

We would like to thank the two anonymous reviewers for their comments. We present below our responses to these comments as well as the modifications we made in the paper following these comments.

In order to respond to the different comments and to make the article more robust, we reran our simulations to diagnose the 5 AAOD and SSA of the different aerosols of the TACTIC scheme. Several figures have been added in the revised article. Figure 7 shows the contribution of the model BrC absorption to the total aerosol absorption. In the light of these results we added in our article the evaluation of the aerosol AAOD and SSA at 350 nm (Figures 8 and 10).

Anonymous Referee 1

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General Overview:

This manuscript describes model-calculated aerosol extinction and absorption optical depth (AOD and AAOD) as well as single scattering albedo (SSA) by the ARPEGE-Climat global climate model. The authors give a detailed description of the modelled processes and parameterizations and evaluate their results by comparison to ground-based and satellite observations.

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General Comments:

The weakest part of the presented work in the evaluation of their results using satellite observations. For this task they have used the POLDER-GRASP aerosol product as well as an OMI aerosol product.

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The accuracy of POLDER-GRASP retrievals has been evaluated by comparisons to AERONET observations as documented in the provided references. As for the OMI aerosol product, the authors seem unaware of the availability of two quite different OMI aerosol products: OMAERO and OMAERUV.

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For the OMI-OMAERO satellite product they have used, the authors do not provide any references on the evaluation of AOD and SSA with ground-based observations. Because of the large differences in retrieved aerosol parameters between the two satellite datasets (POLDER-GRASP and OMI-OMAERO), and between the model and OMI-OMAERO as reported in the different tables and figures, especially over the oceans, it is important to properly document the expected accuracy of the reference satellite data sets.

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Without a literature-supported accuracy analysis of the OMI-OMAERO aerosol data, this satellite-product should not be used as a reference data set. This issue as well as the minor comments below should be addressed for the paper to be published.

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We have clarified the OMI aerosol product used in this study: it is the OMAERUVd satellite product. For clarity we now use the term OMI-OMAERUVd instead of OMI in the article. Its description has therefore been updated and detailed with relevant references on the evaluation of SSA with ground-based observations (Jethva et al. 2014; Drakousis et al. 2020).

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We have added the following paragraph in our manuscript: "- OMI-OMAERUVd (2005-2019, 1°resolution, Torres et al. 2007, 2013) for the AAOD and SSA (350 and 440 nm) and the AOD (550 nm). The OMI (Ozone Monitoring Instrument) OMAERUVd dataset comes from a spectrometer aboard NASA's Earth Observing System's Aura satellite and is archived at the

NASA Goddard Earth Sciences Data and Information Services Center. The level 3 daily global gridded product OMAERUVd-v003, used in this study, is produced with all data pixels which fall in a grid box with quality filtered, based on the pixel level OMI Level 2 Aerosol data product OMAERUV. The OMAERUV data product is an improved version of the TOMS version-8 algorithm that essentially uses ultraviolet radiance data (Jethva et al., 2014). The estimated uncertainty in retrieved SSA is of ± 0.03 for AOD (440 nm) larger than 0.4. This error is largely attributed to the uncertainty in the instrument calibration

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(Dubovik et al., 2000; Jethva et al., 2014). AOD over land is expected to have the same root-mean square error (RMSE) as TOMS retrievals (0.1 or 30% whichever is larger). Over ocean, the AOD RMSE is likely to be two times larger. The RMSE for AAOD is estimated to be 0.01 (OMI User's Guide). An evaluation of the OMAERUVd aerosol SSA data through comparisons against daily SSA products from 541 globally distributed AERONET stations for a 15-year period (2005-2019) was carried out in the study of Drakousis et al. (2020). They show that about 50% of OMI-OMAERUVd - AERONET matchups agree within

50 the absolute difference of 0.03 at 440 nm. However, they also indicate that OMI-OMAERUVd tends to overestimate SSA over areas where biomass burning occurs."

Specific Comments:

55 Pg. 6: Line 29. Although OA's can be considered largely non-absorbing in the visible, they do absorb in the UV. In our BrC parameterization, we separate organic aerosols into aerosols emitted by fossil fuel sources (that we still call organic aerosols or OA) and BrC emitted by biomass burning sources and by biofuel sources. And we make the hypothesis that OA are non absorbing aerosols, which is commonly admitted in modelling studies, and that BrC are absorbing aerosols.

60 We reformulated our text that now reads :" In this parameterization, BrC corresponds to organic aerosols emitted by BB and BF while OA corresponds to organic aerosols emitted by FF. At this stage, we consider our OA aerosol as a non absorbing aerosol, as shown by most observations (Laskin et al., 2015), while BrC is considered as an absorbing aerosol."

65 Pg. 7: Line 9. Add Andreae et al (2019) reference. Done.

70 Pg. 9: Line 19. The availability of recently produced global representation of spectral aerosol absorption from the combined use of AERONET AOD and satellite radiances from OMI and MODIS observations (Kayetha et al., 2022) should be mentioned. A sentence has been added in the revised version: "It can be noted that a global representation of the spectral aerosol absorption in the UV-to-visible wavelength range (340–670 nm) based on a synergy of ground measurements (AERONET AOD) and of satellite observations (near-UV OMI radiances and visible MODIS (Moderate Resolution Imaging Spectroradiometer) radiances) is presented in Kayetha et al. (2022)."

75 Pg. 7: Line 24. What does 'most satisfactory results' mean? There are few studies that investigate the level of BrC absorption decrease over time (bleaching effect). To study this parameter, we carried out 3 tests with several absorption decreases (25, 50 and 75% after one day). Pending further studies on this subject, we decided to take an average BrC absorption decrease over time, 50% after one day. This average value correspond in our sensitivity tests to the most realistic results when compared to our reference datasets.

80 We changed our text that now reads: "The best comparison with our reference data sets was obtained with the 50% value. For clarity reasons..."

85 Pg. 7: Line 27. The source of the data shown in Table A1 should be clearly identified. An additional column indicating the source (i.e., reference) should be added. An additional table (Table A2) with the reference for the refractive index of each aerosol type used in the Mie code to compute optical properties has been added.

90 Pg. 7: Line 30. Provide references (or supporting reasoning) for the choice of rain and snow washout efficiencies as well as for BrC fractions content in cloud-mixtures. Please comment on the overall importance of these assumptions (i.e., sensitivity) on the study results. This sensitivity issue is an interesting one that has not been addressed in this work. Due to the lack of data in literature and for consistency reasons we decided to parameterise BrC deposition (wet and dry) as OA deposition. Indeed, we made this choice in order to observe impacts largely related to changes in optical properties. References for the rain and snow washout efficiencies as well as for the BrC fraction content in cloud-mixtures are Michou et al. (2015) and Bourgeois and Bey (2011). These references have been added in the revised version.

95 Pg. 8: Line 2. Provide a literature reported quantitative estimate on predominance of absorption by primary BrC from BB and BF emissions over that of the absorbing SOA. In their study, Wang et al. 2016 indicate "As the absorption from primary OA (Br-POA) from biofuel and biomass burning typically dominates that of absorbing SOA (Br-SOA) (Martinsson et al., 2015; Laskin et al., 2015), the absorption of Br-SOA is much more challenging to detect than Br-POA in most field measurements." Furthermore, Saleh et al. 2014 say "SOA is less absorptive than primary OA, but has a stronger wavelength dependence". In a previous study (Saleh et al. 2013) state: "For the investigated fuels, SOA is less absorptive than POA in the long visible,

100 but exhibits stronger wavelength-dependence and is more absorptive in the short visible and near-UV." The study of Kumar et al. 2018 also indicates: "The corresponding mass absorption cross section of POA (5.5 m² g⁻¹) was higher than that of SOA (2.4 m² g⁻¹) at 370 nm. However, SOA presents a substantial mass fraction, with a measured average SOA / POA mass ratio after aging of ~ 5 and therefore contributes significantly to the overall light absorption, highlighting the importance of wood-combustion SOA as a source of atmospheric brown carbon."

105 On the basis of the above literature, we have modified the sentence of the article: "One limitation of this study is to neglect absorption by biogenic (Lin et al., 2014; Saleh et al., 2015) and aromatic SOA (Wang et al., 2014; Jo et al., 2016; Wang et al., 2018). Some studies show that the absorption of the primary BrC from BB and BF emissions usually dominates that of the absorbing SOA (Saleh et al., 2013; Martinsson et al., 2015; Wang et al., 2016). However, Kumar et al. (2018) indicate in their study that SOA, after aging, can contribute significantly to the overall absorption."

110 Pg. 8: Line 20. The description of the OMI aerosol product is ambiguous. As described in the Torres et al (2007) reference, there are two different aerosol algorithms: OMAERO and OMAERUV. In this work, it looks like the authors refer to the OMAERO product. **Thank you for asking for clarifications on the OMI aerosol product. We double checked what we used, and in this work, we have used OMAERUVd. The description of the OMI aerosol product has therefore been clarified and detailed with relevant references.**

115 Pg. 8: Line 22. Remove the Ahmad et al (2003) and Jethva et al (2014) references. The Ahmad et al (2003) reference is a pre-launch publication irrelevant in the context of the work presented here. The Jethva et al (2014) reference addresses the evaluation of the OMAERUV aerosol product. **The Ahmad et al (2003) reference has been removed. On the other hand, as we use OMI-OMAERUVd, the reference Jethva et al (2014) has been kept.**

120 Pg. 8: Line 24. Provide references for the quoted AAOD and SSA uncertainties (0.01 and 0.03) of the OMI-OMAERO aerosol product. **Done. References for the SSA are Dubovik et al (2000) and Jethva et al (2014). For the AOD and the AAOD the information comes from the OMI User's Guide.**

125 References

Remove Ahmad et al (2003) reference

Remove Jethva et al (2014) reference

References to add:

130 Andreae, M. O.: Emission of trace gases and aerosols from biomass burning – an updated assessment, *Atmos. Chem. Phys.*, 19, 8523–8546, <https://doi.org/10.5194/acp-19-8523-2019>, 2019.

Kayetha, V., Torres, O., and Jethva, H.: Retrieval of UV-visible aerosol absorption using AERONET and OMI–MODIS synergy: spatial and temporal variability across major aerosol environments, *Atmos. Meas. Tech.*, 15, 845–877, <https://doi.org/10.5194/amt-15-845-2022>, 2022.

135 **Done, except for the Jethva et al (2014) reference which is appropriate to the OMAERUV aerosol product.**

Technical Corrections:

Pg. 9: Line 3. Replace OMI with OMI-OMAERO

Done with OMI-OMAERUVd.

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Pg. 9: Line 21. Replace 'including or not' with 'with or without' **Done.**

Pg. 9: Line 23. Replace 'include two members' with 'two modelling configurations' We have rephrased our sentence: "All simulations consist in 30-year AMIP-type simulations with prescribed monthly sea surface temperature (SST) and sea ice fraction. 145 The period covered is 2000–2014, it is simulated twice for each simulation (by changing the initial state of the atmosphere), so the total number of simulated years is of 30."

Pg. 12: Lines 27 and 32. Replace OMI with OMI-OMAERO [Done with OMI-OMAERUVd](#).

150 Pg.13: Lines 4, 8, 15, 17. Replace OMI with OMI-OMAERO [Done with OMI-OMAERUVd](#).

Pg. 16: Line 27. Replace OMI with OMI-OMAERO [Done with OMI-OMAERUVd](#).

155 Pg. 17: Line 32. Replace OMI with OMI-OMAERO [Done with OMI-OMAERUVd](#).

Pg. 29: Replace OMI with OMI-OMAERO in row 1 and caption of Table 2 [Done with OMI-OMAERUVd](#).

Pg. 31: Replace OMI with OMI-OMAERO in column 1 row 7 and caption of Table 4 [Done with OMI-OMAERUVd](#).

160 Pg 32: Replace OMI with OMI-OMAERO in column 7 row 1 and caption of Table 5 [Done with OMI-OMAERUVd](#).

Pg. 37 to Pg. 44: Replace OMI with OMI-OMAERO in legends and captions of Figures 3 to 10 [Done with OMI-OMAERUVd](#).

Appendix: Replace OMI with OMI-OMAERO in legends and captions of Figures A1 to A3 [Done with OMI-OMAERUVd](#).

165