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Interactive comment on "Chemical composition of PM₁₀ and PM₁ at the high-altitude Himalayan station Nepal Climate Observatory-Pyramid (NCO-P) (5079 m a.s.l.)" by S. Decesari et al.

S. Decesari et al.

s.decesari@isac.cnr.it

Received and published: 27 March 2010

Reply to Anonymous Referee #1

1. Abstract. 2. Introduction. Line 27. What exactly is meant by 'optically-active aerosol'. I would believe that all aerosols are optically active, but I think a key characteristic of an actual aerosol climatic impact is linked with the aerosol loading and relative amount of absorption. I think a clarification is needed, or a more specific definition of 'optically-active'. I also would suggest not referring to the aerosol in this fashion in the abstract. I suspect other papers in the special issue address this point in a more specific way and hence it is not needed in this paper.



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REPLY: We agree to omit the expression "optically-active aerosol" from the abstract and from the text. We decided to replace it with "light-absorbing aerosol" when citing the high EC concentrations observed at the NCO-P site and discussed in this paper.

3. Page 25492, lines 7-8. It would be helpful to include the months that are being discussed as summer to give a better view of the data coverage.

REPLY: We specified the months and quoted the overview paper of the experiment reporting the exact starts and terminations of the seasons at NCO-P.

4. Page 25497, lines 20-25. This discussion is a bit confusing. A clarification is needed with respect to what is meant by 'well-mixed'. Also, ammonium sulphate can not be used as a 'characteristic of an aged pollution' since it can exist in excess near sources as has been clearly documented in a wide range of urban studies. It should also be noted that it is difficult to infer seasonal differences for the 'monsoon' and 'postmonsoon' cases since there are only a limited number of samples (n=6, m=5). With respect to aged airmasses it would also be useful to discuss the WSOC/OC ratios. For example, based on the data in Table 1 the ratios are nearly identical for the 'premonsoon' and 'dry season' cases. Doesn't this suggest similar aging of air masses reaching the site? Also it would appear that WSOC is at times greater than OC (it appears this is the case during monsoon samples) and this needs to be discussed. Could it be linked with gas phase OC artifacts?

REPLY: We dropped the expression "well-mixed" because misleading. We agree that no chemical species discussed in this paragraph are pure tracers of fresh and aged air masses, however their relative concentrations can define the fresh/aged character of pollution in a relative sense. Since NOx oxidation is much faster than for SO2, the much higher nitrate/sulphate ratios in the premonsoon period compared to the dry season indicates fresher pollution in the former season. We rephrased the paragraph to make it clearer. We have extended the discussion of the WSOC/OC data, but we agree that these results do not support any clear seasonal change in the fresh/aged 9, C11779–C11782, 2010

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character of organic matter. However, without knowing the sources of particulate organic matter, it is difficult to use the WSOC/OC ratio for tracing ageing, because some sources, namely biomass burning, provides POA with already a high water-soluble fraction, whereas the POA formed by other sources, like fossil fuel combustion, are mainly insoluble. Therefore, strong or weaker variations in the WSOC fraction can originate from simultaneous or contrasting modifications of ageing state and source contributions. Finally, it should be noted that the WSOC/OC ratio can be relatively low even in very aged air masses (see the study by Aggarwal and Kawamura, Atmos. Environ. 2009, and compare to Kumagai et al., Atmos. Environ. 2009) which is something not really explained by the current paradigms of chemical ageing (Jimenez et al., Science 2009). Without organic tracer data, we decided to leave out from this discussion any topics related to organic source apportionment and also any in-deep discussion of WSOC/OC results. In the new version of the paper, we acknowledge the unexplained results concerning the WSOC fraction and also the possible role, as suggested by the Referee, of sampling artifacts.

5. Page 255023. Lines 20 -30. It would be helpful to include in the discussion the EC/OC and WSOC/OC ratios on the source regions. It would appear that the afternoon/night samples do not vary greatly for these ratios. Does this mean that in both cases longer range transport carries OC and EC to the region or that more local, Nepalese sources dominate both cases?

REPLY: The data reported in Figure 1 and Table 1 clearly show that, in the out-ofmonsoon period, carbonaceous aerosol concentrations are strongly influenced by the valley breeze circulation, and therefore are controlled by the injection of polluted PBL air occurring through upslope breezes. On the basis of the EC/TC and WSOC/OC ratios, summarized in a new table of the manuscript, EC is slightly enriched in night-time compared to daytime during the premonsoon season, but in general the variability in the WSOC/OC and EC/TC ratios in PM10 between different breeze regimes is rather small, indicating that nighttime catabatic air masses actually recirculate carbonaceous

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particles belonging to residual mixing layers and that apparently the contribution from long-range transport is small. Based on our observations, this does not apply to sulphate aerosols. The WSOC/TC ratios in PM1 suggest greater diurnal variations in TC composition, which may be related to shifts in the size distributions. However, in order to fully clarify this point, the chemical composition dataset for PM1 needs to be extended.

6. Figure 5. I am confused as to why the ratios are plotted versus mass concentrations. Certainly there is information in the ratios shown on the Y-axes but I am not sure why there should be a relationship with anthropogenic and/or reconstructed mass. This point seems to be made in what appears to be no meaningful statistical relationships in any of the plots shown in Figure 5. I don't believe the plots are useful and it would be potentially better to simply have a summary table of the values (perhaps with means and standard deviations) discussing the results in prior sections as has already been mentioned above. I would remove the 'nature of the background aerosol' section and have related discussion in the prior relevant sections.

REPLY: We accept the Referee's criticism, and we decided to remove the entire section and to replace Figure 5 with a new table reporting all aerosol chemical indexes, including NO3/SO4, TC/SO4, WSOC/OC, EC/TC. The discussion of the data reported in this table has been integrated in section 3.1.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 25487, 2009.

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