

Response to Referee #2

We thank the referee for their review, which helped us improve the manuscript. Below are the original comments in italics with our responses in bold text.

The manuscript presents the algorithm for analysis of aerosol mixture composition, based on expected EarthCare observations. Authors consider 4 main aerosol components (two types of fine and two types of coarse mode particles) and try to present the observed particle volume as a sum of these components volume, using OEM approach. Atmospheric aerosol is a complicated object and particle parameters are strongly variable, still this looks like a reasonable approach, because number of independent observations from EarthCare will be very limited. Manuscript is well written, contains new important results and is suitable for AMT. For me, personally, was interesting to see, how additional 532 nm measurements will influence the results obtained from UV channels.

Thank you for the positive feedback on our work.

Technical comments.

p.6. Choice of particle model parameters is always an issue. In particular, parameters depend on RH. This especially critical for maritime aerosol and for small non-absorbing particles. Probably authors could mention range of RH, where their model is applicable.

Indeed, the dependence of the microphysical properties on the relative humidity is not mentioned in the paper, because it is explored in detail in Wandinger et al., 2023a. For clarity, we have included the following sentence in the revised manuscript (lines 170-172): “The justification for the choice of microphysical parameters is described in Wandinger et al. (2023a), including a discussion on the relative humidity dependence of the size and refractive index of the particles.”

Table.2. Authors assume that depolarization ratio of small particles is 3%. In practices, however, we often observe smoke and sulfate particles with depolarization up to ~8% or even higher. Small values of assumed depolarization ratio probably may increase contribution of dust particles in the examples provided in Fig.11.

For smoke particles the lidar ratio at 532 nm is usually significantly larger than at 355 nm. Can it be obtained from component parameters in Tab.2? The same question is for dust. In experiment lidar ratio of dust at 355 sometimes significantly higher than at 532.

In HETEAC, the model on which HETEAC-Flex is based, the particle linear depolarization ratio for all three spherical components is 0%. However, the scattering properties that were used in HETEAC-Flex were slightly adjusted to better reflect the experimental observations (Floutsi et al., 2023). These adjustments are also explicitly discussed in lines 169-175 of the original manuscript. Indeed, the particle linear depolarization ratio of the pure fine, spherical non-absorbing (FSNA) component is approx. 3% at 355 and 532 nm. This aerosol component resembles more pollution-related aerosol than smoke (the latter is absorbing and, thus, associated with the FSA component). Our long-term lidar-based collection of intensive aerosol properties indicates that the particle linear depolarization ratio of pollution-related aerosol is around 1.1 ± 0.3 and 2.8 ± 1 % for 355 and 532 nm, respectively (see Table 1 and Sect. 3.1.5 in Floutsi et al., 2023). Therefore, the choice of particle linear depolarization ratio is justified.

A particle linear depolarization ratio of approx. 8% appears to be rather high for spherical particles. In the aforementioned case of smoke, it would indicate the presence of non-spherical soot particles in the aerosol mixture, usually observed in the upper troposphere (e.g., Burton et al., 2015).

However, in the case of elevated particle linear depolarization ratio, as correctly noticed, the contribution of the coarse, non-spherical (CNS) component will increase. One important aspect is that while the CNS component resembles dust particles, it does not necessarily imply the presence of dust. Other non-dust particles may also be coarse mode and non-spherical.

Based on the scattering properties of Table 2, the wavelength dependence of the lidar ratio (higher 532-nm/lower 355-nm) for smoke particles cannot be captured. This wavelength dependence is the most significant signature of aged smoke and stratospheric smoke, rather than fresh tropospheric smoke. In addition, the wavelength dependence would only impose a problem in the HETEAC-Flex retrievals for the retrieval modes that utilize the information from both wavelengths simultaneously, i.e., modes 5 and 6. With that being said, we should mention that we are aware of this issue and that currently there is ongoing research within the framework of the CARDINAL (Clouds, Aerosol, Radiation – Development of INtegrated Algorithms) project, aiming to define the microphysical/optical properties of stratospheric aerosol components (HETEAC-2) for EarthCARE.

The wavelength dependence of the lidar ratio for dust is well captured by HETEAC and can be resolved by HETEAC-Flex.

Ln.231. As I understand, sometimes contribution of a single component may exceed 1.0. Just wonder, if it is possible to add additional constraint, that the sum of all components must be 1.0?

In theory the contribution of a single component may exceed 1, but this is already tackled by a specific constraint, i.e., the constraint described by Eq. 8 prohibits that from happening. As explained in lines 231- 242 of the original manuscript, if the relative volume contribution of an aerosol component exceeds 1, then the penalty function adds a large term to the cost function, and therefore, for this solution the cost function is not minimal.

Ln.238. "... if the total relative volume contribution (sum of the relative volume contribution per aerosol component) is greater than 1, then the state vector is normalized"

This is actually related to my previous question.

Indeed, it is related to the previous question, and is one of the constraints to prevent the issue you raised.

Ln.239. "...There is no constraint in place in the case of a total relative volume contribution that is less than 1."

Why no normalization this case?

As stated in lines 240-241 of the original manuscript version, in case of the total volume contribution of less than 1 we chose to attribute the missing volume contribution to uncategorized aerosol rather than normalizing the state vector. We chose to do so to avoid over-constraining the solution space.

Fig.5. Probably no reason to show lidar ratio and extinction below 1 km.

Thank you, we have updated the Figure accordingly. In addition, due to a suggestion from Referee #3 now Fig. 5 includes the backscatter-related color ratio.

References

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Floutsi, A. A., Baars, H., Engelmann, R., Althausen, D., Ansmann, A., Bohlmann, S., Heese, B., Hofer, J., Kanitz, T., Haarig, M., Ohneiser, K., Radenz, M., Seifert, P., Skupin, A., Yin, Z., Abdullaev, S. F., Komppula, M., Filioglou, M., Giannakaki, E., Stachlewska, I. S., Janicka, L., Bortoli, D., Marinou, E., Amiridis, V., Gialitaki, A., Mamouri, R.-E., Barja, B., and Wandinger, U.: DeLiAn – a growing collection of depolarization ratio, lidar ratio and Ångström exponent for different aerosol types and mixtures from ground-based lidar observations, *Atmos. Meas. Tech.*, **16**, 2353–2379, <https://doi.org/10.5194/amt-16-2353-2023>, 2023.

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