

**Response to Reviewer's comments (MS Ref. No.: acp-2010-98):
Long-term record of aerosol optical properties and chemical composition from a high-altitude site (Manora Peak) in Central Himalaya" by K. Ram et al; Atmos. Chem. Phys. Discuss., 10, 7435–7467, 2010**

Date: 9th July, 2010 (corrections as per the ACPD paper published online)

Referee #2

Received and published: 26 May 2010

The paper of Ram et al. describes long term variability of aerosol chemical and optical properties sampled at a high altitude site in Central Himalaya. Several years of continuous measurements of aerosol composition (including OC, EC, ionic composition, TSP and Water-soluble OC), aerosol mass and aerosol optical depth are presented and the origin of the variability is discussed both in term of transport and processes. The data is original (although some parts have already been presented in another paper –Ram and Sarin, 2010-) and of interest given the low number of studies from this area. The paper provides information about the spatial (vertical) extension of pollution from the Indian Gangetic plains and also investigates the role of desert dust episodes in the long term record. The paper is well suited to the special issue on pollution in the Himalayas and could be published in ACP after corrections indicated below:

Response:

We thank the Reviewer for the constructive comments/suggestions. Our response to reviewers' comments is provided (blue color font) in the following note.

Introduction. The last sentence of the introduction section needs to be rephrased. There is no real discussion on aerosol mixing state and heterogeneous chemistry in the paper and the most obvious reason for sampling at high altitude is rather the spatial representativity of the site rather than process studies. In general, the introduction does not refer very well to the paper itself. My understanding is that monitoring from a high altitude area surrounding a very intense emission zone is to assess to which extent transport is affecting atmospheric composition far away from emission sources. The list of references can be completed with work from Nepal Climate Observatory – Pyramid which is being discussed in the same issue (and with Bonasoni et al., 2008) and with recent work.

Response:

The last para in the introduction section is changed and rephrased to read as “Our strategy of sampling from a high-altitude site is advantageous in order to study the long-range transport of aerosols through free-troposphere”.

Section 2.1. Important information is missing from the document concerning sampling and data reduction methodologies: how many samples have been collected? Which frequency and sampling time resolution (nights vs days)? Are all seasons sampled with similar frequency? Are results corrected for STP? These information should be added in this section.

Response:

The information on the total number of samples, frequency and number of samples collected in each season was provided in the earlier MS (Lines 21-25; P7438 and Line# 1; P7439). The samples were collected during day-time and integrated over 6-8 hrs. The following information has been added in Table caption (Table 1) “The concentrations in standard conditions can be obtained by multiplying the data in table by a factor of 1.3”.

Another general comment concerns referencing to summer/winter/pre-monsoon, monsoon etc.. that is somewhat confusing. Summer is sometime indicated in April-June (which does not correspond to “summer” for many readers), sometimes in September when referring to another study. To avoid this confusion, I recommend sticking to either the “normal” season classification or the Monsoon referenced classification, especially when comparing with previous studies. Perhaps sticking to Bonasoni et al. (this special issue) or Bonasoni et al., 2008) classification would be more appropriate.

Response:

The concept and clarification of seasons in a tropical region is different than the conventional and normal classification for the temperate climate. As per the suggestion, we have referred the time-period of April-June as pre-monsoon/summer, post-monsoon as Oct-Nov. The winter season in north India extends from Dec-March.

I have had some difficulties with organization of section 3.1 where I expected a general overview of the record. §3.1 basically deals with desert dust events more than chemical characteristics in general. The information on TSP provided in the text only refer to min and max values from a 3-year period, which seems quite reductive. I would have expected a longer discussion on variability, i.e. commenting Figure 3 in more details. The highest TSP values of course correspond to dust events but the full record cannot be reduced to discussing these events only. I believe more statistics are needed to characterize seasonal variations of TSP and provide a better statistics of dust events (including frequency) rather than a focus on one single event. Figures like pie charts for each season (or aerosol type) including all aerosol components would be extremely useful also for further use of data and comparison with model outputs. Without further information, it is difficult to check the dominance of carbonaceous aerosol during winter time (7441 line 16-17) for example.

Response:

The section 3.1 is now reorganized as per the suggestion of the Reviewer. The dust storm events and their frequency of occurrence are provided in the revised MS. Furthermore, a figure (Fig. 4) for seasonal aerosol composition is added in the revised MS.

In the same §3.1 (7441, line17-19), statements concerning aerosol emission sources are not fully support neither by Figure 3 nor with appropriate references. In fact, the discussion on emission sources relates more to the following sections than to this one. I am wondering if the paper would not be easier to read keeping the discussion on source origin in a separate section and very general description of aerosol variability first (including all components).

Response:

The section 3.1 is now reorganized and describes the emission sources and seasonal variability in the aerosol chemical composition. Section 3.2 provides information related to frequency of dust storms and changes in the chemical characteristics of aerosols during dust storm events. The added table (Table 3) provides the chemical composition of aerosols during the dust events and normal days. The following sentences have been added in the text:

“The desert-regions of the middle-East and the Thar Desert (in western India) are the major sources of dust over Indian region. The frequency of dust storm in northern India is 6 to 8 per year (Chinnam et al., 2006; Dey et al., 2004; Hegde et al., 2007)”.

Still section 3.1. (7442, line 14-on). There is some confusion between text and figure 3. In Figure 3, the shaded areas are indicated as either dust events or pollution events (on which basis?). I see in all shaded areas the increase in TSP paralleled with increases in Ca^{2+} , WSOC, etc. . . and cannot see that the relative abundance of carbonaceous species shows a decrease. Again, there is a need to further support these assertions by statistical analysis. It is not clear in the same §how the contribution of mineral (85% (by mass I suppose) 7442, line 28) is estimated since not all oxides are being measured. Is it the simple difference between TSP and all other non-dust related aerosol components?

Response:

The dust storm events are identified from the MODIS image (Fig. 1) and are further ascertained from the chemical tracers (example, carbonate carbon and Ca^{2+} concentrations). The occurrence of pollutants is identified based on a simultaneous increase in the concentrations of SO_4^{2-} , NO_3^- , OC and EC. The mass contribution of mineral dust is estimated by taking the difference between TSP and the measured aerosol components (i.e. mineral dust = $\text{TSP} - 1.8 \times \text{OC} - \text{EC} - \text{WSIS}$).

Pie charts or an explicit Table would clearly help the reader to clarify apportionment of aerosol components throughout the paper. Table 1 is important but mixes all events and without further explications, it is difficult to understand if variability is due to a higher frequency of polluted or dust events for example.

Response:

We have now added a figure (Fig. 4) for the seasonal representation of aerosol composition. Furthermore, a table (Table 3) on the aerosol composition during dust events and normal days is provided in the revised MS.

Section 3.2 (7444, line 8). The sentence “the abundances of OC and EC are almost a factor of 2 lower in July 2006 to September 2007 compared to those in other sampling years” cannot be verified neither from figure 3 nor from table 1. It is necessary to improve the quality of presentation. Also, please refer to Decesari et al (this special issue) when comparing to previous work on EC/OC in the Himalaya. Same section 7444, line 16, I do not see why reference to SSA values are given here, since it is repeated in a later section. Comparing EC and BC (from filter-based spectrometers) in terms of absolute concentrations is tricky. I am not sure that the difference between the 2 can be attributed only to atmospheric spatial variability. I would limit comparisons of Manora Peak EC values with other sites using the same technique. The variability of the mass absorption

efficiency observed in the following section is a clear indication that BC and EC cannot be directly compared without correcting the BC signal.

Response:

The OC and EC concentrations and comparison with other high-altitude sites have been now summarized in Table 2. The data of Decesari et al (2010) has been added in the Table 2. The SSA and BC mass concentrations have been deleted from the table and text.

Section 3.3 (7445, line 6-on). Analysis of OC/EC ratio should be made with great care. A number of intercomparison studies are available showing that similar techniques may lead to very different OC/EC. Perhaps to this is to be mentioned in the text. 7445 line 13: this is a confusing sentence since the origin of carbonaceous aerosol is the primary question posed in this section. Yet, it seems that authors already have the information on source apportionment at the site. Differentiating emissions from motor vehicle / industry from other sources cannot be simply made from OC/EC measurements. Please either add references or change or drop the sentence. Change lactation to location also. In the same section (7445, last sentence) the reference to Boundary layer height is not so trivial. Lowering boundary layer height and increased emissions in winter have opposite effects. This is not clear from the sentence. In fact, many studies from high altitude areas in the regions had to discuss the strong influence of local circulation (mountain breeze), even during winter time. The concept of boundary layer height is not appropriate over complex terrain. Please discuss this issue. P. 7446 line 2-on: the whole paragraph is difficult to follow. On one side, it is observed that averaged values of OC/EC in summer are not different than in winter and post monsoon. Instead, the median OC/EC values seem to be seasonal dependent (need to clarify the statistical relevance of the number indicated in parenthesis, by the way. Why not giving standard deviation as well since more than one season is sampled?). I am not sure how to interpret it and did not find any good explanation in the text and finally the OC/EC variability is used for deriving the SOC fraction. I understand the conclusion is that SOC is produced all year long and not only during summer as indicated (7446, line 14). The whole section needs clarification. Also, (7446, line 14), the discussion using WSOC starts before the specific WSOC section 3.4. This is a little bit confusing.

Response:

The concept of using the OC/EC ratio for the source apportionment has been discussed in the revised MS. The entire section has been reorganized and appropriate references are provided. The role of BL dynamics in trapping the aerosols has been deleted. The standard deviation is now added to the average (median) OC/EC ratio and discussed accordingly. Much of the discussion on WSOC/OC ratio has been moved to a newer section – 3.4 (as suggested by the reviewer).

Section 4.1. as mentioned earlier, summer is referred to April-June and May- September periods in the same section. Please clarify because the 2 records seem to show opposite results since summer in Gobbi et al. corresponds to Monsoon in the present paper. Check the construction of sentence “we have compiled. . .(7448, line 10)” which is unclear. Please change Gobi to Gobbi in Table 2. Overall, I am not convinced that the whole section is really necessary in the context of the present paper, at least in this format.

There is no attempt, for example, to relate column to in-situ information or to work on an event basis.

Response:

The time-period of April-June has been referred as pre-monsoon season. We believe that this section is relevant despite the fact that no information on the vertical profile is given. The variability in AOD values clearly indicates a periodicity with higher values during pre-monsoon season (also captured by MODIS, Aqua satellite for the year 2006; Fig. 1).

Section 4.2. The method used to derive absorption and mass-absorption efficiency is described in another paper. However, I am a little bit disturbed by the fact that b_{abs} , σ_{abs} and EC are discussed as totally independent measurements, which is not the case. Perhaps, this could be more clearly mentioned and explained either in §2.3 or in §4.2. Please also compare your results to Marcq et al., from the special issue and complete Table 2.

Response:

The measurement of absorption and mass-absorption efficiency is described in Ram and Sarin (2009). This is mentioned in the very first sentence of section 4.2. These optical parameters are discussed in separate sections as they are used for different purposes. The b_{abs} is used for the estimation of SSA while σ_{abs} is used for the measurement of BC mass concentration in optical instruments. The results from this study are compared with those from Marcq et al. (2010) in the revised MS.

Section 4.3. As mentioned earlier, absolute values of σ_{abs} have to be handled with care given the uncertainty of defining the EC/OC frontier in thermograms. My feeling is that the discussion is going too far without a clear error propagation analysis. Cozic et al. were using independent BC and OC/EC techniques. In fact, I am surprised then to see so little variability associated to seasonally averaged values (7450, line 7), which does not seem to correspond to the uncertainty provided in Figure 7b. Please clarify.

Response:

To avoid the repetition, we have not provided details of the measurements and uncertainties associated with σ_{abs} and b_{abs} . The reference was made to Ram and Sarin (2009) for this purpose and was mentioned in section 2.3 (last sentence of the first paragraph of the original MS. We have now added a sentence “The propagated root-sum-square error for the determination of b_{abs} and σ_{abs} is estimated to be of the order of ~23 and 32%, respectively (Ram and Sarin, 2009)”.

The absorption properties (b_{abs} and σ_{abs}) and EC mass concentrations are derived by a single instrument (the thermo-optical EC-OC analyzer). This is an advantage of our measurement techniques. Unlike the measurements of absorption and EC concentration via two different analytical instruments, all the measured parameters are derived from a single aliquot of aerosol samples. Furthermore, the measurement of absorption was calibrated against the Aethalometer (Please refer to Ram and Sarin, 2009).

The results provided at line 7 (Page 7450) refers to those reported by Cozic et al. (2008). This sentence was made in the continuation of the results obtained by Cozic et al. (2008). We apologize for not clearly mentioning this in the text (now corrected in revised MS). Our data are presented in Table 1 and Fig. 7b (now Fig. 8b).

Conclusion and implications. I believe that the conclusion section (as the introduction) could be improved to really emphasize the specificity of this study. At the present stage, no real implications are discussed, both in term of transport of pollution to remote regions or radiative forcing, for example. The last sentence of the conclusion (7452, line17-19) is not very clear and could be re-written.

Response:

The introduction and conclusion sections have been rewritten and implications of this study are discussed in the revised MS.

References:

- Chinnam, N., Dey, S., Tripathi, S. N. and Sharma, M.: Dust events in Kanpur, northern India: Chemical evidence for source and implications to radiative forcing, *Geophys. Res. Lett.*, 33, L08803, doi:10.1029/2005GL025278, 2006.
- Dey, S., Tripathi, S. N., Singh, R. P. and Holben, B. N.: Influence of dust storms on aerosol optical properties over the Indo-Gangetic basin, *J. Geophys. Res.*, 109, D20211, doi:10.1029/2004JD004924, 2004.
- Hegde, P., Pant, P., Naja, M., Dumka, U. C. and Sagar, R.: South Asian dust episode in June 2006: Aerosol observations in the central Himalayas, *Geophys. Res. Lett.*, 34, L23802, doi:10.1029/2007GL030692, 2007.
- Marcq, S., Laj, P., Roger, J. C., Villani, P., Sellegri, K., Bonasoni, P., Marinoni, A., Cristofanelli, P., Verza, G. P. and Bergin, M.: Aerosol optical properties and radiative forcing in the high Himalaya based on measurements at the Nepal Climate Observatory – pyramid site (5100 ma.s.l), *Atmos. Chem. Phys. Discuss.*, 10, 5627–5663, 2010.
- Ram, K. and Sarin, M. M.: Absorption coefficient and site-specific mass absorption efficiency of elemental carbon in aerosols from urban, rural and high-altitude sites in India, *Environ. Sci. Technol.*, 43, 8233-8239, 2009.