

Authors' replies to the Referee's comments on "Scattering and absorption properties of near-surface aerosol over Gangetic–Himalayan region: the role of boundary layer dynamics and long-range transport"

We would like to express our sincere gratitude to the anonymous referee for his/her insightful and constructive comments on this study. All the comments and suggestions are highly valuable for us to improve the quality of the manuscript. We incorporated and edited whole manuscript as suggested by both reviewers. Our point-by-point reply to the referee's comments is listed below. The comments are indicated in black and our responses in italic blue.

Responses to comments by Anonymous Referee #1

I have two minor comments before the paper can be published in ACP.

1) Table 2 should present references not only to Andrews et al. (2011) but also to the original papers included in Andrews et al. (2011).

These are for example Marcq et al. (2010) for NCO:

Marcq, S., Laj, P., Roger, J. C., Villani, P., Sellegri, K., Bonasoni, P., Marinoni, A., Cristofanelli, P., Verza, G. P., and Bergin, M.: Aerosol optical properties and radiative forcing in the high Himalaya based on measurements at the Nepal Climate Observatory-Pyramid site (5079 m a.s.l.), *Atmos. Chem. Phys.*, 10, 5859–5872, doi:10.5194/acp-10-5859-2010, 2010.

Collaud Coen et al. (2004; 2007) for Jungfraujoch:

Collaud Coen, M., Weingartner, E., Schaub, D., Hueglin, C., Corrigan, C., Henning, S., Schwikowski, M., and Baltensperger, U.: Saharan dust events at the Jungfraujoch: detection by wavelength dependence of the single scattering albedo and first climatology analysis, *Atmos. Chem. Phys.*, 4, 2465–2480, doi:10.5194/acp 4-2465-2004, 2004.

Collaud Coen, M., Weingartner, E., Furger, M., Nyeki, S., Prévôt, A. S. H., Steinbacher, M., and Baltensperger, U.: Aerosol climatology and planetary boundary influence at the Jungfraujoch analyzed by synoptic weather types, *Atmos. Chem. Phys.*, 11, 5931–5944, doi: 10.5194/acp-11-5931-2011, 2011

Moreover, other recent papers describing aerosol optical properties at mountaintop sites not

included in Andrews et al. (2011) should be presented in Table 2.

For example:

Pandolfi, M., Ripoll, A., Querol, X., and Alastuey, A.: Climatology of aerosol optical properties and black carbon mass absorption cross section at a remote high-altitude site in the western Mediterranean Basin, *Atmos. Chem. Phys.*, 14, 6443-6460, doi:10.5194/acp-14-6443-2014, 2014.

Authors: *We updated Table 3 by including retrievals from the suggested papers.*

2) I have already commented this in my previous review. Table 1 still includes a parameter (BAE) which was not presented/commented neither in the text nor in the Figures. Given that the authors present BAE in Table 1 I would like to see more comments on this parameter in the text (not just removing it from the Table). Which is the relationship between BAE and SAE? Which additional information can be drawn using BAE compared to the SAE? Is BAE a parameter which add useful information compared to SAE?

Authors: *In the revised manuscript, we included the seasonal mean values of backscatter coefficient and BAE in a new Table 2. Moreover, the monthly variation of the spectral BAE is shown in the new Figure 3 and discussed in comparison to that of SAE (Fig. 2). We found that SAE and BAE are strongly correlated to each other and spectral BAE has larger sensitivity on the particle size than SAE. Therefore, BAE can also be used for the aerosol type discrimination, but it does not seem to add specific information due to covariance with the SAE spectral dependence.*

Responses to comments by Anonymous Referee #2

“On the other hand”

This phrase is over used and usually is out of context or doesn't add to the sentence content. I suggest deleting all uses of this phrase as it detracts from the paper.

Authors: The phrase “on the other hand” was omitted throughout the revised version, following the reviewer’s suggestion.

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The filter was changed whenever the amount of transmitting light achieved is ~70% of the initial intensity

The filter was changed whenever the site technician was allowed into the trailer and not when the filter transmission dropped to 0.7. The text should read “Absorption data with filter transmissions below 0.7 were rejected in this study.”

Authors: This sentence was rephrased as suggested by the reviewer.

Following the methodology from several previous works (Bond et al., 1999; Ogren, 2010; Virkkula et al., 2011), the raw PSAP data were processed to estimate the σ_{ap} by incorporating the sample area, flow rate and spot size calibrations. Other biases are due to the scattering and multi-sample ...

The correction of Ogren is an additional correction that is applied to the Bond et al., correction. The Virkkula correction is different than that of Bond et al. The data downloaded from the ARM archive uses the Bond and Ogren corrections and not the Virkkula correction.

Authors: We are thankful to the reviewer for this observation. In the revised version, we deleted the reference to Virkkula et al. 2011, since the downloaded ARM dataset did not use this correction.

The angular non-idealities (i.e. truncation error) and non-Lambertian light source were corrected following the methodology described by Anderson and Ogren (1998) and details given in Dumka and Kaskaoutis (2014 and references therein). These corrections are needed to subtract the light scattering by air molecules, the instrument walls and the detector background noise.

The wording in the above paragraph is awkward. The truncation correction is unrelated to the subtraction of background scattering from the walls and filtered air. I suggest removing the sentence “These corrections are needed to subtract the light ...”

Authors: *As suggested, the sentence was removed from the revised manuscript.*

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It should be noted that the properties of $D_{1\mu\text{m}}$ particles are also included in the $D_{10\mu\text{m}}$, but with much lesser contribution. All the examined variables present a slight decreasing trend with wavelength, but the largest differences are seen as a function of the particle size, since $D_{10\mu\text{m}}$ particles exhibit higher scattering, backscattering and absorption as well. On the other hand, the range of all variables is larger for $D_{10\mu\text{m}}$ particles since their size distribution is much more expanded, suggesting larger variability in source regions, mixing processes and optical properties. Although such a behavior is expected for the scattering and backscattering processes via the Mie theory (the larger particles are more efficient scatterers especially at longer wavelengths), the higher absorption by the larger particles is an important finding of the GVAX campaign.

The above sentences should be removed from the discussion. These are obvious and don't need to be stated. Unless there is an instrument error it isn't possible for the subum scattering and absorption values to be greater than those of the sub 10 um aerosol.

Authors: *Following the reviewer's suggestion, the sentences were deleted from the revised version. Moreover, we removed the earlier Figure 2, on which these discussions are referred and it was replaced with the new Table 2 (as suggested by the reviewer). In this Table, we included the seasonal mean values of extensive and intensive aerosol properties. The related text also has been modified accordingly.*

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It should be noted that the difference in absorption coefficient between $D_{10\mu\text{m}}$ and $D_{1\mu\text{m}}$ was found to be larger for higher aerosol loading. On the other hand, the scattering is much larger (~40%) for the $D_{10\mu\text{m}}$, especially at longer wavelengths, while the σ_{bsp} exhibits rather neutral wavelength dependence.

Refer here to the R_{ap} and R_{sp} values. Scattering and absorption values will always be higher for sub 10um than sub 1um aerosol.

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Authors: *This text has been modified in the re-revised version, since the figure has been replaced by a Table. However, we do not emphasize here on the seasonal variation of scattering and absorbing coefficients, since, as stated in the manuscript, this was analyzed in a previous paper (Dumka and Kaskaoutis, 2014). The R_{ap} and R_{sp} values are now included in the new Table 2, so this discussion matches with the Table.*

Lower values of σ_{sp} ($97 \pm 9 \text{ Mm}^{-1}$) and higher of σ_{bsp} ($14 \pm 1 \text{ Mm}^{-1}$) at 550 nm compared to Nainital were found in Anantapur, south India during January – December (Gopal et al., 2014), suggesting dominance of different aerosol types.

This sentence is awkward. Refer instead to the backscatter fraction at both sites to compare aerosol types rather than scattering and backscattering values as these only depend on the aerosol loading and not the aerosol type.

Authors: *This sentence was rephrased as “Higher values of σ_{bsp} ($14 \pm 1 \text{ Mm}^{-1}$) and b (~ 0.14) at 550 nm compared to Nainital were found in Anantapur, south India during January – December 2011 (Gopal et al., 2014), suggesting dominance of different aerosol types associated with lower aerosol loading (mean σ_{sp} of $97 \pm 9 \text{ Mm}^{-1}$).” We maintained in the discussions the values of scattering and back-scattering coefficients at Anantapur for comparison purposes.*

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The SAE follows an anti-correlation with the coarse-to-fine mode ratio exhibiting much higher values for sub-micron aerosols, especially at shorter wavelengths (Andreae et al., 2002).

This is obvious and doesn't need stating.

Authors: *This sentence has been deleted.*

The spectral $D_{1\mu\text{m}}/D_{10\mu\text{m}}$ SAE ratio is nearly constant to 1.68 suggesting that $D_{10\mu\text{m}}$ particles possess higher scattering at longer wavelengths leading to a more neutral spectrum (Manoharan et al., 2014).

This is obvious and doesn't need stating. 10 μm aerosol will always have higher scattering at longer wavelengths than 1 μm aerosol and hence a lower SAE.

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Authors: *This sentence was removed in the re-revised version.*

Thus, despite the fact that $D_{1\mu\text{m}}$ and $D_{10\mu\text{m}}$ particles exhibit similar annual pattern for scattering and absorption (Dumka and Kaskaoutis 2014), the values and the wavelength dependence may be quite different, indicating that the particle size plays a prominent role in altering the aerosol optical properties and wavelength dependence.

Can you restate this sentence, as it doesn't make much sense? It would be better to compare the seasonal variations in R_{ap} and R_{sp} .

Authors: *This sentence has been rephrased as "Previous work (Dumka and Kaskaoutis 2014) showed a similar annual pattern of σ_{sp} and σ_{ap} for both $D_{1\mu\text{m}}$ and $D_{10\mu\text{m}}$ particles. The present analysis revealed different values and wavelength dependence, indicating that the particle size plays a prominent role in altering the aerosol optical properties." The comparison between R_{ap} and R_{sp} as a function of particle size is presented in the new Table 2.*

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The larger SAE that was found in monsoon (Fig. 3) indicates particles of smaller size, which are associated with more isentropic scattering and smaller b values. During post monsoon and winter, the b increases (except a small decrease in November) reaching its highest value in March suggesting more irregular type of scattering and favoring of backscatter, which is characteristic of the dust particles (Liu et al., 2008). The b is larger at longer wavelengths, especially for the $D_{1\mu\text{m}}$ particles, since the backscatter wavelength dependence is lower than that of total scattering (Fig. 2) and, therefore, the backscatter-to-total scattering ratio (b) is more enhanced at longer wavelengths. Slight higher b values are found for the sub-micron particles over Nainital at 0.45 and 0.55 μm , which become significantly higher at 0.7 μm compared to those of $D_{10\mu\text{m}}$ (Fig. 5).

Smaller particles have both higher SAE and higher backscattering fractions. Isotropic scattering is indicative of smaller particles. Forward scattering and smaller backscattering fractions are indicative of larger particles. It would be good to plot Mie calculations of SAE and bsf as a function of particle size to understand how these two variables coincide and when they don't coincide. Lack of a covariance usually indicates a bimodal distribution.

Authors: *This paragraph has been modified in some part, including reviewer's notes about covariance between SAE and b. We could not plot Mie calculations of SAE and b as a function of size, due to non-availability of code with us readily.*

Both fractions are below 1, especially for the scattering as was seen in Fig. 2, suggesting that the particles larger than 1 μm contribute more to scattering and absorption.

There is a fundamental misunderstanding of the measurements that is repeated through the paper. The submicron scattering and absorption values will always be less than 1.0. Sub 10um indicates all particles less than 10um. It contains both the coarse and accumulation mode aerosol, not just the coarse mode. Hence the sub10um scattering and absorption values will always be greater than or equal to the subum values.

Authors: *This sentence was removed in the re-revised manuscript.*

The sub-micron absorption fraction is higher than that of scattering suggesting that the SSA would be higher for D1 μm , as justified (SSA for D1 μm = 0.93, SSA for D10 μm = 0.91) in a previous study

Higher absorption in the subum mode implies a lower SSA, not higher.

Authors: *We thank the reviewer for this observation. We corrected the sentence as well as the values of SSA according to our previous publication (see Fig. 8 in Dumka and Kaskaoutis, 2014).*

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For a certain absorption value, the scattering of D10 μm particles was larger than that of D1 μm for σ_{sp} values above 200 Mm^{-1} indicating that for clean atmospheres the discrimination of the optical properties between super-micron and sub-micron aerosols is really difficult.

Again there is a misunderstanding of the aerosol size modes. Best to compare R_{ap} and R_{sp} as a function of loading.

Authors: *This sentence has been rephrased in the re-revised manuscript. We discussed briefly the results of SAE, AAE vs scattering coefficient as well. As stated in previous parts of the manuscript, the sub-micron absorption and scattering fractions are*

somewhat lower for higher aerosol concentrations (i.e., in November and March). This can be explained by combining Figs. 6 and 7.

Figure 2 can be removed and the information placed in a table.

Authors: *Figure 2 has been removed and replaced with Table 2 in the re-revised version.*

We hope that the above fresh inclusions, modifications and general edits in the re-revised manuscript help in its improvement and would be accepted.