of air quality in the valley.

L96: "CO is a useful tracer of urban air pollution". In the light of large contributions to CO from forest fires and agricultural waste burning (discussed later in the text), you should mention this important source as well.

Thank you for highlighting other important sources of CO other than urban sources. We have included forest and agro-residue waste burning sources of CO in lines 117-119.

Forest fires and agro-residue burning in the IGP and foothills of the Himalaya are other important contributors of CO in the region (Mahata et al., 2017; Bhardwaj et al., 2017).

L122: How does the CO emission estimate by Shrestha et al. (2013) compare with your emission estimate? Please add to the discussion in Section 3.5.

Shrestha et al., (2013) estimated the amount of CO emitted by a fraction of the vehicle fleet in the Kathmandu Valley. They neither estimated the total CO emission from all sources nor the CO fluxes in the Kathmandu Valley. Hence we cannot compare our estimate, which is from all sources, with theirs, which is from only a fraction of vehicle fleet.

L206 and for following sites: Where was the inlet mounted? What is the total height above ground of the inlet? Repeat from table 2.

The suggestion has been incorporated. The new text reads as follows in

Lines 232-233

The inlets of the CO and O_3 analyzers were mounted on the roof top of the temporary lab, 20 m above the ground level.

Line 241

…. building and its inlet was 2 m above ground. An automatic…..

Lines 249-250

…. The inlet of the O_3 analyzer was placed 25 m above the ground.

Lines 257-258

The instruments were kept in a one-story building of the school and its inlet was 5 m above the ground. The AWS….

And line 264

…Nagarkot Health Post and their inlets were 5 m above the ground. The AWS…

L254: These IR CO analyzers usually show a strong drift with lab temperature. Did you assure that lab temperatures varied as little as possible (AC) or did you use some additional drift correction? Once daily zero checks would probably not be sufficient. Can you rule out that part of the observed diurnal cycle of CO is due to instrument errors?

We agree with your concern regarding a drift in IR based CO analyzers due to lab temperature. We didn't use an AC to maintain the temperature. We tried to keep the fluctuation in room temperature as small as possible using fans and windows. The IR-based CO monitor was run simultaneously with a co-located cavity ring down spectrometry based CO analyzer (Picarro CO analyzer) for ~ 3 months. The correlation coefficient and slope between their data are 0.99 and 0.96 respectively. This indicates there was very small drift in IR-based CO values (refer Mahata et al., 2017 for details). Therefore, we do not need to apply a correction to the IR-based CO data, and we can be confident that the observed diurnal cycle is not due to the instrument errors in IR-based CO measurements. We included a line as follows to reinforce this point in lines 283-289.

An IR-based Thermo CO monitor (model 48i-TLE) was run simultaneously with a co-located cavity ring down spectrometry based Picarro CO analyzer for nearly 3 months. The correlation coefficient and slope between the two measurements were found to be 0.99 and 0.96, respectively (Mahata et al., 2017). This indicates that there was very little drift in the IR-based CO values due to room temperature change, within acceptable range (i.e., within the measurement uncertainties of the instruments). Therefore, we did not any apply correction in the IR-based CO data.

L258: What was the result of the span check? Did the instrument drift since the last span check?

Thank you for your suggestion to make clear about the span check and drift of the instrument. We have included more information about it in line 289 as follows

The IR-based CO instruments' span drifts were within a 5 % range.

L283: What are the given uncertainties? Standard deviation of hourly observations? Uncertainty of the mean?

We would like to thank you for pointing out the confusion about the given uncertainties which are the standard deviation of the hourly averaged data, which is now clarified in lines 319-321 as follows:

The CO mixing ratios (measured in parts per billion by volume, hereafter the unit is denoted as ppb) of hourly averaged data over the total observation periods at four sites and their standard deviation were: Bode (569.9 ± 383.5) ppb…..

L302: Can you mention a bit more about what is known about the kind and timing of trash burning? Are these small scale fires (individual households) or larger scale (communities/neighbourhood)? Are there any regulations on this kind of waste treatment? It is mentioned elsewhere that this happens at night? Why? Seems to be a rather simple process to tackle to improve overall air quality.

Thanks for the suggestion. The information on waste type and timing has been added in lines 342-348.

Other studies conducted during the SusKat-ABC campaign have identified garbage (household waste and yard waste) burning as a key source of various air pollutants, such as OC and EC (Kim et al., 2015), PAHs (Chen et al., 2015), and NMVOCs (Sarkar et al., 2016; Sarkar et al., 2017). Garbage burning is often done in small fires and quite sporadic, normally taking place in the evenings and mornings (partly chosen to avoid attention from the responsible authorities). The rate of waste (and also biomass) burning in the morning is higher in winter due to the use of the fires for providing warmth on colder days.

L335: "support turbulent vertical diffusion". Although this statement is absolutely true, this is already reflected by the deeper mixing layer during daytime. I suggest to reformulate in such a way that the reasons for a deeper mixing layer are given in the first sentence (heating of surface by incoming solar radiation and (secondary) higher horizontal wind speeds and turbulence production). Then only mention the flushing effect of the increased wind speeds in the second sentence. In the end, the increased horizontal wind speeds are caused by the growing mixing layer height as well, so buoyancy production of turbulence is the real cause for the increased ventilation of the surface layer, but the above discussions seems to be sufficient.

The paragraph is rephrased according to the suggestion in lines 383-388.

The MLH starts increasing after radiative heating of the surface by incoming solar radiation. The heating of the ground causes thermals to rise from the surface layer resulting in the entrainment of cleaner air from above the boundary layer leading to the dissolution of nocturnal stable boundary layer. Increasing wind speeds (4-6 m s^{-1}) wind speeds (4-6 m s^{-1}) during daytime also support turbulent vertical diffusion, as well as flushing of the pollution …..

Section 3.2.1: You could also comment on the distinctly different shapes of the nighttime increase at Bode and Bhimdhunga. Bode shows an almost linear increase, which may indicate continued emissions into the local stable boundary layer, whereas Bhimdhunga shows a more isolated peak during the morning transition phase. So it would indicate that slope winds bring part of the polluted valley boundary layer up to the pass even in the early morning, which seems well possible considering the east facing slope above which the site is located. The same influence can be seen in O_3 at the site.

We would like to thank you for pointing out nighttime distinct shape of increased CO at Bode and Bhimdhunga. We are agree with your argument that the linear increase of CO at Bode is because of continuous addition of CO emitted from continuous sources nearby, i.e., brick kilns, in the shallow boundary layer. The isolated peak found at Bhimdhunga in morning at a mountain ridge could be due to elevated polluted layer brought up to the site by the up slope winds that start once the east-facing slope is heated by the morning sun. Thus, we have rephrased the paragraph in lines 375-380 as follows:

This is mainly associated with the persistent emissions such as those from brick kilns, which are in close proximity to the Bode measurement site under the stable boundary layer. The isolated peak during the morning transition phase at Bhimdhunga could be due to an elevated polluted layer because of the slope wind (Panday et al., 2009).

L351f: The argument about decreased forest fires and agricultural waste burning should be clarified a bit. Up to this point in the manuscript one had the impression that most of the CO at Bode was due to the brick kilns. But now the big difference between the seasons is explained through the absence of forest fires, etc and the brick kilns are only mentioned at the very end. When and why do they actually stop production? Due to the precipitation in the monsoon season?

The brick kilns are operated seasonally, from January to April every year. They are shut down the summer monsoon rainy period (June-September). We have rephrased the sentence and clarify about the brick kiln closure in monsoon period in lines 402-404 as follows:

Because of the rainfall, the brick production activities are stopped in the valley (usually they are operational from January-May every year). Further, the rainfall also….

L360: Why is this apparent? Even if the kilns operate at night you should show that there is a direct link to the site in terms of advection direction? Isn't residential heating the more likely candidate?

We have removed the confusing word "apparent". Brick kilns are operated even in the night time. Thus, the calm easterly wind brings pollutants from nearby brick kilns to the site (refer supplementary Figure S2 in Mahata et al., 2017; Mues et al., 2018). We have incorporated the suggestion in lines 412-414 as follows:

The nighttime accumulation of CO in Bode during pre-monsoon and winter is due to the influence of nearby brick kilns (Mahata et al., 2017) because of the calm easterly wind (refer supplementary Figure S2 in Mahata et al., 2017).

L368: There is also a distinct shift in the morning peak visible at all 3 sites for the different seasons. Can you please comment on this? Probably it is just due to an earlier onset of the morning transition in Mar-Apr, but maybe changes in local emissions may play a role as well.

Thank you for pointing out the clear shift in the morning peak at all sites in different seasons. We are in agreement with you regarding ca. one hour shift in morning peak from pre-monsoon to winter. This is due to earlier onset of the activities due to an earlier sunrise during the pre-monsoon than in winter. However, one hour shift in morning peak between Bode and Bhimdhunga/Naikhandi in pre-monsoon and winter is associated with commencement of early local emission under the shallow boundary layer at Bode. One hour lag in the morning peak at Bhimdhunga and Naikhandi may be due to uplifting of polluted layer and transport of city

pollutants to the site, respectively, that starts only after the nearby slops are heated by solar radiation. One new paragraph is included in lines 429-435 as follows:

The distinct shift in the morning peak was seen at all 3 sites by season. The one hour shift in the morning peak from the pre-monsoon to winter is due to an earlier onset of the morning transition. However, the one hour difference in the morning peak between Bode (pre-monsoon at 8:00; winter at 9:00) and Bhimdhunga/Naikhandi (pre-monsoon at 9:00; winter at 10:00) in the pre-monsoon and winter is associated with commencement of early local emissions under the shallow boundary layer at Bode. The one hour lag in the morning peak at Bhimdhunga and Naikhandi may be due to transport of city pollutants to the site, respectively.

L393 "data means": Not clear which parameter is referred to here. Mean CO for the whole period or at a specific time of day?

Thank you for your suggestion to make it clear. We have tried to make it clear in lines 460-462.Yes, it is the mean CO of hourly data of the whole period. The rephrased sentences read as follows:

The t-test of the two hourly data means of CO in period I and period II at Bode, Bhimdhunga and Naikhandi (as in Figure 5) were performed at 95% confidence level and the differences were found to be statistically significant ($p < 0.5$).

L473: The comparability of the old time series with the recent may also be hampered by the difference in location and sampling height as well as a general difference in instrument calibration. These points should be mentioned as well.

Thank you for the suggestions. We have included them in the manuscript in lines 573-575.

…we cannot draw any conclusions about trends over the decade between the observations because of the difference in location and sampling height as well as a general difference in instrument calibration. However, a clear…

L499ff: Isn't the prolonged afternoon peak due to the same regional scale transport that was responsible for elevated CO? Free tropospheric contribution alone would not explain the difference between winter and pre-monsoon. Why not carry out the same kind of analysis as for CO in Figure 5.

We agree with you that the prolonged afternoon peak in the pre-monsoon could also be due to regional transport. We also carried out a similar analysis for O_3 (see figure below) as we did for CO in Figure 5, and also estimated the change in ozone mixing ratios (see table Table TS1) for the same two periods P1 and P2 (period 2 is being influenced by regional emissions). However, we don't see any prolonged afternoon peak on O_3 mixing ratios, at least not at Bode and Paknajol in the Valley, as we see in case of CO mixing ratios, likely because of the different reaction pathways of O_3 during night and day, and in different locations (valley floor vs. ridge)

Table. O_3 mixing ratios of period 1 (P1 = Mar 16-30) and period 2 (P2 = Apr 1-15) at Bode Paknajol and Nagarkot. The changes in O_3 mixing ratios and their percentage change at two periods were calculated during 24 hours and day time (8:00-18:00) hours.

	P2-P1	% change	P2d-P1d	% change in day hours
Site	24 hours	24 hours	8:00-18:00	8:00-1800
Bode	13.4	33.8	13.7	26.2
Paknajol	12.7	31.6	19.1	35.0
Nagarkot	16.1	30.5	17.5	27.2

The % change in O_3 between two periods (P2-P1) is almost same at Bode and Nagarkot and highest at Paknajol in both 24 hours and day hours (8:00-18:00) calculations. It is likely that as explained earlier dynamics (horizontal transport, including regional transport and vertical down-mixing from the free troposphere) contributed to observed afternoon ozone mixing ratios, as pointed out by Putero et al. (2015). It is difficult to infer anything how much each process contributed to increase in O_3 values in period 2. In case of CO, it is clearly seen that the upwind sites (Bhimdhunga and Naikhandi), i.e, unwind to the Paknajol, Bode and Nagarkot during

afternoon hours, had higher CO values than Bode in P2, indicating that there was a clear influence of regional transport of CO and hence led to prolonged afternoon peaks.

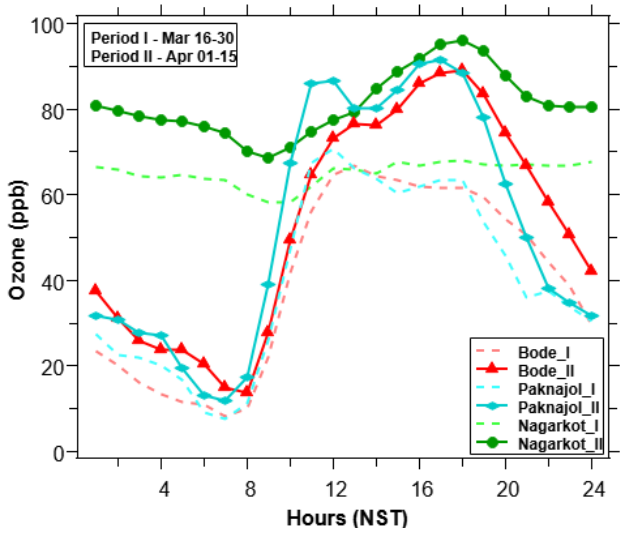

Figure. Comparison of hourly averaged O_3 mixing ratios during normal days (March 16-30) labelled as period I (dash line, faint color) and episode days (April 1-15) labelled as period II (line, dark color) in 2013 at Bode, Paknajol and Nagarkot.

Therfore, we have deleted the part that is poorly justified:
"Which Putero et al. (2015) suggested results in the broader afternoon peak of ozone during the pre-monsoon at Paknajol site, also observed at Bode site (and somewhat at Nagarkot)."
Therefore to reflect all the role of dynamics we have slightly modified the sentences in lines 601-605 as follows:

The typical O_3 maximum mixing ratio in the early afternoon at the urban and semi-urban sites is mainly due to daytime photochemical production as well as entrainment of ozone due to dynamics (both intrusion of ozone rich free tropospheric air into the boundary layer, and regional scale horizontal transport of ozone), as explained in case of Paknajol by Putero et al. (2015)

L505: The dip in O3 in the morning transition hours once more indicates the origin from the polluted stable boundary layer.

The suggestion has been included in the manuscript in lines 611-613

The dip in O_3 at Nagarkot (Figure 7) in the morning transition hours indicates the upward mixing of air from the polluted (and ozone-depleted) nocturnal boundary layer as it is breaking up.

L536 and equation 1: Why give t in hours? Why not just use seconds? Would save the conversion factor in the equation and is a better SI unit anyway!

This is because we used hourly averaged MLH and CO data. It is mentioned in the text (description of eq. 1).

542ff: One additional important limitation of the method is that of regional representativeness. As is said in the text, wind speeds are low so the observed CO increase at Bode may be rather localized and the emission estimate only valid for a small area and not for the whole city or valley. This is especially important when comparing the results with those from emission inventories that average over relatively large grid cells.

We agree with you that emissions are not uniform throughout the valley and thus our estimates may not be regionally representative. This can be checked only when we have a high resolution emission inventory, which (1 km x 1km spatial resolution) is being developed for the Kathmandu Valley and rest of Nepal (Sadavarte et. al., 2018). Therefore as per your suggestion, we have included a fifth point as one of our assumptions in lines 670-673.

v) CO emission is assumed to be uniform throughout the valley; this may not be correct, but cannot be verified until a high resolution emission inventory data is available, which is being developed for the Kathmandu Valley and rest of Nepal with a 1 km x 1km spatial resolution (Sadavarte et. al., 2018).

And we have revised assumption (iv) to make it clear in lines 660-663.

(iv) the vertical mixing of pollutants between the mixing layer and the free atmosphere is assumed to be negligible at night, thus strictly seen is the estimated CO flux calculated with eq. 1 only valid for the morning hours. When applied to the whole day the implicit assumption is that the emissions are similar during the rest of the 24 h period.

We have also reorganized 2nd paragraph of section 3.5 in lines 673-677.

During nighttime assumption (i) might not be entirely correct since the degree of mixing in the nocturnal stable layer and thus the vertically mixing is drastically reduced compared to daytime (and thus the term "mixing layer" is not entirely accurate, but we nevertheless apply it here due to its common use with ceilometer measurements). This adds a degree of uncertainty to the application of ceilometer observations to compute top-down emissions estimates, which will only be resolved once nocturnal vertical profile measurements are also available in order to characterize the nocturnal boundary layer characteristics and the degree to which the surface observations are representative of the mixing ratios throughout the vertical column of the nocturnal stable layer.

L555: Was the method actually applied to every night that had sufficient CO data? Or did you filter for low wind speed, constant MLH conditions? In which case it should be mentioned for how many nights per month the estimate was possible.

Yes, the method was applied when both CO and MLH hourly data were available during the night-morning hours. They were available for almost every day. Wind speed or any other filters were not applied during the flux calculation.

L574: Can you provide a realistic uncertainty for this estimate?

Thank you for the suggestion. Because the calculation of the emission flux is subject of several assumptions and there are no uncertainties given for the mixing layer height. It is not possible to calculate realistic uncertainties for the emission estimates. In order to still give an idea of the variability of the estimated flux numbers the 25th and 75th percentile is shown in Figure 8.

Instead of calculating realistic uncertainty, we have included few points (in lines 729-739) which have important role in flux estimates.

The emission estimates computed here are subject to several further uncertainties which are discussed in detail in Mues et al., (2017). In short, the uncertainties of CO flux estimates arise from (i) the assumptions that Bode site represents the whole atmospheric column and entire valley, which is not possible to verify without having many simultaneous monitoring stations in the valley (measurements at a few sites where CO was monitored for this study show some difference in CO mixing ratios), (ii) the higher variability (unclear minima and maxima during the morning and night hours) in the diurnal cycles of CO from June to October show a much higher variability than other months, that in turn makes it difficult to choose the exact hour of CO minimum and maximum needed for the flux estimation and (iii) the possible impact of wet deposition is not taken into account but would rather cause to generally underestimate the emission rate.

L587, the statement in brackets: Statement unclear? What do you mean by "averaged for the valley as a whole"? Did you apply the method also to other sites? Or just to Bode?

We have rephrased the sentence to avoid the confusion. We only apply this method at Bode. We do not have mixing layer height (MLH) measurements at other sites. The rephrased sentence reads in lines 713-716 as follows:

…..EDGAR HTAP V2.2 emission inventory database for 2010 [note that the CO emission values for the location at Bode and the whole valley (27.65-27.75°N, 85.25-85.40°E) were found to be the same up to two significant figures]….

L633: Again: mention the potential larger scale advection of polluted air masses (as for CO). See comment above.

The suggestion has been included in the conclusion section in lines 769-771. The rephrased sentence is as follows;

….The diurnal cycle showed evidence of photochemical production, larger scale advection of polluted air masses, as well as possible down-mixing of O_3 during the daytime, as also observed by Putero et al., (2015) at Paknajol, with the hourly….

Figure1: Instead of this 3D view, it would be more beneficial to have a plain 2D map with a scale indicator that would allow to identify the distances between sites. In addition, it would be a benefit to see the location of the large point sources (kilns) in such a map as well. Topography could still be included as isolines or shading. Main traffic routes would help as well.

Thank you for the suggestion. We have revised the figure as your suggestion. Please see reviewer 1's comments for the Figure 1above.

References

Sadavarte, P., Rupakheti, M., Shakya, K., Bhave, P.V., and Lawrence, M.G.: Nepal emission (NEEM): A high resolution technology - based bottom-up emissions inventory for Nepal, ACP in preparation, 2018.

Observation and analysis of spatio-temporal characteristics of surface ozone and carbon monoxide at multiple sites in the Kathmandu Valley, Nepal

Khadak Singh Mahata[1,2], Maheswar Rupakheti[1,3], Arnico Kumar Panday [4,5], Piyush Bhardwaj[6], Manish Naja[6], Ashish Singh[1], Andrea Mues[1], Paolo Cristofanelli[7], Deepak Pudasainee[8], Paolo Bonasoni[7], Mark G. Lawrence[1,2]

[1]Institute for Advanced Sustainability Studies (IASS), Potsdam, Germany

[2]University of Potsdam, Potsdam, Germany

[3]Himalayan Sustainability Institute (HIMSI), Kathmandu, Nepal

[4]International Centre for Integrated Mountain Development (ICIMOD), Lalitpur, Nepal

[5]University of Virginia, Charlottesville, USA

[6]Aryabhatta Research Institute of Observational Sciences (ARIES), Nainital, India

[7]CNR-ISAC, National Research Council of Italy – Institute of Atmospheric Sciences and Climate, Bologna, Italy

[8]Department of Chemical and Materials Engineering, University of Alberta, Edmonton, Canada

Correspondence to: Maheswar Rupakheti (maheswar.rupakheti@iass-potsdam.de) and Khadak Singh Mahata (khadak.mahata@iass-potsdam.de)

Abstract

Residents of the Kathmandu Valley experience severe particulate and gaseous air pollution throughout most of the year, even during much of the rainy season. The knowledge base for understanding the air pollution in the Kathmandu Valley was previously very limited, but is improving rapidly due to several field measurement studies conducted in the last few years. Thus far, most analyses of observations in the Kathmandu Valley have been limited to short periods of time at single locations. This study extends ~~on~~ the past studies by examining the spatial and temporal characteristics of two important gaseous air pollutants (CO and O_3) based on simultaneous observations over a longer period at five locations within the valley and on its rim, including a supersite (at Bode in the valley center, 1345 m above sea level) and four satellite sites (at Paknajol, 1380 m asl in the Kathmandu city center, at Bhimdhunga (1522 m asl), a mountain pass on the valley's western rim, at Nagarkot (1901 m asl), another mountain pass on the eastern rim, and Naikhandi (1233 m asl), near the valley's only river outlet). CO and O_3 mixing ratios were monitored from January to July 2013, along with other gases and aerosol particles by instruments deployed at the Bode supersite during the international air pollution measurement campaign SusKat-ABC (Sustainable Atmosphere for the Kathmandu Valley – endorsed by the Atmospheric Brown Clouds program of UNEP). The monitoring of O_3 ~~monitoring~~ at Bode, Paknajol and Nagarkot as well as the CO monitoring at Bode were extended ~~beyond July 2013~~ until March 2014 to investigate their variability over a complete annual cycle. Higher CO mixing ratios were found at Bode than at the outskirt sites (Bhimdhunga, Naikhandi and Nagarkot), and all sites except Nagarkot showed distinct diurnal cycles of CO mixing ratio with morning peaks and daytime lows. Seasonally, CO was higher during ~~the~~ pre-monsoon (March-May) season and winter (December-February) season than during monsoon season (June-September) and post-monsoon (October-November) seasons~~., especially~~ This is primarily due to the emissions from brick ~~kiln~~ industries, which are only operational during this period (January-April), as well as increased domestic heating during winter, and regional forest fires and agro-residue burning during the pre-monsoon season. It was lower during the monsoon due to rainfall, which reduces open burning activities within the valley and in the surrounding regions, and thus reduces ~~the~~ sources of CO. The meteorology of the valley also played a key role in determining the CO mixing ratios. The wind is calm and easterly in the shallow mixing

layer, with a ~~low~~ mixing layer height (MLH) of about 250 m, during the night and early morning. The MLH slowly increases after the sunrise~~s~~ and decreases in the afternoon. As a result, the westerly wind becomes active and reduces the mixing ratio during the day time. Furthermore, there was evidence of ~~some~~ an increase in the O_3 mixing ratios in the Kathmandu Valley as a result of emissions in the Indo-Gangetic Plain~~s~~ (IGP) region, particularly from biomass burning including agro-residue burning. ~~influence of pollution from the greater region around the valley.~~ A top-down estimate of the CO emission flux was made by using the CO mixing ratio and mixing layer height ~~(MLH)~~ measured at Bode. The estimated annual CO flux at Bode was 4.9~~2~~ μg m^{-2} s^{-1}, which is 2-14 times higher than that in widely used emission inventory databases (EDGAR HTAP, REAS and INTEX-B). This difference in CO flux between Bode and other emission databases likely arises from large uncertainties in both the top-down and bottom-up approaches to estimating the emission flux. The O_3 mixing ratio was found to be highest during the pre-monsoon season at all sites, while the timing of the seasonal minimum varied across the sites. The daily maximum 8 hour average O_3 exceeded the WHO recommended guideline of 50 ppb on more days at the hilltop station of Nagarkot (159/357 days) than at the urban valley bottom sites of Paknajol (132/354 days) and Bode (102/353 days), presumably due to the influence of free-tropospheric air at the high-altitude site, as also indicated by Putero et al., (2015) for the Paknajol site in the Kathmandu Valley as well as to titration of O_3 by fresh NOx emissions near the urban sites. More than 78% of the exceedance days were during the pre-monsoon period at all sites. ~~This was due to both favorable meteorological conditions as well as contributions of precursors from regional sources such as forest fires and agro-residue burning~~. The high O_3 mixing ratio observed during the pre-monsoon period is of a ~~high~~ concern for human health and ecosystems, including agroecosystems in the Kathmandu Valley and surrounding regions.

1. Introduction

Air pollution is one of the major health risks globally. It was responsible for premature loss of about 7 million lives worldwide in 2012 (WHO, 2014), with about 1.7 million of these being in South Asian countries (India, Pakistan, Nepal and Bangladesh) in 2013 (Forouzanfar, 2015). The latest report shows that the indoor and outdoor air pollution are each responsible for 4 million

premature deaths every year (http://www.who.int/airpollution/en/). South Asia is considered to be a major air pollution hotspot (Monks et al., 2009) and it is expected to be one of the most polluted regions in the world for surface ozone (O_3) and other pollutants by 2030 (Dentener et al., 2006; IEA 2016; OECD 2016). Past studies have shown that the air pollution from this region affects not only the region itself, but is also transported to other parts of the world, including comparatively pristine regions such as the Himalayas and the Tibetan plateau (Bonasoni et al., 2010; Ming, et al., 2010; Lüthi et al., 2015), as well as to other distant locations such as northern Africa and the Mediterranean (Lawrence and Lelieveld, 2010). ~~Similarly, t~~The pollutants are also uplifted to the tropopause by convective air masses and transported to the extratropical stratosphere during the monsoon season (Tissier and Legras., 2016; Lawrence and Lelieveld, 2010; Fueglistaler et al., 2009; Highwood and Hoskins, 1998). ~~The a~~Air pollution ~~problem~~ is particularly alarming in many urban areas of South Asia, including in the city of Kathmandu and the broader Kathmandu Valley, Nepal (Chen et al., 2015; Putero et al., 2015; Kim et al., 2015; Sarkar et al., 2016; Shakya et al., 2017). This is due to their rapid urbanization, economic growth and the use of poor technologies in the transportation, energy and industrial sectors. In Kathmandu topography also plays a major role: the bowl-shaped Kathmandu Valley is surrounded by tall mountains and only a handful of passes. Topography is a key factor in governing local circulations, where low MLH (typically in the range 250 m to 1,500 m) and calm winds, have been observed particularly during nights and mornings. This in turn results in poor ventilation (Mues et al., 2017). Overall, this is conducive to trapping air pollutants and the deterioration of air quality in the valley. Effectively mitigating air pollutants in the regions like the Kathmandu Valley requires scientific knowledge about characteristics and sources of the pollutants. To contribute to this urgently-needed scientific knowledge base, in this study we focus on the analysis of measurements of two important gaseous species, carbon monoxide (CO)~~, an urban air pollution tracer,~~ and O_3, at multiple sites in and around the Kathmandu Valley. This study analyzes data from January 2013 to March 2014, which includes the intensive phase of an international air pollution measurement campaign – SusKat-ABC (Sustainable Atmosphere for the Kathmandu Valley – Atmospheric Brown Clouds) – conducted during December 2012 - June 2013 (Rupakheti et al., 2018~~7~~, manuscript in preparation, submission anticipated in 1-2 months), with measurements of O_3 and CO at some sites continuing beyond the intensive campaign period (Bhardwaj et al., 2017; Mahata et al., 2017).

CO is a useful tracer of urban air pollution as it is primarily released during incomplete combustion processes that are common in urban areas. Forest fires and agro-residue burning in the IGP and foothills of the Himalaya are other important contributors of CO in the region (Mahata et al., 2017; Bhardwaj et al., 2017). ~~It~~ CO is ~~also~~ toxic at high concentrations indoors and outdoors, but our focus here is on ambient levels. The main anthropogenic sources of CO in the Kathmandu Valley are vehicles, cooking activities (using liquefied petroleum gas, kerosene, and firewood), and industries, including brick kilns, especially biomass co-fired kilns with older technologies, and until recently diesel power generator sets (Panday and Prinn, 2009; Kim et al, 2015; Sarkar et al., 2016; Mahata et al., 2017; Sarkar et al., 2017). Tropospheric ~~ozone~~O_3, which is formed by photochemical reactions involving oxides of nitrogen (NO_x) and volatile organic compounds (VOCs), is a strong oxidizing agent in the troposphere. Because of its oxidizing nature, it is also deleterious to human health and plants already at typically polluted ambient levels (Lim et al., 2012; Burney and Ramanathan, 2014; Feng, 2015; Monks et al., 2015). Tropospheric O_3 is estimated to be responsible for about 5-20 % of premature deaths caused by air pollution globally (Brauer et al., 2012; Lim et al., 2012; Silva et al., 2013). It has also been estimated that high concentrations of O_3 are responsible for a global loss of crops equivalent to $ 11-18 billion annually (Avnery et al., 2011; UNEP and WMO, 2011), a substantial fraction of which is associated with the loss in wheat in India alone (equivalent to $ 5 billion in 2010) (Burney and Ramanathan, 2014). O_3 can also serve as a good indicator of the timing of the breakup of the nighttime stable boundary layer (when the ozone levels increase rapidly in the morning due to downward transport from the free troposphere (Panday and Prinn, 2009; Geiß et al., 2017).

Only a few past studies have reported measurement~~s~~~~d~~ of ambient CO mixing ratios in the Kathmandu Valley. Davidson et al. (1986) measured CO in the city center and found mixing ratios between 1 and 2.5 ppm in the winter (December – February) of 1982-1983. Panday and Prinn (2009) measured similar levels of CO mixing ratios during September 2004 – June 2005, although the main sources of CO shifted from biofuel-dominated air pollutants from cooking activities in the 1980s to vehicle-dominated pollutants in the 2000s. The growth rate in the vehicle fleet has had a substantial influence on air pollution in the valley, including CO and O_3. Out of 2.33 million vehicles in Nepal, ~~~50%~~close to half of them are in the Kathmandu Valley (DoTM, 2015). Shrestha et al. (2013) estimated ~~31 kt of~~ annual emission of CO of 31 kt in 2010

from a fraction of ~~the~~ today's vehicle fleet in the Kathmandu Valley ~~in 2010~~ by using data from a field survey as input ~~data~~ to the International Vehicle Emission (IVE) model. The model simulation considered motorcycles, buses, taxis, vans and three wheelers, but did not include personal cars, trucks and non-road vehicles. The studied fleets covered ~73% of the total fleet (570,145) registered in the valley in 2010, with motorcycles being the most common vehicle (69% of ~~within~~ the total fleet).

Past studies have investigated the diurnal and seasonal variations of CO and O_3 mixing ratios in the Kathmandu Valley. Panday and Prinn (2009) observed distinct diurnal variations of CO mixing ratios and particulate matter concentrations observed during September 2004 – June 2005 at Bouddha (about 4 km northwest of the SusKat-ABC supersite at Bode), with morning and evening peaks. ~~It was~~They found for ~~in~~ the Kathmandu Valley that such peaks were created by the interplay between the ventilation, as determined by the local meteorology, and the timing of emissions, especially traffic and cooking emissions. The morning CO peak was also associated with the recirculation of the pollutants transported down from an elevated residual pollution layer (Panday and Prinn, 2009).

O_3 was observed to have lower nighttime levels in the city center than at the nearby hilltop site of Nagarkot (Panday and Prinn, 2009). Pudasainee et al. (2006) studied the seasonal variations of O_3 mixing ratios based on the observation for a whole year (2003-2004) in Pulchowk in the Lalitpur district, just south of central Kathmandu Metropolitan City (KMC) in the Kathmandu Valley. They reported seasonal O_3 mixing ratios to be highest during the pre-monsoon (March – May) and lowest in the winter (December – February). As a part of the SusKat-ABC Campaign, Putero et al. (2015) monitored O_3 mixing ratios at Paknajol, an urban site in the center of the KMC over a full-year period (February 2013-January 2014). They also observed similar seasonal variations in O_3 mixing ratios in the valley to those observed by Pudasainee et al. (2006), with highest O_3 during the pre-monsoon (1 February – 12 May) season, followed by the monsoon (13 May – 6 October), post-monsoon (7 October – 26 October) and winter (27 October – 31 January) seasons. They found that during the pre-monsoon season, westerly winds and regional synoptic circulation transport O_3 and its precursors from regional forest fires located outside the Kathmandu Valley. In another study conducted as part of the SusKat-ABC Campaign, 37 non-methane volatile organic compounds (NMVOCs) were measured at Bode, with data recording

every second, during winter of 2012-2013; the measurements included isoprene, an important biogenic precursor of O_3 (Sarkar et al., 2016). They found concentrations to vary in two distinct periods. The first period was marked by no brick kiln operations and was associated with high biogenic emissions of isoprene. During the second period nearby brick kilns, which use coal mixed with biomass, were in ~~operation ;~~ they~~that~~ contributed to elevated concentrations of ambient acetonitrile, benzene and isocyanic acid. Furthermore, the authors found that oxygenated NMVOCs and isoprene combined accounted for 72% and 68% of the total O_3 production potential in the first period and second period, respectively.

Prior to the SusKat-ABC campaign there were no studies that simultaneously measured ambient CO and O_3 mixing ratios at multiple sites in the Kathmandu Valley over extended periods of time. Past studies either focused on one long-term site, or on short-term observation records at various sites (Panday and Prinn, 2009), or they investigated the seasonal characteristics of single pollutants such as O_3 at a single site in the valley (Pudasainee et al., 2006). The most comparable past study is by Putero et al. (2015), who described O_3 mixing ratios at one SusKat-ABC site (Paknajol) in the Kathmandu city center observed during the SusKat-ABC campaign, and discussed O_3 seasonal variations. There is also a companion study on regional CO and O_3 pollution by Bhardwaj et al. (2017) which is based on O_3 and CO mixing ratios monitored at the SusKat-ABC supersite at Bode in the Kathmandu Valley for a limited period (January-June 2013) and at two sites in India (Pantnagar in Indo-Gangetic Plain and Nainital in Himalayan foothill). They reported simultaneous enhancement in O_3 and CO levels at these three sites in spring, highlighting contribution of regional emissions, such as biomass burning in northwest Indo-Gangetic Plain (IGP), and regional transport to broader regional scale pollution, including in the Kathmandu Valley. In this study, we document the diurnal and seasonal (where applicable) characteristics and spatial distributions of CO and O_3 mixing ratios based on simultaneous observations at several locations within the valley and on the valley rim mountains over a full year, helping to characterize the pollution within the valley and the pollution plume entering and exiting the valley. We also compute the first top-down estimates of CO emission fluxes for the Kathmandu Valley and compare these to CO emissions fluxes in widely-used emission datasets such as EDGAR HTAP (Janssens-Maenhout et al., 2000), REAS (Kurokawa et al., 2013) and INTEX-B (Zhang et al., 2009).

2. Study sites and methods

The Kathmandu Valley, situated in the foothills of the Central Himalaya, is home to more than 3 million people. The valley floor has an area of about 340 km^2, with an average altitude of about 1300 m above sea level (m asl). It is surrounded by peaks of about 1900-2800 m asl. The valley has five major mountain passes on its rim: the Nagdhunga, Bhimdhunga and Mudku Bhanjhyang passes in the west, and the Nala and Nagarkot passes in the east, as shown in Figure 1. The passes are situated at altitudes of 1480-1530 m asl. There is also one river outlet (the Bagmati River) towards the south~~west~~, which constitutes a sixth pass for air circulation in and out of the valley (Regmi et al., 2003; Panday and Prinn, 2009). We selected five measurement sites, including two on the valley floor (Bode and Paknajol), two on mountain ridges (Bhimdhunga and Nagarkot) and one near the Bagmati River outlet (Naikhandi) to characterize the spatial and temporal variabilities of CO and O_3 mixing ratios in the Kathmandu Valley. A short description of the measurement sites is presented here and in Table 1, ~~and~~ while details on instruments deployed at those sites for this study are presented in Table 2. Further details of the measurement sites are described in the SusKat-ABC campaign overview paper (Rupakheti et al., 2017, manuscript in preparation).

Bode (27.69°N and 85.40°E, 1344 m asl): This was the supersite of the SusKat-ABC Campaign. Bode ~~(27.69°N and 85.40°E, 1344 m asl)~~ is located in the Madhyapur Thimi municipality in the ~~eastern~~ just east of the geographic center~~part~~ of the valley. It is a semi-urban site surrounded by ~~scattered~~ urban buildings and residential houses ~~in~~ scattered across agricultural lands. Within 4 km ~~T~~there are 10 brick kilns and the Bhaktapur Industrial Estate towards the southeast ~~direction~~, ~~within 4 km distance from the site~~ (refer to Sarkar et al., 2016; Mahata et al., 2017 for details). The O_3 and CO instruments at Bode site were placed on the fifth floor of a 6-story building, the tallest in the area. The inlets of the CO and O_3 analyzers were mounted on the roof top of the temporary lab, 20 m above the ground level.

Bhimdhunga: This site (27.73°N, 85.23°E, 1522 m asl) is located on the Bhimdhunga pass on the western rim of the valley. It is one of the lowest points on the north-south running ~~sits on the~~ mountain ridge between the Kathmandu Valley to the east and a valley of a tributary of the Trishuli River to the west. It is situated about 5.5 km from the western edge ~~(Sitapaila)~~ of the

KMC (Kathmandu Metropolitan City), in a rural setting with ~~only a~~very few scattered rural houses nearby. The CO instrument was placed on the ground floor of a small one-story building and its inlet was 2 m above ground. ~~a~~An automatic weather station(AWS) (Hobo Onset, USA) was set up on the roof of another one-story building at a distance of ca. 15 m from the first building.

Paknajol: This site (27.72°N, 85.30°E, 1380 m asl) is located at the city center in the KMC, near the ~~most~~ popular touristic area of ~~(~~Thamel~~)~~. It is in the western part of the valley and about 10 km distance from the Bode supersite. The O_3 and meteorological instruments relevant to this study were placed on the top floor and rooftop of a 6-story building, the tallest in the area (details in Putero et al., 2015; note that CO was not measured here). The inlet of the O_3 analyzer was placed 25 m above the ground.

Naikhandi: This site (27.60°N, 85.29°E, 1233 m asl) is located within the premises of a school (Kamdhenu Madhyamik Vidhyalaya) located at the southwestern part of the valley (~7 km south from the nearest point of the Ring Road). The school premise is open, surrounded by sparsely scattered rural houses in agricultural lands. The nearest village (~75 houses) is about 500 m away in the southwest direction. There are 5 brick kilns within 2 km distance (2 to the north and 3 to the northeast) from the site. The instruments were kept in a one-story building of the school and its inlet was 5 m above the ground. The AWS (Hobo Onset, USA) was installed on the ground near the Bagmati River, ~100 m away from the main measurement site.

Nagarkot: This site is located on a mountain ridge (27.72°N, 85.52°E, 1901 m asl), ca. 13 km ~~away to the~~ east ~~from~~ of Bode, in the eastern part of the valley. The site faces the Kathmandu Valley to the west and small rural town, Nagarkot, to the east. The instruments were set up in a 2-story building of the Nagarkot Health Post and their inlets were 5 m above the ground. ~~t~~The AWS (Vaisala WXT520, Finland) was set up on the roof of the building.

Bhimdhunga Pass in the west and Naikhandi near the Bagmati River outlet in the southwest are the ~~most~~ important ~~pass and river outlet for the valley~~places for interchange of valley air with outside air. The Bhimdhunga and Naikhandi sites are approximately 5.5 and 7 km away from the

nearest edge of the ~~major~~ city, respectively. Similarly, Bode is located downwind of the city centers and thus receives pollution outflow from nearby city centers ~~such as~~ of Kathmandu/~~and~~ Lalitpur due to strong westerly and southwesterly winds (4-6 m s^{-1}) during the day time, and emissions from the Bhaktapur area to the east and southeast direction by calm easterly winds (< 1 m s^{-1}) during the night (Sarkar et al., 2016; Mahata et al., 2017).

A freshly calibrated new CO analyzer (Horiba APMA-370, Japan) was deployed for the first time at Bode. ~~It~~ This instrument is based on the IR absorption method at 4.6 µm by CO molecules. Before field deployment at Bode, it was compared with the bench model of the Horiba (APMA-370), and the correlation (r) between them was 0.9 and slope was 1.09. The instrument was regularly maintained by running auto-zero checks (Bhardwaj et al., 2017). Similarly, another CO analyzer (Picarro G2401, USA) which is based on cavity ring-down spectroscopy technique (CRDS) was also a new factory calibrated unit, and was deployed in Bode along with the Horiba APMA-370. An IR-based Thermo CO monitor (model 48i-TLE) was run simultaneously with a co-located cavity ring down spectrometry based ~~CO analyzer~~ ~~(~~Picarro CO analyzer~~, model G2401)~~ for nearly 3 months. The correlation coefficient and slope between the two measurements were found to be 0.99 and 0.96, respectively (Mahata et al., 2017). This indicates that there was very little drift in the IR-based CO values due to room temperature change, within acceptable range (i.e., within the measurement uncertainties of the instruments). Therefore, we did not any apply correction in the IR-based CO data. ~~The three-month inter-comparison between the Horiba and Picarro CO measurements had a correlation coefficient of 0.99 and the slope was 0.96 (Mahata et al., 2017).~~ All other CO analyzers (Thermo Scientific, 48i-TLE, USA), which are also based on IR absorption by CO molecules, deployed at Bhimdhunga, Naikhandi and Nagarkot, were set up for regular automatic zero checks on a daily basis. In addition, a span check was also performed during the observations by using span gas of 1.99 ppm (Gts-Welco, PA, USA) on March 8, 2013 at Naikhandi and Nagarkot, and on March 9 at Bhimdhunga. The IR-based CO instruments' span drifts were within a 5 % range.

For the O_3 monitor (Teledyne 400E, USA) at Bode, ~~the~~ regular zero and span checks were carried out using the built-in O_3 generator and scrubber (Bhardwaj et al., 2017). This unit was used in Bode from 01 January 2013 to 09 June 2013. New factory-calibrated O_3 monitors

(Thermo Scientific, 49i, USA) were used for the rest of the measurement period (18 June 2013 to 31 December 2013) at Bode, and for the full year of measurements at Nagarkot. A Thermo Environmental O_3 analyzer (Model 49i, USA) was used at the Paknajol site (Putero et al., 2015) with the same experimental set up as described in Cristofanelli et al. (2010). The working principle of all of the O_3 instruments is based on the attenuation of UV radiation by O_3 molecules at ~254 nm.

In order to characterize ~~the~~ observations across the seasons, we considered the following seasons as defined in Shrestha et al. (1999) and used in other previous studies in the Kathmandu Valley (Sharma et al., 2012; Chen et al., 2016; Mahata et al, 2017): Pre-monsoon (March, April, May); Monsoon (June, July, August September); Post-monsoon (October, November); and Winter (December, January, February).

3. Results and discussion

3.1 CO mixing ratio at multiple sites

Figure 2 shows the time series of the hourly average CO mixing ratios at the four sites (Bode, Bhimdhunga, Naikhandi and Nagarkot). Fluctuations in CO mixing ratios were higher during the winter and pre-monsoon than during the monsoon season at all sites. The monsoon rain generally starts in Nepal around mid-June. In 2013, however, there were more frequent rain events in the month of May than in previous years. The CO mixing ratios (measured in parts per billion by volume, hereafter the unit is denoted as ppb) of hourly averaged data over the total observation periods at four sites and their standard deviation were: Bode (569.9 ± 383.5) ppb during 1 January - 15 July, Bhimdhunga (321.5 ± 166.2) ppb during 14 Jan - 15 July, Naikhandi (345.4 ± 147.9) ppb during 3 January - 6 June and Nagarkot (235.5 ± 106.2) ppb during 13 February - 15 July (except 4 April to 7 June). Nagarkot had only about 3 months of CO data (due to a problem in zeroing fail ~~tests~~ of the instrument) during the observation period. For the measurement period, the CO mixing ratio at Nagarkot (~13 km far from Bode) showed small fluctuations compared with the other sites. High CO values in the Kathmandu Valley during the dry season (November-May) were also reported by Panday and Prinn (2009) based on their measurements during September 2004-May 2005 at Bouddha (~ 4 km in northwest from Bode). The simultaneous episodes of high CO observed ~~in~~ from April ~~(~~1to15~~)~~ in Bhimdhunga, Bode and

Naikhandi indicate the influence of regional sources, in addition to local sources. This is discussed further in section 3.2.3.

3.2 Diurnal and seasonal variations of CO

3.2.1 Diurnal pattern of CO at multiple sites

Figure 3 shows the diurnal cycles of CO mixing ratios at four sites (plotted for the period of 13 February to 3 April 2013, when the data were available from all four sites). The variation in the mixing ratios during the day was characterized by a pronounced morning peak, a weaker evening peak, and a daytime low; except at Nagarkot where peaks are less visible. Multiple sources contribute to the morning and evening peaks, especially emission from vehicles, residential burning (fossil fuel and biomass), brick kilns and trash burning (Kim et al., 2015; Sarkar et al., 2016; Mahata et al., 2017). Other studies conducted during the SusKat-ABC campaign have identified garbage (household waste and yard waste) burning as a key source of various air pollutants, such as OC and EC (Kim et al., 2015), PAHs (Chen et al., 2015), and NMVOCs (Sarkar et al., 2016; Sarkar et al., 2017). Garbage burning is often done in small fires and quite sporadic, normally taking place in the evenings and mornings (partly chosen to avoid attention from the responsible authorities). The rate of waste (and also biomass) burning in the morning is higher in winter due to the use of the fires for providing warmth on colder days.

The observed diurnal cycle of CO is similar to that reported in a previous study (Panday and Prinn, 2009), and is also similar to the diurnal pattern of black carbon (BC) in the Kathmandu Valley (Sharma et al., 2012; Mues et al., 2017). The diurnal cycles of these primary pollutants are closely coupled with the valley's boundary layer height, which is about 1200 m during daytime, and falls to approximately 200 m at nighttime in Bode (Mues et al., 2017). Nagarkot and Bhimdhunga, both on mountain ridges, are generally above the valley's boundary layer, especially at night, and thus the diurnal profile especially at Nagarkot is distinct compared to other three sites, being relatively flat with small dip during 12:00-18:00.

~~Clear~~ Distinct morning peaks were observed in Bode, Bhimdhunga and Naikhandi at 08:00, 09:00, and 10:00, respectively, i.e., the morning peak lags by 1-2 hours in Bhimdhunga and Naikhandi compared to Bode. Bhimdhunga on the mountain ridge may receive the Kathmandu

Valley's pollution due to upslope winds (~2 m s^{-1}) from the ~~south~~east direction in the morning hours after the dissolution of the valley's boundary layer due to radiative heating of the mountain slopes. On the other hand, Naikhandi is in close proximity to brick kilns and could be impacted by their plumes carried to the site by northerly winds in the early morning (ca. 07:00-10:00, not shown). The evening peak values at Bode and Bhimdhunga were less pronounced compared to the~~ir~~ morning maxima. The morning peak at Bode was influenced by nighttime accumulation of CO along with other pollutants from nearby brick kilns (Sarkar et al., 2016; Mahata et al., 2017; Mues et al., 2017) and recirculation of air from above (Panday and Prinn, 2009). Similarly, the local pollution from the nearby village and city area due to upslope winds from the valley floor is expected to contribute to the morning peak at Bhimdhunga. The evening peak at Naikhandi was at 21:00 and was closer to the morning values in comparison to the large difference between morning and evening peaks at Bode and Bhimdhunga. A nighttime build-up (linear increase) of various pollutants compared to the afternoon minimum was typically observed in Bode during the SusKat-ABC measurement period, including the main campaign period (Sarkar et al., 2016; Mahata et al., 2017; Mues et al., 2017). This is mainly associated with the ~~nocturnal~~ persistent emissions such as those from brick kilns, which are in close proximity to the Bode measurement site under the ~~decrease in height of the planetary~~ stable boundary layer.~~, along with persistent emissions such as those from brick kilns, which are in close proximity to the Bode measurement site.~~ The isolated peak during the morning transition phase at Bhimdhunga could be due to an elevated polluted layer because of the slope wind (Panday et al., 2009). There appears to be less influence of nighttime polluting sources at Naikhandi and Bhimdhunga than at Bode.

The MLH starts increasing after radiative heating of the surface by incoming solar radiation. The heating of the ground causes thermals to rise from the surface layer resulting in ~~Tthe low daytime CO mixing ratios observed at all sites were partly due to the evolution of mixing layer and~~ the entrainment of cleaner air from above the boundary layer ~~after~~ leading to the dissolution of nocturnal stable boundary layer. Increasing wind speeds (4-6 m s^{-1}) during daytime also support turbulent vertical diffusion, as well as flushing of the pollution by less polluted air masses from outside the valley, with stronger horizontal winds allowing significant transport of air masses into the valley. In addition, reduced traffic and household cooking activities during daytime compared to morning and evening rush hours contribute to the reduced CO mixing ratios.

3.2.2 CO diurnal variation across seasons

Due to the lack of availability of simultaneous CO data at all sites covering the entire sampling period, a one-month period was selected for each season to examine the diurnal variation across the seasons, and to get more insights into the mixing ratios at different times of the day, as reported in Table 4. Figure 4 shows the diurnal variation of CO mixing ratios in Bode, Bhimdhunga, and Naikhandi during the selected periods for the three seasons.

The diurnal cycles during each season had different characteristics. The most prominent distinction was that the CO mixing ratio was low during the monsoon period over all sites (Figure 4, Table 4) as a result of summer monsoon rainfall in the valley, which is 60 - 90% of the 1400 mm rainfall for a typical year (Nayava, 1980; Giri et al., 2006). Because of the rainfall, the brick production activities are stopped in the valley (usually they are operational from January-April every year). Further, ~~T~~the rainfall also diminishes many burning activities (forest fires, agro-residue and trash burning~~, and the brick kilns~~) within the valley and surrounding region, and thus reduces CO emissions. Afternoon CO mixing ratios were higher in the pre-monsoon season than in the other two seasons in Bode, Bhimdhunga and Naikhandi (also see Table 4), with the most likely sources being emissions from forest fires and agro-residue burning arriving from outside the valley during this season (this will be discussed further in section 3.2.3). Nighttime accumulation was observed in Bode and Bhimdhunga, but not at Naikhandi, where the mixing ratio decreased slightly from about 20:00 until about 04:00, after which the mixing ratios increased until the morning peak. The nighttime accumulation of CO in Bode during pre-monsoon and winter is ~~apparently~~ due to the influence of nearby brick kilns (Mahata et al., 2017) because of the calm easterly wind (refer supplementary Figure S2 in Mahata et al., 2017). Previous studies carried out at the Bode site during the SusKat-ABC campaign have attributed over a dozen brick kilns located near Bode as strong sources of BC and EC (Kim et al., 2015; Mues et al., 2017), NMVOCs (Sarkar et al, 2016; Sarkar et al., 2017), SO2 (Kiros et al., 2016) and CO (Mahata et al., 2017), and the enhanced concentrations were observed during nighttime and mornings when winds blew from east and southeast bringing emissions from the location of the brick kilns to the observation site.

Bhimdhunga is not near any major polluting sources such as brick kilns, and it is unclear whether the nighttime CO accumulation in Bhimdhunga is primarily due to ongoing local residential pollution emissions, and/or to pollution transported from remote sources. The transition of the wind from westerlies during the day to easterlies during the night, with moderate wind speed (~2-4 m s^{-1}) at Bhimdhunga, may bring polluted air masses westwards which were initially transported to the eastern part from the Kathmandu Valley during the daytime (Regmi et al., 2003; Panday and Prinn, 2009; Panday et al., 2009).

The distinct shift in the morning peak was seen at all 3 sites by season. The one hour shift in the morning peak from the pre-monsoon to winter is due to an earlier onset of the morning transition. However, the one hour difference in the morning peak between Bode (pre-monsoon at 8:00; winter at 9:00) and Bhimdhunga/Naikhandi (pre-monsoon at 9:00; winter at 10:00) in the pre-monsoon and winter is associated with commencement of early local emissions under the shallow boundary layer at Bode. The one hour lag in the morning peak at Bhimdhunga and Naikhandi may be due to transport of city pollutants to the site, respectively.

Across the seasons, the afternoon (12:00-16:00) CO mixing ratio was higher during the pre-monsoon than in the winter at all three stations (p value for all sites < 0.5) (Table 4), although the mixing layer was higher in the pre–monsoon season than in the winter in Bode (and presumably at the other sites as well). This is not likely to be explained by local emissions in the valley, since these are similar in the winter and pre-monsoon periods. Putero et al. (2015) suggested instead that this reflects an influx of polluted air into the valley due to large synoptic circulation patterns during the pre-monsoon season. Such regional influences are explored further in the next section.

3.2.3 Regional influence on CO in the valley

Recent studies have indicated the likelihood of regional long-range transport contributing to air pollution in different parts of Nepal (Marinoni et al., 2013; Tripathee et al., 2014; Dhungel et al., 2016; Rupakheti et al., 2016; Lüthi et al., 2016; Wan et al., 2017), including the Kathmandu Valley, especially during the pre-monsoon period (Panday and Prinn, 2009; Putero et al., 2015; Bhardwaj et al., 2017). During the pre-monsoon season, frequent agro-residue burning and forest fires are reported in the IGP region including southern Nepal and the Himalayan foothills in

India and Nepal (Ram and Sarin, 2010; Vadrevu et al., 2012), and in the Kathmandu Valley. This season is also characterized by the strongest daytime local wind speeds (averaging 4-6 m s^{-1}) in the Kathmandu Valley (Mahata et al., 2017). Our study also observed several episodes of days with both elevated CO mixing ratios (Figure 2) and O_3 mixing ratios (also measured in parts per billion by volume, hereafter the unit is denoted as ppb) (Figure 5) during April and May, especially during the late afternoon period. The influence of regional pollutants was investigated by comparing a 2-week period with normal CO levels (16–30 March (hereafter "period I") with an adjacent two week period (1-15 April) with episodically high CO mixing ratios (hereafter "period II"), which nicely fit with the "burst" in regional fire activity presented by Putero et al. (2015) in their Figure 9. The t-test of the two hourly data means of CO in period I and period II at Bode, Bhimdhunga and Naikhandi (as in Figure 5) were performed at 95% confidence level and the differences were found to be statistically significant ($p < 0.5$).

Figure 5a shows the diurnal cycle of CO mixing ratios during period I (faint color) and period II (dark color) at Bode, Bhimdhunga and Naikhandi. The difference between two periods was calculated by subtracting the average of period I from average of period II. The average CO mixing ratios during period II were elevated with respect to period I by 157 ppb at Bode, 175 ppb at Bhimdhunga, and 176 ppb at Naikhandi. The enhancements in mixing ratios at the three sites were fairly similar from hour to hour throughout the day, with the exception of the late afternoon when the enhancement was generally greatest. This consistency across the sites suggests that the episode was caused by a large-scale enhancement (regional contribution) being added onto the prevailing local pollution levels at all the sites. A large-scale source would also be consistent with the greater enhancements of CO at the outskirt sites, which would be most directly affected by regional pollution, compared to the central valley site of Bode, with strong local sources. The enhancement during the period II is substantial (statistically significant as mentioned above), representing an increase of approximately 45% at the outskirt sites of Bhimdhunga and Naikhandi (which start with lower CO levels), and 23% at Bode. During both periods I and II, local winds from west (the opposite direction from the brick kilns, which were mostly located to the southeast of Bode) were dominant during daytime at Bode (Figure 5b, c). This suggests that the elevation in CO levels was caused by additional emissions in period II in the regions to the west and southwest of the Kathmandu Valley, e.g., large scale agricultural burning and forest fires during this period, as also noted by Putero et al. (2015) (see their Figure

9). Far away, in Lumbini in the southern part of Nepal (Rupakheti et al., 2016), and Pantnagar in northern IGP in India (Bharwdwaj et al., 2017), about 220 km (aerial distance) to the southwest and 585 km to the west, respectively, of the Kathmandu Valley, CO episodes were also observed during the spring season of 2013, providing a strong indication that the episode in period II was indeed regional in nature.

3.3 O_3 in the Kathmandu Valley and surrounding areas

Figure 6 shows the hourly average and daily maximum 8-hour average of O_3 mixing ratios at Bode, Paknajol, and Nagarkot from measurements during the SusKat campaign and afterwards, along with O_3 mixing ratios from a previous study (November 2003 - October 2004; Pudasainee et al., 2006) at the Pulchowk site (4 km away from Paknajol) in the Latitpur district. The daily maximum 8-hour average O_3 was calculated by selecting the maximum O_3 mixing ratio from 8 hour running averages during each day. The nighttime mixing ratio of hourly O_3 drops close to zero in Bode, Paknajol and Pulchowk in the winter season. This is a typical characteristic of many urban areas where reaction with NO at night depletes O_3 from the boundary layer (e.g., Talbot et al., 2005). In the pre-monsoon and monsoon months, the titration is not as strong and the hourly O_3 falls, but generally remains above 10 ppb. Similar patterns of ozone mixing ratios were observed at other sites in northern South Asia. For example, higher O_3 mixing ratios were observed in the afternoon (84 ppb) and lower during the night and early morning hours (10 ppb) at Kullu Valley, a semi-urban site located at 1154 m asl, in the North-western Himalaya in India (Sharma et al. 2012). A similar dip in O_3 value in the dark hours was observed at Ahmedabad, India by Lal et al. (2000). Nagarkot, in contrast, is above the valley's boundary layer and has lesser NO for titration at night at this hill station as has been observed in another hill station in Himalayan foothills (Naja and Lal, 2002). Thus, the O_3 level remains above 25 ppb during the entire year at Nagarkot. As also shown in Table 3, at all sites, the O_3 mixing ratios were highest in the pre-monsoon, but the timing of the lowest seasonal values varied across the sites: post-monsoon in Bode, winter in Paknajol and monsoon in Nagarkot. Such differences in minimum O_3 across the sites can be anticipated due to the locations of the sites (e.g., urban, semi-urban, rural and hilltop sites, with differing availabilities of O_3 precursors from different emission sources). The seasonal variations of O_3 observed at Bode in this study are consistent with Putero

et al. (2015) and Pudasainee et al. (2006), who also observed O_3 maxima during the pre-monsoon, but O_3 minima during the winter season.

The daily maximum 8-hour average O_3 mixing ratio (solid colored circles in Figure 6) exceeded the WHO recommended guideline of 50 ppb (WHO, 2006, black dotted line in Figure 6) most frequently during the pre-monsoon period and the winter. During the observation period, the daily maximum 8-hour average O_3 exceeded the WHO guideline on 102 out of 353 days of observation (29%) at Bode, 132/354 days (37%) at Paknajol and 159/357 days (45%) at Nagarkot. The higher exceedance rate at Nagarkot is because it is at higher altitude, which results in (i) greater exposure to large-scale regional pollution, especially from forest fire in the Himalayan foothills and agro-residue burning in the IGP region, outside the Kathmandu ~~v~~Valley (Sinha et al., 2014, Putero et al., 2015), (ii) less titration of O_3 by NO_x, being farther away from the main pollution sources, and (iii) exposure to O_3 rich free tropospheric air, including influences from stratospheric intrusions. The diurnal profiles of O_3 mixing ratios (Figure 7) at three sites Bode and Pakanajol in the Valley and Nagarkot, a hilltop site normally above the Kathmandu Valley's boundary layer shows, notably in the morning hours, that the residual layer above the Kathmandu Valley's mixing layer contains a significant amount of ozone. Based on the surface ozone data collected at Paknajol during 2013-14, Putero e al. (2015) concluded that downward mixing of ozone from the residual layer contributes to surface ozone in the Kathmandu Valley in the afternoon hours (11:00-17:00 local time). It is likely that the same source has also contributed to higher ozone mixing ratios at Nagarkot. Such mixing has been observed at other sites as well. Wang et al. (2012) reported the increase in downward mixing of O_3 from the stratosphere to the middle troposphere (56%) and the lower troposphere (13%) in spring and summer in Beijing. The downward flux was highest in the middle troposphere (75%) in winter. Similarly, Kumar et al. (2010) reported that more than 10 ppb of stratospheric contribution at a high altitude site (in Nainital) during January to April. However, there were no significant stratospheric intrusions seen in spring and summer (seen only in winter) at Nepal Climate Observatory-Pyramid (Putero et al., 2016).

During the SusKat-ABC campaign in 2013 and later in 2014, passive sampling of ~~various~~ gaseous pollutants (SO_2, NO_x, NH_3 and O_3) was ~~also~~ carried out at fourteen sites including urban/semi-urban sites (Bode, Indrachowk, Maharajganj, Mangal Bazar, Suryabinayak,

Bhaisepati, Budhanilkantha, Kirtipur, and Lubhu) and rural sites (Bhimdhunga, Naikhandi, Sankhu, Tinpiple, and Nagarkot) in the Kathmandu Valley (Kiros et al., 2016). Similar to this study, they also observed higher O_3 mixing ratios in rural areas than the urban/semi-urban sites in the Kathmandu Valley. Exceedances of the WHO standard are most common during the pre-monsoon season, occurring 78% (72/92 days), 88% (78/89 days) and 92% (85/92 days) of the time at Bode, Paknajol and Nagarkot, respectively. A study by Putero et al., (2015), based on O_3 mixing ratio measurements at Paknajol in the Kathmandu Valley, as a part of the SusKat-ABC campaign, has reported that the dynamics (both by horizontal and vertical winds) plays a key role in increased O_3 mixing ratios in the afternoon in the Kathmandu Valley. They estimated that the contribution of photochemistry varied as a function of the hour of the day, ranging from 6 to 34 %. Unfortunately, no viable NOx measurements were obtained at any site in the Kathmandu Valley and surrounding mountain ridges during the SusKat-ABC campaign. Speciated VOCs were measured at Bode only for about 2 months but NOx was not available for the same period. Therefore we were not able to discern quantitatively proportional contributions of NOx, VOCs and intrusion (chemistry vs. dynamics) from the free troposphere or lower stratosphere to observed O_3 concentrations at Nagarkot, Bode and other sites in the Valley. ~~Thus, i~~In the context of protecting public health, crops and regional vegetation, the O_3 mixing ratios in the Kathmandu Valley and surrounding areas clearly indicate the urgent need for mitigation action aimed at reducing emissions of its precursor gases NOx and VOCs. However, air quality management plans need to consider carefully the reduction strategies of NMVOCs or NOx while aiming at mitigating the O_3 pollution in the Kathmandu Valley. If the correct strategy (NMVOCs vs. NOx) is not applied, then O_3 mixing ratios could increase, for example, as seen in Huszar et al. (2016) where they reported that reducing NMVOCs in urban areas in central Europe leads to O_3 reduction whereas the focus on NOx reduction results in O_3 increase.

The SusKat-ABC O_3 data can be compared to observations made about a decade ago by Pudasainee et al. (2006) at the urban site of Pulchowk, not far from Paknajol, as plotted in Figure 6d. The daily maximum 8-hour average O_3 had exceeded the WHO guideline at Pulchowk for 33% (95/292 days) of days during the observation from November 2003 to October 2004. The exceedance was 38% (133/354 days) of days at Paknajol during Feb 2013 - March 2014. Due to inter-annual variability and differences in the seasonal observation time periods at Pulchowk and

Pakanajol, we cannot ~~make~~draw any conclusions about trends over the decade between the observations because of the difference in location and sampling height as well as a general difference in instrument calibration. However, a clear similarity between the observations is that most of the exceedance took place during pre-monsoon season, during which both studies have observations throughout the season (~90 days). The percentage of exceedance at Pulchowk during the pre-monsoon season in 2003-2004 was 70% (63/90 days) and at Pakanajol in 2013 it was 88% (78/89 days). However, just like for the annual fraction of exceedances, due to inter-annual variability we cannot say that the 18% (or ca. 15 days) difference in the exceedances is significant. A longer term O_3 record would be needed to really establish if there is a trend in the ozone concentrations.

3.4 O_3 seasonal and diurnal variation

The seasonal average O_3 mixing ratios at Bode, Nagarkot and Paknajol are shown in Table 3. For comparison, the O_3 mixing ratios measured at two sites in India, (i) Manora Peak (1958 m asl), ca. 9 km from Nainital city, a site in rural mountain setting and (ii) Delhi, a highly-polluted urban setting in northwest IGP are also listed in the Table, based on results from Kumar et al. (2010) and Ghude et al. (2008). There is a strong similarity between the urban and semi-urban sites in Nepal (i.e., Bode, Pakanajol) and India (i.e., Delhi), as well as between the rural and mountain sites in Nepal (i.e., Nagarkot) and India (i.e., Manora Peak), with small differences. The peak mixing ratios were in the pre-monsoon period: at the rural and mountain sites the peak ozone mixing ratio values were very similar (64 and 62 ppb for Nagarkot and Manora Peak, respectively) and are due to influences discussed earlier for Nagarkot; at the sub-urban and urban sites the pre-monsoon values are significantly lower (ca. 40, 42, 33 ppb for Bode, Paknajol, Delhi, respectively) due to fresh NOx emissions near the urban sites and the consequent titration of ozone with NO. The lowest O_3 seasonal values at rural and mountain sites typically occur in the monsoon months while for semi-urban and urban sites, the minimum was observed during post-monsoon (Bode) and winter (Paknajol).

Figure 7 shows the diurnal variation of O_3 mixing ratios at Bode, Paknajol and Nagarkot in the different seasons. The typical O_3 maximum mixing ratio in the early afternoon at the urban and semi-urban sites is mainly due to daytime photochemical production as well as entrainment of ozone due to dynamics (both intrusion of ozone rich free tropospheric air into the boundary

layer, and regional scale horizontal transport of ozone), as explained in case of Paknajol by Putero et al. (2015). ~~which Putero et al. (2015) suggested results in the broader afternoon peak of ozone during the pre-monsoon at Paknajol site, also observed at Bode site (and somewhat at Nagarkot).~~

The ozone mixing ratios are relatively constant throughout the day at Nagarkot (~1901 m asl), which, being a hilltop site, is largely representative of the lower free tropospheric regional pollution values, ~~but~~ however, it is also affected by ozone production from precursors transported from the Kathmandu Valley due to westerly winds during the afternoon hours. The dip in O_3 at Nagarkot (Figure 7) in the morning transition hours indicates the upward mixing of air from polluted (and Ozone-depleted) nocturnal boundary layer as it is breaking up.

3.5 CO emission flux estimate

It is possible to determine a top-down estimate of the average CO emission flux for the morning hours for the region around the Bode site by applying an approach that was developed and used in Mues et al. (2017) to estimate the emission fluxes of BC at Bode. The analysis of Mues et al. (2017) found BC fluxes for the Kathmandu Valley that were considerably higher than the widely-used EDGAR HTAP emission database (Version 2.2). Support for this top-down estimate was found by considering the BC concentrations and fluxes for the Kathmandu Valley in comparison to Delhi and Mumbai; although the observed BC concentrations were similar in all three locations, the EDGAR HTAP V2.2 emissions of BC for the Kathmandu Valley are much lower than those for Delhi and Mumbai, while the top-down emissions estimate for the Kathmandu Valley were similar to the emissions from EDGAR HTAP V2.2 for Delhi and Mumbai (Mues et al., 2017).

Here we apply the same method as developed in Mues et al. (2017) to estimate the CO fluxes based on the observed CO mixing ratio and ceilometer observations of the mixing layer height (*MLH*) in Bode for the period of ~~a~~1 year (March 2013-February 2014). ~~It is important to note that the term "mixing layer", applied generally to ceilometer observations, is not entirely accurate, since the degree of mixing in the nocturnal stable layer is drastically reduced versus daytime. This adds a degree of uncertainty to the application of ceilometer observations to compute top-down emissions estimates, which will only be resolved once nocturnal vertical profile~~

~~measurements are also available in order to characterize the nocturnal boundary layer characteristics and the degree to which the surface observations are representative of the mixing ratios throughout the vertical column of the nocturnal stable layer.~~

Using the approach used by Mues et al. (2017), the CO fluxes can be calculated from the increase in CO concentrations during the nighttime period when the *MLH* is nearly constant, using:

$$FCO(t_x, t_y) = \frac{\Delta CO \times ave(MLH(t_x), MLH\ (t_y))}{\Delta t \times 3600} \times \frac{MLH(t_y)}{MLH\ (t_x)} \quad (1)$$

where $FCO\ (t_x, t_y)$ is the CO emission flux (in µg m^{-2} s^{-1}) between time t_x and t_y (in hours), ΔCO is the change in CO mixing ratio (in µg m^{-3}) between time t_x and t_y, $ave(MLH(t_x), MLH(t_y)$ are average of the mixing layer heights (in m) between time t_x and t_y, Δt is the time interval between t_x and t_y, and $MLH(t_y)/MLH(t_x)$ is mixing layer ~~compression~~collapse factor, accounting for the small change in MLH ~~during the observation period~~ between the night and the morning hours. The calculation of the emission flux is based on mean diurnal cycle per month of CO and MLH and tx and ty represent the time with the minimum (tx) and the maximum (ty) CO concentration in the night and morning (see Mues et al., 2017 for details).

This method of calculating the CO emission flux is based on ~~four~~five main assumptions: (i) CO is well-mixed horizontally and vertically within the mixing~~ed~~ layer in the region immediately surrounding the Bode site; (ii) the *MLH* remains fairly constant during the night so that the product of the CO concentration (µg m^{-3}) and the *MLH* (m) represents CO mass per unit area within the column, and any change in ~~this~~ mass per unit area represents the net flux into the column; (iii) the transport of air pollutants into and out of the stable nocturnal boundary layer of the valley is negligible, which is supported by the calm winds (<1 m s^{-1}) during the night and morning hours at the site (Mahata et al., 2017), (iv) the vertical mixing of pollutants between the mixing layer and the free atmosphere is assumed to be negligible at night, thus strictly seen is the estimated CO flux calculated with eq. 1 only valid for the morning hours. When applied to the whole day the implicit assumption is that the emissions are similar during the rest of the 24 h

663 period. ~~the CO emissions during the daytime are similar to those at night,~~ An assumption that is
664 viable on average for some sources like brick kilns which operate day and night, but which does
665 not apply to all sources, e.g., the technique will tend to underestimate emissions due to traffic,
666 which are typically much stronger during the day than at night, while it will overestimate
667 emissions due to waste burning, which is typically more prevalent during the night and early
668 morning (pre-sunrise) than during the daytime. Assumption (iv) is made because equation 1
669 only works well for calculating the CO flux at night-morning period, when there is a relatively
670 constant *MLH* and limited vertical and horizontal mixing; and v) CO emission is assumed to be
671 uniform throughout the valley; this may not be correct, but cannot be verified until a high
672 resolution emission inventory data is available, which is being developed for the Kathmandu
673 Valley and rest of Nepal with a 1 km x 1km spatial resolution (Sadavarte et. al., 2018). During
674 nighttime assumption (i) might not be entirely correct since the degree of mixing in the nocturnal
675 stable layer and thus the vertically mixing is drastically reduced ~~versus~~ compared to daytime
676 (and thus the term "mixing layer" is not entirely accurate, but we nevertheless apply it here due
677 to its common use with ceilometer measurements). This adds a degree of uncertainty to the
678 application of ceilometer observations to compute top-down emissions estimates, which will
679 only be resolved once nocturnal vertical profile measurements are also available in order to
680 characterize the nocturnal boundary layer characteristics and the degree to which the surface
681 observations are representative of the mixing ratios throughout the vertical column of the
682 nocturnal stable layer.

683 It is not possible to directly compute the emission flux for a full 24-hour day using this top-down
684 method, since the emissions during the day could be either greater or smaller than at night, and
685 because the other assumptions do not hold (in particular there is considerable vertical mixing
686 with the free troposphere and stronger horizontal transport during the daytime). Thus the top-
687 down computation only provides a useful indicative value. However, while it is also not possible
688 to estimate how much different the daytime emissions are, it is possible to determine an absolute
689 lower bound for the CO flux (FCO_{min}) by making the extreme assumption that the CO emissions
690 are non-zero only during the hours which were used in the calculation, and that they were zero
691 during the rest of the day (this provides a lower bound to the emissions since the daytime
692 emissions physically cannot be negative). This lower bound of the flux (FCO_{min}) is thus

calculated by scaling back the 24-hour flux to only applying over the calculation time interval (Δt), using:

$$FCO_{min.} = FCO \times \frac{\Delta t}{24} \quad (2)$$

Figure 8 shows the estimated monthly CO emission flux, along with its 25th and 75th percentile values as an indication of the variability of the estimated flux in each month; the lower bound of the CO flux based on Equation 2 is also shown. The estimated annual mean CO flux at Bode is 4.9~~2~~ µg m^{-2} s^{-1}. Seasonally, the emissions are computed to be highest during December to April (3.6~~4~~-8.~~43~~6 µg m^{-2} s^{-1}), coinciding with the brick kiln operation period, which resulted in elevated concentrations of most pollutants at Bode (Kim et al., 2015; Chen et al., 2016; Sarkar et al., 2016; Mahata et al., 2017; Mues et al., 2017), including CO (Bhardwaj et al., 2017; Mahata et al., 2017), while the emissions were generally lower during the remaining months (0.5~~4~~-5.~~43~~7 µg m^{-2} s^{-1}). The uncertainty in the top-down CO emissions estimate will be largest during June to October, due to the greater diurnal and day-to-day variability with the minimum and maximum CO mixing ratio values during the night and early morning used in Equation 1 often being less distinct than in the other months.

Comparing the annual mean top-down estimated CO emission flux at Bode (4.9~~2~~ µg m^{-2} s^{-1}) with available global and regional emission inventories, the top-down estimated CO flux is twice the value, 2.4 µg m^{-2} s^{-1}, for the Kathmandu Valley in the EDGAR HTAP V2.2 emission inventory database for 2010 [note that the CO emission values for the location at ~~of~~ Bode and the whole averaged for the valley ~~as a whole~~ (27.65-27.75°N, 85.25-85.40°E) were found to be the same up to two significant figures]. The estimated CO flux was 6.5-8 times as high as in the REAS database (0.63-0.76 µg m^{-2} s^{-1}, based on the 2008 values in Kurokawa et al., 2013), and between 3 and 14 times higher than the values in the INTEX-B database for 2006 (0.35-1.77 µg m^{-2} s^{-1}; Zhang et al., 2009). The large differences between our estimated CO emission flux and these emission databases is not likely to be due to the comparison of data for different years; rather, it indicates the substantial uncertainties in both the top-down and bottom-up approaches to estimating the emission flux. Although our approximation of the emission flux relies on several

assumptions, the fact that the lower bound value that we calculate is still as high as or higher than the values in some of the published emission datasets likely indicates that the bottom-up emissions are missing or underestimating some important sources, which will be important to examine carefully and improve as a basis for interpreting future modelling studies of CO pollution in the Kathmandu Valley and surrounding regions, as well as for assessing possible mitigation options.

The emission estimates computed here are subject to several further uncertainties which are discussed in detail in Mues et al., (2017). In short, the uncertainties of CO flux estimates arise from (i) the assumptions that Bode site represents the whole atmospheric column and entire valley, which is not possible to verify without having many simultaneous monitoring stations in the valley (measurements at a few sites where CO was monitored for this study show some difference in CO mixing ratios), (ii) the higher variability (unclear minima and maxima during the morning and night hours) in the diurnal cycles of CO from June to October show a much higher variability than other months, that in turn makes it difficult to choose the exact hour of CO minimum and maximum needed for the flux estimation and (iii) the possible impact of wet deposition is not taken into account but would rather cause to generally underestimate the emission rate.

4. Conclusions

Ambient CO and O_3 mixing ratios were measured in the framework of the SusKat-ABC international air pollution measurement campaign at five sites (Bode, Paknajol, Bhimdhunga, Naikhandi and Nagarkot) in the Kathmandu Valley (Table 1) and its fringes, initially during January to July 2013, and later extended to one year at three sites (Bode, Paknajol and Nagarkot) to better understand their seasonal characteristics. The observed CO and O_3 levels at all sites except Nagarkot were characteristic of highly-polluted urban settings, with the particular feature that the bowl-shaped valley and resulting meteorology had several effects on the pollution levels.

At all sites, the CO mixing ratios were higher during the early morning and late evening, especially an observation connected to the interplay between the ventilation of the boundary layer and the diurnal cycles of the emission sources. Under calm wind conditions that limited mixing within, into and out of the Kathmandu Valley, the morning CO peak tended to be more

pronounced due to the buildup of pollution at night in the shallow planetary boundary layer. This nocturnal buildup was especially strong during January to April at Bode, with the mean CO mixing ratio increasing by about a factor of 4 in the 12 hours from 20:00 to 08:00, especially due to operation of nearby brick kilns continuing through ~~the~~ night. During the daytime, the wind becomes stronger and the horizontal and vertical circulation dilutes and transports pollution around and out of the valley. Although normally the pollution levels are presumed to be higher in the heavily populated valley than in the immediate surrounding region, occasionally the synoptic circulation will transport in CO and O_3-rich air, especially influenced by forest fires and agro-residue burning in the IGP region and Himalayan foothills, as was observed on a few episode~~s~~ ~~days~~ in the pre-monsoon season.

The observed O_3 mixing ratio was highest in the pre-monsoon season at all sites, and the daily maximum 8-hour average O_3 exceeded the WHO guideline of 50 ppb on about 80% of the days during this season at the semi-urban/urban sites of Bode and Paknajol, while at Nagarkot (which is in the free troposphere, i.e., above valley's boundary layer most of the time, especially during nighttime) it exceeded the WHO guideline on 92% of the days in pre-monsoon season. During the whole observation period, the 8 hour maximum average O_3 exceeded the WHO recommended value on 29%, 37% and 45% of the days at Bode, Paknajol and Nagarkot, respectively. The diurnal cycle showed evidence of photochemical production, larger scale advection of polluted air masses as well as possible down-mixing of O_3 during the daytime, as also observed by Putero et al., (2015) at Paknajol, with the hourly mixing ratio at the polluted site increasing from typically 5-20 ppb in the morning to an early afternoon peak of 60-120 ppb (Putero et al., 2015; Bhardwaj et al., 2018).

These high O_3 levels have deleterious effects on human health and ecosystems, including agro-ecosystems in the Kathmandu Valley and surrounding regions, thus justifying mitigation measures to help reduce the levels of O_3 (its precursors VOCs and NOx), CO and other pollutants. Determining the most effective mitigation measures will be challenging due to the complicated interplay of pollution and meteorology as well as local and regional pollution sources. This study has provided information on current ambient levels and the diurnal/seasonal variations. This will be helpful in the design of future policies, both as a baseline for evaluating the effectiveness of mitigation measures, as well as giving insight into the connections between

various pollutant sources (e.g., brick kilns) and their impacts on seasonally elevated CO levels, especially at nighttime. One particular contribution has been the development of a top-down estimate of the total emission flux of CO at Bode, which was found to be 4.9~~2~~ µg m^{-2} s^{-1}. This is several times higher (by a factor of 2-14 times) than the CO emission fluxes for the Kathmandu Valley in state-of-the-art inventories such as EDGAR-HTAP, REAS, and INTEX-B. This points out the need for the development of updated comprehensive emission inventory databases for this region. The improved emission inventory is necessary~~, in order~~ to provide more accurate input to model simulations ~~needed~~ to assess air pollution processes and mitigation options for the Kathmandu Valley and the broader surrounding region.

While the high levels of particulate pollution in the Kathmandu Valley have caught the main attention of the public and policymakers, due to their immediately visible nature, our paper points out that ozone is also a serious problem here. In fact, its higher levels on the nearby mountaintop location of Nagarkot, which is much more representative of regional air pollution, point to an ozone problem in the wider foothills of the Himalayas. In fact ~~that~~ the extent of ozone pollution in the large surrounding Himalayan foothills has been insufficiently recognized until our study. This ~~, and that~~ needs monitoring and research to identify feasible mitigation options.

Data Availability

The observational data collected for this study will be made public through the SusKat website of IASS. They are also available upon direct request sent to maheswar.rupakheti@iass-potsdam.de and khadak.mahata@iass-potsdam.de.

Acknowledgement

We are thankful to the funders of the IASS – the German Ministry of Education and Research (BMBF) and the Brandenburg State Ministry of Science, Research and Culture (MWFK) – for their generous support in making these measurements and their analysis possible. This study was partially supported by core funds of ICIMOD contributed by the governments of Afghanistan, Australia, Austria, Bangladesh, Bhutan, China, India, Myanmar, Nepal, Norway, Pakistan,

Switzerland, and the United Kingdom, as well as by funds from the Government of Sweden to ICIMOD's Atmosphere Initiative. The authors would like to thank Bhupesh Adhikary, Bhogendra Kathayat, Shyam Newar, Dipesh Rupakheti, Nirjala Koirala, Ashish Bhatta, Begam Roka, Sunil Babu Khatry, Giampietro Verza, and several staff members at the Kamdhenu Madhyamik Vidhyalaya, Naikhandi who assisted in the field measurements, Siva Praveen Puppala for initial data processing, and Pankaj Sadavarte for helping with the emission databases. We are grateful to the Department of Environmental Sciences, University of Virginia, USA, for making available CO and O_3 instruments for the measurements. We also thank the staff at Real Time Solutions (RTS), Lalitpur, Nepal for providing an automatic weather station.

References

Avnery, S., Mauzerall, D. L., Liu, J., and Horowitz, L. W.: Global crop yield reductions due to surface ozone exposure: 1. Year 2000 crop production losses and economic damage, Atmos. Environ., 45, 2284–2296, doi:10.1016/j.atmosenv.2010.11.045, 2011.

Bhardwaj, P., Naja, M., Rupakheti, M., Panday, A. K., Kumar, R., Mahata, K., Lal, S., Lawrence, M. G., Chandola, H. C.: Variations in surface ozone and CO in the Kathmandu Valley during SusKat-ABC international field campaign, Atmos. Chem. Phys. Discus., https://doi.org/10.5194/acp-2017-306, 2017.

Bonasoni P., P. Laj, A. Marinoni, M. Sprenger, F. Angelini, J. Arduini, U. Bonafè, F. Calzolari, T. Colombo, S. Decesari, C. Di Biagio, A. G. di Sarra, et. al.: Atmospheric brown clouds in the Himalayas: first two years of continuous observations at the Nepal Climate Observatory-Pyramid (5079 m). Atmos. Chem. Phys., 10, 7515-7531, 2010.

Brauer, M., Amman, M., Burnett, R. T., Cohen, A., Dentener, F., Zenati, M., Henderson, S. B., Krzyzanowski, M., Martin, R. V., Van Dingenen, R., van Donkelaar, A., and Thurston, G. D.: Exposure assessment for estimation of the global burden of disease attributable to outdoor air pollution, Environ. Sci. Technol., 46, 652–660, doi:10.1021/es2025752, 2012.

838 Burney, J., and Ramanathan, V.: Recent climate and air pollution impacts on Indian agriculture,
839 Proceedings of the National Academy of Sciences of the United States of America, 111, 16319-
840 16324, doi:10.1073/pnas.1317275111, 2014.

841 Chen, P., Kang, S., Li, C., Rupakheti, M., Yan, F., Li, Q., Ji, Z., Zhang, Q., Luo, W., Sillanpää,
842 M.: Characteristics and sources of polycyclic aromatic hydrocarbons in atmospheric aerosols in
843 the Kathmandu Valley, Nepal, Sci. Total Environ., 538, 86-92, doi:
844 10.1016/j.scitotenv.2015.08.006, 2015.

845 Cristofanelli, P., Bracci, A., Sprenger, M., Marinoni, A., Bonafè, U., Calzolari, F., Duchi, R.,
846 Laj, P., Pichon, J. M., Roccato, F., Venzac, H., Vuillermoz, E., and Bonasoni, P.: Tropospheric
847 ozone variations at the Nepal Climate Observatory- Pyramid (Himalayas, 5079ma.s.l.) and
848 influence of deep stratospheric intrusion events, Atmos. Chem. Phys., 10, 6537–6549,
849 doi:10.5194/acp-10-6537-2010, 2010.

850 Dentener, F., Stevenson, D., Ellingsen, K., Van Noije, T., Schultz, M., Amann, M., Atherton, C.,
851 Bell, N., Bergmann, D., and Bey, I.: The global atmospheric environment for the next
852 generation, Environ. Sci. Technol., 40, 3586-3594, 2006.

853 Davidson, C. I., Lin, S.-F., and Osborn, J. F.: Indoor and outdoor air pollution in the Himalayas,
854 Environ. Sci. Technol., 20(6), 561 – 566, doi:10.1021/es00148a003, 1986.

855 Dhungel, S., Kathayat, B., Mahata, K., and Panday, A.: Transport of regional pollutants through
856 a remote trans-Himalayan valley in Nepal, Atmos. Chem. Phys., ~~Discuss., 2016, 1-23,~~
857 ~~doi:10.5194/acp-2016-824, 2016~~18, 1203-1216, https://doi.org/10.5194/acp-18-1203-2018,
858 2018.

859 Department of Transport Management (DoTM).: Annual report of Ministry of Labor and
860 Transport Management, Government of Nepal, 2015.

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

Fowler, D., Flechard, C., Cape, J. N., Storeton-West, R. L., and Coyle, M.: Measurements of ozone deposition to vegetation quantifying the flux, the stomatal and nonstomatal components, Water Air Soil Pollut., 130, 63–74, doi:10.1023/a:1012243317471, 2001.

Fueglistaler, S., Dessler, A. E., Dunkerton, T. J., Folkins, I., Fu, Q., and Mote, P. W.: Tropical tropopause layer, Rev. Geophys., 47, RG1004, doi:10.1029/2008RG000267, 2009.

Geiß, A., Wiegner, M., Bonn, B., Schäfer, K., Forkel, R., von Schneidemesser, E., Münkel, C., Chan, K. L., and Nothard, R.: Mixing layer height as an indicator for urban air quality?, Atmos. Meas. Tech. Discuss., 2017, 1-32, doi:10.5194/amt-2017-53, 2017.

Giri, D., Murthy, K., Adhikary, P., Khanal, S.: Ambient air quality of Kathmandu Valley as reflected by atmospheric particulate matter concentrations (PM10), Int. J. Environ. Sci. Technol. 3, 403–410, 2006.

Highwood, E. J. and Hoskins, B. J.: The tropical tropopause, Q. J. Roy. Meteorol. Soc., 124, 1579–1604, doi:10.1002/qj.49712454911, 1998.

Huszar, P., Belda, M., and Halenka, T.: On the long-term impact of emissions from central European cities on regional air quality, Atmos. Chem. Phys., 16, 1331–1352, doi:10.5194/ acp-16-1331-2016, 2016.

International Energy Agency (IEA).: Energy and air pollution, World Energy Outlook Special Report 2016, International Energy Agency, 2016.

Janssens-Maenhout, G., Dentener, F., van Aardenne, J., Monni, S., Pagliari, V., Orlandini, L., Klimont, Z., Kurokawa, J., Akimoto, H., Ohara, T., Wankmüller, R., Battye, B., Grano, D., Zuber, A., and Keating, T.: EDGAR-HTAP: a harmonized gridded air pollution emission dataset based on national inventories, Tech. Rep. JRC68434, Publications Office of the European Union, doi:10.2788/14102 (online), http://publications.jrc.ec.europa.eu/repository/handle/JRC68434, 2000.

Kiros, F., Shakya, K. M., Rupakheti, M., Regmi, R. P., Maharjan, R., Byanju, R. M., Naja, M., Mahata, K., Kathayat, B., and Peltier, R. E.: Variability of Anthropogenic Gases: Nitrogen oxides, sulfur dioxide, ozone and ammonia in Kathmandu Valley, Nepal, Aerosol Air Qual. Res., 16: 3088–3101, 2016.

894 Kumar, R., Naja, M., Venkataramani, S., and Wild, O.: Variation in surface ozone at Nainital: A
895 high-altitude site in the central Himalayas, J. Geophys. Res., 115 (D16),
896 doi:10.1029/2009JD013715, 2010.

897 Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T.,
898 Kawashima, K., and Akimoto, H.: Emissions of air pollutants and greenhouse gases over Asian
899 regions during 2000-2008: Regional emission inventory in Asia (REAS) version 2, Atmos.
900 Chem. Phys., 13, 11 019–11 058, doi:10.5194/acp-13-11019-2013, 2013.

901 Lal, S., Naja, M., and Subbaraya B. H.: Seasonal variations in surface ozone and its precursors
902 over an urban site in India, Atmos. Environ., 34, 2713-2724, doi: 10.1016/S1352-
903 2310(99)00510-5, 2000.

904 Lawrence, M., and Lelieveld, J.: Atmospheric pollutant outflow from southern Asia: a review,
905 Atmos. Chem. Phys., 10, 11017-11096, 2010.

906 Lim, S. S., Vos, T., Flaxman, A. D., Danaei, G., Shibuya, K., Adair-Rohani, H., Amann, M.,
907 Anderson, H. R., Andrews, K. G., Aryee, M., Atkinson, C., Bacchus, L. J., Bahalim, A. N.,
908 Balakrishnan, K., Balmes, J., Barker-Collo, S., Baxter, A., Bell, M. L., Blore, J. D., Blyth, F.,
909 Bonner, C., Borges, G., Bourne, R…and Ezzati, M.: A comparative risk assessment of burden of
910 disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990-2010:
911 a systematic analysis for the global burden of disease study 2010, Lancet, 380, 2224–2260, 2012.

912 Lüthi, Z. L., Škerlak, B., Kim, S. W., Lauer, A., Mues, A., Rupakheti, M., and Kang, S.:
913 Atmospheric brown clouds reach the Tibetan Plateau by crossing the Himalayas, Atmos. Chem.
914 Phys. 15, 6007-6021, doi:10.5194/acp-15-6007-2015, 2015.

915 Mahata, K. S., Panday, A. K., Rupakheti, M., Singh, A., Naja, M., and Lawrence, M. G.:
916 Seasonal and diurnal variations of methane and carbon dioxide in the Kathmandu Valley in the
917 foothills of the central Himalaya, Atmos. Chem. Phys. Discuss., 2017, 1-55, doi:10.5194/acp-
918 2016-1136, 2017.

919 Marinoni, A., Cristofanelli, P., Laj, P., Duchi., R., Putero, D., Calzolari, F., Landi., T. C.,
920 Vuillermoz, E., Maione, M., and Bonasoni, P.: High black carbon and ozone concentrations

921 during pollution transport in the Himalayas: Five years of continuous observations at NCO-P
922 global GAW station, J. Environ. Sci., 25(8) 1618–1625, 2013.

923 Ming, J., Xiao, C., Sun, J., Kang, S.-C, and Bonasoni, P.: Carbonaceous particles in the
924 atmosphere and precipitation of the Nam Co region, central Tibet, J. Environ. Sci.-CHINA,
925 22(11), 1748-1756, 2010.

926 Monks, P. S., Granier, C., Fuzzi, S., Stohl, A., Williams, M. L., Akimoto, H., Amann, M.,
927 Baklanov, A., Baltensperger, U., Bey, I., Blake, N., Blake, R. S., Carslaw, K., Cooper, O. R.,
928 Dentener, F., Fowler, D., Fragkou, E., Frost, G. J., Generoso, S., Ginoux, P., Grewe, V.,
929 Guenther, A., Hansson, H. C., Henne, S., Hjorth, J., Hofzumahaus, A., Huntrieser, H., Isaksen, I.
930 S. A., Jenkin, M. E., Kaiser, J., Kanakidou, M., Klimont, Z., Kulmala, M., Laj, P., Lawrence, M.
931 G., Lee, J. D., Liousse, C., Maione, M., McFiggans, G., Metzger, A., Mieville, A.,
932 Moussiopoulos, N., Orlando, J. J., O'Dowd, C. D., Palmer, P. I., Parrish, D. D., Petzold, A.,
933 Platt, U., Poeschl, U., Prevot, A. S. H., Reeves, C. E., Reimann, S., Rudich, Y., Sellegri, K.,
934 Steinbrecher, R., Simpson, D., ten Brink, H., Theloke, J., van derWerf, G. R., Vautard, R.,
935 Vestreng, V., Vlachokostas, C., and von Glasow, R.: Atmospheric composition change – global
936 and regional air quality, Atmos. Environ., 43, 5268–5350, doi:10.1016/j.atmosenv.2009.08.021,
937 2009.

938 Mues, A., Rupakheti, M., Münkel, C., Lauer, A., Bozem, H., Hoor, P., Butler, T., and Lawrence,
939 M.: Investigation of the mixing layer height derived from ceilometer measurements in the
940 Kathmandu Valley and implications for local air quality, Atmos. Chem. Phys., doi:10.5194/acp-
941 17-8157, 2017.

942 Naja, M., and Lal, S.: Surface ozone and precursor gases at Gadanki (13.5°N, 79.2°E), a tropical
943 rural site in India, J. Geophys. Res. 107 (D14), ACH 8-1-ACH 8–13, doi:10.1029/2001jd000357,
944 2002.

945 Nayava, J. L.: Rainfall in Nepal, the Himalayan Rev. Nepal, Geographical Society, 12:1– 18,
946 1980.

947 Organisation for Economic Co-operation and Development (OECD): The economic
948 consequences of outdoor air pollution, OECD Publishing,
949 doi: http://dx.doi.org/10.1787/9789264257474-en, 2016.

950 Panday, A. K., and Prinn, R. G..: Diurnal cycle of air pollution in the Kathmandu Valley, Nepal:
951 Observations, J. Geophys. Res., 114 (D9), doi:10.1029/2008JD009777, 2009.

952 Panday, A. K. Prinn, R. G., and Schär, C.: Diurnal cycle of air pollution in the Kathmandu
953 Valley, Nepal: 2. Modeling results, J. Geophys. Res., 114 (D21), doi:10.1029/2008JD009808,
954 2009.

955 Pudasainee, D., Sapkota, B., Shrestha, M. L., Kaga, A., Kondo, A., and Inoue, Y.: Ground level
956 ozone concentrations and its association with NOx and meteorological parameters in Kathmandu
957 Valley, Nepal, Atmos. Environ., 40(40), 8081–8087, doi:10.1016/j.atmosenv.2006.07.011, 2006.

958 Putero, D., Cristofanelli, P., Marinoni, A., Adhikary, B., Duchi, R., Shrestha, S.D., Verza, G.P.,
959 Landi, T.C., Calzolari, F., Busetto, M., Agrillo, G., Biancofiore, F., Di Carlo, P., Panday, A. K.,
960 Rupakheti, M., and Bonasoni,P.: Seasonal variation of ozone and black carbon observed at
961 Paknajol, an urban site in the Kathmandu Valley, Nepal. Atmos. Chem. Phys., 15(24), 13957-
962 13971, doi:10.5194/acp-15-13957-2015, 2015.

963 Putero, D., Cristofanelli, P., Sprenger, M., Škerlak, B., Tositti, L., and Bonasoni, P.: STEFLUX,
964 a tool for investigating stratospheric intrusions: application to two WMO/GAW global stations.
965 Atmos. Chem. Phys., 16, 14203–14217, doi:10.5194/acp-16-14203-2016, 2016.

966 Ram, K., and Sarin, M.: Spatio-temporal variability in atmospheric abundances of EC, OC and
967 WSOC over Northern India, J. Aerosol Sci., 41, 88–98, 2010.

968 Regmi, R. P., Kitada, T., and Kurata, G.: Numerical simulation of late wintertime local flows in
969 Kathmandu Valley, Nepal: Implication for air pollution transport, J. Appl. Meteorol., 42, 389-
970 403, 2003.

971 Rupakheti, M., Panday, A. K., Lawrence, M. G., Kim, S. W., Sinha, V., Kang, S. C., Naja, M.,
972 Park, J. S., Hoor, P., Holben, B., Sharma, R. K., Mues, A., Mahata, K. S., Bhardwaj, P., Sarkar,
973 C., Rupakheti, D., Regmi, R. P., and Gustafsson, Ö.: Air pollution in the Himalayan foothills:

974 overview of the SusKat-ABC international air pollution measurement campaign in Nepal,
975 Atmos. Chem. Phys. Discuss., in preparation, 2017.

976 Rupakheti, D., Adhikary, B., Praveen, P. S., Rupakheti, M., Kang, S.-C., Mahata, K. S., Naja,
977 M., Zhang, Q., Panday, A. K., and Lawrence, M. G.: Pre-monsoon air quality over Lumbini, a
978 world heritage site along the Himalayan foothills, Atmos. Chem. Phys., 17, 11041-111063,
979 https://doi.org/10.5194/acp-17-11041-2017, 2017.

980 Sadavarte, P., Rupakheti, M., Shakya, K., Bhave, P.V., and Lawrence, M.G.: Nepal emission
981 (NEEM): A high resolution technology - based bottom-up emissions inventory for Nepal, ACP
982 in preparation, 2018.

983 Sarkar, C., Sinha, V., Kumar, V., Rupakheti, M., Panday, A., Mahata, K.S., Rupakheti, D.,
984 Kathayat, B., and Lawrence, M.G.: Overview of VOC emissions and chemistry from PTR-TOF-
985 MS measurements during the SusKat-ABC campaign: high acetaldehyde, isoprene and isocyanic
986 acid in wintertime air of the Kathmandu Valley, Atmos. Chem. Phys., 16, 3979-4003, 2016.

987 Sarkar, C., Sinha, V., Sinha, B., Panday, A. K., Rupakheti, M., and Lawrence, M. G.: Source
988 apportionment of NMVOCs in the Kathmandu Valley during the SusKat-ABC international field
989 campaign using positive matrix factorization, Atmos. Chem. Phys., 17, 8129-8156, 2017.

990 Shakya, K. M., Rupakheti, M., Shahi, A., Maskey, R., Pradhan, B., Panday, A., Puppala, S. P.,
991 Lawrence, M., and Peltier, R. E.: Near-road sampling of PM2. 5, BC, and fine-particle chemical
992 components in Kathmandu Valley, Nepal, Atmos. Chem. Phys., 17, 6503-6516,
993 https://doi.org/10.5194/acp-17-6503-2017, 2017.

994 Sharma, P., Kuniyal, J. C., Chand, K., Guleria, R. P., Dhyani, P. P. and Chauhan, C.: Surface
995 ozone concentration and its behavior with aerosols in the northwestern Himalaya, India. Atmos.
996 Environ. 71, 44-53, doi:10.1016/12.042, 2013.

997 Shrestha, A. B., Wake, C. P., Mayewski, P. A., and Dibb, J.E.: Maximum Temperature Trends in
998 the Himalaya and Its Vicinity: An Analysis Based on Temperature Records from Nepal for the
999 Period 1971–94, J. Climate, 12, 2775-2786, 1999.

Shrestha, S. R., Kim Oanh, N. T., Xu, Q., Rupakheti, M., and Lawrence, M. G.: Analysis of the vehicle fleet in the Kathmandu Valley for estimation of environment and climate co-benefits of technology intrusions, Atmos. Environ., 81, 579-590, 2013.

Silva, R. A., West, J. J., Zhang, Y., Anenberg, S. C., Lamarque, J.-F., Shindell, D. T., Collins, W. J., Dalsoren, S., Faluvegi, G., Folberth, G., Horowitz, L. W., Nagashima, T., Naik, V., Rumbold, S., Skeie, R., Sudo, K., Takemura, T., Bergmann, D., Cameron- Smith, P., Cionni, I., Doherty, R. M., Eyring, V., Josse, B., MacKenzie, I. A., Plummer, D., Righi, M., Stevenson, D. S., Strode, S., Szopa, S., and Zeng, G.: Global premature mortality due to anthropogenic outdoor air pollution and the contribution of past climate change, Environ. Res. Lett., 8, 034005, doi:10.1088/1748-9326/8/3/034005, 2013.

Talbot, R., Mao, H., and Sive, B.: Diurnal characteristics of surface level O_3 and other important trace gases in New England, J. Geophys. Res., 110 (D9), doi:10.1029/2004JD005449, 2005.

Tripathee, L., Kang, S.-C., Huang, J., Sharma, C., Sillanpaa, M., Guo, J., and Paudyal, R.: Concentrations of trace elements in wet deposition over the central Himalayas, Nepal, Atmos. Environ., 95, 231–238, 2014

Sinha, V., Kumar, V., and Sarkar, C.: Chemical composition of pre-monsoon air in the Indo-Gangetic Plain measured using a new air quality facility and PTR-MS: high surface ozone and strong influence of biomass burning, Atmos. Chem. Phys., 14, 5921-5941, doi:10.5194/acp-14-5921-2014, 2014.

Tissier, A.-S. and Legras, B.: Convective sources of trajectories traversing the tropical tropopause layer, Atmos. Chem. Phys., 16, 3383–3398, doi:10.5194/acp-16-3383-2016, 2016.

Vadrevu, K., Ellicott, E., Giglio, L., Badarinath, K., Vermote, E., Justice, C., Lau, W.: Vegetation fires in the Himalayan region - aerosol load, black carbon emissions and smoke plume heights, Atmos. Environ., 47, 241–251, 2012.

Wang, Y., Konopka, P., Liu, Y., Chen, H., Müller, R., Plöger, F., Riese, M., Cai, Z., and Lü, D.: Tropospheric ozone trend over Beijing from 2002–2010: Ozonesonde measurements and modeling analysis, Atmos. Chem. Phys., 12, 8389–8399, doi:10.5194/acp-12-8389-2012, 2012.

World Health Organization (WHO): WHO Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide, Global update 2005, Summary of risk assessment, WHO Press, Geneva, Switzerland, 2006.

World Health Organization (WHO).: 7 million premature deaths annually linked to air pollution, 2014 (http://www.who.int/mediacentre/news/releases/2014/air-pollution/en/).

Zhang, Q., Streets, D., Carmichael, G., He, K., Huo, H., Kannari, A., Klimont, Z., Park, I., Reddy, S., Fu, J., Chen, D., Duan, L., Lei, Y., Wang, L., and Yao, Z.: Asian emissions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys., 9, 5131–5153, 2009.

Table 1. Information on the sampling sites (of the SusKat-ABC campaign) used in this study with sampling carried out during 2013-2014 in the Kathmandu Valley. The altitude is in meter above mean sea level (m asl)

Site	General setting of site	Location, altitude (m asl)
Bode	Sub-urban, tallest building with scattered houses surrounded by agricultural fields	27.69°N, 85.40°E, 1345
Bhimdhunga	Rural. On the ridge, close to the pass separating the Kathmandu Valley from a valley of a tributary the Trishuli River to the west	27.73°N, 85.23°E, 1522
Paknajol	Urban, city-center, the tallest building in the neighborhood	27.72°N, 85.30°E, 1380
Naikhandi	Rural, at outlet of Bagmati River in Southwest corner of the Valley	27.60°N, 85.29°E, 1233
Nagarkot	Mountain rural. Mountain top site of the eastern valley rim, north facing towards the Kathmandu Valley	27.72°N , 85.52°E, 1901

Table 2. Details of the instruments deployed at different sites during the observation period during January 2013-March 2014 in the Kathmandu Valley.

Location	Instrument	Parameters	Inlet/sensor height (above ground)	Duration	Group
1. Bode	a. Horiba APMA-370	CO	20 m	1 Jan-7 Jun 2013	ARIES
	b. Teledyne 400E	O_3	20 m	1 Jan-7 Jun 2013	ARIES
	c. Thermo Scientific 49i	O_3	20 m	18 Jun-31 Dec 2013	IASS
	d. Picarro G2401	CO	20 m	6 Mar 2013-5 Mar 2014	ICIMOD
	e. Campbell AWS	T, RH, SR, WS, WD, RF	22 m	1 Jan-30 Mar 2013	IASS
	f. Davis AWS (Vantage Pro2)	T, RH, P, RF	21 m	30 May-Jul 2013	UVA
	g. Ceilometer (Vaisala CL31)	MLH	20 m	01 Mar 2013- 28 Feb 2014	JGUM
2. Bhimdhunga	a. Thermo Scientific 48i	CO	2 m	1 Jan-15 Jul 2013	UVA
	b. AWS Hobo Onset	T, RH, SR, WS, WD, P	5 m	1 Jan-30 Jun 2013	UVA
3. Naikhandi	a. Thermo Scientific 48i	CO	5 m	3 Jan- 6 Jun 2013	UVA
	b. 2B Tech. Model 205	O_3	5 m	1 Feb-25 May 2013	UVA
	c. AWS Hobo Onset	T, RH, SR, WS, WD, P	2 m	3 Jan-25 Apr 2013	UVA
4. Nagarkot	a. Thermo Scientific 48i	CO	5 m	13 Feb-Apr 3 2013; 8 Jun-15 Jul 2013	UVA
	b. Thermo Scientific 49i	O_3	5 m	9 Jan-30 Jun 2013	UVA
	c. Campbell AWS	T, RH, SR, WS, WD, RF	7 m		IASS
	d. AWS (Vaisala WXT 520)	T, RH, SR, WS, WD, RF, P	7 m	10 Feb-30 Jun 2013	RTS
5. Paknajol	a. Thermo Environmental (49i)	O_3	25 m	1 Feb 2013-30 Jan 2014	EV-K2-CNR
	b. AWS (Vaisala WXT 425)	T, RH, SR, WS, WD, RF, P	25 m	1 Feb 2013-30 Jan 2014	EV-K2-CNR

Note: T - temperature, RH - relative humidity, SR- solar radiation, WS - wind speed, WD - wind direction, RF- rainfall, P – pressure and MLH – Mixing layer height; ARIES - Aryabhatta Research Institute of Observational Sciences, India; ICIMOD - International Center for Integrated Mountain Development, Nepal; IASS - Institute for Advanced Sustainability Studies, Germany; UVA- University of Virginia, USA; JGUM – Johannes Gutenberg University Mainz, Germany; RTS - Real Time Solutions, Nepal; Ev-K2-CNR - Everest-Karakorum - Italian National Research Council, Italy.

Table 3. Summary of the monthly average ozone mixing ratios (ppb) [average (Avg), standard deviation (SD), minimum (Min.) and maximum (Max.)] at four sites* in the Kathmandu Valley, Nepal during 2013-2014 and two sites (Manora Peak and Delhi) in India

	Bode	Paknajol	Nagarkot	Manora[a] Peak	Delhi[b]
Month	Avg ± SD [Min., Max.]	Avg ± SD [Min., Max.]	Avg ± SD [Min., Max.]	Avg ± SD	Avg [Min., Max.]
January	23.5 ± 19.9 [1.4, 87.1]	16.9 ± 18.3 [0.1, 71.7]*	46.7 ± 5.7 [36.4, 73.7	37.3 ± 14.8	19.3 [10, 14.7]
February	25.6 ± 20.4 [1.2, 94.5]	24.2± 20.1 [1.6, 91.7]	47.5 ± 7.5 [28.2, 83.6]	43.8 ± 16.8	25.3 [10.9, 55.7]
March	37.4 ± 24.3 [1.2, 105.9]	37.7 ± 23.8 [1.6, 95.8]	62.4 ± 9.5 [40.5, 98.9]	56.6 ± 11.4	29.7 [13.8, 58]
April	43.4 ± 26.6 [1.4, 116.2]	46.7 ± 26.8 [1.0, 115.5]	71.5 ± 15.5 [40.1, 121.0]	63.1 ± 11.7	33 [13.7, 64.3]
May	38.5 ± 21.2 [2.0, 111.1]	42.8 ± 20.6 [6.7, 103.3]	59.0 ± 20.6 [15,0, 124.5]	67.2 ± 14.2	35.4 [19.8, 62]
June	27.8 ± 12.0 [1.7, 68.4]	27.5 ± 17.0 [0.6, 90.7]	34.2 ± 9.1 [4.6, 72.0]	44.0 ± 19.5	25.6 [12.8, 46.4]
July	21.1 ± 9.5 [1.7, 82.0]	20.5 ± 13.4 [2.0, 77.9]	25.9 ± 6.2 [11.1, 48.0]	30.3 ± 9.9	19.1 [9.4, 37.1]
August	20.3 ± 9.9 [2.0, 70.9]	20.1 ± 12.6 [0.8, 73.1]	28.3 ± 5.8 [15.5, 62.9]	24.9 ± 8.4	14.3 [9.7, 29.5]
September	23.3 ± 14.9 [0.5, 85.9]	24.9 ± 17.4 [0.4, 108.1]	34.8 ± 9.6 [16.1, 79.7]	32.0 ± 9.1	17.7 [7.7, 37.7]
October	19.4 ± 13.8 [0.1, 70.9]	22.6 ± 17.0 [0.6, 83.5]	35.2 ± 10.2 [18.0, 73.8]	42.4 ± 7.9	21.7 [9, 56.9]
November	18.6 ± 15.1 [0.3, 67.7]	22.4 ± 20.9 [0.1, 84.0]	40.1 ± 8.1 [25.6, 73.3]	43.9 ± 7.6	22.6 [9, 55.1]
December	21.7 ± 17.8 [1.0, 96.6]	19.5 ± 19.7 [0.1, 82.0]	43.8 ± 9.0 [24.8, 85.11]	41.6 ± 6.3	20.2 [9.1, 40.3]
Season:					
Winter	24.5 ± 20.1 [1.2, 94.5]	20.2 ± 19.6 [0.1, 91.7]	45.8 ± 7.8 [24.8, 85.1]	40.9	21.6 [9.1, 55.7]
Pre-monsoon	39.8 ± 24.2 [1.2, 116.2]	42.4 ± 24.0 [1.0, 115.5]	64.3 ± 16.7 [14,9, 124.5]	62.3	32.7 [13.7, 64.3]
Monsoon	22.7 ± 12.0 [0.5, 85.9]	23.2 ± 15.5 [0.4, 108.1]	30.8 ± 8.7 [4.6, 79.7]	32.8	19.2 [7.7, 46.4]
Post-monsoon	19.0 ± 14.5 [0.1, 70.9]	22.5 ± 18.9 [0.1, 84.0]	37.6 ± 9.5 [18.0, 73.8]	39.4	22.2 [9, 56.9]

[a] Kumar et al. (2010), [b] Ghude et al. (2008). * O_3 data of Paknajol on January was of 2014.

Table 4. Averege CO mixing ratio (ppb) at different time of the day (daytime - 12:00 – 16:00), and nighttime - 23:00 – 03:00) and the monthly average (total) at four sites in the Kathmandu Valley.

	Winter (16 Jan-15 Feb)			Pre-monsoon (16 Mar-15 Apr)			Monsoon (16 Jun-15Jul)			Post-monsoon (16 Oct-15 Nov)		
Sites	daytime	nighttime	Total	daytime	nighttime	total	Daytime	nighttime	total	daytime	nighttime	total
Bode	405.35	927.21	819.17	430.91	839.17	770.52	210.59	230.08	241.34	269.10	453.95	397.24
Bhimdhunga	324.62	354.23	374.27	374.64	479.37	471.33	196.61	202.85	198.40			
Naikhandi	280.97	356.14	380.40	382.71	425.17	449.83						
Nagarkot							141.68	158.78	160.41			

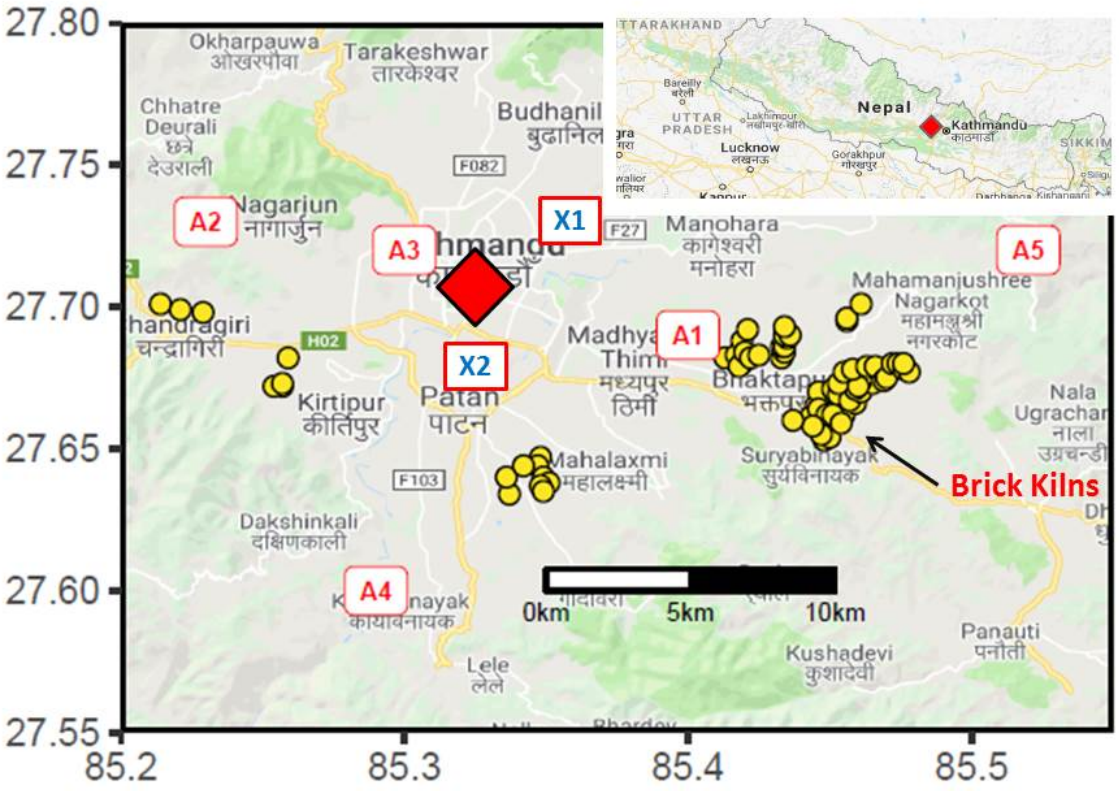

Figure 1. Observation sites in the SusKat-ABC international air pollution campaign during 2013-2014 in the Kathmandu Valley. A1 = Bode, A3 = Paknajol, and A4 = Naikhandi were selected within the valley floor and A2 = Bhimdhunga and A5 = Nagarkot on the mountain ridge. Naikhandi site is also near the Bagmati River outlet. ~~Major passes of the Kathmandu Valley are (a) Mudku Bhanjhyang pass, (b) Bhimdhunga pass and (c) Nagdhunga pass in the west, and (d) Nagarkot and (e) Nala pass in the east and only (f) river outlet in the valley are shown in the Figure~~. Past study sites, ~~(~~Bouddha (X1) and Pulchowk (X2), which are referred in the manuscript, are also shown in the Figure. Source: Google Maps.

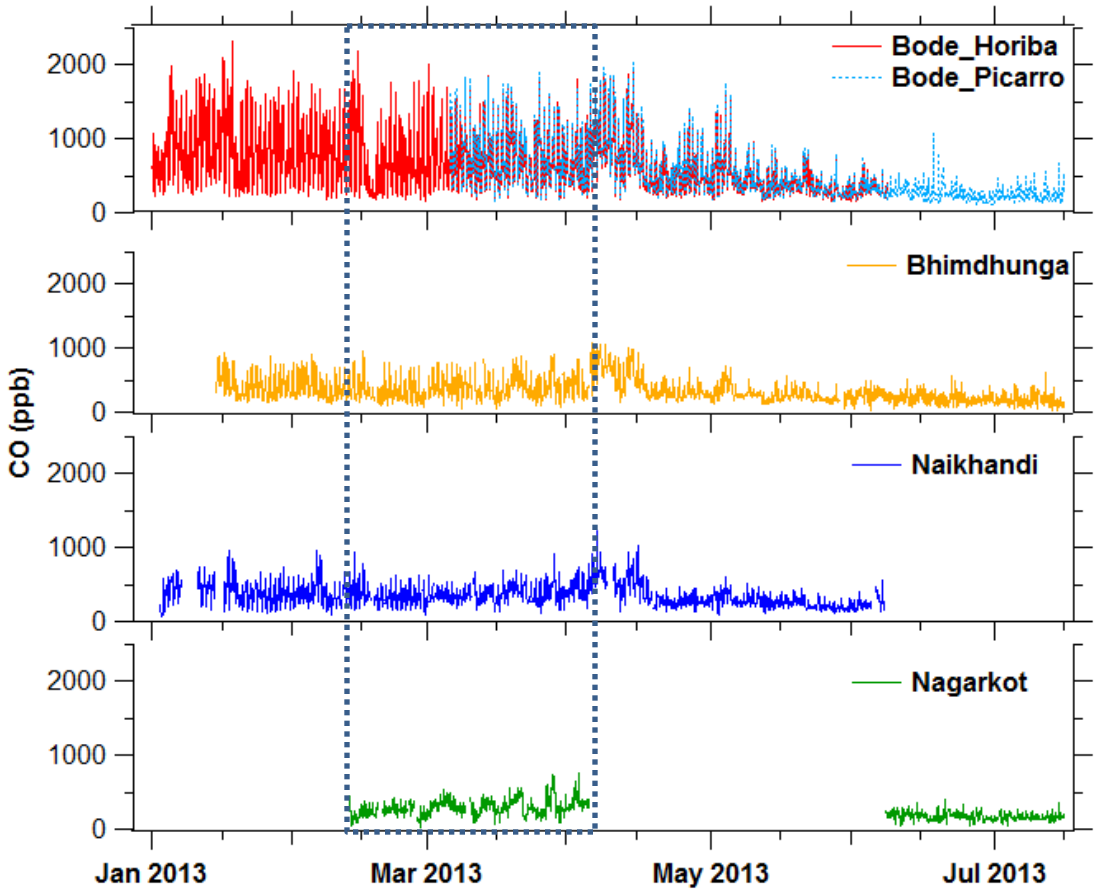

Figure 2. Hourly average CO mixing ratios observed at supersite (Bode) and three satellite sites (Bhimdhunga, Naikhandi and Nagarkot) of the SusKat-ABC international air pollution measurement campaign during January to July 2013 in the Kathmandu Valley. The dotted box represents a period (13 February - 03 April, 2013) during which data for all four sites were available.

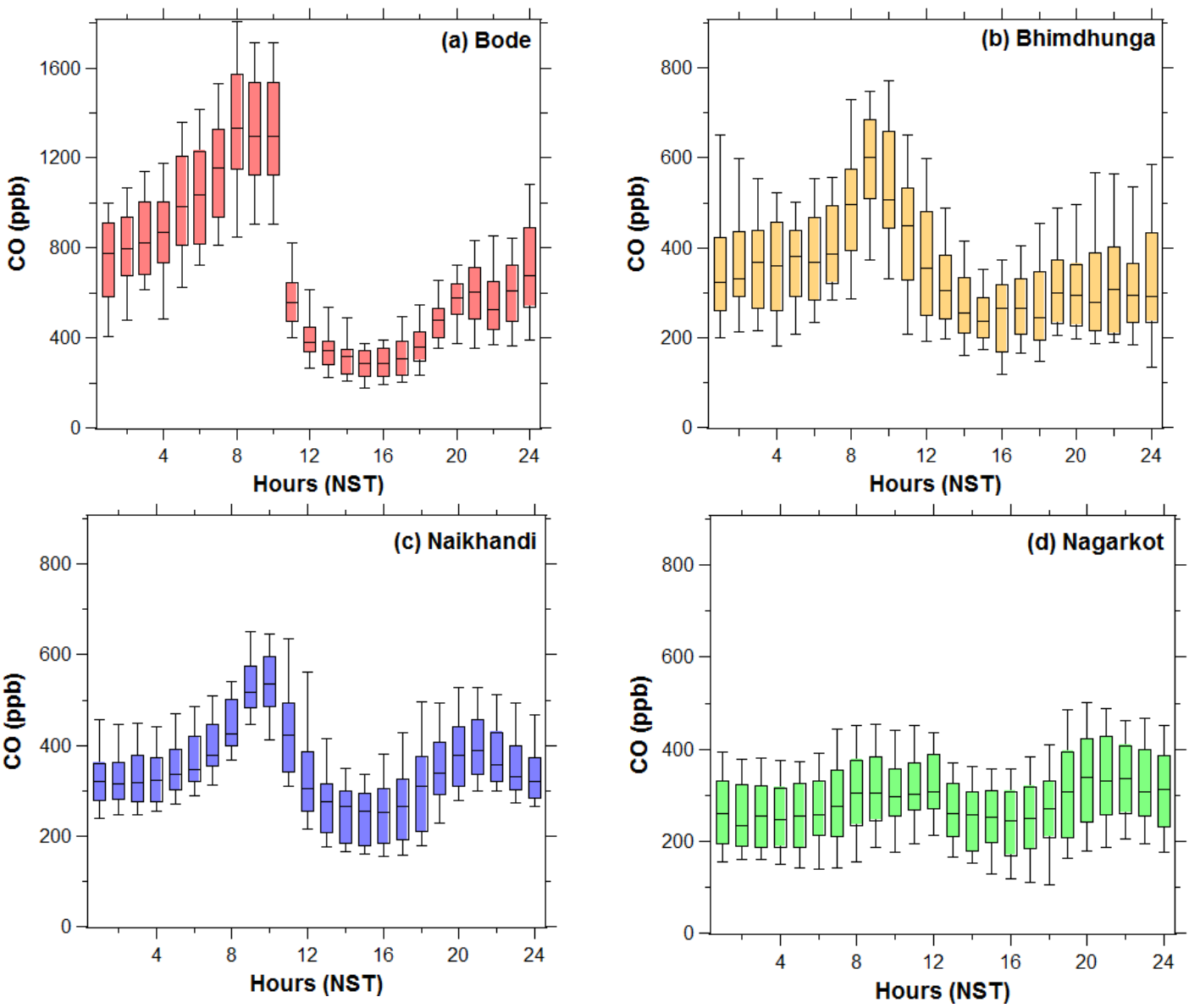

Figure 3. Diurnal variations of hourly average CO mixing ratios during the common observation period (13 February–03 April, 2013) at Bode, Bhimdhunga, Naikhandi and Nagarkot. The lower end and upper end of the whisker represents 10th and 90th percentile, respectively; the lower end and upper end of each box represents the 25th and 75th percentile, respectively, and the black horizontal line in the middle of each box is the median for each month. Note: the y-axis scale of Bode is twice that of the other three sites.

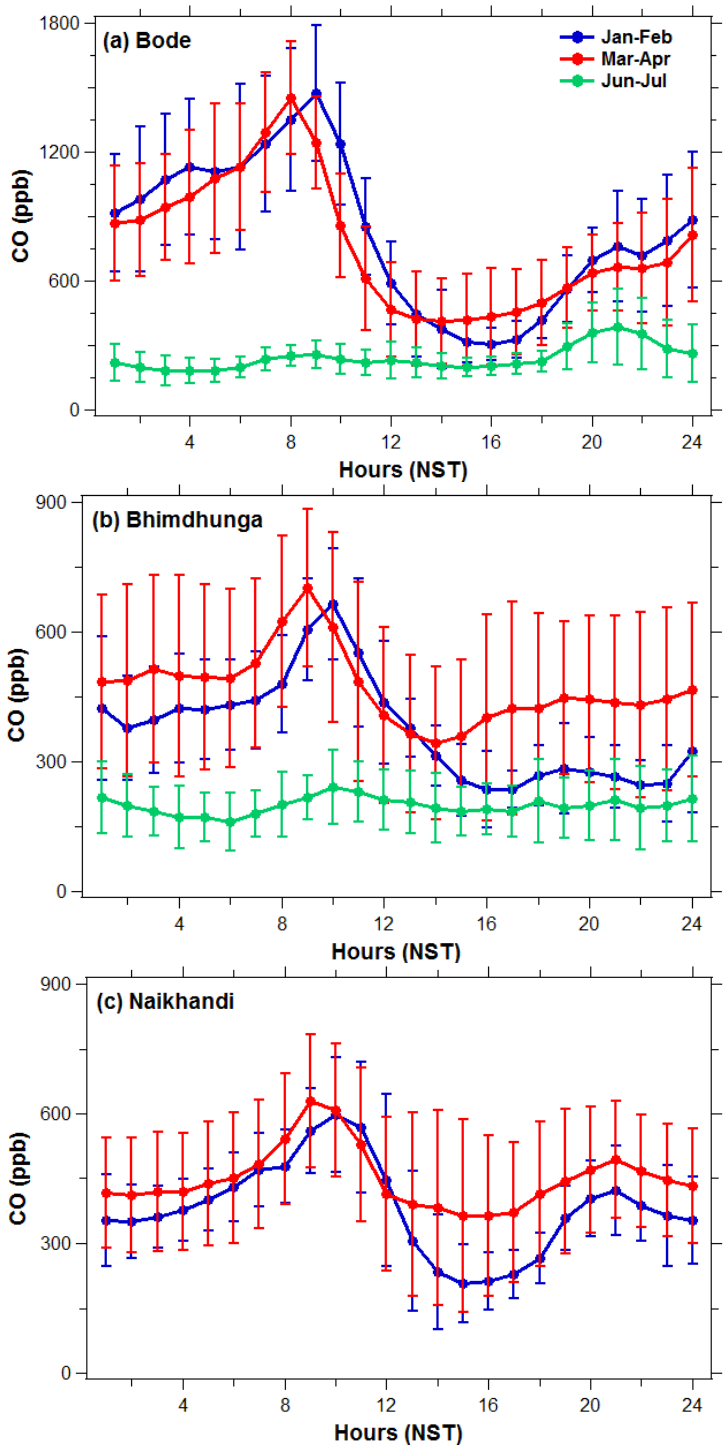

Figure 4. Comparison of diurnal variation of hourly average CO mixing ratios for four seasons at Bode,Bhimdhunga and Naikhandi. Due to the lack of continuous data at some sites, data of one month in each season were taken for comparison as representative of the winter (16 Jan – 15 Feb), pre-monsoon (16 Mar – 15 Apr) and monsoon (16 Jun – 15 Jul) season of 2013. Note: y-axis scale of the top panel (Bode) is double than lower two panels (Bhimdhunga and Naikhandi).

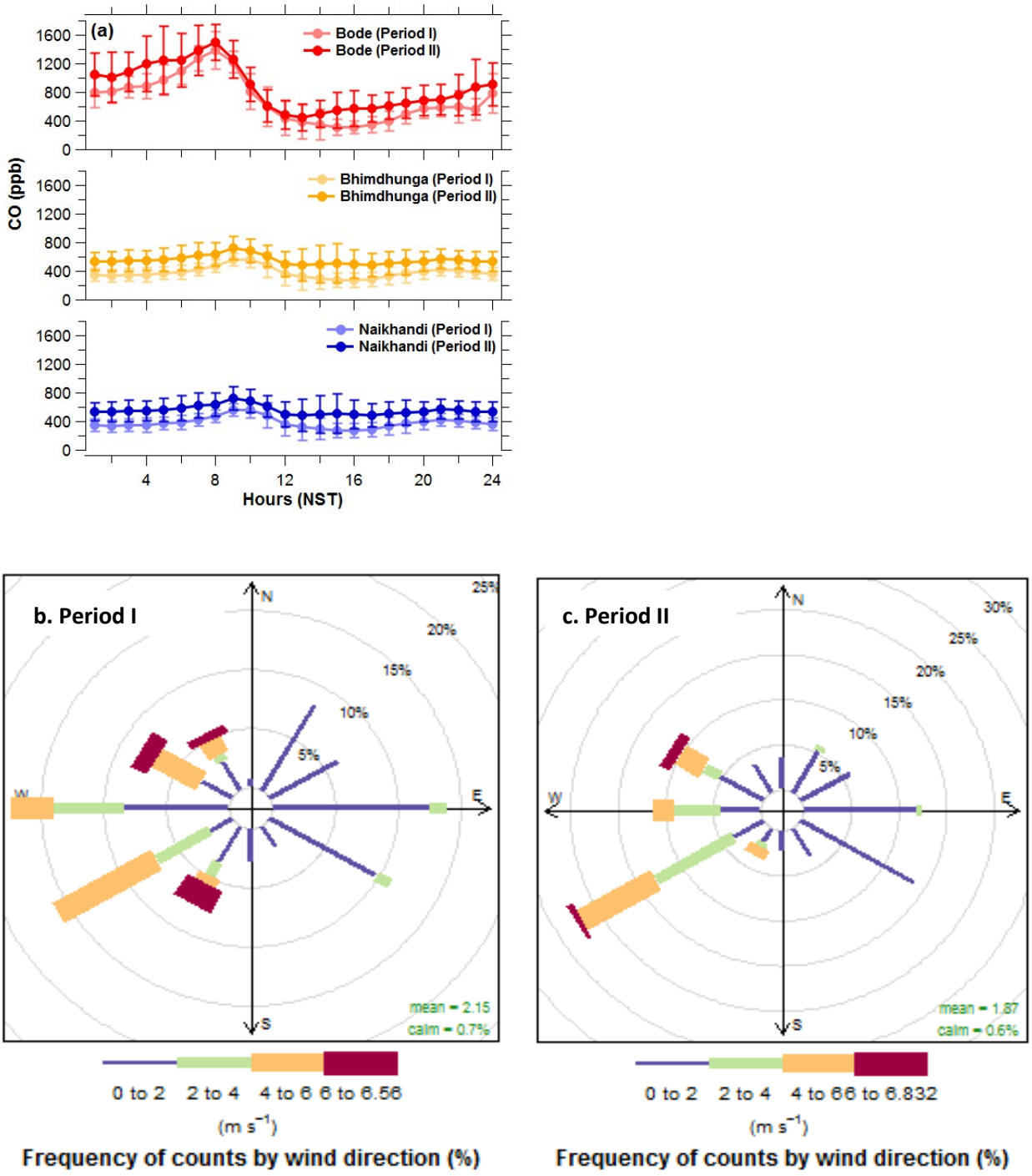

Figure 5. Comparison of hourly average CO mixing ratios during normal days (March 16-30), labelled as period I (faint color) and episode days (April 1-15), labelled as period II (dark color) in 2013 at (a) Bode, Bhimdhunga and Naikhandi in the Kathmandu Valley. The wind roses at Bode corresponding to two periods are also plotted (b) period I and (c) period II respectively.

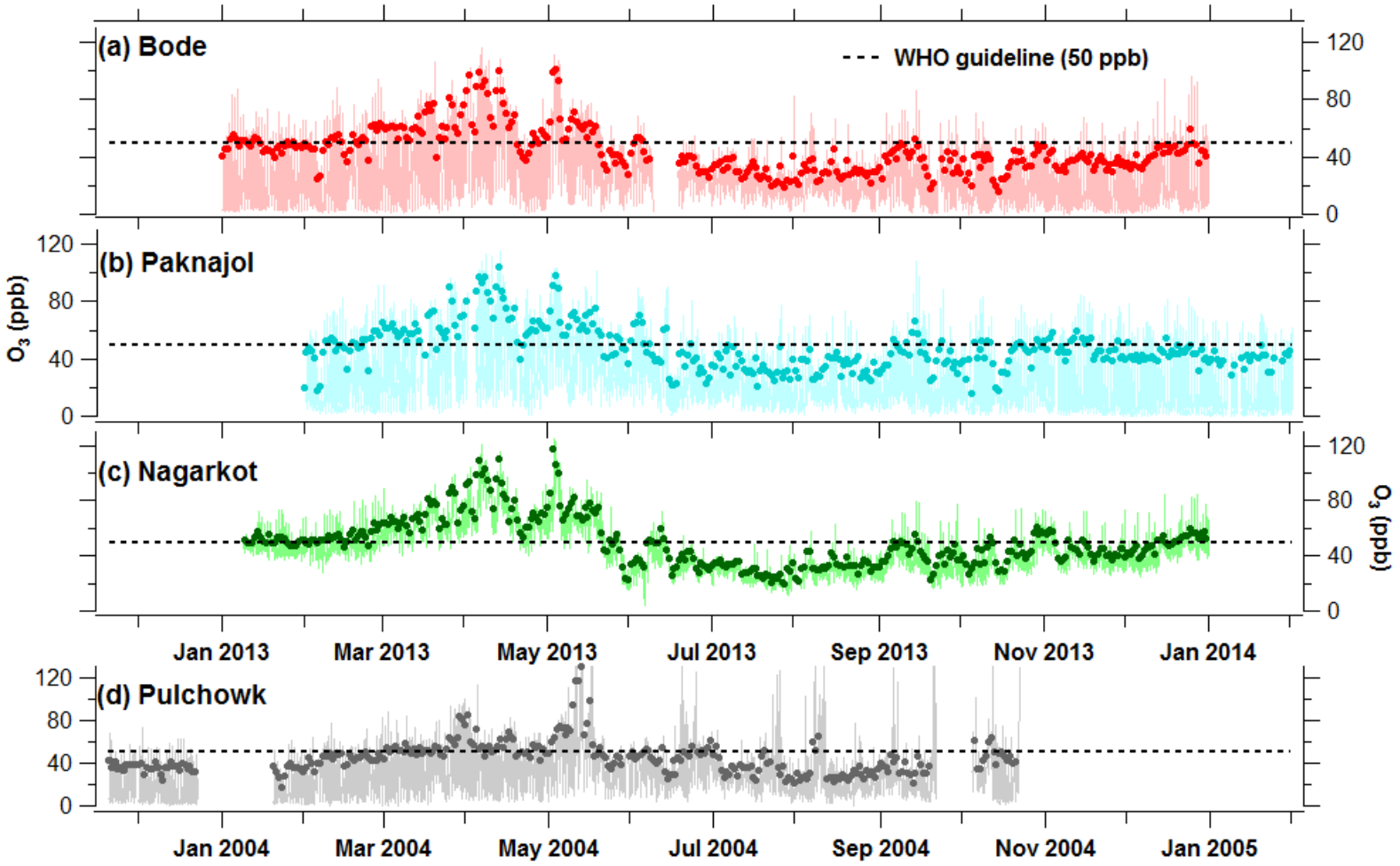

Figure 6. Time series of hourly average (faint colored line) and daily maximum 8-hr average (solid colored circle) O_3 mixing ratio at (a) Bode (semi-urban), (b) Paknajol (urban)and (c) Nagarkot (hilltop) observed during 2013-2014, and (d) Pulchowk (urban) observed during November 2003-October 2004 in the Kathmandu Valley. Black dotted line represents WHO guideline (50 ppb) for daily maximum 8-hour average of O_3.

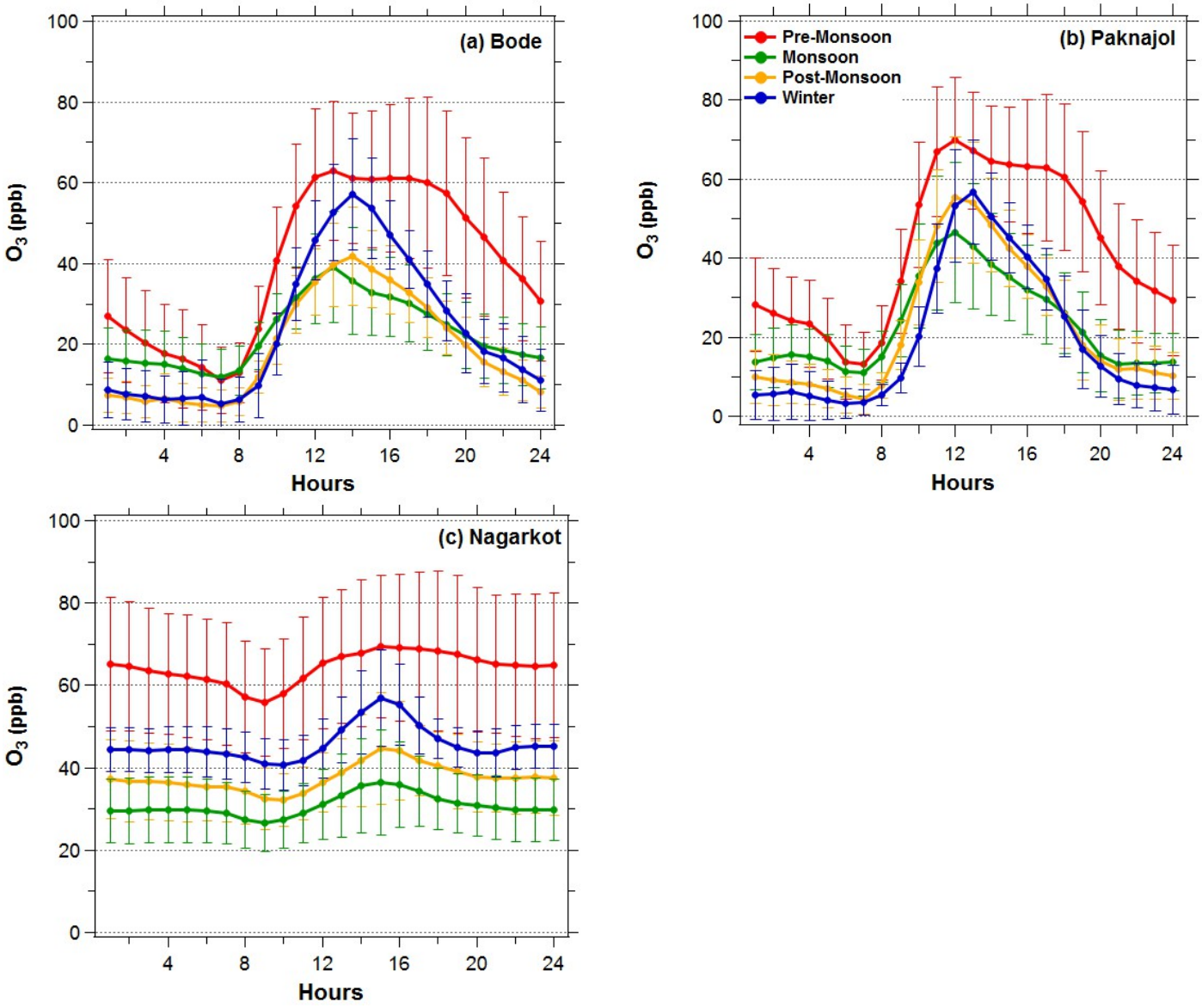

Figure 7. Diurnal pattern of hourly average O_3 mixing ratio for different seasons during January 2013-January 2014 at (a) Bode, (b) Paknajol, and (c) Nagarkot in the Kathmandu Valley. The four seasons (described in the text) are defined as: pre-monsoon (Mar-May), monsoon (Jun-Sep), post-monsoon (Oct-Nov), winter (Dec-Feb).

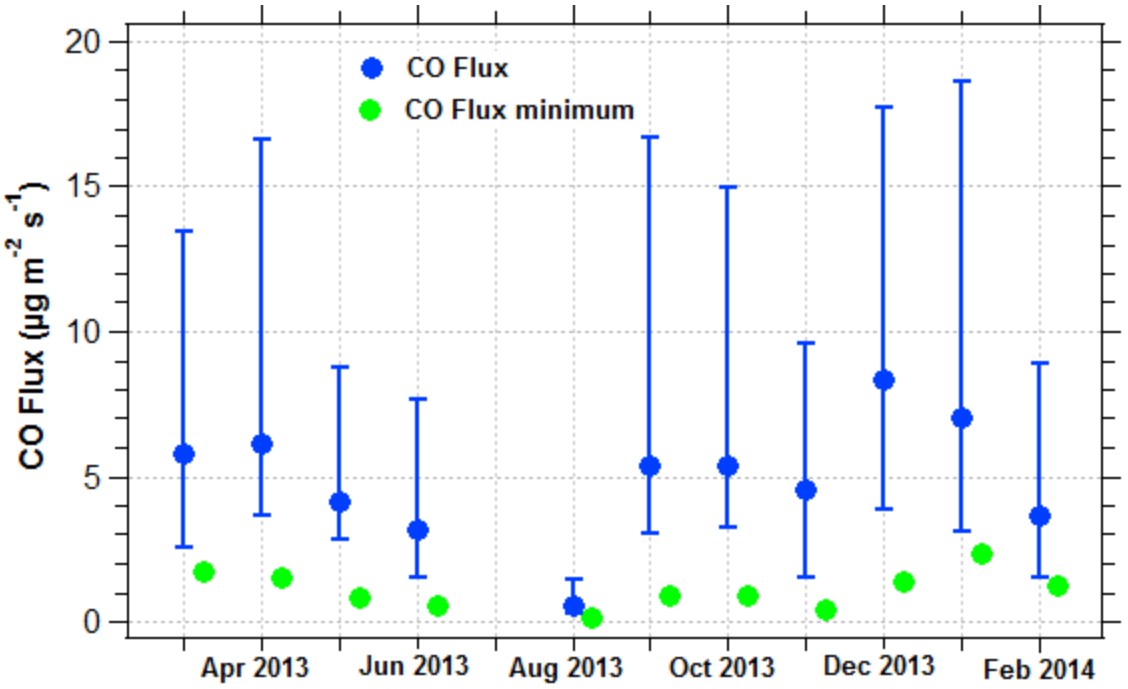

Figure 8. The estimated monthly average CO emission flux, which is based on the mean diurnal cycle of CO mixing ratios of each month for two conditions: (i) with data of all days (CO Flux) (blue dot) with lower and upper ends of the bar representing 25th and 75th percentile respectively, and (ii) with data of morning hours (CO Flux minimum (green dot) in which zero emission is assumed for the other hours of the day. The fluxes for July were not estimated as there were insufficient (less than 15 days) of concurrent CO and mixing layer height data. It is expected that the F_{CO} and $F_{COmin.}$ for July should fall between values for June and August 2013.