

Response to Referee comments

**Interactive comment on “Transport of regional pollutants through a remote trans-Himalayan valley in Nepal” by Shradda Dhungel et al.**

*Dear Dr. Stone,*

*We would like to thank the referees for their constructive comments, which have helped us to further improve the clarity and quality of the manuscript. Specifically, we have: 1) performed a substantial formatting and re-working of the manuscript tables and figures, 2) clarified the methods related the estimates of net daily flux, and 3) included additional cautionary statements regarding the conclusions that can (and cannot) be drawn from our study and findings. The responses to the referees are included below. Each referee comment is followed by our response to that comment (in italics). With these improvements incorporated into our manuscript, we now feel that our manuscript is now suitable for publication in ACP.*

*Thank you for your continued consideration,*

*Shradda Dhungel\*, Bhogendra Kathayat, Khadak Mahata, and Arnico Panday*

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**Referee #1**

This is my third review of this manuscript. I can say that the authors made a good work with this last revision. The manuscript presentation is significantly improved and the results are discussed in an appropriate and balanced way. Thus, I can recommend publication but some technical corrections must be considered.

Line 14-16: something appear to be missed in this sentence. Please, check. Maybe: "...to the higher Himalayas. Nevertheless, observational evidences of the transport of polluted air-masses across Himalayas has been lacking to date."

*We have clarified the text to read: “Models and satellite imagery suggest that strong wind systems within deep Himalayan valleys are major pathways by which pollutants from the IGP are transported to the higher Himalaya. However, observational evidence of the transport of polluted air masses through Himalayan valleys has been lacking to date.”*

Line 22-23, page 3: I do not agree: e.g. Bonasoni et al. (2010) showed clearly the effect of thermal wind circulation on the transport of pollutants along an Himalayan valley.

*We have added Bonasoni et al. 2010 reference.*

Line 33, Page 3: too much "previously"!

*We have removed the latter use of ‘previously’.*

Line 8-9, page 5: please check the wind sectors (are they inverted?)

*We have made the correction as indicated by the referee.*

Line 20-27, page 5: the formula is not clear to me. Why did you express wind speed in m/hr (and not in m/s)? Is not clear to me, the reason for using the factor 1/6. On which basis did you perform the summation? I suppose over time but please, explicitate it! Please, revise this formula and correct it accordingly....

*We have now included a  $\Delta t$  term (equal to 600 seconds) within the formula and ensured that the our time units are in seconds that we explicitly state how the summation was performed. The modified text now reads: "Up-valley and down-valley BC fluxes were calculated separately and determined using wind direction measurements. Instantaneous flux at time  $t$  was calculated as:*

$$j_t = BC_t ws_t$$

*where  $BC_t$  is the BC concentration ( $mg BC m^{-3}$ ) at time  $t$  and  $ws_t$  is the wind speed ( $m s^{-1}$ ) at time  $t$ . From this, net daily mass transport ( $M_d$ ) for day  $d$  was estimated as:*

$$M_d = A \left( \sum (j_{t,up} \Delta t) - \sum (j_{t,down} \Delta t) \right)$$

*where  $A$  is the cross-sectional area of the valley at Jomsom ( $1.41 \times 10^6 m^2$ ),  $j_{t,up}$  and  $j_{t,down}$  are the instantaneous up-valley and down-valley fluxes, respectively, occurring during day  $d$ , and  $\Delta t$  is the length of each time step in seconds (i.e., 600 seconds). The cross-sectional area was estimated as a trapezoid with a height of 800 m (the difference in elevation between the two Jomsom stations) and cross-valley distances of 800 m (at JSM\_1) and 2720 m (at JSM\_2). The average of these two cross-valley distances (1760 m) multiplied by the height (800 m) yields the cross-sectional area of  $1.41 \times 10^6 m^2$ . Thus, net daily flux was calculated as the difference between the summation of instantaneous up-valley fluxes for day  $d$  and the summation of instantaneous down-valley fluxes for day  $d$ ."*

Line 29, page 6: Fig 5b? It is correct?

*We have corrected the figure citation from Fig. 5b to Fig. 3b.*

Line 30, page 7: Maybe better Fig.6

*We have corrected the figure citation from Fig.5 to Fig. 6.*

Line 20, pag. 8: "(Fig. 4 and 6)"

*We have corrected the text as advised.*

Pag 10, line 5: only 1 number after decimal for O3.

*We have corrected the text as advised.*

## Referee #2

This is my first time to review this MS. I totally agree with comments from those previous referees. I find that numbers of problems still existed in the new version of the MS. I suggest the MS a major revision and the following are some more suggestions to the authors.

P3 Line 3 reference of (Kopacz et al., 2011) should be replaced by (Zhang et al., 2015) because the results of the former overlooked the potential contribution of local Tibet, which is out of date.

*We now also reference Zhang et al., 2015.*

2. P3 Line 31 “no pollution data from Jomsom have been previously reported previously”. The authors overlooked one just published work last year that has done 14C study of BC at Jomsom, which showed the influence of BC from South Asia to the Himalaya and the local biomass burning sourced BC to valleys of the Himalaya (Li et al., 2016). In addition, please add (Chen et al., 2017) and (Luthi et al., 2015), which are also two related article of this study.

*We have now included all three references as suggested.*

3. I am sorry to say that the figures and tables are not typical for both ACP and other scientific journals, which have been pointed out by other referees. However, the authors make little changes and still have lots of problems. For instance, you should at least add longitude and latitude to the figure 1 so that the readers know where your research site located. You also need to point out where is South Asia, where is the Tibetan Plateau at this figure.

*We have improved the clarity of the figure and its caption. We have included an inset in Panel (a) to show the location of the NASA image. We have also indicated the location of the Kali Gandaki Valley, the Indo-Gangetic Plain, the Himalaya, and the Tibetan Plateau. The updated caption now reads: “Figure 1. (a) NASA Worldview image from November 4<sup>th</sup>, 2014 depicting thick haze intruding the Himalayan foothills with red arrow over the KGV. Inset map in the top-right corner of panel (a) shows the location of the Worldview image. Dashed box shows the location of panel (b). (b) Expanded scale of the KGV showing station locations of Lete (LET\_AWS) near the entrance of the valley (yellow star); Marpha (MPH-AWS) in the core region (gray star); the Jomsom (JSM\_STA) sampling station for BC and O<sub>3</sub> and the two associated AWS sites (JSM\_1 and JSM\_2) in the core region (orange star); and Eklobhatti (EKL\_AWS) near the exit (blue star). (c) The atmospheric observatory at Jomsom (JSM\_STA) (28.87° N, 83.73° E, 2900 m asl). (d) Valley cross-sectional elevation profile at the station locations.”.*

Figure 2 looked a little strange and is useless. Instead, the words that point out four sites should be moved to right photo of figure 1. The table of Figure 2 should be moved to supporting information.

*We have removed Figure 2. The site information previously included therein has been moved to the figure caption of Figure 1, and the table has been moved to supporting information as the new Supplementary Table 1.*

The expression of tables is not typical, which should be “Three line table”. Please change the format of tables.

*We have updated all of the tables to be “three line tables”.*

Finally and honestly, the English of the article is not fluent, please improve it by expert English editor.

*The grammar and flow of the entire text has been thoroughly checked by multiple native English speakers. We leave it to the Editor to decide.*

### Referee #3

This is my third review of the manuscript. In comparison to my previous review the authors have partly addressed my three main points of concern although not necessarily in as much detail as I had hoped for. However, they have removed some more speculative parts of the discussion of the pollution transport towards the valley, so that this part is not hindering publication anymore. Nevertheless, I still have some further comments concerning the last additions and I make some additional recommendations concerning the presentation of figures and tables, which in my view are still not up to ACP standards. Once these issues are addressed publication in ACP could be considered.

#### Local influences

The authors have replied to my concerns about local pollution episodes by adding a sentence on the use of the local air strip which suggests that this will only have minor influences. However, the authors did not follow the suggestion of further screening their data based on local wind direction and location of sources, but argue that the vertical distance to the town is large enough to minimise the influence. Without further analysis and data this remains hard to judge, but there seems to be sufficient mention of local sources now to create the required cautiousness when interpreting the results.

*We agree that it is important to account for local sources and agree that the existing text conveys sufficient caution to the reader. The text in Section 3.4 not only cautions the reader that not all of the BC that we measure necessarily originates from the IGP but also explicitly lists the active local sources that may partially contribute to the diurnal patterns that we observe.*

#### Flux calculation

The authors added a whole section explaining the calculation of net upvalley BC fluxes and also explain the limitations of their approach. One of the main assumptions of the approach is that of constant depth of the upvalley or downvalley flows which is clear oversimplification of the flow especially during nighttime. The authors should add another sentence on the nighttime flow. It should be mentioned that their assumption is most likely an overestimation of the nighttime flow depth and, hence, nighttime downvalley flux. Therefore, their net upvalley flux is most likely an underestimation and at least the conclusion that there is net upvalley mass flux seems to be supported by the approach, whereas the absolute numbers should definitely be taken with caution. I would also suggest a rewrite of the equation of the daily net mass transport. Instead of using  $1/6$  in front of each term, it would be more logical to just multiply with  $\Delta t$  at the end of the equation. This would immediately make clear that we are looking at an integration over time. The value of  $\Delta t$  can then be given in the text.

*We have modified the equation as suggested and included a statement indicating how our choice of polluted boundary layer depth likely affects our daytime and nighttime flux estimates. The added text now reads: "Supplementary Figure 1 shows that JSM\_2 is well within the polluted haze layer during daytime/upvalley flows and that some BC is almost certainly transported above 800 m elevation above the valley floor. In addition, it is important to note that – by assuming a constant polluted boundary layer depth throughout the day – to a certain degree we likely underestimate up-valley flux during daytime and overestimate down-valley flux during nighttime. We therefore ensure that our estimates are conservative."*

## Trajectories

The previously criticised section on transport towards the valley mouth discussed by the analysis of air mass trajectories was removed from the manuscript. Therefore, my suggests for improving this kind of analysis were not followed, however, the manuscript does not draw any vague conclusions on this point anymore.

*While we agree with the referee that this would be important for better understanding the pollutant sources, we ultimately decided that such an analysis was beyond the scope of our study. Future work will investigate this in detail.*

## Presentation of figures and tables

Some of the figures and tables still need improvement before publication in ACP.

*We have improved the formatting of all tables based on the suggestions of the other referees. Figure 1 has been improved based on the suggestions of one of the other referees. The previous Figure 2 has been removed, and the information from its table has been moved to the supplementary materials. The figure showing daily net flux (now Figure 6) has also been improved.*

Figure 2 should be split into a figure and a table! I think I mentioned this before in an earlier review ...

*We have moved the information from the table to the supplementary materials (now Supplementary Table 1). The image has been removed, and the information on station location and name has been included in Figure 1.*

Table 1 looks more like a figure than a table. Please check if this agrees with ACP guidelines for tables!

*We have made the previous Table 1 into a Figure (now Figure 2).*

Figure 3: Maybe wind-roses are just not the best way to present the wind statistics in the case of such a clear bidirectional flow. Maybe a pdf of wind speed for 3 different wind sectors would be much clearer. The figure right now consists mostly of white space and the interesting information makes up only a very small fraction of the plot and is hard to see.

*We prefer to keep wind roses for this figure, as we feel they adequately convey the changes in wind speed and direction both throughout the day and between seasons. To minimize the space demanded by the figure, we have moved the wind roses for the Monsoon and Post-Monsoon periods to the supplementary materials, keeping only the wind roses for the Pre-Monsoon season in the main text.*

Figure 7: In general the figure quality could be improved by not squeezing everything together in the vertical. Make the figure less wide, but add some vertical space between individual panels. I would also suggest to use a different y-axis scale for the monsoon period. Like it is presented now one cannot even tell the magnitude of the fluxes.

*We have improved Figure 7 as suggested.*

# Transport of regional pollutants through a remote trans-Himalayan valley in Nepal

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**Abstract.** Anthropogenic emissions from the combustion of fossil fuels and biomass in Asia have increased in recent years.

High concentrations of reactive trace gases and light-absorbing and light-scattering particles from these sources ~~over the Indo-Gangetic Plain (IGP) of southern Asia~~ form persistent haze layers, also known as atmospheric brown clouds, ~~over the Indo-Gangetic Plain (IGP)~~ from December through early June. Models and satellite imagery suggest that strong wind systems within

deep Himalayan valleys are major pathways by which pollutants from the IGP are transported to the higher Himalaya. ~~However, Nevertheless, yet~~ observational evidence of ~~the transport of polluted air masses~~ through Himalayan valleys has

been lacking to date. To evaluate this pathway, we measured black carbon (BC), ozone (O<sub>3</sub>), and associated meteorological conditions within the Kali-Gandaki Valley (KGV), Nepal, from January 2013 to July 2015. BC and O<sub>3</sub> varied over both diurnal and seasonal cycles. Relative to nighttime, mean BC and O<sub>3</sub> concentrations within the valley were higher during daytime when

the up-valley flow (average velocity of 17 m s<sup>-1</sup>) dominated. BC and O<sub>3</sub> concentrations also varied seasonally with minima during the monsoon season (July to September). Concentrations of both species subsequently increased post-monsoon and peaked during March to May. Average concentrations for O<sub>3</sub> during ~~the seasonally representative months of~~ April, August, and November were 41.7 ppbv, 24.5 ppbv, and 29.4 ppbv, respectively, while the corresponding BC concentrations were 1.17

μg m<sup>-3</sup>, 0.24 μg m<sup>-3</sup>, and 1.01 μg m<sup>-3</sup>, respectively. Up-valley fluxes of BC were significantly greater than down-valley fluxes

during all seasons. In addition, frequent episodes of BC concentrations two to three times higher than average persisted from several days to a week during non-monsoon months. Our observations of increases in BC concentration and fluxes in the valley, particularly during pre-monsoon, ~~support the hypothesis~~ ~~provide evidence~~ that trans-Himalayan valleys are important conduits for transport of pollutants from the IGP to the higher Himalaya.

*Keywords: black carbon, ozone, trans-Himalayan valleys, pollutant pathways, long-range transport, regional transport episodes, short-lived climate forcers.*

## 1. Introduction

Persistent atmospheric haze, often referred to as Atmospheric Brown Cloud (ABC) (Ramanathan and Crutzen, 2003), affects broad geographic regions including the Indo-Gangetic plain (IGP) in southern Asia (Ramanathan and Carmichael, 2008), eastern China (Ma et al., 2010), southeast Asia (Engling and Gelencser, 2010), sub-Saharan Africa (Piketh et al., 1999), Mexico (Vasilyev et al., 1995), and Brazil (Kaufman et al., 1998). In southern Asia, the haze covers extensive areas particularly during the period of mid-November to mid-June preceding the summer monsoon season. Major combustion sources ~~– including – (primarily anthropogenic) – including~~ the anthropogenic burning of agricultural waste, garbage, biofuel, and fossil fuels as well as wildfires ~~– emit – emit~~ volatile and particulate-phase compounds to the atmosphere that contain oxidized and reduced forms of sulfur, nitrogen, and organic carbon (OC) together with elemental (black) carbon (BC) and other species. These emissions are intermixed and chemically interact with mechanically produced aerosols (e.g., sea salt and mineral dust). Important secondary pollutants such as ozone (O<sub>3</sub>) also form from photochemical reactions involving nitrogen oxides, ~~and volatile organic species are also produced.~~ Together, this mixture of atmospheric species constitutes the ABC or ~~the brown haze~~ over South Asia (Ramanathan et al., 2005; Gustafsson et al., 2009). These optically thick layers include high concentrations of light-absorbing and light-scattering particles (Menon et al., 2002) that modulate radiative transfer. Light-absorbing aerosols (primarily BC and crustal dust) contribute to warming of the atmospheric warming while light-scattering aerosols (primarily S-, N-, and OC-dominated particles) drive cooling at the surface. The combined effects of light-absorbing and light-scattering aerosols from anthropogenic sources serve to reduce UV and visible wavelength radiation at the surface (i.e., surface forcing), increase ~~the warming of the tropospheric warming~~ (i.e., atmospheric forcing), and change the net top of the atmosphere solar flux (i.e., top-of-the-atmosphere forcing) (Andreae and Crutzen, 1997; Kaufman et al., 2002, Ramanathan et al., 2005). Light-absorbing anthropogenic pollutants like BC also significantly influence global warming, in terms of direct radiative forcing (Jacobson, 2001; Bond et al., 2013), and; regional influences from such pollutants close to sources are greater than those on the global scale (Ramanathan et al., 2007b).

The elevated concentrations of aerosols in the anti-cyclone have also been shown to weakens ~~the~~ circulation patterns and reduces total monsoon precipitation over southern India (Ramanathan et al., 2005; Fadnavis et al., 2013) while intensifying the monsoon over the foothills of the Himalaya (Lau et al., 2006). In addition to warming the atmosphere, the rising concentrations of BC and O<sub>3</sub> over southern Asia (e.g., Ramanathan and Carmichael, 2008) have detrimental impacts on human health. ~~The effects~~ Recent work has shown that elevated levels of these two key pollutants can compromise BC on cardiopulmonary and respiratory ~~problems health are greater than those of PM 2.5 or 10 particles (Krupnick et. al. 1990;~~ Janssen et al., 2011). In addition, O<sub>3</sub> ~~also compromises pulmonary function (Krupnick et. al, 1990)~~ and is a leading pollutant contributing causing to biodiversity loss (Royal Society, 2008) as well as declining in crop yields by directly damaging leaves (Auffhammer et al., 2006).

~~Most H~~haze over the IGP often reaches heights of more than 3 km above sea level via convection and advection, and the Himalaya range forms a 2500 km long, 8 km high complex topographic barrier along the northern edge of the IGP (Singh et

al., 2004; Dey and Di Girolamo, 2010; Gautam et al., 2011, [Lüthi et al., 2015](#)). Numerous studies have investigated the transport of pollutants from the IGP to the Himalayan foothills, the immediate source region for potential transport to the Tibetan Plateau (TP) (Pant et al., 2006; Dumka et al., 2008; Komppula et al., 2009; Hyvärinen et al., 2009; Ram et al., 2010; Brun et al., 2011; Gautam et al., 2011; Srivastava et al., 2012). Further, a suite of studies involving satellite imagery (Ramanathan et al., 2007a, Brun et al., 2011), back trajectories (Lu et al., 2011), model calculations (Kopacz et al., 2011; [Zhang et al., 2015](#)), ice core analyses (Lee et al., 2008, Kang et al., 2010), and measurements in the higher Himalaya (Bonasoni et al., 2010, [Chen et al., 2017](#)) strongly suggest that pollutants are efficiently transported from the IGP to the higher Himalaya and onto the Tibetan Plateau, especially during spring prior to the monsoon. ~~This transport north of the Himalayas is potentially concerning as the TP plays a vital role in regulating the regional climate due to its effect on the Asian summer monsoon (ASM) and the hydrologic cycle. In the TP, light-absorbing aerosols may not only serve warm the atmosphere at high altitudes but – when and, when deposited onto snow and ice surfaces – may also, decrease albedo and thereby substantially increasing the melting rates of snow and glaciers and snow melting (Kang et al., 2010). Indeed, Model simulations (Qian et al., 2011) have shown that these light-absorbing aerosols change the surface radiative flux in the higher Himalaya and the TP by 5 to 25 W m<sup>-2</sup> during the pre-monsoon months of April and May. The TP plays a vital role in regulating the regional climate due to its effect on the Asian summer monsoon (ASM) and the hydrologic cycle.~~ The interrelated perturbations of the ABC on radiative transfer, air quality, the hydrologic cycle, and crop yields have important long-term implications for human health, food security, and economic activity over southern Asia. However, to date, there has been little observational research to directly demonstrate the role of mountain valleys in the transport of air pollutants from the IGP and the Himalayan foothills to higher elevations within and north of the Himalayan range.

In other studied mountain ranges, it is well known that flow patterns carry polluted air masses up valleys to higher elevations by providing a path of least resistance between tall mountains. In the European Alps, prevailing wind systems in the mountain river valleys funnel polluted air from peripheral source regions to high elevations in a phenomenon known as “Alpine Pumping” (Weissmann et al., 2005). Under fair weather conditions during daytime, the upslope winds are capable of transporting significant pollutants and moisture into the free troposphere (Henne et al., 2004). Relative to air over plains, the air within the valley heats and cools more quickly (Steinacker, 1984). The resultant differences in temperature create gradients in pressure and density, which in turn drive the transport of air from the plains to higher elevations during the daytime (Reiter and Tang 1984, Whiteman and Bian, 1998; Egger et al., 2000; [Bonasoni et al., 2010](#)). Such direct evidence of a Himalayan mountain valley wind system and its role in pollution transport has yet to be observed. Here we present 2.5 years of measurements of BC, O<sub>3</sub>, and associated meteorological data from one of the deepest trans-Himalayan valleys, the Kali-Gandaki Valley. We examine seasonal and diurnal patterns of BC and O<sub>3</sub>, investigate potential episodes of enhanced pollution transport up-valley, and make a preliminary estimate of BC mass transport. In doing so, we seek to provide the first observational evidence of trans-Himalayan valleys acting as conduits for pollution transport from the IGP to the higher Himalaya.



## 2. Measurement Sites and Methods

### 2.1 Measurement Sites and Instrumentation

This paper presents data from an atmospheric measurement station in Jomsom (28.87° N, 83.73° E, 2900 m asl) within the core region of the KGV along with four other automated weather stations up and down the valley from Jomsom (Figure 1). With the exception of aerosol optical depth measured as part of AERONET (AErosol RObotic NETwork) (Xu et al., 2015) and a <sup>14</sup>C study at several sites in the Himalaya and TP, including Jomsom, (Li et al., 2016); no pollution data from Jomsom have previously been reported. Here we report diurnal and seasonal trends in two important SLCPs – BC and O<sub>3</sub> – to evaluate the role of trans-Himalayan valleys as pathways for the transport of polluted air from the IGP to the higher Himalaya.

The KGV is located in the Dhaulagiri zone of western Nepal (Fig. 1(a)). The KGV floor changes elevation from approximately 1100 m to 4000 m above sea level (asl) over a horizontal distance of 90 km (Fig. 1(b)). Passing between the two eight thousand meter peaks of Dhaulagiri and Annapurna, it forms one of the deepest valleys in the world. The valley is a narrow gorge at the lower end and opens up into a wider, arid basin (Fig. 1(b)) with a maximum width of approximately one kilometer. The orientation of the KGV varies from the entrance to the exit. The general orientation of the valley is from SW (the mouth of the valley) to NE (the head of the valley) (Fig. 1). Approximately 13,000 inhabitants in several small settlements sparsely populate the valley. Emission sources within the valley include biofuel combustion for cooking and fossil-fuel combustion by off-road vehicles. There is an airport in Jomsom which operates only in the morning, as the narrow valley width and high up-valley wind speeds make it dangerous for small aircraft to land at other times during the day. A total of 5245 vehicles have been registered in Dhaulagiri Zone since 2008, but most are based in the southern towns of Kusma, Baglung, and Beni, below 1 km altitude, at the lowest left corner of the map in Figure 1(b) (DOTM, 2016).

The atmospheric observatory at Jomsom (JSM\_STA) is equipped with instruments to measure BC, O<sub>3</sub>, and meteorology (Figure 2 Supplementary Table 1). The observatory is located on the southeast corner of a plateau jutting out from an east-facing slope about 100 m above the valley floor and with no major obstructions either up or down the valley. Equivalent black carbon (hereafter referred to as BC) was measured with a Thermo Multiangle Absorption photometer (MAAP), model 5012 that uses a multi-angle photometer to analyze the modification of radiation fields – as caused by deposited particles that entered through a straight, vertical inlet line – in the forward and back hemisphere of a glass-fiber filter (GF-10). MAAP was operated at a flow rate of 20 L min<sup>-1</sup> and measured BC at a 1-minute frequency. We note that Hyvärinen (2013) illustrates the artifact in MAAP measurements in environments with high aerosol loading with an underestimation of concentrations above 9 µg m<sup>-3</sup>. Since the median monthly concentrations for the duration of the measurement were less than 1 µg m<sup>-3</sup> and 90<sup>th</sup> percentile measurements were below 2 µg m<sup>-3</sup> (and therefore below this threshold), MAAP corrections were not applied. O<sub>3</sub> was measured with a 2B Tech model 205 via the attenuation of ultraviolet light at 254 nm passing through a 15 cm long absorption cell fitted with a quartz windows. The instrument was operated at a flow rate of 1.8 L min<sup>-1</sup>. For instrument calibration, the BC instrument performed an automatic span and zero checks every 24 hours, while zero checks

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on the O<sub>3</sub> instrument were performed every 7 days. Wind speed and direction were measured by an automated weather station (JSM\_2) installed on a ridge 8900 m (JSM\_2) above the sampling site for BC and O<sub>3</sub> (JSM\_1).

## 2.2 Data summary

135 The observatory operated from January 2013 through July 2015, but periodic power disruptions caused occasional data gaps (Figure 2). Unless otherwise noted, data reported herein correspond to periods when BC, O<sub>3</sub>, and meteorological data were available simultaneously. Data were binned by season as follows: monsoon (July-September), post-monsoon (October-February) and pre-monsoon (March-June). Time of days corresponds to Nepal's local time zone (LT) (UTC + 5.75 h). From March 2015 to May 2015, four additional automated weather stations (10m in height) were operated along a longitudinal  
140 transect roughly 10 meters above and in the center of the valley floor where wind speeds are typically the highest: near the entrance of the valley at Lete (LET), within the core at Marpha (MPH) and Jomsom (JSM\_2), and near the valley exit at Eklobhatti (EKL) (Figure 1b). Power outages, instrument malfunctions, and a major earthquake in Nepal on April 25<sup>th</sup>, 2015 (and its aftershocks) limited the duration of records at all sites. However, between 1<sup>st</sup> and 14<sup>th</sup> May, all stations operated simultaneously, and the resulting data provide information with which were used to evaluate the diurnal variability of wind  
145 fields along the valley.

BC, O<sub>3</sub>, and meteorological data were averaged over 10 minute intervals. Up-valley (southwesterly) flows are defined as between 215° and 235° 35° and 55° while down-valley (northeasterly) flows include data between 35° and 235° 55°. Data for all days for which complete data were available over entire 24-hour periods were binned by season. The statistical significance of differences between up-valley and down-valley flow conditions during different seasons were evaluated  
150 using the non-parametric Kruskal Wallis and Mann-Whitney tests.

To normalize for the influence of day-to-day variability in absolute concentrations, relative diurnal variability in O<sub>3</sub> and BC concentrations measured during a given month were normalized to a common scale ranging from 0 to 1 (see e.g., Sander et al., 2003; Fischer et al., 2006). Normalized BC values for month  $m$  were calculated as:

$$BC_{n,t} = \frac{BC_t - \min BC_m}{\max BC_m - \min BC_m}$$

155 where  $BC_t$  is the BC concentration at time  $t$  in the month  $m$ , and  $\max BC_m$  and  $\min BC_m$  are the maximum and minimum BC concentrations observed during month  $m$ . The normalized data were then binned into twenty-four, hourly increments. These calculations were repeated for O<sub>3</sub>.

## 2.3 Preliminary flux estimates

Up-valley and down-valley BC fluxes were calculated separately and determined using wind direction measurements.

160 Instantaneous flux at time  $t$  was calculated as:

$$j_t = BC_t ws_t$$

where  $BC_t$  is the BC concentration ( $\text{mg BC m}^{-3}$ ) at time  $t$  and  $ws_t$  is the wind speed ( $\text{m s}^{-1}$ ) at time  $t$ . From this, net daily mass transport ( $M_d$ ) for day  $d$  was estimated as:

$$M_d = A \left( \sum \left( \frac{1}{6} j_{t,up} \Delta t \right) - \sum \left( \frac{1}{6} j_{t,down} \Delta t \right) \right)$$

where  $A$  is the cross-sectional area of the valley at Jomsom ( $1.41 \times 10^6 \text{ m}^2$ ),  $j_{t,up}$  and  $j_{t,down}$  are the instantaneous up-valley and down-valley fluxes, respectively, occurring during day  $d$ , and  $\Delta t$  is the length of each time step in seconds (i.e., 10 minutes divided by 60 minutes/600 seconds). The cross-sectional area was estimated as a trapezoid with a height of 800 m (the difference in elevation between the two Jomsom stations) and cross-valley distances of 800 m (at JSM\_1) and 2720 m (at JSM\_2). The average of these two cross-valley distances (1760 m) multiplied by the height (800 m) yields the cross-sectional area of  $1.41 \times 10^6 \text{ m}^2$ . Thus, net daily flux was calculated as the difference between the summation of instantaneous up-valley fluxes for day  $d$  and the summation of instantaneous down-valley fluxes for day  $d$ .

If we assume that (1) the polluted boundary layer within the valley at Jomsom is 800 m deep (i.e., the approximate elevational difference between the two AWS sites at Jomsom), (2) BC within the polluted boundary layer is well mixed, and (3) wind velocities do not vary significantly with altitude through the polluted layer, the mass flux of BC through a vertical plane across the valley can be estimated. Supplementary Figure 1 shows that JSM\_2 is well within the polluted haze layer during daytime/upvalley flows and that some BC is almost certainly transported above 800 m elevation above the valley floor. In addition, it is important to note that – by assuming a constant polluted boundary layer depth throughout the day – to a certain degree we likely underestimate up-valley flux during daytime and overestimate down-valley flux during nighttime. In this way we therefore ensure that our estimates are conservative. The long atmospheric lifetime of particulate BC against deposition (several days to a week or more) – coupled with turbulent flow within the valley – supports the assumption that BC is well mixed. Supplementary figure 2 shows that the potential temperature ( $\theta$ ) gradient is less than zero between JSM\_1 and JSM\_2 which illustrates that the air within the valley is unstable. In addition, Egger et al. (2000) used theodolite measurements to demonstrate uniform wind speeds within the bottom 1000 m above the KGV floor at Jomsom and other locations. While we observed differences in wind speed of  $\sim 5 \text{ m s}^{-1}$  between the two Jomsom stations during the limited times when data were available from both, we were unable to determine whether this pattern persisted throughout the year. These data limitations also prevented us from a more in-depth assessment of potential nighttime decoupling. For these reasons, our only option was to follow the findings of Egger et al. (2000) and assign the wind speed at JSM\_2 to the entire flux plane.

### 3. Results and Discussion

#### 3.1 Evolution of local wind systems in the KGV

An understanding of the local wind regime is essential for analyzing pollution transport through mountain valleys.

Measurements from JSM\_2 show the diurnal evolution of wind at Jomsom in each season. ~~All data collected were used to analyze the diurnal pattern of wind at Jomsom (Figure 3-3a2a, Supplementary Figure 4-3b2b, 3e2e). Wind roses illustrate the temporal evolution of up- and down-valley flows at JSM\_2 for each season.~~ At JSM\_2, up-valley flows are southwesterly and dominant during daytime, with peak velocities above  $15 \text{ m s}^{-1}$  between 0900 LT to 1800 LT. Wind velocities decreased substantially after 1800 LT, with variable wind direction until midnight, followed by northeasterly winds (during pre- and post-monsoon seasons (Figure 3-3a2a and 3e2eSupplementary Figure 4b)). The wind patterns during monsoon appear strongly influenced by the monsoon anticyclone, ~~an~~ this observation ~~which~~ is in agreement with wind direction measurements from other Himalayan valleys (Bonasoni et al., 2010; Ueno et al., 2008) (Fig-5Supplementary Figure 4a2b).

195  
200 Although wind velocities at JSM\_2 varied over the year, non-monsoon months exhibited similar diurnal patterns ~~that evolved seasonally~~ as a function of sunrise and sunset (Figure 3-3a2a, 3b2b, 3e2eand Supplementary Figure 4); ~~characteristics which were exhibited in the monsoon season.~~ As discussed below, this alternating pattern in wind direction from strong daytime flows to weak nighttime flows during dry months ~~results~~ in a net transport of pollutants up the valley.

205 Our measurements at the four AWS stations on the valley floor illustrated the evolution of surface wind velocities along the length of the KGV (Supplementary Figure 3). In general, wind speeds along the valley floor were strongest within the core of the valley at MPH, JSM\_1 and JSM\_2 and were ~~lower-weaker~~ in the entrance (LET) and exit (EKL) regions (Supplementary Figure 3). The ~~observation of these duration of~~ strong ~~daytime~~ wind speeds within the valley during ~~daytime~~ is consistent with the hypothesis that wind patterns are modulated by ~~the a~~ pressure gradient created ~~as a result offrom the~~ differential heating of the arid valley floor relative to the mouth of the valley (Egger et al, 2000). In addition, ~~our~~ comparison  
210 of measurements at JSM\_1 and JSM\_2 (Figure 3-3a2a, and Supplementary Figures 3, 4a, and 4b) provided information regarding vertical variability in wind speed. Velocities at the higher elevation site of JSM\_2 were about  $5 \text{ m s}^{-1}$  and  $3 \text{ m s}^{-1}$  greater than those near the valley floor during daytime and nighttime, respectively. -The two sites exhibited similar diurnal cycles with the exception of a relatively stronger northeasterly wind at JSM\_2 from 0300 to 0900 LST.

### 3.2 Diurnal variability in BC and O<sub>3</sub>

215 Based on median values, O<sub>3</sub> peaked during daytime and dropped to minimal levels before sunrise during all three study seasons (pre-monsoon, monsoon, and post-monsoon). However, in April 2013 (pre-monsoon period), O<sub>3</sub> peaked in the late afternoon whereas in November 2014 (post-monsoon), it peaked in the early afternoon. In addition, the normalized diurnal excursions were greater during the pre- and post-monsoon periods relative to the monsoon period – represented by August 2014. In contrast, BC concentrations increased rapidly in the early morning, decreased during late morning, and then rose  
220 again ~~through during~~ the afternoon and early evening hours (Figure 4-43); ~~this pattern was consistent across all seasons, with R~~ relative diurnal variability ~~was being~~ somewhat greater during post-monsoon relative to the pre-monsoon and monsoon periods. Across all seasons, the lower normalized distributions for BC relative to O<sub>3</sub> reflect infrequent periods of high BC concentrations.

Several factors likely contributed to differences in timing of the daily peaks in O<sub>3</sub> and BC concentrations. These include potentially different source regions for BC and O<sub>3</sub> precursors, the timing of the photochemical production and destruction of O<sub>3</sub>, and contributions of O<sub>3</sub> and its precursors from non-combustion sources like stratospheric ozone and biogenic hydrocarbons from vegetation. The early morning BC peak during all seasons suggests contributions from the local combustion of biofuels for cooking and heating, which are most prevalent during early morning. The secondary peak in the afternoon and early evening occurs when the local anthropogenic emission sources are at a minimum in the KGV.

### 3.3 Seasonal variability in BC and O<sub>3</sub>

All data generated during the measurement period were binned by month to evaluate the seasonal patterns of BC and O<sub>3</sub> (Figure 5-54). In addition, individual months with the most complete data coverage during the pre-monsoon (April 2013), monsoon (August 2014), and post-monsoon (November 2014) seasons were selected to evaluate aspects of temporal variability in greater detail (Figure 6-55). We divided the seasons into dry (pre-monsoon, post-monsoon) and wet (monsoon) seasons to understand the transport of pollutants via Himalayan valleys in the presence and absence of wet deposition processes. Based on median values, the highest concentrations of both species occurred during the months preceding the monsoon and the lowest were during months of the monsoon. The significantly lower concentrations of both species during the monsoon reflected the combined influences of synoptic easterly airflow that transports a cleaner marine air mass over the region, reduced agricultural residue burning (Sarangi et al., 2014), and more efficient aerosol removal via wet deposition (Dumka et al., 2010). In addition to the possible active wet deposition of O<sub>3</sub> precursors during the monsoon season, increased cloudiness during monsoon may also reduce O<sub>3</sub> production (Lawrence and Lelieveld, 2010).

The post-monsoon timing of peak BC concentrations observed in previous studies performed in the IGP and Himalayan foothills (see e.g., Tripathi et al, 2005; 2007; Ramchandran et al., 2007; Putero et al, 2015) differs from our observations in the KGV, where we see heightened BC during the pre-monsoon season (Figure 5-54). These findings are generally in agreement with other high altitude observations. For example, the Nepal Climate Observatory-Pyramid (NCO-P) station at the 5079 m asl in the Himalaya has also shown high seasonal differences for BC and O<sub>3</sub> between pre-monsoon (0.444 (±0.443) µg m<sup>-3</sup> BC; 61 (±9) ppbv O<sub>3</sub>) and monsoon (0.064 (±0.101) µg m<sup>-3</sup> BC; 39 (±10) ppbv O<sub>3</sub>) (Cristofanelli et al., 2010, Marinoni et al., 2013) (Table 1). Our results therefore indicate a lagged peak in BC and O<sub>3</sub> within the KGV and presumably other deep Himalayan valleys, as compared to sites within the IGP.

### 3.4 Local winds as drivers of BC and O<sub>3</sub> transport in the KGV

Figure 6-5 shows the time series of BC and O<sub>3</sub> during an individual month in each season (April/pre-monsoon, August/monsoon, November/post-monsoon). For April, BC concentrations peaked at 0700 LST (Figure 6-65) when wind velocities were low (Figure 6-65). This peak occurred about an hour later in November, with both periods experiencing a decrease in BC over the rest of the morning as wind speeds increased and diluted local emissions (Figure 4-43). Thereafter, BC concentrations increased over the afternoon and early night, reaching secondary peaks near midnight LST in April and

several hours earlier in November and August (Figures 4 and 6-4-3 and 6-5). Distinct morning and afternoon peaks in BC concentration ~~were~~ seen in the post-monsoon season when the up-valley wind speeds ~~were~~ relatively weaker than in pre-monsoon season (Figures 4 and 6-4-3 and 6-5). The bimodal diurnal distribution of BC concentration in Jomsom ~~was~~ similar to that observed in Kathmandu (Putero et al. 2015) but unlike a singular late afternoon/evening peak seen at high elevation sites (Bonasoni et al., 2010) during non-monsoonal seasons. This illustrates that deep Himalayan valleys are susceptible to diurnal pollution similar to that of urban areas like Kathmandu. The morning peak in BC ~~was~~ most likely due to local pollutants (from household and morning aircraft traffic) in Jomsom and settlements downwind of Jomsom, while the afternoon peak ~~was~~ likely primarily associated with long range ~~transport~~ in addition to local pollutants. At the same time, O<sub>3</sub> exhibited ~~a~~ distinct minimum in the early morning with concentrations increasing towards an early afternoon peak – occurring well before BC's afternoon peak. The O<sub>3</sub> minimum in the morning ~~provided~~ further ~~evidencesupports~~ that the morning BC peak originated ~~s~~ from local sources, as O<sub>3</sub> is only formed downwind of pollution sources. Further, the Jomsom station measuring BC and O<sub>3</sub> (JSM\_STA) ~~was~~ located more than 100 m above the valley floor – ~~where -where~~ the village of Jomsom sits. As such, we do not expect that local evening emissions would ~~have reached~~ the stations at the cessation of up-valley flows and that evening drainage flows, following the valley floor, would remove these local evening emissions down-valley.

Percentage distributions of up-valley and down-valley BC concentrations, fluxes, and net daily fluxes are depicted in Figure 7-6 and summarized in Supplementary Table 2-1. ~~The differences in Up-valley and down-valley BC concentrations between up-valley and down-valley flows were quite similar – with down-valley concentrations being slightly higher – but were found to be~~ statistically significantly different for all seasons, ~~with higher down-valley concentrations~~ (Figure 7a7a6a). However, because wind velocities were relatively ~~higher-stronger~~ during up-valley daytime flows and the durations of up-valley flows ~~was~~ ~~ere~~ ~~modestly~~ longer than those of down-valley flow, the corresponding up-valley fluxes of BC during daytime were markedly and significantly greater than down-valley fluxes, ~~during across~~ all seasons (Figure- 7b7b6b). These results suggest an oscillatory movement of polluted air within the valley, where polluted air masses are pushed up-valley during daytime and retreat a shorter distance during nighttime. These differences between up-valley and down-valley fluxes yielded significant net daily up-valley fluxes of BC during all seasons (Figure- 7c6e). Because heating would have driven growth of the boundary layer – ~~thereby enhancing the and thus greater~~ ventilation and dilution of pollutants during daytime relative to night – ~~we, we~~ infer that the calculated differences ~~in-between~~ up-valley ~~versus and~~ down-valley fluxes correspond to ~~the~~ lower limits for net BC fluxes.

Positive up-valley fluxes ~~were~~ consistent with an “alpine pumping” mechanism in the Himalayan valleys and thereby support the hypothesis that these valleys are important pathways for pollution transport. In addition, we estimated ~~d~~ substantial net daily mass transport of BC up the valley during the pre-monsoon season: 1.05 kg day<sup>-1</sup> (based on the average net daily flux) and 0.72 kg day<sup>-1</sup> (based on the median net daily flux). While preliminary, these estimates provide the first semi-quantitative constraints on ~~the~~ mass transport of BC from the IGP to the high Himalaya through deep valleys, ~~and,~~

More generally, these fluxes have important implications for the regional cycling of BC over southern Asia.

### 290 3.5 Evidence of regional transport episodes in valley concentration

Along with the regular diurnal and seasonal variability driven by local winds as described above, we also observed anomalous periods – lasting several days to more than a week – during which BC concentrations were significantly greater than the 90<sup>th</sup> percentile for corresponding annual averages (Table 2). These extended periods of high BC at JSM\_STA were evidence of large-scale transport from the IGP to the Himalayan foothills in conjunction with local valley winds (Figure 8-87). Elevated O<sub>3</sub> concentrations did not always accompany these long-duration periods of high BC concentrations above 90<sup>th</sup> percentile. Different atmospheric lifetimes, chemical reactivity, source location, and sinks of BC and O<sub>3</sub> may have contributed to these differences in BC and O<sub>3</sub> concentrations detected in the valley. Table 2 reports the number of days in which O<sub>3</sub> concentration was above 90<sup>th</sup> percentile within each episode.

We partitioned these episodes into three characteristic patterns based on the relative variability of BC. Pattern A was characterized as a fluctuating daily maximum in BC with peaks that repeatedly exceeded the 90<sup>th</sup> percentile but with daily minima below the 90<sup>th</sup> percentile (Figure 8-87[1a]). Pattern B was characterized by a steady buildup of BC concentration over the period of the regional transport episode, with peak BC concentrations over the 90<sup>th</sup> percentile but without a low daily minimum (Figure 8-87b). Pattern C exhibited a combination of both Patterns A and B during a single regional episode (Figure 8-87c). A total of 34 regional episodes were identified from January 2013 through June 2015, 47% of which were categorized as Pattern A, 32% as Pattern B, and 21% as Pattern C. The wind speeds at Jomsom during these transport episodes exhibited diurnal variability similar to those during other periods (Figure 8-78[II]). During the regional transport period in November 2014 (Pattern A), average daily BC concentration was 1.29-3 μg m<sup>-3</sup> which is over the 75<sup>th</sup> percentile (0.88-9 μg m<sup>-3</sup>) of the BC concentration for the measurement duration. The maximum daily concentration during the period was 3.04-1 μg m<sup>-3</sup>. However, the corresponding average O<sub>3</sub> was only 28.07-1 ppbv, slightly below the average (29.48-5 ppbv) for the entire data set (Figure 8-78-[1a]). The mean BC concentration during a May 2014 pattern-Pattern B transport episode, one example of which occurred in May 2014, was 1.77-8 μg m<sup>-3</sup> (Figure 8-87[1b]). It was above the 90<sup>th</sup> percentile (1.49-5 μg m<sup>-3</sup>) for the entire measurement period while O<sub>3</sub> concentrations were at 49.74 ppbv, slightly below the 90<sup>th</sup> percentile (52.9 ppbv). One of the Pattern C- type transport episodes was identified in May 2013, when the average concentration was well above the 90<sup>th</sup> percentile for both BC (2.09-1 μg m<sup>-3</sup>) and O<sub>3</sub> (57.549 ppbv) (Figure- 8-87[1c]). The diurnal wind pattern in the KGV was conserved during the Pattern A example, but a longer period of up-valley flows occurred during the examples for Patterns B and C (Figure- 8-87).

### 4. Conclusion

This study provides new in-situ observational evidence of the role of a major Himalayan valley as an important pathway for transporting air pollutants from the IGP to the higher Himalaya. We found that concentrations of BC and O<sub>3</sub> in the KGV exhibited systematic diurnal and seasonal variability. The diurnal pattern of BC concentrations during the pre- and post-

monsoon seasons were modulated by the pulsed nature of up-valley and down-valley flows. Seasonally, pre-monsoon BC concentrations were higher than in post-monsoon season. We also found that morning and afternoon peaks in the post-monsoon season were more pronounced than those of pre-monsoon season, likely due to the relatively lower wind speeds during post-monsoon. Significant positive up-valley fluxes of BC were measured during all seasons, and preliminary flux estimates (which require a more robust estimate in future work) show the efficiency and magnitude of pollutant transport up the valley. During episodes of regional pollution over the IGP, relatively higher concentrations of BC and O<sub>3</sub> were also measured in the KGV.

The frequency and magnitude of pollution events highlighted in the paper need to be studied for a longer period in order to understand the associated interannual variability. In addition, future work should focus on understanding the vertical and horizontal distribution of particulate matter and ozone in the Himalayan region, and their impacts on the radiative budget, the ASM, and regional climate.

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**Table 1. Comparison of BC and O<sub>3</sub> concentrations (mean ±standard deviation) between high elevation Himalayan site NCO-P CNR and JSM\_1.**

<u>Sites</u>	<u>Altitude (m)</u>	<u>Co-ordinates</u>	<u>Season</u>	<u>BC (µg m<sup>-3</sup>)</u>	<u>O<sub>3</sub> (ppb)</u>
<u>NCO-P</u>	<u>5079</u>	<u>27.95°N, 86.81° E</u>	<u>Pre-monsoon</u>	<u>0.32 (±0.34)</u>	<u>60.9 (±8.4)</u>
<u>(Bonasoni et al., 2010)</u>			<u>Monsoon</u>	<u>0.05 (±0.06)</u>	<u>38.9 (±9.0)</u>
			<u>Post-monsoon</u>	<u>0.14 (±0.08)</u>	<u>46.3 (±5.0)</u>
<u>JSM_1</u>	<u>2800</u>	<u>28.87°N, 83.73° E</u>	<u>Pre-monsoon</u>	<u>0.90 (±0.45)</u>	<u>39.5 (±8.23)</u>
			<u>Monsoon</u>	<u>0.21 (±0.24)</u>	<u>25.1 (±6.48)</u>
			<u>Post-monsoon</u>	<u>0.71 (±0.42)</u>	<u>31.4 (±4.50)</u>

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Table 2. List of enhanced BC episodes observed at JSM\_STA and the concurring regional sources from MODIS (\* Data is from January-July; + Incomplete O<sub>3</sub> data).

2013 (90 <sup>th</sup> percentile : BC = 1.53 µg m <sup>-3</sup> and O <sub>3</sub> = 49.5 ppbv)			
Month	Episode Length	Episode type	Days with O <sub>3</sub> mixing ratio above 90 <sup>th</sup> percentile
Jan	6 <sup>th</sup> – 15 <sup>th</sup>	C	1
	28 <sup>th</sup> – 31 <sup>st</sup>	C	1 <sup>+</sup>
Feb	Jan 28 <sup>th</sup> – Feb 2 <sup>nd</sup>	C	1 <sup>+</sup>
Mar	1 <sup>st</sup> – 3 <sup>rd</sup>	A	0
	6 <sup>th</sup> – 9 <sup>th</sup>	A	0
Apr	11 <sup>th</sup> – 13 <sup>th</sup>	B	0
	18 <sup>th</sup> – 27 <sup>th</sup>	B	4
	6 <sup>th</sup> – 10 <sup>th</sup>	B	5
	12 <sup>th</sup> – 14 <sup>th</sup>	B	3
May	27 <sup>th</sup> – 30 <sup>th</sup>	A	4
	3 <sup>rd</sup> – 11 <sup>th</sup>	B	9
Oct	28 <sup>th</sup> – 30 <sup>th</sup>	A	No data
Nov	1 <sup>st</sup> – 5 <sup>th</sup>	A	No data
Dec	23 <sup>rd</sup> – 30 <sup>th</sup>	A	0 <sup>+</sup>
	17 <sup>th</sup> – 24 <sup>th</sup>	C	0 <sup>+</sup>
2014 (90 <sup>th</sup> percentile: BC = 1.60 µg m <sup>-3</sup> and O <sub>3</sub> = 56.8 ppbv)			
Month	Episode Length	Episode type	Days with O <sub>3</sub> mixing ratio above 90 <sup>th</sup> percentile
Jan	13 <sup>th</sup> – 15 <sup>th</sup>	A	No data
Feb	Jan 31 <sup>st</sup> – Feb 1 <sup>st</sup>	A	No data
Mar	14 <sup>th</sup> – 16 <sup>th</sup>	A	No data
Apr	5 <sup>th</sup> – 8 <sup>th</sup>	B	2
	10 <sup>th</sup> – 30 <sup>th</sup>	C	15 <sup>+</sup>
May	Apr 10 <sup>th</sup> – May 1 <sup>st</sup>	C	15 <sup>+</sup>
	8 <sup>th</sup> – 13 <sup>th</sup>	B	6
	17 <sup>th</sup> – 23 <sup>rd</sup>	C	5 <sup>+</sup>
Jun	5 <sup>th</sup> – 8 <sup>th</sup>	B	3
	10 <sup>th</sup> – 17 <sup>th</sup>	B	8
Nov	15 <sup>th</sup> – 30 <sup>th</sup>	A	0
Dec	Nov 15 <sup>th</sup> – Dec 8 <sup>th</sup>	A	0
2015 (90 <sup>th</sup> percentile: BC = 1.47 µg m <sup>-3</sup> and O <sub>3</sub> = 49.7 ppbv)			
Month	Episode Length	Episode type	Days with O <sub>3</sub> mixing ratio above 90 <sup>th</sup> percentile
Jan	15 <sup>th</sup> – 17 <sup>th</sup>	A	No data
	21 <sup>st</sup> – 25 <sup>th</sup>	A	No data
Feb	21 <sup>st</sup> – 26 <sup>th</sup>	A	No data
May	6 <sup>th</sup> – 9 <sup>th</sup>	A	0 <sup>+</sup>
	18 <sup>th</sup> – 26 <sup>th</sup>	A	8
Jun	29 <sup>th</sup> – 31 <sup>st</sup>	B	3
	6 <sup>th</sup> – 12 <sup>th</sup>	B	7

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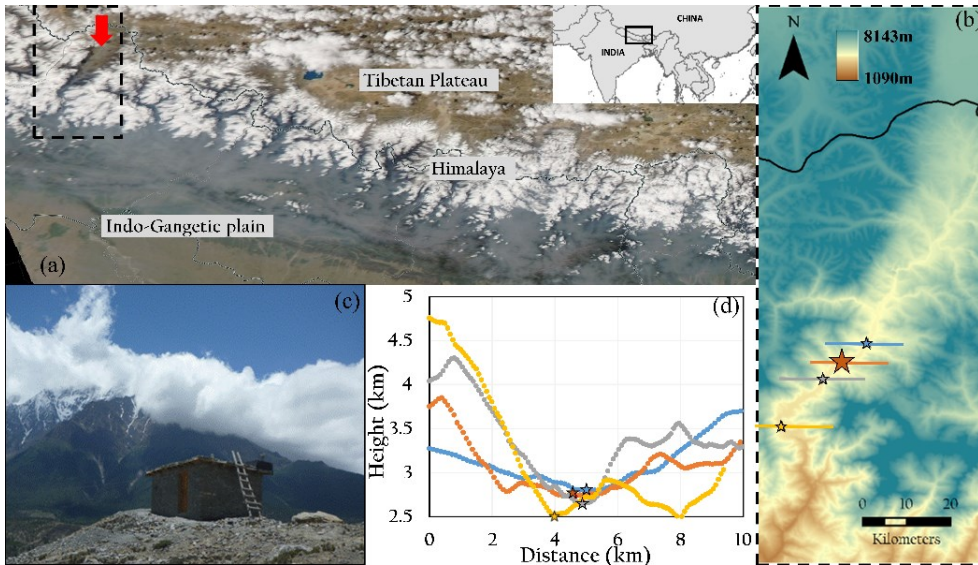
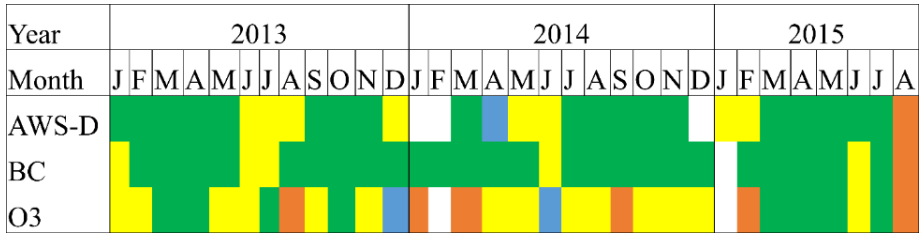


Figure 1. (a) NASA Worldview image from November 4<sup>th</sup>, 2014 depicting thick haze intruding the Himalayan foothills with red arrow over the KGV. Inset map in the top-right corner of panel (a) shows the location of the Worldview image. Dashed box shows the location of panel (b). (b) Expanded scale of the KGV showing station locations of Lete (LET\_AWS) near the entrance of the valley (yellow star); Marpha (MPH\_AWS) in the core region (gray star); the Jomsoms (JSM\_STA) sampling station for BC and O<sub>3</sub> and the two associated AWS sites (JSM\_1 and JSM\_2) in the core region (orange star); and Eklobhatti (EKL\_AWS) near the exit (blue star). (c) The atmospheric observatory at Jomsom (JSM\_STA) ([28.87° N, 83.73° E, 2900 m asl](#)). (d) Valley C cross-sectional elevation profile at the indicated station locations.

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625 Figure 2. Data timeline for JSM\_2 AWS, BC and O<sub>3</sub> measurements in Jomsom. Green indicates complete data, blue is with few data points missing, yellow is more than 15 days data, orange is less than 15 days and blank is no data.

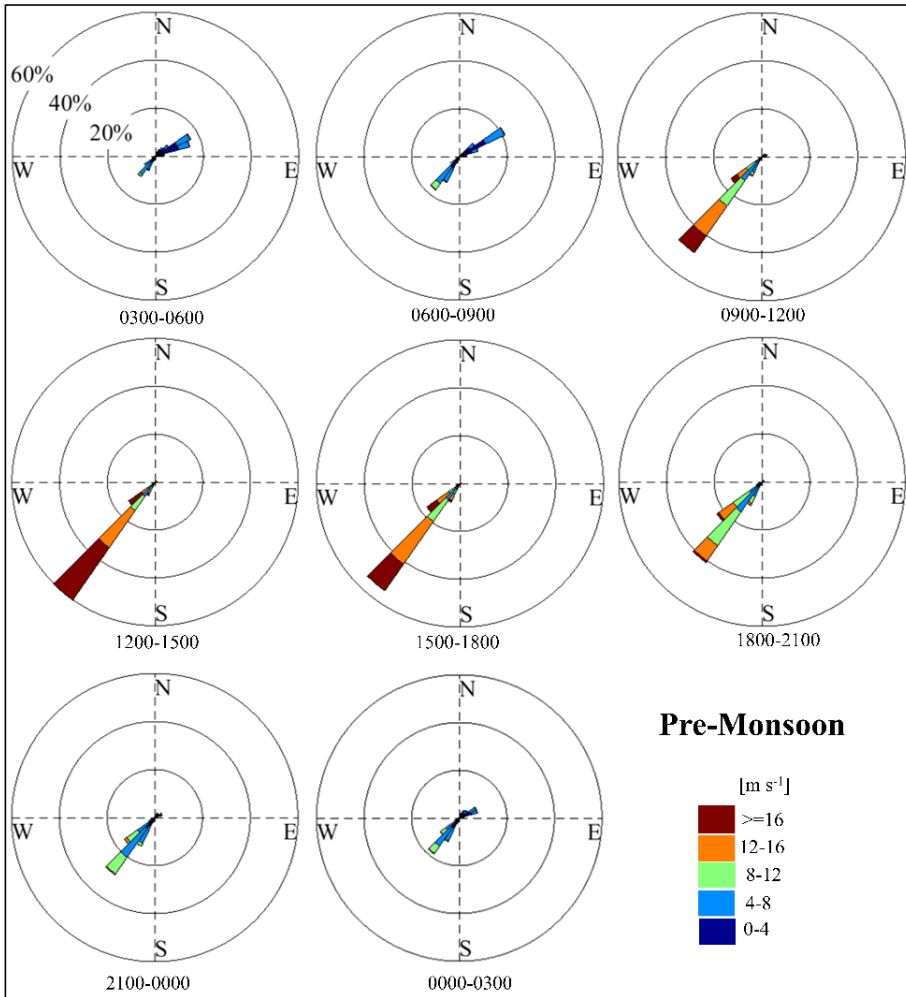
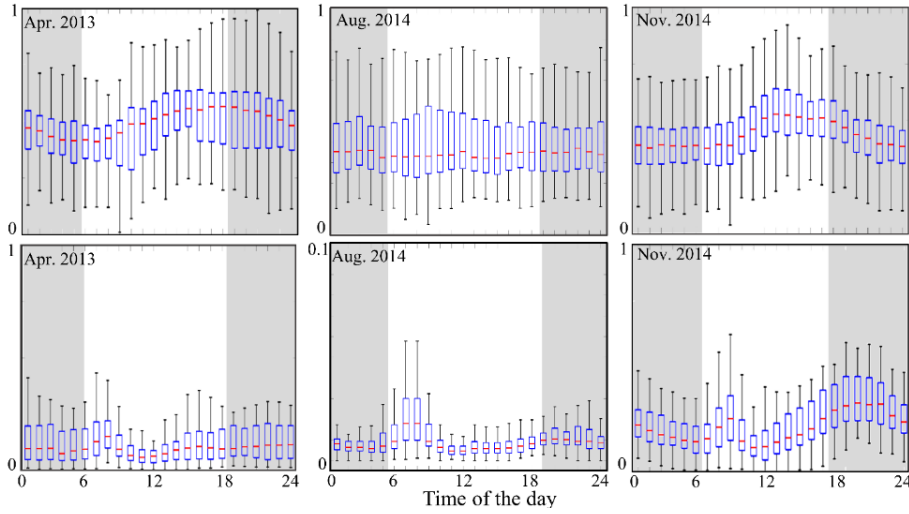


Figure 3. Wind rose for pre-monsoon season – binned into 3-hour increments depicting diurnal evolution in wind speed and direction at JSM\_2.

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635 **Figure 4.** Box and whisker plots depicting the 90<sup>th</sup>, 75<sup>th</sup>, 50<sup>th</sup>, 25<sup>th</sup>, and 10<sup>th</sup> percentiles for normalized diel variability in O<sub>3</sub> (upper panels) and BC (lower panels) at JSM\_STA during April 2013 (pre-monsoon), August 2014 (monsoon), and November 2014 (post-monsoon). Scale for August 2014 is from 0 to 0.1.

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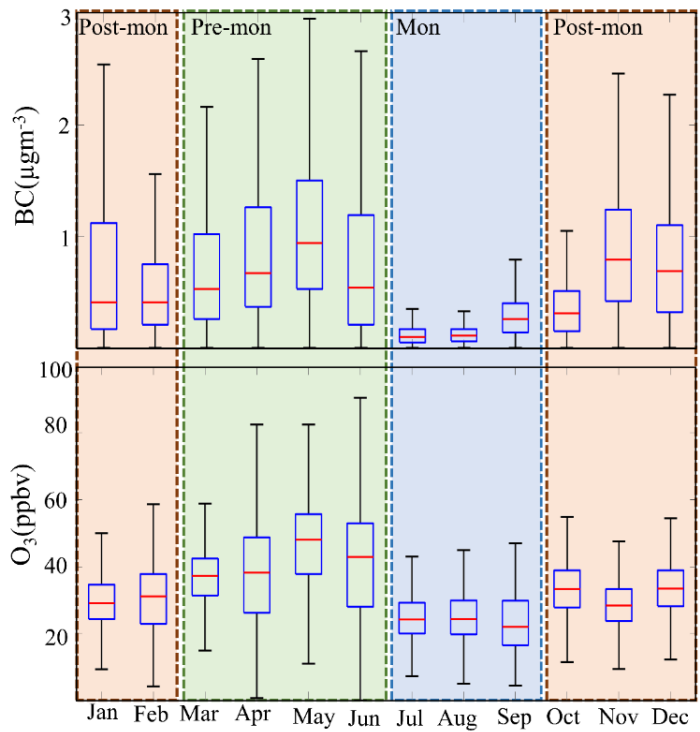
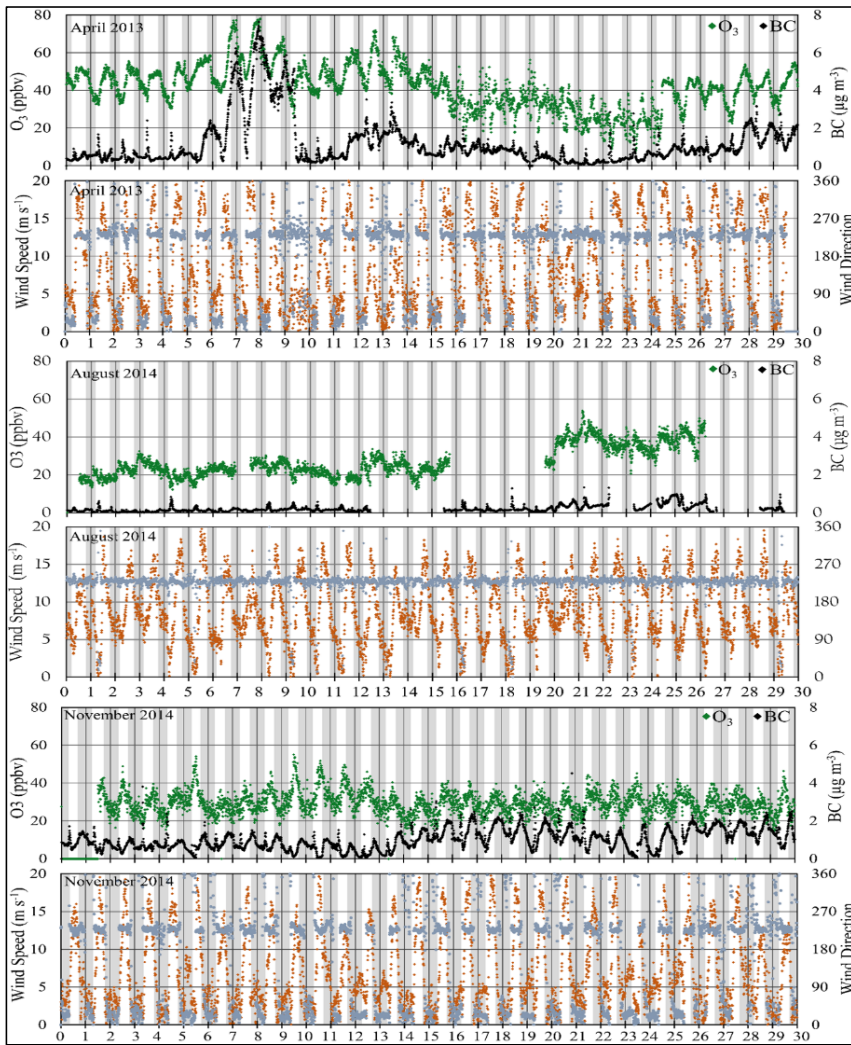


Figure 5. Box and whisker plots depicting the 90<sup>th</sup>, 75<sup>th</sup>, 50<sup>th</sup>, 25<sup>th</sup>, and 10<sup>th</sup> percentiles for monthly concentrations of BC (upper panel) and O<sub>3</sub> (lower panel) between January 2013 and August 2015 at JSM\_STA. Orange shaded areas indicate the post-monsoon season (Post-mon), green shaded area indicates the pre-monsoon (Pre-mon) and blue are indicates the monsoon (Mon) season.

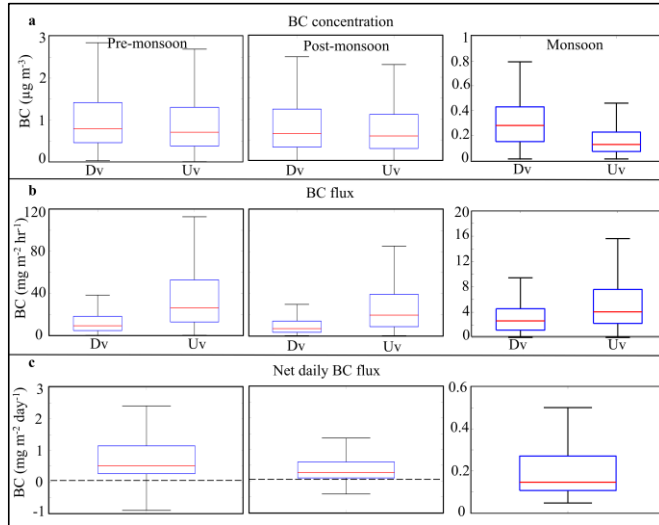
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Figure 6. Variation in O<sub>3</sub>, BC, and associated wind speed and direction at JSM\_STA during (a) April 2013 (pre-monsoon), (b) July 2015 (monsoon), and (c) November 2014 (post monsoon). The grey shaded area denotes night.



660 **Figure 7. (a) BC concentration distribution with down-valley (Dv) and up-valley (Uv) flows in Jomsom, (b) calculated Dv and Uv**  
**flux for each season, (c) Net daily flux per season. The dotted line in panel c marks  $0 \text{ mg m}^{-2} \text{day}^{-1}$ . The red line represents 50<sup>th</sup>**  
**percentile, the edge of the box 25<sup>th</sup> and 75<sup>th</sup> percentile while the whiskers show maximum and minimum values. Scale for monsoon**  
**season is  $0$  to  $1 \mu\text{g m}^{-3}$  for BC concentrations,  $0$ - $20 \text{ mg m}^{-2} \text{hr}^{-1}$  for BC flux and  $0$  to  $0.6 \text{ mg m}^{-2} \text{day}^{-1}$  for net daily BC flux.**

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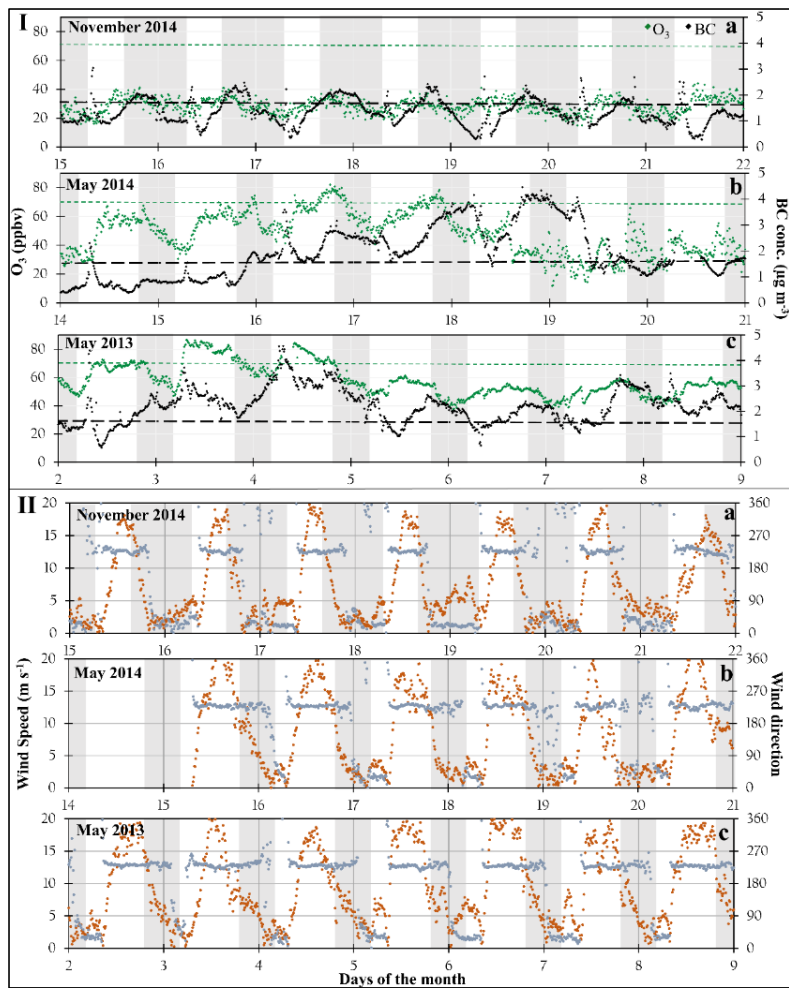


Figure 8. (I) Examples of extended periods with relatively high BC and O<sub>3</sub> concentrations at JSM\_STA during November 2014 (Pattern A) and May 2014 (Pattern B) and May 2013 (Pattern C). The dashed black and green lines depict two-year averages for BC and O<sub>3</sub>, respectively. (II) Corresponding wind direction and wind speed during the high BC and O<sub>3</sub> episodes. The orange dots represent wind speed, and the blue dots represent wind direction.

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