

## Reply on RC1

Lei Li et al.

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Author comment on "Climatology of aerosol component concentrations derived from multi-angular polarimetric POLDER-3 observations using GRASP algorithm" by Lei Li et al., Earth Syst. Sci. Data Discuss., <https://doi.org/10.5194/essd-2022-108-AC1>, 2022

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This study presents the climatology of aerosol mass burden for different components based on a newly developed algorithm GRASP/Component with the input of POLDER-3 multi-channel radiance observations. Global observations of aerosol component concentration are crucial for both the estimation of aerosol effective radiative forcing from satellite observations and also the evaluation of the performance of global aerosol models, but retrieving those information from satellite has been challenging. Most of previous studies just used the column aerosol optical properties (e.g., AOD/AODf/AI) to estimate aerosol forcings, it seems somewhat rough but is the only feasible option. The dataset presented here is thus very valuable and useful for our scientific community. This manuscript is well organized, the analysis methods are technically sound. I personally think this manuscript is suitable for publication after a minor revision.

**Response:** Thank you very much for the time and efforts you have put into reviewing the manuscript. We are very grateful for your positive evaluations and helpful comments on our work, which have enabled us to improve the manuscript. Here are our point-by-point responses to the comments:

### Detail comments:

- Figures showing annual mean concentrations of different aerosol components (2005 - 2013) are quite interesting, but not much mentioned in the main text. Many recent studies revealed the aerosol effective forcing changed during last two decades due to the change in anthropogenic emissions (<https://doi.org/10.5194/acp-2022-295>;

<https://doi.org/10.5194/essd-12-1649-2020>). I feel that it could be quite interesting to plot the long-term trends of different aerosol components over main industrial regions (West Europe, East U.S., China and India) and discuss the potential implication or linkage to the observed radiative forcing trend.

**Response:** We appreciated the reviewer's comment and suggestion very much. As the referee mentioned, the linear trends of different aerosol components and also the potential implication or link to the observed radiative forcing trend are highly interesting

and merit a more focused effort. We also agree that a regionally zoomed analysis of the linear trends of components, as well as their potential implications, should be in scope of future studies. At present, due to very large volume of produced data in this publication and limited resources, we have added only a global overview of linear trends of components. We also added a discussion on potential implications, including two citations (see below), in order to highlight the scientific significances of the presented in our study component retrievals.

We have added the text in the abstract of the revised version (lines 43-45): "The extensive satellite-based aerosol component dataset is expected to be useful for improving global aerosol emissions and component-resolved radiative forcing estimations."

Also, lines 807-821 and 865-870 in the revised version: "More importantly, the data assimilation of this extensive satellite-based aerosol component dataset can importantly contribute to improving global aerosol emissions estimation and further improvement of accuracy of the estimated aerosol radiative forcing in general and per aerosol component in particular. For instance, the AOD and the AAOD products derived from POLDER-3 observations have been already used to constrain GEOS-Chem inverse modeling for the improvement of global black carbon, organic carbon and desert dust aerosol emissions (Chen et al., 2018, 2019). Using the presented in this study additional satellite-based aerosol component products, the further improvement of global aerosol emissions estimation is thus expected. The presented efforts are also in line with the studies suggesting that the employment of satellite-constrained anthropogenic and natural aerosol emissions by the climate models is required to improve the accuracy of aerosol radiative forcing estimations (e.g., Bellouin et al., 2020; Quaas et al., 2022). That is, the linear trends in column concentration of the main aerosol components, such as BC, BrC, CAI and CNAI as shown in Fig. S1 in the supplement, can provide a better global scale satellite-measured constraints on the properties of anthropogenic (BC and BrC) and natural (CAI and CNAI) aerosols and will contribute to improving the accuracy of anthropogenic aerosol radiative forcing estimations."

Fig S1. Linear trends in column concentration of BC, BrC, CAI and CNAI components with the criteria of AOD (440 nm) > 0.2 and for BC (> 1 mg/m<sup>2</sup>), BrC (> 10 mg/m<sup>2</sup>), CAI (> 2 mg/m<sup>2</sup>), and CNAI (> 50 mg/m<sup>2</sup>).

#### Reference:

Bellouin, N., Davies, W., Shine, K. P., Quaas, J., Mülmenstädt, J., Forster, P. M., Smith, C., Lee, L., Regayre, L., Brasseur, G., Sudarchikova, N., Bouarar, I., Boucher, O., and Myhre, G.: Radiative forcing of climate change from the Copernicus reanalysis of atmospheric composition, *Earth Syst. Sci. Data*, 12, 1649–1677, doi: 10.5194/essd-12-1649-2020, 2020.

Quaas, J., Jia, H., Smith, C., Albright, A. L., Aas, W., Bellouin, N., Boucher, O., Doutriaux-Boucher, M., Forster, P. M., Grosvenor, D., Jenkins, S., Klimont, Z., Loeb, N. G., Ma, X., Naik, V., Paulot, F., Stier, P., Wild, M., Myhre, G. and Schulz, M.: Robust evidence for reversal in the aerosol effective climate forcing trend, *Atmos. Chem. Phys. Discuss.*, 1–25, doi: 10.5194/acp-2022-295, 2022.

- Line 281: How can the authors conclude 'the GRASP/Component provided the overall most consistent both total and detailed aerosol properties' based on the findings by Zhang et al. (2021)? I don't understand the causal link. Could the authors develop a bit on this?

**Response:** We appreciated the reviewer's suggestion very much. We added more descriptions and explanations in the revision (Lines 311-317): "Indeed, Zhang et al. (2021) validated GRASP/Component optical properties against AERONET data and concluded that generated total AOD values have minimal bias both over land (-0.02 for 550 nm) and ocean (0.01 for 550 nm), similar to total AOD provided by GRASP/Models, while the detailed properties such as AE, AODF and AODC have similarly good validation metric as GRASP/HP. This suggests that the GRASP/Component products provide the overall most consistent both total and detailed aerosol properties (e.g., spectral AODF, AODC, and SSA etc.) with respect to previous GRASP (Models, HP, and Optimized) products."

- Figure 2: I guess STD here is calculated from daily data? Anyway, it is better to specify how the authors do the calculation.

**Response:** Thank you for this comment. We added more descriptions and explanations in the revised version (Lines 346-348): "Figure 2 shows the corresponding standard deviations (STD, calculated from all months data during the period 2005–2013, same for other components in the following) divided by the mean of BC concentration."

- Also, in my opinion, the manuscript would benefit from editing help from someone with full professional proficiency in English.

**Response:** We appreciated the reviewer's conscientious correction very much and we also have checked the grammar and typos carefully and corrected them in the revised manuscript.

#### **Minor corrections:**

- Lines 24: "an intermediate retrievals" -> "intermediate retrievals"

**Response:** Done.

- Line 30: "concentration" -> "concentrations"

**Response:** Done.

- Line 32: "aerosol" -> "aerosols"

**Response:** Done.

- Line 45: the full name of IPCC is needed when first mentioned.

**Response:** It was added in the revised version.

- Line 58: "the importance of having sufficient spectral resolution of measurements to capture the differences" -> "the importance of sufficient spectral resolution of measurements on capturing the differences"

**Response:** Done.

- Line 85: I feel that "and sometimes the same" is not necessary here.

**Response:** It was removed in the revised version and more explanations were added to make it clear.

- Lines 101: "has" -> "have"

**Response:** Done.

- Line 106: "the impact of satellite polarimetry on aerosol monitoring remains fairly." I do not quite understand this sentence. Can the authors rephrase it?

**Response:** It is rephrased and clarified (Lines 115-123): "However, due to rather limited amount of available multi-angle polarization observations and the complexity in their interpretation, the added value of satellite polarimetry on aerosol monitoring remains questionable. Indeed, the polarimetry has enhanced sensitivities to numerous atmospheric parameters and inversion algorithms are required to consider all these sensitivities adequately. Partially due to this complexity, the practical advantages of multi-angular polarimetric retrieval were not convincingly exhibited by the available operational multi-angular polarimetric products in the past for a long time and only recently the advanced aerosol products (including present study) make the advantages of the polarimetry for aerosol remote sensing more evident (e.g., see discussion in Dubovik et al., 2019, 2021b)."

- Lines 122-123: the full names of MAP and SRON are missing.

**Response:** We added the full names of MAP and SRON in the revision.

- Line 147: "This is significant" -> "This is a significant"

**Response:** Done.

- Line 160: ". This study" -> ", this study"

**Response:** Done.

- Line 183: "has" -> "have"

**Response:** Done.

- Line 223: "0.1° and 1°" did the authors mean 0.1° and 0.1°?

**Response:** There are two different spatial resolution for Level 3 products: 0.1° and 1°. We rewrote the sentence in the revision (Line 244) to clarify that as "There are two different spatial resolution of 0.1° and 1° for the Level-3 products".

- Line 225: It is better to clarify how the authors transform from original resolution to MERRA-2 resolution, by interpolation or aggregation?

**Response:** We added more descriptions and explanations to clarify it in the revision.

- Line 234: "contribution" -> "contributions"

**Response:** Done.

- Line 239: "factions" -> "fraction"

**Response:** Done.

- Line 241: Can authors explain "fractions of 6 components" a bit more? Is it the mass fraction or extinction fraction?

**Response:** We added more descriptions and explanations in the revision (Lines 267-272): "Thus, the main conceptual difference of GRASP/Component from GRASP Optimized and High Precision is the retrieval of volume fractions of six components (black carbon, brown carbon, fine- and coarse-mode non-absorbing insoluble, coarse-mode insoluble absorbing, mainly representing iron oxides in mineral dust, and relative humidity for the host calculation) instead of direct retrieval of the real and imaginary parts of complex refractive index at each wavelength (12 parameters in GRASP Optimized and High Precision)".

- Line 253: "seem" -> "seems"

**Response:** Done.

- Line 277: "and considering the apparent of aerosol composition climatological patterns." this sentence is difficult to read with grammar error (apparent of). Can the authors rephrase it?

**Response:** Sorry for this mistake, the revised version is (Lines 308-310): "Therefore, the main focus of this study is on the analysis/verifications of aerosol composition (fractions) retrievals and considering the apparent climatological patterns of aerosol composition. apparent of aerosol composition climatological patterns".

- Line 310: "columnar" -> "column"

**Response:** Done, also through the whole manuscript.

- Line 619: "Moreover, possible the hygroscopicity of aged dust." Can the authors rephrase this sentence to a more readable format?

**Response:** Thank you for your attention, the revised version is (Lines 672-680): "The aerosol water presence over ocean during the dust transport and near southern Africa during the biomass burning is also rather logical because of higher atmospheric relative humidity. Moreover, a significant increase of mineral dust hygroscopicity was attributed in several studies to the aging processes and the dust mixing with soluble hygroscopic material (Sullivan et al., 2009; Tang et al., 2016). Impact of such mixtures on remote sensing observations was also observed (Derimian et al., 2017; Falkovich et al., 2004)."