## **Responses to Referee 1 Comments**

This manuscript presents findings from measurements of aerosol particles at a remote site in the central Himalayan Mountains. Atmospheric measurements at high elevation sites are important for the assessment of long-range transport processes and regional-scale pollution characteristics, while few such studies have been conducted in South Asia. The study presented here included detailed speciation of the carbonaceous aerosol fraction with a focus on water-soluble dicarboxylic acids and related compounds. A strong seasonal dependence was observed for most organic species, while notable diurnal variations were found for only some compounds. During summer, the dicarboxylic acids and related compounds were much more abundant and mainly derived from anthropogenic activities, such as fossil fuel combustion, in the Indo-Gangetic plain, based on characteristic ratios of selected diacids. On the other hand, during winter biomass burning influence was more significant. Interestingly, photochemical aging and secondary organic aerosol formation appeared to be enhanced in the winter, which was likely related to the prevailing meteorological conditions and air mass history.

This paper addresses relevant scientific issues and fits well within the scope of ACP. The study appears to have been carried out with sufficient QA/QC measures, and the presentation of the methods and findings is concise and clear for the most part. Therefore, I recommend publication of this manuscript in ACP, following a few suggestions listed below, including a final check of the English grammar.

Technical corrections:

1. Please, check the correct use of the definite article "the" throughout the entire manuscript (i.e., it is missing in many cases), e.g., on p. 936, line 25; p. 937, line 4; p. 938, lines 5, 8, 10, 18 and 20; p. 939, line 22; etc.

Response: The authors appreciate the reviewer's comments on our work. Following the reviewer's suggestion, we have taken all the above mentioned corrections in the revised manuscript.

2. p. 936, lines 25-27: The sentence is not worded well and should therefore be changed slightly; specifically the word "ever" is not proper here, and the phrase "reveal enormous pollution transport pathways" should be reworded.

Response: This part of the text is modified in the revised manuscript (page 3, lines 31-37);

The Indian Ocean Experiment (INDOEX) was an international, multiplatform field campaign to assess the climatic and chemical influence of anthropogenic aerosols toward the Indian Ocean (Satheesh and Ramanathan, 2000). From the INDOEX campaign it was established that, during winter, this region would be covered by anthropogenic aerosols of sulphate, nitrate, organics, soot and fly ash via long-range transport of air pollution from South and Southeast Asia (Lelieveld et al., 2001).

3. p. 939, line 6: Change "climatology" to "climate".

Response: As suggested "climatology" is replaced by "climate" (page 5, line 100).

4. p. 941, line 15: Isn't the filter punch placed into a quartz tube rather than a "quartz boat" in the Sunset analyzer (in contrast to the DRI carbon analyzer)?

Response: Yes, it is the same one (filter punch placed into a quartz tube). Now, we have replaced the word "boat" to "tube" (please see page 8, line 165).

5. p. 942, line 3: Change "chromatography" to "chromatograph".

Response: Changed (please see page 8, line 180).

6. p. 943, line 6: Shouldn't it say "increases" instead of "decreases"?

Response: Sorry for the mistake. Now, we have corrected the sentence (please see page 9, line 208);

As the day advances, the boundary layer height increases, thereby capping inversion breaks and pollutants from the valley region (including Indo Gangetic plains) get readily transported to higher levels.

7. p. 943, line 17 Insert "with" before "each".

Response: The sentence is modified accordingly (please see page 10, lines 218-220);

Overall, three carbonaceous components (OC, EC and WSOC) generally correlate well with each other, showing minima during winter and maxima during summer (~ 2 times).

8. p. 944, lines 25-26: Perhaps the authors may want to change this part of the sentence "... the regional air quality will considerably change ..." to "... the regional air quality will considerably be influenced by ...".

Response: The sentence is modified as follows (page 11, lines 251-253);

Regional air quality over the northwestern part of India during winter season will considerably be influenced by the regular practice of crop harvesting. But, wood/coal burning for domestic use will continue throughout the year.

9. p. 945, line 2: These observations were made during spring rather than summer.

Response: The text is modified as follows (page 11, lines 255-256);

".....over a remote part of the Tibetan Plateau (Yunnan Province) during spring (April and May), reported higher OC/EC ratios  $(4.3 \pm 2.1)$ ."

10. p. 945, lines 5-7: This sentence is not worded clearly, while the contents are correct.

Response: The text is modified as follows (page 11, lines 260-261);

The major part of aerosol carbon is derived from smoldering process in the form of watersoluble organics and it can also act as cloud condensation nuclei (CCN)

11. p. 945, lines 17-18: It is not clear why the correlation between WIOC and EC "can be explained by that WIOC has various origins". The second part of the sentence makes sense and should be emphasized as the main reason for the good correlation, i.e., WIOC and EC have common sources, which are mainly the primary emissions from fossil fuel combustion.

Response: We agree with the reviewer's opinion. The text is modified accordingly (page 12, lines 270-272);

A good correlation between WIOC and EC was also apparent (except for winter night samples), indicating the influence of fossil fuel combustion over the study region.

12. p. 948, lines 20-23: I am not convinced that the lower diacid-C/TC ratios clearly indicate that the aerosol is influenced by hydrophobic carbonaceous material. For example, there are many other polar organic species (such as carbohydrates), aside from diacids, which contribute to TC as well. If the authors are convinced of their claim, then a better, i.e., more clear, explanation should be given here.

Response: We agree that many other polar organic compounds can also significantly contribute to TC. In the revised manuscript, we have changed the sentence, as follows (page 16, lines 385-389);

The observed lower diacid-C/TC ratios during summer indicate not only less photochemical aging but also the enhanced contribution from hydrophobic carbonaceous aerosols from combustion sources (Aggarwal and Kawamura, 2008; Jung et al., 2010). However, many other polar organic species (e.g., carbohydrate-like substances) also contribute to TC cannot be ruled out (Stone et al., 2009).

13. p. 955, lines 4-11: Please, provide references to support these implications.

Response: Supporting reference is included in the revised manuscript (page 24, lines 571-572);

The study conducted by Kawamura and Kaplan (1987) reported that diesel fuel vehicular exhaust shows a higher  $Ph/C_6$  ratio (6.58) than that from gasoline fuel vehicle (2.05).

References:

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- Jung, J., Tsatsral, B., Kim, Y. J., and Kawamura, K.: Organic and inorganic aerosol compositions in Ulaanbaatar, Mongolia, during the cold winter of 2007 to 2008:

Dicarboxylic acids, ketocarboxylic acids, and  $\alpha$ -dicarbonyls, J. Geophys. Res., 115, D22203, doi:10.1029/2010JD014339, 2010.

- Kawamura, K. and Kaplan, I. R.: Motor exhaust emission as a primary source of dicarboxylic acids in Los Angeles ambient air, Environ. Sci. Technol., 21, 105-110, 1987.
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- Satheesh, S. K. and Ramanathan, V.: Large differences in tropical aerosol forcing at the top of the atmosphere and Earth's surface, Nature, 405, 60–63, 2000.
- Stone, E. A., Hedman, C. J., Sheesley, R. J., Shafer, M. M., and Schauer, J. J.: Investigating the chemical nature of humic-like substances (HULIS) in North American atmospheric aerosols by liquid chromatography tandem mass spectrometry, Atmos. Environ., 43, 4205– 4213, doi:10.1016/j.atmosenv.2009.05.030, 2009.