

Reviewer #1:

The authors present measurements of BC (light absorption), ozone, and aerosol particles (PM1 and PM10) obtained over approximately a year in the Kathmandu Valley. The year is broken up into 4 periods, pre-monsoon, monsoon, post-monsoon, and winter. Diurnal pattern of the quantities measured appear to be typical of that found in many other locations, dominated by the time dependence of emissions, photochemistry, and boundary layer dynamics. There is some dependence of ozone on wind direction and the high ozone season coincides with the season for regional vegetation fires. What is present appears to be basically correct, though not very exciting because similar features have been seen before and with a limited suite of instruments there is only so much that can be said about cause and affect.

We thank the Reviewer for his/her valuable suggestions and his/her encouraging evaluation. In the following, we report our point-to-point replies to each of the raised points. Modifications to the text are performed in the revised version of the manuscript and are marked in red color.

The experimental setup was defined taking into account both the scientific goals of the SusKat campaign (i.e. achieving a more comprehensive understanding of the dynamics of air pollution and related emissions in the Kathmandu Valley, and constituting the scientific basis in order to support the local implementation of mitigation actions) and the complex logistic conditions in Kathmandu. Since a long duration experiment was planned, state-of-art instrumentation was selected, characterized by a well-known robustness and relatively low necessity of human interventions. It should be also kept in mind that the Kathmandu urban area is characterized by severe problems of power distribution and that black-out can often occur for several hours per day. Even though an UPS system was installed at Paknajol, the installation of complex instrumentation characterized by high power demand (e.g. AMS, SMPS/DMPS) was prevented. However, to our knowledge, this experimental suite still represents the most complete setup ever installed at a single measurement site in the Kathmandu city center.

We believe that having kept these “basic” instruments running for almost two years up to date has been a success, useful for providing a unique dataset that can be used for the implementation of mitigation measurements.

An opportunity that is missed is the determination of relations between ozone, BC, and aerosol particles (accumulation and coarse mode). Correlations between these quantities are presented in Table 2, but because all data is used it is not possible to distinguish chemical effects from boundary layer dynamics. I suggest that this paper could interest a wider audience with a partial repeat of Table 2, limiting the data to convective hours. A very interesting quantity to look for is secondary aerosol production. Insight can be gained by looking at the regression of ozone vs. PM1. Also BC vs. PM1. I'm guessing that relations would be lost by doing regressions on a data subset defined by season or trajectory location. I would do regressions on a day by day basis and see what drops out. Perhaps the slopes of these plots will depend on solar radiation.

I recommend publication with revisions, though I believe the authors would be well served by seeing whether an approach such as given above is productive.

We thank the Reviewer for his/her perspicacious advice on performing correlations on a limited subset only. Unfortunately, our PM₁ values were collected on a daily basis (24h resolution), thus a direct comparison with a subset of O₃ or BC hourly values was not possible. Keeping also in mind that accumulation and coarse particles were available for two seasons only (pre-monsoon

and monsoon), we added a new Table (Table S1, Supplement), where correlations are performed on a subset between 11:00 and 17:00 LT. This time range corresponds, according to the average diurnal variations for wind speed and solar radiation, to the convective hours. By looking at Table S1, the computed r between BC and the number of accumulation particles did not differ much from that of Table 2, indicating the important role of primary emissions (traced by BC) in determining aerosol particle number; nevertheless, the slight decrease in such correlation might indicate the presence of other processes (such as secondary aerosol production). This was further highlighted by the change in the correlation coefficient between O_3 and the accumulation fraction during the convective hours. O_3 can be considered a tracer of secondary pollution processes; moreover, as also shown in the paper, at Kathmandu the high O_3 values in the afternoon are the consequence of transport of aged air masses from the upper residual layers and/or horizontal advection.

Table S1. Correlation coefficients (r) between several parameters (BC, O_3 , accumulation and coarse particles, WS, T and RAD) for hourly and daily (in parentheses) values, over the whole sampling period, computed during convective hours only (i.e. between 11:00 and 17:00).

	O_3	BC	Acc.	Coarse	WS	T	RAD
O_3	-	0.20 (0.17)	0.62 (0.61)	0.51 (0.52)	0.16 (0.49)	0.18 (0.16)	0.50 (0.53)
BC	0.20 (0.17)	-	0.82 (0.81)	0.79 (0.71)	-0.35 (-0.24)	-0.44 (-0.58)	-0.10 (-0.36)
Acc.	0.62 (0.61)	0.82 (0.81)	-	0.79 (0.79)	-0.03 (0.15)	-0.13 (-0.13)	0.03 (-0.01)
Coarse	0.51 (0.52)	0.79 (0.71)	0.79 (0.79)	-	-0.06 (0.21)	-0.03 (0.03)	0.03 (0.04)
WS	0.16 (0.49)	-0.35 (-0.24)	-0.03 (0.15)	-0.06 (0.21)	-	0.28 (0.37)	0.11 (0.72)
T	0.18 (0.16)	-0.44 (-0.58)	-0.13 (-0.13)	-0.03 (0.03)	0.28 (0.37)	-	0.34 (0.50)
RAD	0.50 (0.53)	-0.10 (-0.36)	0.03 (-0.01)	0.03 (0.04)	0.11 (0.72)	0.34 (0.50)	-

Despite adding this new Table, we further analyzed, on a day-by-day basis, the diurnal variations for O_3 , accumulation particle number, BC and RAD. In the following, we report two case studies, very different one from each other. In the first case, we observed a situation dominated by primary pollution, as testified by the high correlation between BC and accumulation particles: for this case, the correlation between O_3 and accumulation particle number during the convective hours was fairly poor ($r = 0.25$). During the second analyzed day, we observed higher correlation ($r = 0.97$) between O_3 and the accumulation fraction. For this case, we supposed that aged air-masses richer in secondary pollutants (i.e. O_3 and aerosol) have been mixed and transported to the measurement site within the afternoon mixed layer.

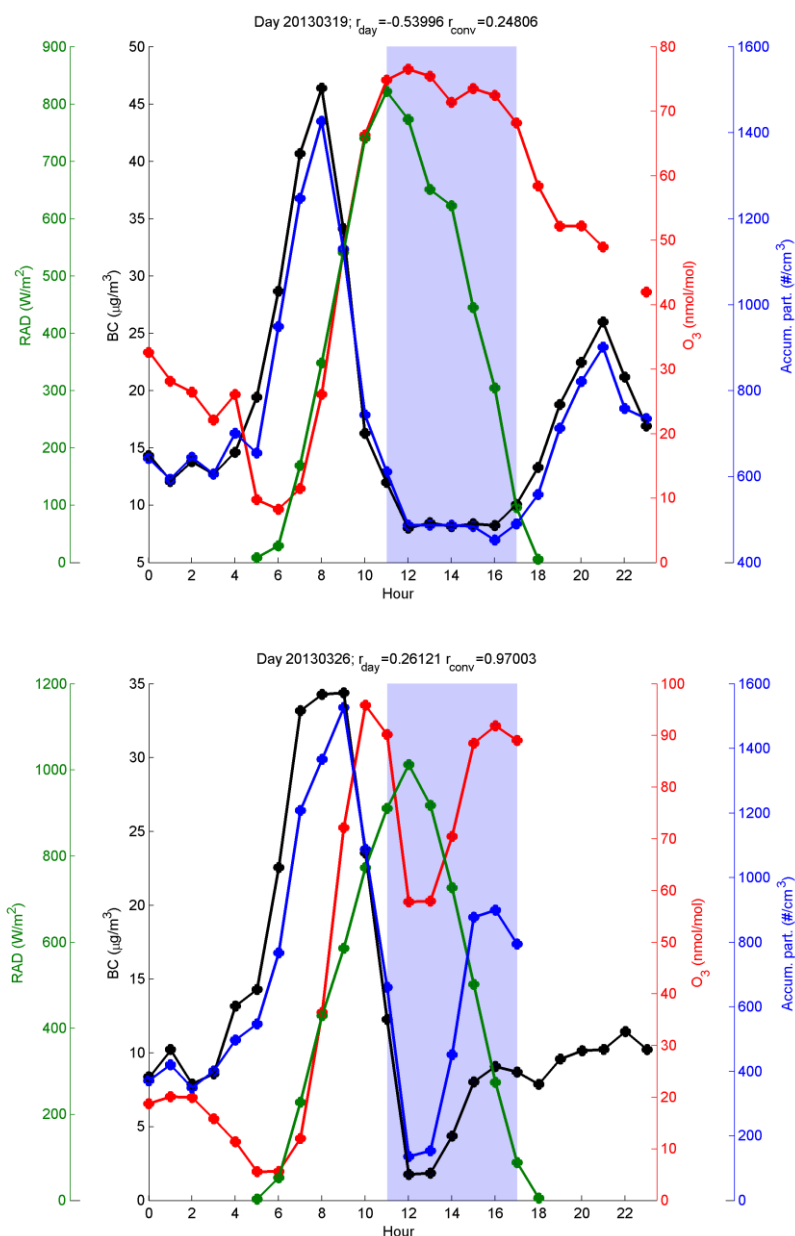


Figure R1. Diurnal variations for BC, O₃, accumulation particles and RAD, for two specific days: dominated by primary pollution (top) and likely influenced by other processes (bottom). Shaded areas represent the convective hours, i.e. between 11:00 and 17:00 LT.

The text was modified as follows (Page 22543, Line 23): “In order to distinguish the chemical effects from the boundary layer dynamics, we also computed correlation coefficients limiting the data to convective hours only (i.e. between 11:00 and 17:00, according to the wind speed and solar radiation diurnal variations). The slight weaker correlation between BC and accumulation particle number and, on the other hand, the increase in correlation between O₃ and accumulation particle number may indicate the role of other processes (e.g. secondary aerosol production) in the air-masses which characterize this specific time span (Table S1, Supplement). In particular, we suppose that aged air-masses rich in secondary pollutants (i.e. O₃ and aerosol) can be transported to the measurement site in the afternoon mixed layer.”.

General Comments:

The paper presents many numerical values of concentration in the text. These numbers would be much easier for a reader to find if they were instead reported in a Table. I realize that there is a diversity of time periods and meteorological conditions, such that the number of Table headings could be unwieldy. I would urge the authors to select for a Table as large a subset as makes sense for comparisons and reserve for the text, a discussion of comparisons, etc.

We decided to insert a new Table, encompassing average and standard deviation values of the pollutants for every season, to decrease the amount of numerical values in the text and enhance its readability. Therefore, most of these numerical values have been removed in Section 3.2, where lines are now referring to Table 2 (Table 2 of the previous version of the manuscript has now become Table 3). Moreover, a new sentence has been added (Page 22537, Line 6): "...the seasons, while seasonal average values are presented in Table 2."

Table 2. Average values (\pm standard deviation) of the pollutants, computed for the different seasons selected by the periods of Table 1.

	O ₃ (nmol/mol)	BC ($\mu\text{g}/\text{m}^3$)	Accum. ($\#/ \text{cm}^3$)	Coarse ($\#/ \text{cm}^3$)	PM ₁ ($\mu\text{g}/\text{m}^3$)	PM ₁₀ ($\mu\text{g}/\text{m}^3$)
Pre-monsoon	38.0 \pm 25.6	14.5 \pm 10.4	668 \pm 383	4.2 \pm 2.5	98 \pm 83	241 \pm 134
Monsoon	24.9 \pm 16.5	6.3 \pm 3.8	250 \pm 141	1.9 \pm 1.1	32 \pm 12	107 \pm 37
Post-monsoon	22.8 \pm 17.0	6.2 \pm 3.9	-	-	26 \pm 10	101 \pm 38
Winter	20.0 \pm 19.8	18.3 \pm 14.1	-	-	74 \pm 26	320 \pm 75
All	27.0 \pm 21.3	11.6 \pm 10.7	505 \pm 372	3.3 \pm 2.4	48 \pm 42	169 \pm 113

Page 22538, line 21-25. Ratio of daily to hourly standard deviations are an interesting quantity. However, I have not read Chevalier et al (2005) and don't know how to interpret these numbers other than the sweeping statement that daily and hourly variations are both important. A concern is that seasonal variations are clearly affecting BC and to a lesser extent ozone. A different way to look at data would be to use relative standard deviations that could be defined e.g., for the diurnal case by normalizing a day of measurements by the average of that day and for the daily case, normalizing by the average over a time period which could be a year or could be one of 4 periods defined or could be periods of defined length, such as a month.

By following Chevalier et al. (2007), the ratio daily/hourly variability aims at identifying what is the contribution of boundary-layer processes compared to that due to day-to-day changes (e.g. changing weather conditions, transport at synoptic scale) in determining O₃ and BC variations. Since no clear signal is obtained for what concerns O₃ and BC measurements (0.54 for O₃ and 0.59 for BC), we stated that both processes account the same in attributing these pollutants' variability. However, under the Reviewer's suggestion, we also computed relative standard deviations, i.e. defined by normalizing each hourly value by the daily average and each daily value by the seasonal average to which that day belongs. For BC, the relative standard deviations (daily/hourly) ratio is 0.54, thus almost equal to what obtained in the previous analysis; for O₃ it drops a bit more (0.42), further indicating the importance of diurnal photochemistry/local dynamics in determining O₃ variations at Kathmandu.

Regarding the conclusion that pollutants are mainly of local origin: This is further supported by the ratio of BC to PM1. That ratio is too high to be due to wildland fires.

We agree with the Reviewer concerning this point. A new sentence has been added (Page 22548, Line 3): "...at Paknajol, further supported by the high ratio BC/PM₁".

Significant figures: Aerosol concentrations are given to 0.1 particle per cm³ out of a total of hundreds to more than a thousand. Actual accuracy is of order 10%. The decimal digit should at least be removed for values greater than 100.

This has been corrected. All decimal digits for accumulation particles and for PM concentrations have been removed.

Term "correlation coefficient": It appears from the negative values in Table 2 that you have followed correct usage and are reporting r, not r². It would be useful to have an explicit statement, which could be done as simply as adding (Pearson's r) or even (r) at the point first mentioned.

The expression "(r)" has been added in the new version of the manuscript (Page 22542, Line 24) and in the caption of Table 3.

Specific Comments:

Page 22532 - 22533 Are instrument averaging times for instruments described in bullets 1 to 4 given in text?

All instruments described in bullets 1 to 4 in Section 2.1 sample at 1-minute time resolution. Measurements are validated on a 1-min basis and then averaged in order to obtain hourly values. This is stated in Page 22534, Lines 1-3.

page 22532, line 24 "These are referred to the SRP15 reference scale ..." please explain.
SRP15 stands for Standard Reference Photometer 15. It is the instrument used within WMO/GAW to propagate the NIST ozone measurement scale. For clarification, the manuscript has been modified and a new reference has been added (Page 22532, Line 24): "These are referred to the WMO/GAW reference scale (SRP#15, see Klausen et al., 2003) hosted at the GAW World Calibration Centre (WCC) at EMPA (Switzerland)".

Klausen, J, Zellweger, C, Buchmann, B, and Hofer, P, 2003. Uncertainty and bias of surface ozone measurements at selected Global Atmosphere Watch sites. Journal of Geophysical Research 108 (D19), 4622, doi:10.1029/2003JD003710.

page 22533, line 18 coincidence errors Are number concentrations high enough that coincidence errors are a concern? What was the maximum dilution factor used? Dilution flow rate is given but not sample flow rate.

The maximum concentration that avoids coincidence error is 500000 L⁻¹, as declared by the OPC manufacturer. The sample flow rate was kept constant at 1 L min⁻¹; the highest dilution factor used was 5, even though the most used was 4 (being the dilution flow often 3 L min⁻¹) that thus raises the coincidence error limit to 2·10⁶ L⁻¹. This limit was exceeded rarely: only few hours at the end of March and at the beginning of April 2013. Therefore, we can consider negligible its impact on the dataset.

page 22534, line 25 recurrent neural network How does a recurrent neural network take into account the multi-day effects of meteorology? There is a reference but a simple explanation would aid the reader.

The Elman network used in this work is composed by three layers: the input layer, the hidden layer and the output layer. In the Elman network some nodes of the input layer are set with the status of the neurons in the hidden layer, in this way the signal is backpropagated from the hidden layer to the input layer. The effect is that the neurons in the second layer contain information of parameters at the previous time step. This mechanism takes into account the multi-day effects of meteorology. This information is fully presented in Biancofiore et al. (2015).

page 22535, line 4 meteorological effects usually last for more than one day You are implicitly defining meteorological effects to exclude diurnal boundary layer cycles.

Yes, the Reviewer is partially right: there are boundary layer cycles but also meteorological conditions that last for more than a day. In our model architecture, both are taken into account. In the revised version of the manuscript we will highlight this point (Page 22535, Line 6): "...of meteorology, as well as diurnal boundary layer cycles".

page 22538, line 26-27. Fig 3 shows the shape of the ozone diurnal cycle was similar during all seasons. I disagree. The pre-monsoon ozone is clearly different in the late afternoon.

This sentence was meant to highlight the fact that the maxima and minima in the O₃ diurnal cycle occurred at the same moment of the day, independently on the season. The sentence has been rephrased: "O₃ diurnal variation is shown in panel b of Fig. 3: a peak in O₃ mixing ratios characterized the central part of the day (between 11:00 and 13:00), while a minimum was observed in the morning (between 5:00 and 6:00)."

page 22541, line 4 "We magnified ..." magnified is not the correct word. perhaps you call attention to something, but it is not made larger

The term "magnified" has been changed to "highlighted".

page 22543 line 9, direct pollution What is the meaning of direct in this context? Term not commonly used as a synonym of emitted or primary.

The term "direct" has been changed to "primary".

Page 22543 – 22544 Discussion of Fig. 6 hard to follow. Before I accept the conclusion that high ozone at 16:00 is due to dynamical effects (attributed to upper residual ozone in one place and residual ozone and/or horizontal advection in another place), I would want to see a diurnal cycle predicted using only RAD.

The different simulations reported in Fig. 6 show that, when in the model is used only wind speed as input data, the model simulates well only the afternoon high level of O₃, missing completely the peak before noon. In contrast, by modelling O₃ using as input parameters wind speed and solar radiation, the model reproduces well the peak before noon and the high levels of afternoon-evening O₃. Putting together the results of these two simulations, we can conclude that the high level of O₃ during the afternoon is mainly due to dynamics, for the following two reasons: (i) in the model, the wind speed used as input is enough to reproduce the afternoon concentrations of O₃ and (ii) the inclusion of solar radiation does not improve the agreement between measured

and modelled O₃ during the afternoon, but enhances substantially the agreement between measurements and simulations before noon, when photochemistry, as expected, plays a larger role.

This paragraph has been rephrased to make this part clearer (Page 22543-22544, Lines 27-29 and 1-9): “The simulation that included all the proxies reproduced quite well the observed O₃ mixing ratios for all hours of the day, whereas a simulation that included only wind speed (a good proxy of atmospheric dynamics) reproduced with accuracy the afternoon (after 15:00) and evening levels of O₃, missing completely the main O₃ peak before noon. In contrast, by using as input parameters both wind speed and solar radiation, the model reproduced well the peak before noon and the high levels of afternoon-evening O₃. Putting together the results of these two simulations, we can conclude that the high level of O₃ during the afternoon is mainly due to dynamics (vertical intrusion from upper atmospheric layers and/or horizontal advection), for the following two reasons: (i) in the model, the wind speed used as input is enough to reproduce the afternoon concentrations of O₃ and (ii) the inclusion of solar radiation does not improve the agreement between measured and modelled O₃ during the afternoon, but substantially enhances the agreement between measurements and simulations before noon, when photochemistry, as expected, plays a larger role.”.

page 22546 line 8 much polluted regions please rephrase

The word “much” was a misprint, thus has been removed.

Table 9 Time axis. Split into hard-to-visualize intervals. Suggest monthly or bi-monthly tics. A shading scheme in plot C that delineates 4 meteorological periods would help the reader follow text. *Figure 9 has been modified, by changing the x-axis using monthly ticks (same as Fig. 2). Moreover, in order to distinguish the different seasons, four shading areas have been added to panel c. Colors for these refer to the ones used in Figure 3. The caption has been modified accordingly.*

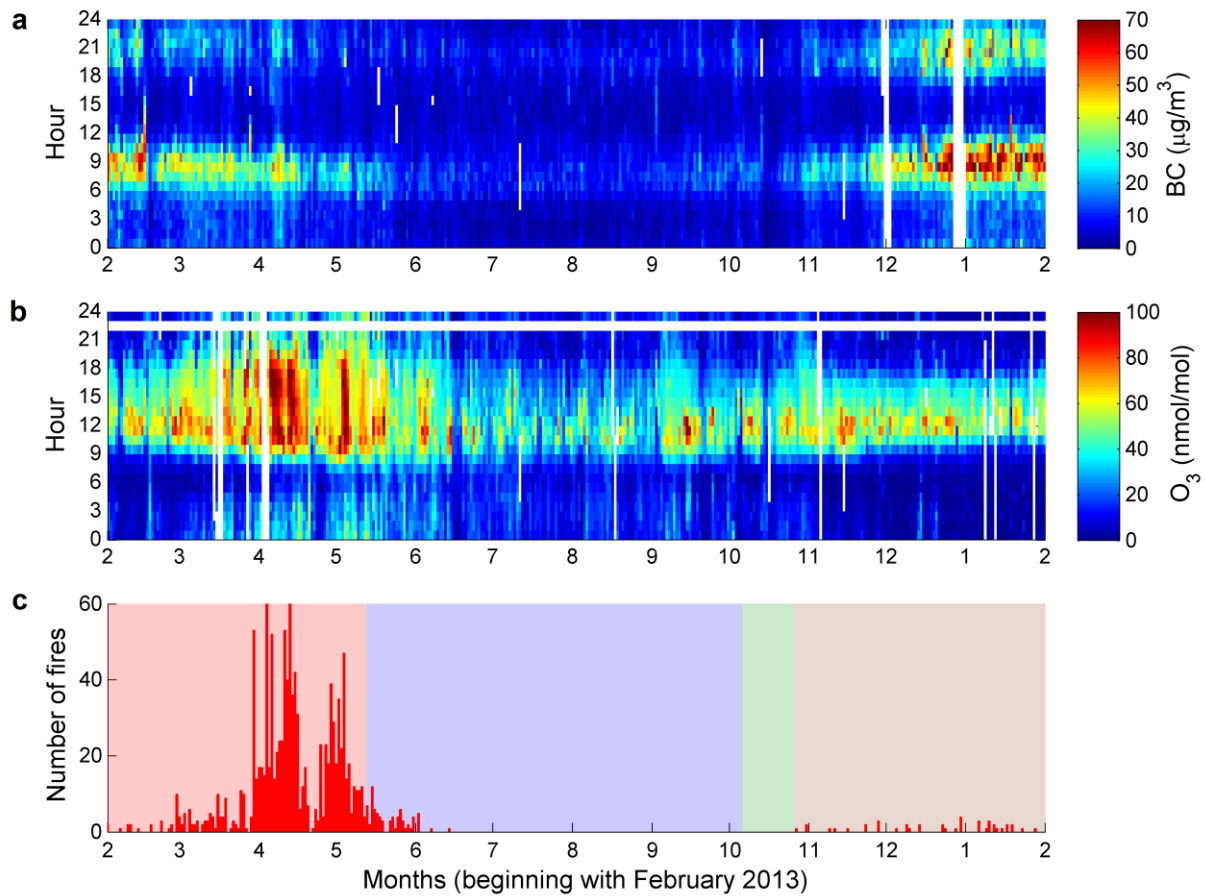


Figure 9. BC (panel a) and O_3 (panel b) diurnal variations over the entire sampling period. The color scale has been set to a maximum of $70 \mu\text{g}/\text{m}^3$ and $100 \text{ nmol}/\text{mol}$ for BC and O_3 , respectively. Panel c shows the total daily number of fires found in the Southern Himalayas box (see Putero et al., 2014); note that the y-axis has been limited to a maximum value of 60. Shaded areas in panel c indicate the different seasons (red: pre-monsoon, blue: monsoon, green: post-monsoon and brown: winter).