Object: Response to comments on “Atmospheric Brown Clouds in the Himalayas: first two year of continuous observations at the Nepal-Climate Observatory at Pyramid (5079 m)”, by P.Bonasoni et al.

Dear Editor,

Please find below the responses to the specific comments raised in the 2 reviews. We believe all comments have been addressed and we followed all the suggested changes. Modifications with respect to the original manuscript are clearly highlighted in this letter.

We hope the manuscript now meets ACP scientific standards for publication.

Sincerely,

Paolo Bonasoni

Anonymous Referee #1

- *The authors thank the Referee for the valuable comments, which have enabled us to improve the paper and gain a better focus on the results. All remarks were accurately evaluated and discussed point-by-point in our answer.*

The paper focuses on the contribution of regional scale and large scale pollutant transport to the Nepal-Climate Observatory at Pyramid. It first of all gives an overview over the problematic of brown clouds in the Himalayas, and describes the set of instrumentation that is used at the Pyramid site which is suitable to characterize the aerosol load ozone and black carbon and the contribution of the ‘brown cloud’ at the site. To characterize the transport phenomena a mixture of modelling, back trajectories and local measurements to characterize the regional advection in the valley wind system is necessary. To describe the brown cloud the parameters, ozone (as a secondary product of biogenic and anthropogenic emissions) black carbon as a product of burning processes and the number of coarse particles (> 1 um) is used. Modelling is based on WRF simulations and LAGRANTO backtrajectories. The analyses are done on a seasonal basis comparing pre-monsoon, monsoon and winter conditions. The paper is supposed to be part of the special issue “Atmospheric Brown Cloud in the Himalayas” ,the objective of this paper is to evaluate the influence of the Asian Brown Cloud on the Himalayan atmosphere composition.’: With the aim of providing basic information useful for other research activities conducted at NCO-P and presented in detail in the companion papers, the meteorological conditions at the station during the first two years of activity (March 2006–February 2008) are presented, identifying the seasonal transitions as a function of local weather regime and discussing the local and large-scale air-mass circulation that characterised the measurement site.

General comments:

The manuscript includes a comprehensive summary of the brown cloud problematic and according to the instrumentation list is based on a large experimental data base with instrumentation well suitable for the proposed research. However, most of the manuscript is based on model results analyses which are not supported by the appropriate observational data either on the brown cloud composition or the composition of the air masses encountered during other large scale advection processes.

- *In the previous paper, model elaborations were used to interpret experimental observations that represented the paper's "core". However, in agreement with the Referee, the observational data analysis has been further integrated in the manuscript (also adding PM1 and aerosol scattering) which now includes an investigation of the behaviour of several atmospheric compounds, such as black carbon, aerosol mass (PM1), scattering coefficient, coarse particles (used as marker for dust) and ozone concentration.. This permits a better characterization of the atmospheric properties at NCO-P in order to study the ABC influence at this Himalayan site.*
  Considering this last aspect, it should be borne in mind that the unambiguous definition of Atmospheric Brown Cloud concerns the seasonal values of AOD > 0.3 with absorption contribution larger than 10% (Ramanathan et al., 2008). This it is clearly not suitable for a high altitude site where the reduced integrated atmospheric columnar path (e.g. 5 km of difference between NCO-P and Indo-Gangetic Plains) makes the identification of a brown cloud hot spot problematic, also considering that the ABC is usually confined to altitudes lower than 5000 m. This identification of brown clouds at NCO-P, linked to the presence of ABC hot spots over IGP, is made possible thanks to continuous in situ observations. In particular, we identified time periods possibly affected by direct ABC transport by selecting in-situ observational data characterised by significantly enhanced BC, aerosol scattering coefficient and PM1, and with large AOD (>0.4) in the Himalayan foothills and IGP as well. With this aim, we defined a threshold of brown cloud influence for all the in-situ observations, considering the values greater than the background levels plus 2σ associated to positive component (Vy)of southerly valley breeze The background conditions at the site were derived from nighttime values related to back trajectories ending at NCO-P after travelling for 5 days at altitude higher than NCO-P. The typical levels of the compounds linked to direct transport of brown cloud to the site are now calculated and inserted in a new table, Tab.2, together with the values of coarse aerosol and surface ozone.
  In order to provide a detailed presentation of this new analysis that better focuses on results of this study and to avoid an excessive length of paper, we have removed part of the modelling investigation presented in the ACPD manuscript. In particular, regional-scale WRF simulations (Sect. 2.4 and analyses in Sect. 4.4 in the ACPD paper) were moved to the supplementary material. The detailed analysis of atmospheric compound variations as a function of different synoptic air-mass circulations (Sect. 4.2.2 in the ACPD paper) will be the object of a further, specific, paper.

The different source regions of atmospheric compounds should have a different composition signature.

- *As already shown in the ACPD paper the different clusters of backtrajectories are associated with significant differences in atmospheric concentrations. However, to better focus on the direct transport of ABC constituents to NCO-P, we have removed this analysis from the revised paper. It will be discussed in detail in a future specific paper.*

The composition of the brown cloud is can be described for example using the optical effective aerosols plus the total number of aerosols to account for the high number of burning aerosols which are too small to interact directly with radiation but possibly carry a significant fraction of black carbon.

- *In order to better describe periods influenced by brown cloud, in the new version of the paper we implement the parameters used for a complete aerosol overview: black carbon, aerosol mass (PM1), aerosol scattering coefficient at 700 nm, coarse particle number and ozone. As discussed in Venzac et al. (2008) and Sellegri et al (2010), since the total number of particles is strongly influenced by nucleation and Aitken particles, due to a very high frequency of new particle formation episodes at the site, CN cannot be considered an unambiguous parameter for characterizing the ABC influence at such a high-altitude measurement site.*

As a proxy for the optical active particles the number of particles larger than 1 um is not sufficient.

- *We discussed particles >1μm as tracers for mineral dust transport from desert areas, while high concentrations of particles smaller than 1μm indicate the presence of pollution. From the number size distribution of particles smaller than 1μm, the OPC calculates the aerosol mass of PM1, which is one of the parameters now used to identify the influence of brown clouds in the revised paper.*

The instrument installed measures also the size distribution starting from 300 nm. This is approximately the lowest size of aerosols that are measurable with an optical light scattering instrument. The total number of particles above this diameter would be a better representation of the optically active particles.

- *Following the Referee's suggestion, we now use the scattering coefficient at 700 nm as a better representation of the optically active particles; moreover the PM1 is calculated from number concentrations of accumulation particles (diameter between 300nm and 1um).*

Also the total mass as PM2.5 or PM10 could be derived from this instrument. These numbers would be helpful to compare to other locations.

- *A complete description of aerosol mass variations at NCO-P and comparison with other sites in Asia and other continents have been performed in the companion paper "Aerosol mass and black carbon concentrations, two year-round observations at NCO-P (5079 m, Southern Himalayas)" by A. Marinoni et al.(2010). For this reason only a few findings are reported in this paper, as it goes beyond the scope of the manuscript. A deep comparison of PM1/PM10 chemical composition with Himalayan and other sites where it is available, is also provided by Decesari et al. (2010).*

The number of small (ultrafine) particles can be derived from the SMPS or the TSI3010 counter at the station. Additionally the manuscript mentions also an integrating nephelometer which gives more information about the intensity of the 'brown clouds'. Although in the instrument list none of these experimental data are used in the manuscript.

- *Following the Referee's suggestions, in the revised version of the paper, we also present aerosol scattering coefficient at 700 nm from the integrating nephelometer, to better investigate the possible influence of brown cloud on the atmospheric properties at the measurement site. As stated above, the total particle number* *derived from TSI3010 is not completely suitable for tracing ABC transport due to the high frequency of nucleation and Aitken particles*

The results of modelling and backtrajectories are given in Figures 6, 8 and 9 with a resolution that does not allow to identify most of the features described in the text.

- *The section concerning the modelling analysis has been considerably shortened. Fig. 6, has been redrawn and separated into 4 single plates, now reported in the supplementary material (Fig S2, S3). Figure 8 has been eliminated, while Figure 9 (Figure 6a,b,c in the revised manuscript) resolution has been enlarged, in line with the Referee's suggestion.*

The manuscript would benefit from a more detailed description of the advection processes especially for the regional scale processes linked to direct transport of brown cloud components to the site AND the corresponding typical levels of particle number, mass and optical properties and black carbon in by far more detail than it is presented currently as seasonally average values.

- *In the revised manuscript the advection processes on the local/regional scale are used to provide a specific identification of periods affected by direct transport of brown cloud at NCO-P (Sect. 4.1) and for the purpose of providing estimates of "the corresponding typical levels" of ABC constituents directly transported to NCO-P (as also reported in the first answer). Moreover, the WRF simulations describing the regional circulation are presented in* supplementary material. *The authors maintain that a sufficient description of the along-valley circulation was already provided in the original paper (Section 3.4 and 4.2.1, in particular).*

The different advection patterns should be described with better graphic presentation. For example, the exchange of the air between the Ganges valley and the Tibetian plateau is invisible in Fig. 6. I would recommend also a graph of the valley contour along the transport pathway.

- *Even though this part has been moved to the supplementary materials, in the Fig. S2 and S3 the valley contour is now inserted. The picture resolution has also been increased, and the temperature field colours removed to improve the readability of wind vectors.*

Finally an analysis of aerosol mass and black carbon mass, transported with the different advection systems depending on the season would be a good addition for the interpretation, which anthropogenic activities are responsible for the 'brown cloud' at the station.

- *In Section 4.1, we discuss the typical values observed during direct ABC influenced periods for BC, PM1 and aerosol scattering coefficient.*
  The attempt of an overview analysis of different advection systems as a function of different seasons has been removed and will be presented in detail in a future specific paper. As suggested by both Referees, the present paper is now better focused on the brown cloud influence at the measurement site. However, the analysis of aerosol (mass and size distribution) and black carbon mass variation with respect to different air mass synoptic seasonal circulation is described in Marinoni et al (2010) and Sellegri et al. (2010). Concerning the source apportionment, some hypotheses are given in Decesari et al. (2010), even if it requires a very deep chemical characterization: aerosol mass and BC concentration are not enough to assume any anthropogenic activity responsible for brown cloud influence.

As long as the main focus of the paper is based mainly on model analysis, title, abstract and introduction are not appropriate and should be modified to match the content.

- *In accordance with the manuscript improvements, the revised paper is now more focused on the observations, and better reflects the original title, also considering the revised abstract, introduction and conclusion.*

The data presented in the figures are not always in agreement with the text (e.g. Fig.5 c).

- *Right. In the text related to Figure 5c, the words "in the noon-afternoon" were omitted due to a typesetting error, which has now been corrected.*