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"Sunphotometry of the 2006–2007 aerosol optical/radiative properties at the Himalayan Nepal Climate Observatory – Pyramid (5079 m a.s.l.)" by G. P. Gobbi et al.

Authors Reply to the Referee #2

General Comments (in blue the author's response, AR)

The primary objective of this proposal is to present and discuss sun photometer measurements at a site high in the Himalaya (the NCO-P site) and a location on the Indo-Gangetic plain (Gandhi College). Overall, the data set is potentially important in that there have only been a handful of other studies that have looked at climate relevant aerosol properties in the Himalaya. But with that said, there are several weaknesses in the authors approach, that are discussed in detail below. In particular, the sun photometer data has clearly not been carefully screened for clouds and hence determining trends in aerosol optical depth (not including the contribution from clouds) is problematic. Overall, I think this paper is not suitable for publication in present form and the specific points below must be clearly addressed, and significant changes to the manuscript made before the paper can be considered for publication.

AR: As specified in both the abstract and conclusions, one of the interesting findings of our paper is the persistent contribution (after cloud screening) of <u>cirrus-like</u> thin clouds to the AOD record at NCO-P. Conversely, Referee #2, interprets this signal as a (liquid) cloud-contamination, caused by incomplete cloud-screening. Most of the objections raised by the Referee are linked to this point. In our answers below we shall provide evidence of why we believe the data cloud-screening is robust enough and why the cirrus-like (not cloud-like) contamination remains the best explanation to the conditions encountered at NCO-P. In fact, the results obtained while checking the data to respond to the referees objections provided further support to our hypothesis.

Specific Comments

1. Introduction. I would suggest referencing and discussing the paper by Carrico et al. (*Atmospheric Environment*, 2003) at several locations in the Introduction. The paper by Carrico et al. (2003) includes several key points about aerosols in Nepal discussed in this paper (i.e. AOD, black carbon, organic carbon, and dust annual variability with links to source regions via back trajectory analysis). The authors also need to discuss the relationship between their results and this past work where appropriate throughout the paper.

AR: This suggestion is definitely useful. We shall include the Carrico et al. (2003) reference in the introduction and in relevant sections of the manuscript.

2. P. 1195, Line 25. It should be mentioned that direct radiative forcing also depends on aerosol phase function. The references discussing radiative forcing are not complete and exclude earlier work that set the stage for the references cite. I would suggest at least adding, and discussing the papers by Charlson et al. (*Science*, 1992) and Schwartz (*J. Aerosol Science*, 1996) 3. P. 1200,

AR: We shall mention in the paragraph the direct radiative forcing depends also on aerosol phase function. For the sake of brevity we cited the Haywood and Boucher review paper. We shall add the two references suggested by the Reviewer in the revised manuscript.

Section 3.2. A brief discussion on the cloud-screening process is necessary, since this is a critical factor in discerning general pollution, and dust aerosol particles from clouds

AR: In the revised manuscript we shall insert a short description of the cloud-screening procedure and of its robustness as described in the next paragraphs. This represents a further support to the cirrus-like explanation of the AOD contamination.

The cloud screening applied to our data.

AERONET cloud-screening (level 1.5) is fully described in Smirnov et al. (2000, Cloud screening and quality control algorithms for the AERONET database", Remote Sensing of the Environment, 73, 337-349) and briefly summarized hereafter. The algorithm is based on two major, sequential threshold criteria both related to temporal variations and intended to mask unrealistic high and low frequencies changes in the AOT: 1) 'the triplet stability criterion' and 2) 'the smoothness criterion' (see also the scheme reported below).

1) The first criterion is applied to screen high frequencies variability (of the order of one minute). In fact, the direct sun measurement consists of three acquisitions, each made 30 seconds apart, to yield a triplet observation per wavelength. The measurement is discarded if the AOT range within a triplet is higher than 0.02 (or higher than 0.03*AOT, in cases of high AOT conditions as Biomass burning or haze).

2) If the standard deviation of the daily averaged 500 nm-AOT is less than 0.015 (after triplet variability screening), all measurements are accepted (diurnal stability check). If this is not the case a 'smoothness criteria' is imposed on the time series. This imposes limitations on the AOT derivative with time. Taking into account that the measurements of optical depth (AOT or τ) are taken in n discrete moments of time (t), differences are used instead of analytical derivatives. For operational purposes, an index D (first derivatives difference) is defined as:

$$D = \sqrt{\frac{1}{(n-2)}} \sum \left[\frac{\ln \tau_i - \ln \tau_{i+1}}{t_i - t_{i+1}} - \frac{\ln \tau_{i+1} - \ln \tau_{i+2}}{t_{i+1} - t_{i+2}} \right]^2 \le 16$$

where the threshold $D \le 16$ has been founded on experimental data in different aerosol conditions. If D > 16 the algorithm finds the term with the maximum input to D and eliminate the maximum optical depth associated with it. The diurnal stability check (above) is then applied again and repeated every time a measurement rejection is made by the smoothness criteria. When the number of measurements in a day is reduced to only one or two, that day is completely rejected. Finally, for those measurements passing the 'smoothness criterion' a '3-standard deviation (3 σ) criteria' is applied. In this step the algorithm checks if any measurements fall outside of the 3 σ range about the mean of AOT (τ) at 500 nm as well as for the Angstrom parameter α (estimated using least-square regression in the 440-870 nm range), taken over the entire day (i.e. τ (500 nm) \pm 3 σ and $\alpha \pm$ 3 σ).

In Kaufman et al, 2006 (GRL, 33, L07817, doi:10.1029/2005GL0254782006) we have shown how such <u>AERONET cloud screening tends to overcorrect the aerosol dataset, i.e., to interpret aerosol conditions as clouds rather than the opposite.</u> In particular, that paper demonstrates how at Kampur (Indo Gangetic plains, i.e., location affected by the monsoon), both the average AOD and Angstrom coefficient remain the same even relaxing the AERONET cloud screening by using instead the proposed Spectral Variability Method (which increases from 54 to 74% the percent of measurements accepted as cloud-free, e.g., Table1).

On the basis of the above discussion we believe the AERONET cloud-screening to be a robust, even too selective, one. Hereafter we address the effects such cloud screening has on our NCO-P data:

The frequency distribution of the AOD measurements at NCO-P in the four seasons (pre-monsoon, monsoon, post monsoon, and dry season) is presented in figure A1 below. Here the stars and circles represent level 1.5 and level 1 measurements, respectively, colour scale reports the relevant Angstrom coefficient. The integral frequency of L1 and L1.5 observations are reported by the red and blue lines, respectively. This figure shows that 95% of the cloud-screened measurements have AODs below: 0.25 in the pre-monsoon, 0.15 in the monsoon, 0.06 in the post monsoon, and 0.07 in the dry season. All these levels are well (over one order of magnitude) below the typical AOD of liquid clouds but fully compatible with thin cirrus clouds. Furthermore, most of the cloud screening results to take place in the pre-monsoon and dry seasons not in the monsoon season. In fact, since cloud contamination is identified evaluating the AOT temporal variations, stable uniform clouds, as thin cirrus, might pass the algorithm thresholds and be misclassified as "cloud free". This analysis further supports the cirrus-like rather than the cloud-like contamination of our AOD data.

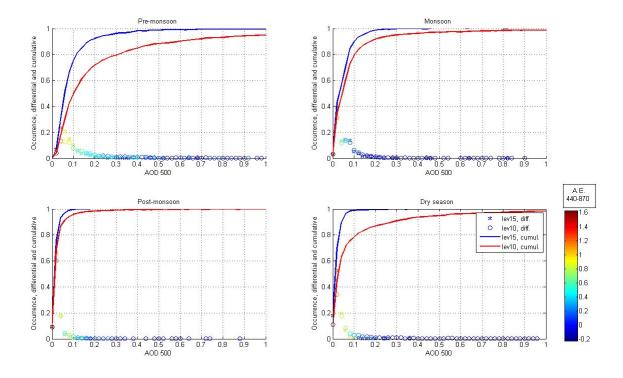


Figure A1

To summarize, on the basis of the points addressed so far we believe that for the purposes of this paper the AERONET cloud-screened, Level 1.5 data we employ here is a good AOD dataset to work with and that the AOD contamination is generated by cirrus-like clouds. These conclusions were already reached in the manuscript. As initially mentioned, the points discussed above will be synthesized in the revised manuscript.

4. Section 3.2. It is not clear why the fine and coarse particle fractions are estimated and shown in Figure 4. These fractions are based on model estimates of the sun photometer data that have inherent uncertainty based on model assumptions. I would suggest the authors discuss the total AOD and plot the Angstrom Exponent for both stations (which allows for the qualitative discussion of aerosol size distribution) rather than presenting the problematic coarse and fine AOD fractions (note that the angstrom exponent for the NCO-P site is plotted in Figure 6b).

AR: The AODs we discuss are the Total and the Fine fraction derived by the AERONET direct-sun observations. Indeed the fine fraction AOD is retrieved by using modelled aerosol properties in interpreting Angstrom coefficients spectral dependence. Still, the Angstrom coefficient-based interpretation suggested as alternative by the Referee #2 is based, as well, on the "aerosol model": high Angstrom = fine particles, low Angstrom = coarse particles. The method employed by the AERONET (O'Neill, et al., 2003; Spectral discrimination of coarse and fine mode optical depth, Vol.. 108, J. Geophys. Res., No. D17, 4559-4573, 10.1029/2002JD002975) provides a quantitative information on the fine mode contribution to the AOD the Angstrom method alone cannot provide. In the presence of contamination, as in our case, this can quantify the importance of the coarse particle contribution, separating at the same time the remaining fine aerosol AOD. In fact, Figure 4 shows the fine mode AOD at NCO-P to decrease in the monsoon season, exactly the way Referee #2 would expect. Still, coarse, dust aerosols cannot be distinguished from cirrus particles by this method. However, as shown in Fig. 2, airmass trajectories in this season mostly do not proceed from arid regions, in addition, precipitation would enhance the removal of such particles. Thanks to this Fine mode vs. Total AOD approach we can then attribute the high monsoon AODs to cirruslike contamination. The benefit of separating fine from total AOD seems to be clear to us.

All these points were already listed in the manuscript abstract (..."the Ev-K2-CNR Pyramid is shown to be affected by the advection of pollution aerosols from the populated regions of southern Nepal and the Indo-Gangetic plains. Such an impact is observed along most of the period April 2006–March 2007 addressed here, with a minimum in the monsoon season. Backtrajectory-analysis indicates long-range transport episodes occurring in this (whole year) period to originate mainly in the West Asian deserts. At this high altitude site, the measured aerosol optical depth is observed to be: 1) about one order of magnitude lower than the one measured at Gandhi College (60m a.s.l.), in the Indo-Gangetic basin, and 2) maximum during the monsoon period, due to the presence of elevated (cirrus-like) particle layers. Assessment of the aerosol radiative forcing results to be hampered by the persistent presence of these high altitude particle layers, which impede a continuous measurement of both the aerosol optical depth and its radiative properties from sky radiance inversions.").

Also, this would allow for better assessment of cloud contamination in the monsoon season AOD retrievals. Generally speaking, it is very hard to believe that the monsoon season measurements are not primarily of cloud droplets (i.e. aerosol particles that exist at RH > 100%). And in fact this point is supported by the authors.

AR: This is not correct, the authors keep supporting the cirrus-like contamination of the AOD.

Given that particulates are readily removed by precipitation scavenging during the monsoon the AOD signal is very likely not from particulates related to dust, biomass burning, and other pollution sources but from condensed liquid water.

AR: Cirrus layers are made of condensed water and are optically thin, clouds are optically thick. The referee does not give quantitative reasons to prefer the cloud droplet explanation to the cirrus-like particle one. We have shown above the cloud explanation to be rather unlikely.

I think plotting Angstrom Exponents will illuminate this issue.

AR: The Angstrom exponent is already plotted in Figure 6. Still, low Angstrom coefficients can be due to coarse aerosols, cirrus clouds and liquid clouds, irrespectively. As discussed above, differentiation cannot be performed by using this parameter alone.

From a direct radiative forcing perspective it is additional light extinction due to the presence of anthropogenic pollutants that is the issue and given the high AOD's during the monsoon season I think the story about direct anthropogenic forcing during that time is misleading.

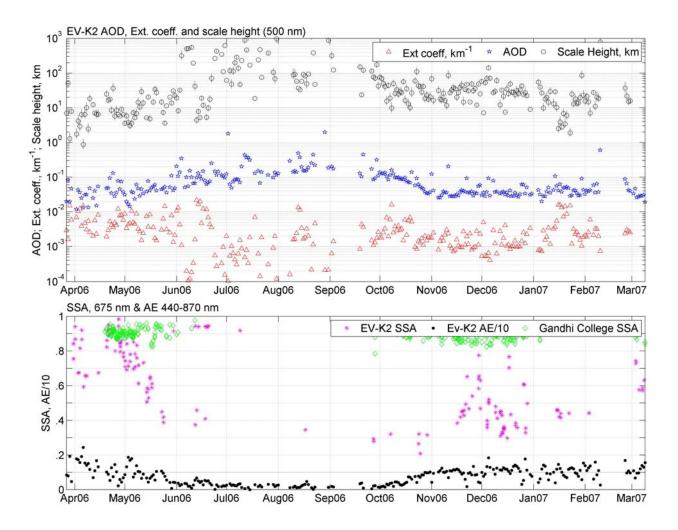
AR: It is not clear which is the "story" the Referee is thinking about. We never attributed high monsoon AODs to anthropogenic aerosols. We specifically state (see again point 2 of the manuscript abstract also reported above) that 'assessment of the aerosol radiative forcing results to be hampered by the persistent presence of these high altitude particle layers', e.g., the cirrus-like contamination.

In summary, I think it is critical that the data be looked at closely for cloud contamination, and the data impacted by clouds removed from the discussion. It is also worthwhile to add that the AOD measurements during the monsoon season are much less frequent and hence may not truly represent the average anthropogenic AOD's during that time. I suspect that at both stations the monsoon season AOD's represent primarily clouds and not RH < 100% aerosols. I would suggest a more rigorous, and clearly described cloud screening process be applied to the data before it is discussed.

AR: The effectiveness of our cloud-screening process has been checked and widely discussed above. We have also shown how separation of fine from total AOD provides a good way to interpret the (cirrus) contamination. Note also that 1) the decrease in number of measurements during the monsoon is well quantified in figure 3; and 2) in no case the sunphotometer data refer to the 'anthropogenic AOD' (indicated by the Referee), since no distinction between anthropogenic and natural aerosol can be done on the basis of such aerosol optical properties. As specified before, we will improve the revised manuscript by adding further information on the cloud-screening process.

5. Page 1202. Line 15. A more detailed explanation is needed of the assumptions made in the Mie Model. In particular, how can the refractive index of 1.5 - 0.001i be justified, particularly given the mix of aerosol sources (i.e biomass burning, dust, other anthropogenic sources) and varying complexity of the aerosol chemical composition and shape? How much uncertainty can be expected in the calculations?

AR: The Referee is right, choice of a 'mean' aerosol (e.g. D'Almeida et al., 1991) refractive index m=1.5 - 0.001i might appear arbitrary. To address this objection we performed a sensitivity study calculating the extinction coefficients for m=1.40-0.0001 and m=1.60-0.01i, a range which includes most of the average aerosol conditions encountered in the atmosphere. The variability generated by such range was then reported as error bars in the scale height plot of the new Figure 6 we propose (shown below). It is now clearly visible that such variability does not impact at all the conclusions drawn in our analysis.



6. Page 1202. The statement that the 'comparison provides information on the altitude dependence of the aerosol content' is incorrect. The AOD is the integral of the extinction with height and the assumption of a perfectly vertically mixed aerosol (i.e. constant extinction coefficient with height) must be made. This is rarely the case, and can not be justified particularly at such a high elevation site impacted by long range transport. The authors instead use an assumption of an exponential decrease in aerosol extinction with height which is also not justifiable. I suggest the authors remove figure 6a and the related discussion. It should be added that as the authors point out, a scale height of 100 km is at times estimated, which is totally unrealistic. This also goes back to another point, which is the data during the monsoon season is likely contaminated by clouds.

AR: Our high cloud (cirrus-like) contamination hypothesis is also supported by the analysis summarized by this figure. The figure is then important to the paper goals and must be kept. Hereafter we answer the Referee's objections to demonstrate the appropriateness of our analysis. The exponential decrease of the extinction coefficient with height is a commonly encountered climatological feature of atmospheric aerosols. This is both observed by lidars and predicted by models. To provide evidence for this, we show below two examples based on long-term climatologies: one by Ferrare et al., (The Vertical Distribution of Aerosols Over the Atmospheric Radiation Measurement Southern Great Plains Site Measured versus Modeled, *Fifteenth ARM Science Team Meeting Proceedings, Daytona Beach, Florida, 2005*) and another by Guibert et al, 2005 (The vertical distribution of aerosol over Europe—synthesis of one year of EARLINET aerosol lidar measurements and aerosol transport modeling with LMDzT-INCA) Atmos Env., 2005.

Our ten-year lidar climatologies collected in Rome confirm the exponential behaviour even at a site where Saharan dust advections often affect the aerosol column (as in Fig 6b of Guilbert (2005) or in Gobbi et al., Atmos. Chem. Phys. (2004).

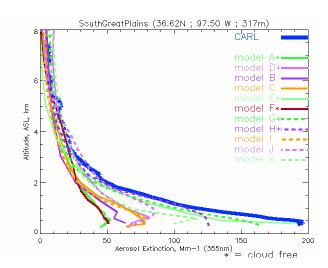


Figure 3. (Ferrare et al. 2005) Average annual extinction profile from the various models and from the Climate Research Facility Raman lidar data. Note the differences in the average vertical distributions, even among models that have similar average aerosol optical thickness.

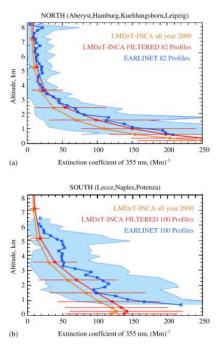


Fig. 6. (Guibert et al. 2005). Comparison of the average lidar aerosol extinction coefficient vertical profile (blue curve), with indication of the 16–84% percentiles (blue area), with two average modeled profiles (filtered for the presence of measurements with indication of the 16–84% percentiles—red thick curve and horizontal red line—and unfiltered all year mean—orange curve). Mean for (a) the northern European stations, i.e. Aberystwyth, Hamburg, Kühlungsborn and Leipzig, and (b) the southern European stations, i.e. Lecce, Naples and Potenza.

Furthermore, it should be noticed that the scale height H_a used in our work to describe the exponential decrease of the extinction $(ext(z)=ext(0) exp(-z/H_a))$ can also be seen as the height of the column a homogeneously mixed aerosol (with constant extinction ext(0) from ground to H_a) would reach: both these vertical distributions of the extinction have as integral the same AOD. This "box distribution" of the aerosols is the condition expected by Referee #2. Even so, scale or column heights are always of the order of a fraction of km or few km, not tens or hundred km as encountered mainly (but not only) in our monsoon observations (e.g., Fig. 6 and related discussion). These latter conditions can only be generated by elevated layers with AOD larger than the aerosol AOD. We then believe this analysis is a good one to attribute the high monsoon coarse particles AODs over the NCO-P to the presence of elevated, thin cirrus-like layers.

7. Figure 6a. As previously mentioned I suggest showing the plots of Angstrom Exponent for each station along with AOD plots. The low values of AE during the monsoon need to be more clearly discussed with respect to cloud contamination.

AR: The importance of keeping the Fine mode and Total AOD analysis rather than single Angstrom analysis alone has been discussed at point 4 above.

8. Page 1204. I think some additional explanation is needed with respect to what the authors mean by radiative forcing in light of the statement 'cirrus-like clouds appear to be more important than aerosols at determining the AOD and the consequent radiative forcing...'. I assume the authors do not mean *anthropogenic* radiative forcing? In general the forcing of interest with respect to aerosols is anthropogenic forcing and it is unclear what the link is between cirrus clouds and aerosols during the monsoon. Hence I am not sure of the utility of this finding.

AR: The Referee is right. We meant the aerosol <u>radiative</u> effect cannot be easily addressed because of such contamination. Note also, that we always refer to the aerosol load as a whole because, as also highlighted at point 4 above, we cannot separate anthropogenic from natural contributions Where necessary, we shall change the revised manuscript to properly address radiative effects from forcings.

9. Figure 7. The CALIOP data seems out of place in this paper. It does, once again, highlight the existence of clouds and their potential impact on AOD retrievals. I am not sure the analyses belong in this

AR: The CALIOP data highlight the existence of high, depolarizing (as described in the manuscript this indicates frozen/non-spherical rather than liquid/spherical droplets) clouds over the Himalayas (in particular the monsoon transect of line "b". These transects also illustrate the extent and patterns of aerosols over the region in the various seasons we address. We believe these plots as very useful at supporting the height-integrated vision provided by the sunphotometer and discussed in the paper.