

## ***Interactive comment on “Chemical composition and aerosol size distribution of the middle mountain range in the Nepal Himalayas during the 2009 pre-monsoon season” by P. Shrestha et al.***

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Reply to Anonymous Referee #1

1. This paper is a good example for the very different observations that can be done when applying two distinct techniques for aerosol observation. The conclusions of the SMPS measurements are: a) that the boundary layer evolution connected with the valley wind system is the key phenomenon controlling the aerosol concentrations at the two sites, and b) that diurnal cycles are superimposed to a background aerosol of accumulation mode particles which “are always there”. On the other hand, filter analysis shows that aerosol mass concentration is controlled by the variation in the level

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of such background particles, which is driven mainly by the synoptic circulation. The papers seems split into two disconnect sections, and this is clearly reflected into the abstract. I strongly suggest the Authors to make more efforts for integrating these two bodies of observations. This can be done looking at the volume aerosol concentrations retrieved by SMPS observations, and trying to identify factors driving the variability between days with more precision: e.g., regional pollution increasing the background of accumulation mode particles in the middle of the day? Synoptic weather affecting the valley breezes system?

REPLY: The reviewer's recommendation for further analysis is well taken. A qualitative analysis using volume concentration measured from SMPS data was already included in Section 3.4 of the original manuscript in the context of the characterizing regional influence on aerosol spectra for Site B where the synoptic scale wind was also discussed.

Figure 1 (new Figure 14 in the revised manuscript) shows the time series of mean volume concentration of aerosols observed at specific times of the day. The effect of regional pollution is more visible when the number/volume concentration is at its minimum, usually during the night time or in the afternoon. The analysis was further extended to Site A as well, using the observed mean volume concentration during the afternoon (12:00 to 16:00 LT) and night time (23:00 to 03:00 LT). The interval range was selected such that minimum volume concentration was observed during that period of the day (Figure 1). As discussed in the original manuscript (Page 15645, Lines 1~4), synoptic scale haze was observed during 15th, 17th and 22nd May 2009 based on MODIS rapid response images from Terra. From 23rd May onwards, due to increased cloud cover, it was impossible to ascertain whether there was any haze event. Besides, significant rainfall events were observed on 15th, 16th, 20th, 21st and 25th May 2009. In general, from Fig. 1a, we observe that the mean vol. concentration during the afternoon, as well as the night, increases in the presence of haze event relative to previous day (eg. 15th, 17th ), while the concentration decreases after the rainfall

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event (16th, 20th and 21st May). A corresponding increase and decrease in elemental carbon and inorganic aerosol composition due to haze event and rainfall was also observed relative to previous day (Figure 10a). Further, the standardized anomalies of inorganic species (IN), elemental carbon (EC), afternoon mean volume concentration (Nv) were estimated to study the linkages between aerosol volume concentration and the aerosol mass concentration of different species, in the presence of regional scale haze event. Figure 2 shows strong in-phase relationship between observed mean afternoon volume concentration and aerosol species concentrations at both sites. The mean volume concentration during the night time was not used as it had some missing values. Specifically, a clear peak is observed during the synoptic haze events followed by dry periods [Site A: 15th, 17th~19th, 23rd and 25th May 2009, and Site B: 3rd to 4th June 2009]. A clear reduction in aerosol mass concentration and volume concentration is observed after a strong rainfall event in both the sites as well. Thus the above analysis indicates that in the presence of regional scale haze and dry periods, the mean volume aerosol concentrations during the period, when it approaches minimum (afternoon/night), increases and so does the aerosol mass concentration. (See the last paragraph of Section 3.4 of the revised manuscript for the updated text). The issue of synoptic weather affecting the valley breeze system is out of the scope of this paper, as the complete picture of actual valley wind circulation patterns in the sampled region needs further investigation through future field campaigns, and it would require area specific high resolution numerical simulations to study those effects, which will be examined in a forthcoming paper using a cloud-resolving model (<1km grid) based on the sounding data over Besisahar from MOHPREX campaign (Barros and Lang 2002).

2. The comparison of the aerosol chemical composition between the two stations is challenging because of the non overlapping sampling periods and because of the different sampling protocols. However, are there any correction factors that the Authors might envisage to make such comparison more quantitative? Is it possible to provide statistics differentiating between “polluted” and “background” conditions (based on Figures 9 and 10)? In this referee’s opinion, and given the dependence of aerosol mass

concentrations on synoptic weather regimes, the comparison between simple averages for the two stations/periods (Table 2) does not make any sense.

REPLY: The difference in the sampling periods was discussed in the original manuscript regarding the interpretation of the chemical composition of aerosols at the two sites ( Page 15645, lines 16~21). Table 2 has been used to simply report the values. Further, a note has been added to the description of the table stating the different sampling periods: “Table 2: Ionic and Organic concentrations of aerosols sampled in Site A (May 4th to May 25th 2009) and Site B (May 28th to June 06th) during the late pre-monsoon season 2009. All units are in ug/m3 .” Figures 9 and 10 along with Figure 2c and 2d show that during haze events, the aerosol composition tends to increase, gradually accumulating before plummeting after a rainfall. Such periodic increase and decrease in aerosol composition due to alternation of haze events and rainfall is a feature of sampling at Site A, unlike Site B, which had only one haze event and one big precipitation event after the haze. In this context, “background” and “polluted” statistics estimates appear to be more simpler for Site B, but not trivial for Site A.

3. Overall, I find the discussion of the link between boundary layer evolution and the variability of accumulation mode and Aitken mode particle concentrations quite convincing. The Authors infer aerosol transport patterns associated with the diurnal cycle of the boundary layer starting from their SMPS observations. But they should rather acknowledge the two papers by Panday and Prinn (JGR 2009) and Panday et al., (JGR 2009), discussing the boundary layer evolution for the Kathmandu area in details, and reporting diurnal cycles for CO analogous to those reported in this paper for aerosol particles. The importance of these two papers for the understanding of the aerosol measurements reported in this study should be clearly acknowledged in Section 3.2.2, in the abstract and in the conclusions of the manuscript.

REPLY: The Reviewer’s comment is well taken. Findings by Panday and Prinn (JGR 2009) were discussed along with Rissler et al. (2006) in the explanation of diurnal cycle of observed aerosol number concentrations at both sites in the original manuscript.

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Following the reviewer's suggestion, Panday et al. (JGR 2009) has also been appropriately cited in that section. Section 3.2.2 has been updated to acknowledge the importance of the two papers. Appropriate references were also added to the Introduction and Conclusions section.

“Previously, Panday and Prinn (2009) also explained the observed semi-diurnal peak of carbon monoxide (CO) at Kathmandu valley (bowl shaped valley, see Fig.1) in terms of diurnal variations in emissions and ventilation. They suggested that strong ventilation from westerly winds lowered CO level during the afternoon in Kathmandu Valley, while CO concentration reduction at night time was explained by the lifting of polluted air due to formation of cold pool in the valley. Further, Panday et al. (2009) used MM5 to simulate the Kathmandu Valley's meteorology and study the dynamics of basins nocturnal cold air pool, using a nested grid with a highest resolution of 1km. They suggested that subsidence warming was present in both summer and winter simulation over Kathmandu Valley for the dissipation of the nocturnal cold air pool. They used back-of-the envelope calculation to confirm the subsidence in their simulation, which also supports the role of mixing of elevated aerosols in residual layers during boundary layer growth in the morning. “(See Section 3.2.2 for the updated text in the revised manuscript)

4. The Authors provide an interesting comparison between their findings and the results from observations at high Himalayan stations. However, it should be emphasized that the present study reports observations from a three weeks period whereas some of the past studies (e.g., Carrico et al., 2003) discuss climatologies and long-term measurements. Especially when dealing with the concentrations of chemical compounds, which have been shown to be affected by synoptic weather, the short measurement period may have introduced biases with respect to the average concentrations for the premonsoon season at the two sites, therefore caution must be taken when comparing with experiments involving different sampling schemes and showing a different data coverage. I do not mean that this part of the paper should be dumped. On the con-

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trary, I suggest to the Authors to make it more circumstantiated, but to reinforce it, and to extend the comparison to most recent studies included in the same ACP special issue (e.g., Sellegri et al., ACPD 2010; Ram et al., ACPD 2010).

REPLY: The Reviewer's comment is well taken. As suggested, comparison has been made with studies from Sellegri et al., ACPD 2010; Ram et al., ACPD 2010.

“Recently, Sellegri et al. (2010) report an integrated (nucleation, Aitken and accumulation mode) aerosol concentration of  $1480\text{cm}^{-3}$  (10:00 to 18:00 LT) and  $675\text{cm}^{-3}$  (03:00 to 08:00 LT) during the pre-monsoon season at NCO-P (May 2006 to May 2008). The corresponding volumetric concentration over the accumulation mode was  $2.51\text{um}^3\text{cm}^{-3}$  (10:00 to 19:00 LT) and  $1.47\text{um}^3\text{cm}^{-3}$  (03:00 to 08:00 LT). Both number and volume concentration indicated lower concentration during the night time. In this study, we also found lower concentration of aerosols at night time at both study sites (23:00 to 04:00 LT). However, the night time mean concentrations at Site A and B are respectively on the order of 6 and 3 times the measured mean nighttime concentration at NCO-P, which is representative more of a tropospheric background.” (See Section 3.2.1 of revised manuscript, end of the 3rd paragraph of revised manuscript)

“More recently, chemical properties of aerosols over the Himalayas were also reported by Ram et al. (2010) for field measurements carried out at Manora Peak, 2000m a.m.s.l. from Feb. 2005 to July 2008. Their reported mean monthly concentrations during May of  $0.8\pm 0.5\text{ugC/m}^{-3}$  for EC and  $4.7\pm 2.0\text{ugC/m}^{-3}$  for OC are much lower than the measured values for EC and OC during this field campaign. Here also, we need to note that the results are compared between two different temporal (short term and long term) measurements. The mean fractions of WSOC/OC for Site A and B were very similar at and respectively. These values are much lower than the average WSOC/OC ratio of  $0.51\pm 0.12$  at Manora Peak for the month of May.” (See Section 3.3, second paragraph of revised manuscript). Also, cautionary notes were added explicitly in the text reminding the readers that we are comparing data with different temporal averages (short term compared to long term).

## 5. Specific comments:

- Page 15642, line 22. I would rather define the 20 nm particles as “fresh particles from local sources or formed in neighbouring areas” rather than “local or less processed background particles”.

REPLY: Line 22 has been rephrased as: “The dominant smaller mode is located around 20 nm, and could be due fresh particles from local sources or formed in nearby areas.”

- Page 15643, line 11. Add a reference for the times of the day when the boundary layer develops and collapses in this area of Nepal for this period of the year. – Page 15646, line 25. A WSOC/OC ratio of 0.2 is not a high fraction. See the data compilation reported by Jaffrezo et al. (ACP 2005).

REPLY: The times of the day when boundary layer develops and collapses were estimated from the vertical profiles of virtual potential temperature obtained from the radiosonde sounding in Besisahar (Site B) during MOHPREX (Barros and Lang 2003). We agree that WSOC/OC ratio of 0.2-0.3 is not high as compared to most values reported by Jaffrezo et al. 2005 or Ram et al. (2010). However, the values are in line with previous values reported in China and for ACE-Asia.

Section 3.3 discussion along with Fig. 11 in the revised manuscript were updated.

“The light absorbing properties of EC (Elemental Carbon) play a crucial role in atmospheric heating and surface cooling and hence are relevant to the radiative properties of aerosols (Ramanathan et al. 2007). The average EC concentration at Sites A and B are and respectively. The mean EC/OC ratio for Site A and B were and , respectively (Fig. 11). These ratios are much smaller than previously reported values of by Venkataraman et al. (2002 ) for Bombay, India during Feb-March 1999. More recently, chemical properties of aerosols over the Himalayas were also reported by Ram et al. (2010) for field measurements carried out at Manora Peak (Fig. 1), 2000m a.m.s.l from Feb. 2005 to July 2008. Their reported mean monthly concentrations of  $0.8 \pm 0.5$

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$\mu\text{gC}/\text{m}^3$  for EC and  $4.7 \pm 2.0 \mu\text{gC}/\text{m}^3$  for OC during May are much lower than the measured values for EC and OC during this field campaign (Table 2). Note that the sampling durations of the two studies were different ( $\sim 2$  weeks in our case;  $\sim 4$  months in 4 different years for Ram et al.). The mean fractions of WSOC/OC for Site A and B were very similar at and respectively (Table 2). These values are much lower than the average WSOC/OC ratio of  $0.51 \pm 0.12$  at Manora Peak for the month of May. However, the concentration of WSOC reported by Ram et al. for the month of May is  $2.7 \pm 1.3 \mu\text{gC}/\text{m}^3$ , which is similar to the WSOC concentration in Site A and B (Table 2). The OC concentration at Manora Peak is only  $4.7 \pm 2 \mu\text{gC}/\text{m}^3$ , thus much lower than in this study (Table 2) which explains the different WSOC/OC fractions. Jaffrezo et al., (2005) summarized the average data for WSOC/OC fractions from previous work that ranged from a low of 0.12 up to a maximum of 0.8. The reported values of WSOC/OC fraction were  $0.28 \sim 0.32$  (winter 1992) and  $0.32 \sim 0.55$  (summer 1992) for Tokyo (urban),  $0.32 \pm 0.15$  for ACE-Asia, and  $0.17 \sim 0.44$  for Nanjing, China. The WSOC/OC fractions reported in this study are in line with those values. Jaffrezo et al. (2005) also discuss the dependence of WSOC on temperature for measurements in the Alps. In the current study, similar dependence was exhibited by WSOC mass concentration and WSOC/OC fraction as a function of air temperature, which probably explains much of the variance in the time series of WSOC for Site A (Fig. 11). However, it is difficult to infer any such temperature influence in WSOC/OC fraction at Site B. Previous studies by Facchini et al. (1999), Mayol-Bracero et al., (2002), Ervens et al. (2005), Mircea et al. (2005), and Decesari et al., (2005) have showed that large fractions of WSOC can potentially influence the aerosol hygroscopic growth and activation. The dominance of particulate organic matter in the PM<sub>2.5</sub> filter samples, and significant WSOC/OC ratios at both sites in this study suggest possible influence of WSOC in the CCN activation for this region, which needs to be further examined. “

- Page 15646, line 26. There are now several models available to predict the CCN activity of aerosol particles made up of both organic and inorganic components. Therefore, if the Authors believe that the WSOC fraction is high enough to significantly alter the

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CCN activity in this specific environment, they should provide full reasoning for that. Otherwise, sentences like “suggest possible influence of WSOC in the CCN activation in this region” are gratuitous and should be omitted.

REPLY: We acknowledge the reviewer’s concern. We are well aware of the current available models to predict the CCN activity of aerosol particles made up of organic and inorganic compounds. We are currently working with the simple Köhler model with insoluble core, and ammonium sulphate coating along with WSOC to investigate the observed diameter growth factor of aerosols from JAMEX. The effect of WSOC on CCN, if any will be examined in a forthcoming paper. Line 25 was rephrased.

- Page 15647, line 8. When comparing the abundance of organic nitrogen in the aerosol from different environments, it is important to use the same metric. WSON/TN values cannot be readily compared to the contribution of “WSON compounds” (molecular mass? nitrogen mass?) to PM2.5.

REPLY: In this manuscript and the references, only Nitrogen concentration has been used, except for the data from Davis, CA which used nitrogen compound. So, we have removed that reference to avoid any further confusion.

- Page 15647, lines 12-15. This is another gratuitous statement, because only in specific ecosystems the supply of airborne nitrogen is a limiting factor for plant productivity.

REPLY: We agree with the Reviewer’s comment. Lines 12-14 were removed from the manuscript.

- Page 15648, line 6. The Authors argue that the rainfall events washed out the aerosol from the low troposphere. However, this explanation can hold or not depending on type and extension of precipitation. In general, it is more frequent that in a convective atmosphere the aerosol concentrations decrease because of the ventilation of the boundary layer rather than due to the aerosol scavenging by the raindrops.

REPLY: The aerosol concentration could also decrease because of ventilation of the

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boundary layer, but in a recently published article in ACPD in the same special issue, Sellegri et al. 2010, also argue that the rainfall washes the aerosol in the region, and further Nishita et al. (2007) also observed a clear decrease of the number concentration of particles in the accumulation mode at Mt Norikura, in Japan, which linearly correlated with the cumulative precipitation amounts during the 24 h preceding the sampling time. The decrease in particle number concentrations of the accumulation mode was found to be mostly explained by in-cloud scavenging at Mt Norikura.

- Last line of the conclusions. What is the rationale for the extension of the transport regime discussed in this paper up to the Tibetan plateau?

REPLY: The line has been rephrased in the current manuscript: “The synoptic scale wind during that period, along with the observed diurnal profile of surface wind, suggests that daytime upvalley wind might be responsible for transporting aerosols from the low lying plains to the inner valleys of the central Himalayas.”

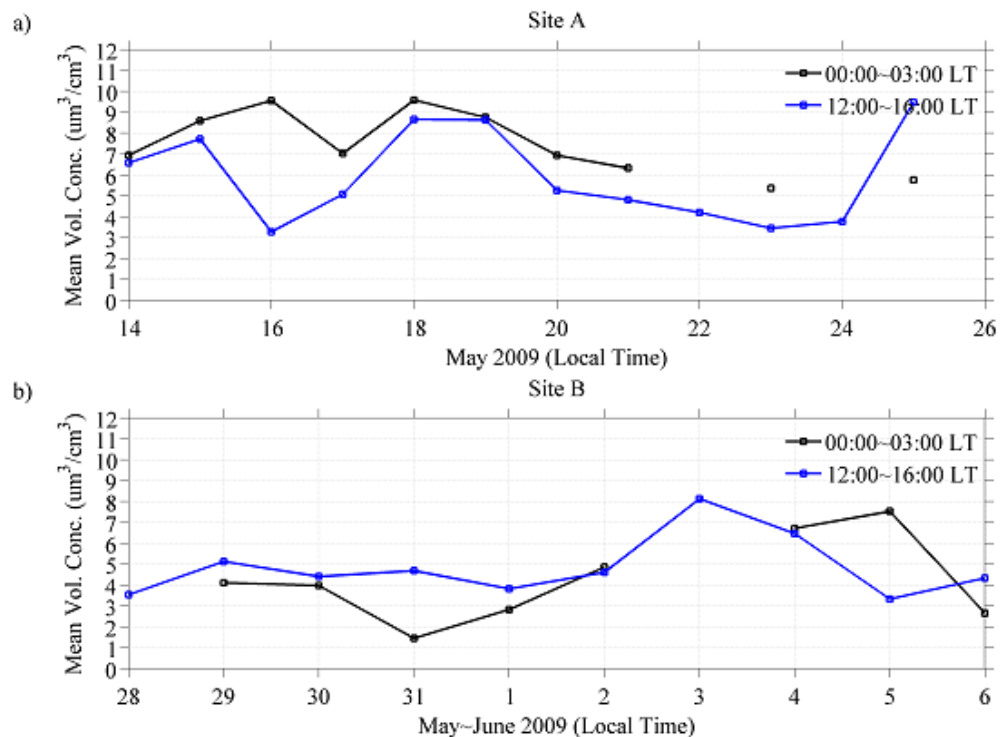
- The mass budget in Figure 8 is wrong: There must be overlaps between OC, WSOC and oxalate, therefore they cannot be plotted in the same pie chart.

REPLY: We removed the oxalate from the plot, but the data are still given in Table 2, and appropriate corrections and labeling has been done to remove any confusion created earlier.

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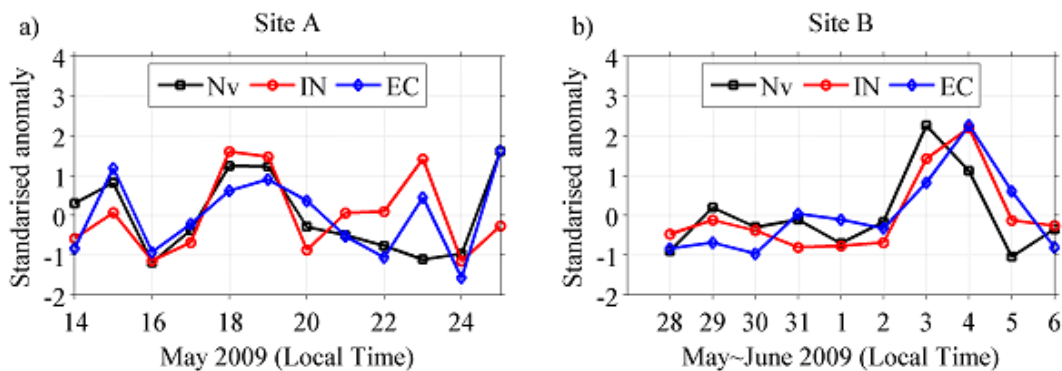
Interactive comment on Atmos. Chem. Phys. Discuss., 10, 15629, 2010.

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**Fig. 1.** Time series of mean volume concentration measured during the night time (23:00 to 03:00 LT) and afternoon (12:00 to 16:00 LT) from the SMPS. The missing values are due to electricity outages.

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**Fig. 2.** Time series of standardized anomalies of afternoon mean volume concentration (Nv), inorganic species (IN) and elemental carbon (EC).

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