

**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)

We thank the Anonymous Referee #1 for his constructive comments/suggestions. Our response to reviewers' comments is provided (blue color font) in the following note.

Referee #1

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The manuscript presents the results of long-term measurements of the aerosol chemical and optical properties at a high-altitude (ca. 2000 m asl) site in north India, in a strategic location for studying the transport of pollutants and desert dust towards the Himalayas. The data presented here are of great value in the context of the research about the impact of natural and anthropogenic aerosols on the atmospheric transparency in south Asia. However, the quality of the paper can certainly be improved, because the discussion is often confused, there are many sentences with an unclear syntax, and the comparison with the results from literature studies is done indifferently with past observations at the same station, with other sites in the Indian subcontinent and with mountain sites of any altitude around the world distracting the reader from the actual purposes of the paper. Overall, the section dealing with the optical properties is better conceived compared to the discussion of the chemical data.

I list below my specific comments:

-- The main result of this study, i.e., natural dust sources account for the greatest fraction of TSP and of AOD but at the same time anthropogenic combustion sources are responsible for almost all aerosol light absorption, should be better emphasized in the abstract and in the conclusions.

Response:

This suggestion has been incorporated as a concluding sentence in the abstract (last line), as per the revised MS.

-- The Authors attribute the source areas of dust particles to Thar desert and as far as in the Middle East, but they do not provide any data to support this statement.

Response:

This statement in the earlier and revised MS is supported by the appropriate references, (Hegde et al., 2007; Prasad and Singh, 2007; Ram et al., 2008) (Lines 24-25 of Page 7439). The reference made to Fig. 1 also provides a better visualization of a dust storm over western India and its subsequent progression to central Himalaya (as sampled in this study from Manora Peak)

-- Seasonal cycles are reported according to the time periods: summertime (April – June), monsoon (July – August), post-monsoon (Sept – Nov), winter (Dec – Mar). However, the summertime and monsoon seasons overlap with the Northern Hemisphere spring and summer, respectively. It would be less ambiguous to refer simply to the phases of monsoon circulation, so that the period April-June would be better defined as the premonsoon period.

Response:

In order to compare the data from the winter season, we are of the opinion that it is appropriate to refer the other major season as summer in the tropical climate. There may not be any ambiguity as long as the time period of a particular season is defined in the text. Also, it is relevant to state that seasons and their respective time period (this study) are defined based on two important meteorological parameters (ambient temperature and the south-west monsoon phase). Nevertheless, as per the suggestion, we have referred the time-period of April-June as summer or pre-monsoon.

-- The comparison of OC and EC concentrations at Manora Peak with those at mountain stations in very different geographical locations (page 7444) leads to contradicting statements: “more or less similar” concentrations (page 7444, line 17) or “significantly higher compared to other high-altitude sites in the world” (page 7452, line 2)? Please, clarify.

Response:

The concentrations of OC and EC (this study) and their comparison with other high-altitude sites are now summarized in Table 2. Also, as suggested by the Reviewer #2, BC mass concentration, derived from optical methods, has been deleted from the text.

-- The conclusion of section 3.3, i.e., the variations of OC/EC ratio can be explained by the transport of biomass burning aerosols with a varying but significant amount of SOC, does not follow clearly from the discussion. The summertime formation of SOC is supported by the OC/EC data alone but not by the WSOC/OC ratios, which instead do not show any clear seasonal cycle. For this reason, the Authors’ statement in the conclusion about a significant contribution by SOC is not convincing.

Response:

The WSOC/OC ratios do not exhibit pronounced seasonal variability unlike the variability in OC/EC ratios. This can be explained based on the contribution of OC and WSOC from different sources. It is likely that water-soluble component of OC can vary on temporal basis (depending on the source variability and source strength); whereas WSOC may be dominated by SOAs. Recently, Favez et al, 2008 have made similar observation for the variability in WSOC/OC ratios.

--The presentation of the results is often confused and difficult to follow. Some examples: Total carbonaceous aerosols are defined and treated in a specific section (3.2), however some results are anticipated in section 3.1 dedicated to mineral aerosols. WSOC/OC ratios have a dedicated section (3.4) but some data are anticipated in section 3.3 dealing with the OC/EC ratios. Data collected at Manora Peak are treated together with results of previous studies at unknown locations (e.g., page 7450, line 3, when quoting the study of Cozic et al.).

Response:

The presentation of results is now suitably organized in the revised MS. Total carbonaceous aerosols are now defined in section 3.1. Much of the discussion on WSOC/OC ratios has been removed from the section dealing with OC/EC ratios and these ratios are now discussed separately. The discussion of Cozic et al. (2008) is modified in latter part of the text.

--There are numerous grammatical errors. The quality of English is inadequate to an international journal and must be improved.

Response:

We have avoided grammatical errors and have improved the quality of presentation.

Minor comments:

-- Abstract, line 10: What is the meaning of “temporal variability in the abundance pattern”? Do the Authors refer to the geographical pattern of the concentrations or what else?

Response:

Abstract and line 10 have been suitably corrected by referring to the seasonal variability in concentrations of carbonaceous species.

-- Page 7438, line 8: “sea” not “see”.

Response:

This typographical error is regretted and corrected.

-- Page 7438, lines 10 – 14: “Our strategy” etc.. This statement is gratuitous. It is not clear why sampling at a mountain site is a better option than sampling elsewhere in respect to characterize all processes listed here (aerosol ageing, change in mixing state, heterogeneous reactions).

Response:

We had referred to the advantage of sampling from a high-altitude site (and not particularly from the mountain site) to study the long-range transport of aerosols through troposphere. Nevertheless, much of the introduction section has been reorganized, and reference made to the aging and changes in the mixing state of aerosols has been deleted.

- “rainfall” is more commonly used than “rain fall”.

Response:

The error is corrected throughout the text.

- Page 7740, line 1: Please, specify that such determinations of the PBL thickness were done at Manora Peak, to clarify to the reader that a PBL height of 1300 m corresponds to an altitude of 3300 m a.s.l. for the top of the PBL.

Response:

As per the suggestion, the suitable information is now provided for the boundary layer height at the sampling site (Manora Peak).

-- Page 7442, first line: a proper reference should be added here to support the presented composition of mineral dust. The study of Cong et al. was focused on TSP not specifically on mineral dust.

Response:

Appropriate references, (Kumar and Sarin, 2009; Rastogi and Sarin, 2009), have been added for the composition of mineral dust.

-- Page 7442, line 10. The displacement of carbonate by sulphuric and nitric acids is an acid/base reaction which leaves unaltered the concentrations of Ca²⁺ and Mg²⁺ in the aerosol. It is the solubility product of the salts which changes. Moreover, why such reactions should alter the mixing state of aerosol compounds?

Response:

The transport/aging of the mineral dust in the atmosphere increases the hygroscopicity due to acid-uptake. The liquid coating enhances further uptake of acids. Thus, changes in the morphology and phase of the dust (calcite) particle changes their optical properties and ability to act as CCN. Nevertheless, much of this discussion has been modified in the text.

-- Page 7442, line 14: “The chemical composition during dust storm events is dominated by mineral aerosols” sounds pleonastic.

Response:

The sentence is modified to read as “The chemical composition during dust storm events is marked by a relative increase in the carbonate carbon (CC) and Ca²⁺ concentrations”.

-- Page 7442, last line: “normal days” should be better called “out-of-dust conditions”.

-- Page 7443, line 2: same as above.

Response:

A table (Table 3) has been added to support the chemical composition during the dust events and normal days.

-- Page 7444, line 17: “..more or less similar”. Please use a less colloquial expression.

Response:

These sentences have been corrected in the revised MS. To avoid repetition, all OC and EC concentrations and comparison with other high-altitude sites have been summarized in Table 2.

-- Page 7445, line 13: “Manora Peak is located at an altitude of 2000 m and represents a typical remote site”. The absence of in situ emissions makes the sampling station a background site, but not necessarily remote, given the proximity of sources in the plains beneath the peak.

Response:

As per the suggestion, this sentence is modified.

-- Page 7445, line 6 contradicts line 16.

Response:

These sentences are modified in the revised MS.

-- Page 7447, line 5: $R^2 = 0.57$ means a positive correlation but not a good correlation.

Response:

The word “good” is replaced by “significant linear correlation”.

-- Page 7448, line 3: “The dust storm” or “Dust storms”?

Response:

“The dust storm” is replaced with “Dust storms”.

-- Page 7450, line 3: What is the sense of reporting at this point of the discussion that in the study of Cozic et al. the absorption coefficient was highly correlated with the EC concentrations?

Response:

This sentence is deleted in the revised MS.

-- Page 7450, line 17: “greater coating of BC due to photochemical activity”. Please, explain better.

Response:

An explanation is provided with appropriate references.

“The internal and/or external mixing highly depends on aerosol chemical composition (mainly SO_4^{2-} , NO_3^- and WSOC), hygroscopicity and chemical processing in the atmosphere. In the presence of photochemistry, secondary aerosol formation and their coating can lead to an increase in the absorption signal for the same amount of absorbing EC (Bond et al., 2006; Moffet and Prather, 2009)”.

-- Figure 3. Indicating the boundaries between the seasons would help the reader in following the yearly cycles of the chemical species.

Response:

The suggestion is accepted and seasons are marked in the figure.

References:

- Bond, T. C., Habib, G. and Bergstrom, R. W.: Limitations in the enhancement of visible light absorption due to mixing state, *J. Geophys. Res.*, 111, D20211, doi:10.1029/2006JD007315, 2006.
- 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.
- Kumar, A. and Sarin, M. M.: Mineral aerosols from western India: Temporal variability of coarse and fine atmospheric dust and elemental characteristics, *Atmos. Environ.*, 43, 4005–4013, 2009.
- Moffet, R. C. and Prather, K. A.: In-situ measurements of the mixing state and optical properties of soot with implications for radiative forcing estimates, *Proc. Natl. Acad. Sci. U. S. A.*, 106, 11872–11877, doi/11810.11073/pnas.0900040106, 2009.

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- Ram, K., Sarin, M. M. and Hegde, P.: Atmospheric abundances of primary and secondary carbonaceous species at two high-altitude sites in India: Sources and temporal variability, *Atmos. Environ.*, 42, 6785-6796, 2008.
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