RC2: Review of: Combining POLDER-3 satellite observations and WRF-Chem numerical simulations to derive biomass-burning aerosol properties over the Southeast Atlantic region.


RC2: Manuscript number:

RC2: Recommendation: Accept with Minor Revisions

RC2: This is an interesting study, in which the authors delve into the representation of the SSA and AE in the WRF-Chem model, and use POLDER/GRASP retrievals to improve the model representations, and then examine the impact of changing OC/BC ratios, refractive indices, etc. in the model. It’s a good idea and brings to the forefront the model representations, which can otherwise seem like a black box in publications. My comments are relatively small.

RC2: A main one is that I do not see any support for the idea that dust is present in the marine atmosphere in July. It’s included in WRF-Chem, and the satellite retrievals produce a coarse mode aerosol that could be dust or sea salt, but the authors do not authenticate its presence. Does CALIOP identify dust over the ocean? How about the Haslett or Denjean papers examining the southerly July flow? If the evidence for dust is slim, why not experiment with a model dust representation that excludes dust? I would suggest the authors do so if they cannot better support that the presence of dust is real.

AC: We investigated the vertical profiles of the modeled main aerosol volume concentrations and the aerosol extinction coefficient at 550 nm along the transport of the biomass burning aerosol (BBA) plume over the Southeast Atlantic (SEA) region averaged over July 2008. We plotted the evolution of the chemical composition of carbonaceous aerosols (BC in black and OC in brown), DUST (in yellow) and SEAS (in blue) and the aerosol extinction at 550 nm (COEFF_EXT_550, dotted red curves) over the whole atmospheric column along the transport of the BBA plume over the SEA region, as shown in Fig. A (not shown in the article, see below).
Figure A: Vertical profiles of the aerosol extinction coefficient at 550 nm ($\sigma_{\text{ext}}$ in km$^{-1}$, red dotted curves) and the volume concentrations (in $\mu g\ m^{-3}$) of carbonaceous aerosols, i.e. black carbon (BC, black solid curves) and organic carbon (OC, brown solid curves), desert dust (DUST, yellow solid curves) and sea salt (SEAS, blue solid curves) simulated by the WRF-Chem reference configuration along the path of the biomass burning aerosol plume, from its emission to its long-distance transport over the Southeast Atlantic region, on average for July 2008. Vertical profiles are plotted for the pixels in Fig. 10 respectively for emission sources (left, blue star), halfway transport (middle, red star) and long-range transport (right, orange star).

Figure A firstly shows that the altitude of the BBA plume gradually decreases with a maximum of the aerosol volume concentration located at around 1.5 km altitude (halfway transport) and then 500 m altitude (long-range transport) over the Southeastern Atlantic Ocean. Figure A secondly shows that volume concentration of sea salt aerosols (SEAS, blue solid curves) and desert dust aerosols (DUST, yellow solid curves) gradually increase between the emission sources and the transport of the BBA plume off the African southern coasts. Besides, variations in the aerosol extinction coefficient at 550 nm (COEFF_EXT_550, dotted red curves) appear to be little influenced by the presence of mineral dust from the emission to the removal of the BBA plume. For example, the peak concentration of desert dust aerosols located at about 2.5 km altitude (yellow curve) is not associated with a significant increase in the aerosol extinction coefficient at 550 nm (red dotted curve) at the level of the long-range transport of the BBA plume. This would suggest that the influence of desert dust aerosols on the aerosol optical properties appears weak at 550 nm at least unlike carbonaceous aerosols (black carbon and organic carbon) constituting the BBA plume. We concluded desert dust aerosols are little present within the BBA plume but they become predominant as the BBA plume moves westward and dissipates very far from the southern African coasts.

This characteristic is supported by the recent work of Deaconu et al. (2019) over the same region using a set of satellite observations from the A-Train constellation. They showed, for the 2006-2009 period (including July months), the presence of dust transported above clouds off the coasts of the Southeast Atlantic Ocean (SEAO). The work of Denjean et al. (2020) also confirmed the presence of mineral dust transported from Sahara and Sahel in the norther part of our domain (e.g., near the coasts of Ghana and Ivory coast).

In the paper, after the following sentence in page 11 at lines 291-292: “Desert dust emissions from North African sources are also considered as they may contribute to the total aerosol load over our studied area.”, we added the
following sentence: “The transport of mineral dust off the coast of the SEAO (see figure 1) is notably supported by recent satellite and airborne observations performed in the northern part of our domain (Deaconu et al. 2019, Denjean et al. 2020).”

RC2: An additional comment is that the authors underemphasize the present of BBA in the boundary layer. In July, a large percentage of the total BBA is in the boundary layer, as opposed to above the clouds. The Haslett and Denjean papers document this (as likely others coming out of DACCIWA although I am not as familiar with their literature) as do publications focused on Ascension Island (see, e.g., Zhang and Zuidema, 2021, ACPD and Zuidema et al 2018). ORACLES observations from the month of August also suggest this is likely the case, e.g., Kacarab 2020 ACP and Redemann ACP 2021.

AC: The reviewer is right and this point is now mentioned in the revised manuscript by adding the following sentence at line 60, page 2: “It is worth noting that the transport of BBA in the boundary layer has also been reported by recent airborne observations (Zuidema et al. 2018, Haslett et al. 2019, Denjean et al. 2020, Kacarab et al. 2020, Redemann et al. 2021, Zhang and Zuidema, 2021).”

RC2: Another comment is that the literature references did not make full use of the new results that have come out as a result of ORACLES/CLARIFY/LASIC/DACCIWA, and to the extent that they do, the references tend to be present later in the paper, as opposed to helping to establish the context within the introduction. I have listed some at the end of this, and either mention in the specific comments or as part of the references how I think they contribute.

AC: This is an interesting remark. The bibliography has been now updated in the revised manuscript by adding most of the recent papers you mentioned. An outlook of the current study could consist in applying our methodology to the 2016-2018 period, which corresponds to the recent observation campaigns (AEROCLO-sA, ORACLES, CLARIFY, LASIC and DACCIWA), to take full advantage of the new results on the BBA properties and additional constraints on their radiative and climate effects.

RC2: A small comment is that the English in places sounds labored, using extra words that a native English speaker would leave out. I make a note of a few such spots below.

AC: We would like to thank you for your comments which are helpful to improve the English.

RC2: More minor/specific comments:

RC2: Title: the title is not entirely accurate I feel, as the study is more about using satellite retrievals to constrain the WRF-Chem aerosol representation. Perhaps the authors want to reconsider.

AC: This is an interesting suggestion. However, we prefer to keep the term “combining” instead of using the term “constraining”. Indeed, our approach is fully based on the coupled use of numerical simulations and satellite retrievals, over a specific period and region. The term “constraining” might be wrongly interpreted, as a more global approach of the WRF-Chem aerosol representation.

RC2: Introduction, lines 33-35: worth noting is that southern Africa, which is the focus of this study, by itself produces one-third of the global annual carbon emissions from BB, according to the cited Werf paper.
AC: Thank you for this comment. It has been included in the revised version of the manuscript.

RC2: Intro, line 38: Costantino and Breon 2013 is not really the right reference for documenting the aerosol transport. The African Easterly Jet-South is not yet active in July. Knippertz et al. 2017 might be a better fit for this, or references within.

AC: As suggested, we replaced the Costantino and Breon (2013) reference with that of Knippertz et al. (2017).

RC2: Intro line 38-40: I am not sure the anticyclonic circulation responsible for long-range transport off of the continent is well established by July. Adebiyi and Zuidema 2016 suggests it isn’t. And neither does Fig. 4. Fig. 4 does show an anticyclonic circulation in a couple of places but I don’t see those affecting the regions selected for this study, shown in Fig. 2. Given that the authors have the model winds at their fingertips, perhaps they could say more about the circulations affecting their selected regions? It seems like the land domain might also be influenced by the west African monsoon? Do any of the cited papers discuss July? I am not sure they do.

AC: This is an interesting remark. According to the study of Gargstang et al. (1996), anticyclonic circulation reaches an 80 % daily occurrence in July over the southern Africa. Subsidence also controls the horizontal (and the vertical) transport of aerosols. This point is now clarified in the revised version of the manuscript.

RC2: introduction, 2nd paragraph, p.2: portions of this discussion feel dated, through the focus on the aerosol above clouds and neglect of the boundary layer BBA. Try to update.

AC: Thank you for this remark. We added the following sentence in the revised manuscript at line 60, page 2: “It is worth noting that the transport of BBA in the boundary layer has also been reported by recent airborne observations (Zuidema et al. 2018, Haslett et al. 2019, Denjean et al. 2020, Kacarab et al. 2020, Redemann et al. 2021, Zhang and Zuidema, 2021).

RC2: p.3: This is a nice literature overview. I’m confused why the Denjean, Taylor, Pistone papers aren’t included here. It’s also a very long paragraph, could it be split into two?

AC: Thanks, we split the third long paragraph into two: from line 62 to 79 and from line 80 to 102. We added the suggested papers at line 83, page 3 in the revised manuscript.

RC2: p. 3 line 93: CLARIFY and ORACLES references should be included here.

AC: We agree with the reviewer and we added the Redemann et al. (2021) reference for ORACLES and the Haywood et al. (2021) reference for CLARIFY at lines 93-94, page 3 in the revised manuscript.

RC2: p. 3 line 95: what does ANR stand for?

AC: ANR stands for the French National Research Agency. We replaced “ANR” by “French National Research Agency” at line 95, page 3 in the revised manuscript.

RC2: P.4 line 106: remove ‘to perform’

AC: Thanks, we removed “to perform” at line 106, page 4 in the revised manuscript.
RC2: P. 4 lines 115-118: would be nice to see more documentation of this, is this based on the authors’ own analysis?

AC: We added the Waquet et al. (2020) reference in the revised manuscript. This is based on the POLDER-3/AERO-AC data.

RC2: p. 7 fig 1: why include August and September? They are not used. I would suggest removing these panels.

AC: We included August and September 2008 because the July-August-September quarter is the heart of the dry season. We wanted to qualitatively assess the biomass burning emission inventory of Turquety et al. (2014) used in our study in terms of fire locations from the literature. Figure 1 thus provides a first general, and larger view of BBA emissions over our region of interest.

RC2: p. 11 lines 291-292: on what basis do the authors believe that desert dust emissions from north Africa may significantly contribute to the total aerosol load?

AC: Thank you for this question. Deaconu et al. (2019) clearly showed the presence of desert dust above clouds with mean Angstrom exponent values down to 0.4 and associated mean AOD of 0.2 over the northern part (between 0° and 5°N) of the Southeast Atlantic Ocean studied from May to October 2006 to 2009 (see their Fig. 2-ab). We removed “significantly” in the revised manuscript (line 291, page 11) which could be overstated.

RC2: p. 13, figure 4: is the ECMWF reanalysis the Interim analysis or ERA5? also, the wind vectors are very difficult to read. Please replot with fewer and thicker vectors. It also seems to me, based on Fig. 9, that the fields at 500 hPa could be removed from Figures 4-6 without loss.

AC: The ECMWF reanalysis is the ERA-Interim. This point is now clarified in the revised manuscript. According to the reviewer's suggestion, we replotted Fig. 4 with fewer and thicker vectors to get a better reading of the wind speed and direction (see below).

![Figure 4: Monthly averaged wind speed (m. s⁻¹) and direction at 850 hPa (left), 700 hPa (middle) and 500 hPa (right) from ECMWF reanalysis (top) and simulated with the WRF-Chem initial configuration (bottom) for July 2008.](image)
We kept the meteorological fields at 500 hPa (about 5.5 km altitude) because BBA are lifted up to an altitude of 6 km over land, on average for July 2008. Therefore, the meteorological fields at 500 hPa also seem important to assess as they may condition the BBA transport.

RC2: p. 14-15, lines 347-349: I cannot see winds capable of a westward aerosol transport in Fig. 4. I think the authors may be confusing the meteorology of September, which I suspect is what the cited papers focus on, with that of July.

AC: This is an interesting remark. Thanks to the reploting of the wind vectors in Fig. 4, we can now better see the north-westward aerosol transport with south-westerly/westerly winds.

RC2: p. 16 lines 384-386: given the finding that GOCART seems to raise too much dust, why not include an experiment in which its presence is reduced? (also ‘dusts’ -> ‘dust’)

AC: The reviewer is right. As explained in the revised manuscript, the average values of AOD are highly overestimated in the WRF-Chem initial configuration (mean $\text{AOD}_{\text{mod},565} \geq 1.5$) compared to the POLDER-3/GRASP retrievals (mean $\text{AOD}_{\text{obs},565} \approx 1.0$) over the northern half of Africa in the Sahara and Sahel region. The GOCART AFWA desert dust emission module used in WRF-Chem (Jones et al. 2010, 2012), based on the Marticorena and Bergametti (1995) scheme, thus seems to raise too much mineral dust over the Sahara/Sahel area. According to the recommendations of Flaounas et al. (2016), who made an assessment of atmospheric dust modelling performance by WRF-Chem (version 3.6) against MODIS observations on arid and semi-arid regions around the Mediterranean, including North Africa, we have applied the adjustment coefficient of 0.5 on the desert dust emission surface fluxes in the GOCART AFWA scheme.

RC2: p. 17 line 404: did the Koffi papers evaluate July explicitly? There is a strong seasonal cycle to the aerosol vertical structure, see, e.g. Redemann 2021, ACP

AC: Koffi et al. (2012, 2016) studied the mean aerosol extinction profiles retrieved by CALIOP and modelled by 12 global models from the AeroCom project over the 2007-2009 period for 12 key worldwide regions, including South Africa (SAF). They especially separated the results into four period: March-April-May, June-July-August, September-October-November and December-January-February. We qualitatively compared the mean aerosol extinction coefficient simulated by WRF-Chem for July 2008 with the one observed by CALIOP during the June-July-August quarter over SAF. In their study, Koffi et al. (2012, 2016) also showed a strong seasonal cycle to the aerosol vertical structure.

RC2: p. 18 line 435: “on the opposite” -> “In contrast”

AC: Thanks, we took into account this correction in the revised manuscript.

RC2: p. 18 line 439: are the authors suggesting an aerosol invigoration effect on shallow clouds? This seems very unlikely to me and I see no reference. I would suggest just attributing the cloud parametrization, although it confuses me the parameterization cited (Lin) is a microphysical parameterization according to Table 1. Wouldn’t the boundary layer scheme be the more likely cause?

AC: The reviewer is right. The planetary boundary layer scheme used in our study (YSU, Hong et al. 2006) would be the more likely cause. We modified the sentence at lines 437-440, page 18, as follows: “The difference between
the model and the POLDER-3 retrievals could come from the planetary boundary layer scheme (YSU, Hong et al. 2006) used in the WRF-Chem reference configuration.”

RC2: p. 18 line 464: sea salt mixed with smoke I would think. What does the model say?

AC: According to the model, the aerosol layer located between 0 and 0.8 km corresponds to sea salt aerosols (purely scattering coarse mode aerosol) located in the boundary marine layer (Deaconu et al. 2019; Peers et al, 2019). We modified the sentence at lines 462-464, page 19, as follows: “For July 12, 2008, the aerosol layer located between 0 and 0.8 km above sea level, generally detected by CALIOP and simulated with the WRF-Chem partially constrained configuration, corresponds to sea salt aerosols in the model, as usually observed in the boundary marine layer (Deaconu et al. 2019; Peers et al, 2019).”

RC2: P. 18 lines 467-468: see also Shinozuka 2020 ACP, which shows many models share these aerosol layer altitude biases.

AC: Indeed, this is an excellent study that we could build on in a future study. The suggested reference has been added in the revised manuscript.

RC2: p. 19, fig. 10 left panel+discussion: is this for the free-tropospheric aerosol layer? An average over the full column? I’m confused by this, and how the coarse mode is increasing with distance. Incidentally the orange star is placed near Ascension Island, and some assessment could be done using the LASIC datasets if interested.

AC: The aerosol volume size distributions are vertically integrated over the whole atmospheric column. We added “whole atmospheric column” in parenthesis after “vertically integrated” at line 474, page 19, and at line 478, page 20 in the revised manuscript. We concluded that the increase in the coarse mode during the aerosol transport, especially during the long-range transport, is due to the more pronounced presence of sea salt and desert dust aerosols (see Fig. A at the beginning). The reviewer is right that some assessment could be done using the LASIC datasets but only a qualitative comparison due to the non-matching period of study.

RC2: p. 22 lines 532-534: no mention of dust in the DACCIWA data description, further leading me to think an experiment should be done where it is removed from WRF-Chem and a further assessment done to see what additional changes have to be incorporated, for WRF-Chem to match the satellite retrievals.

AC: The reviewer is right. We only compared the main aerosol compounds of biomass burning with those measured during the DACCIWA airborne campaign. We conducted three numerical experiments with WRF-Chem to assess the possible influence of desert dust aerosols over the Southeast Atlantic region and their effects on the spectral dependence of SSA from aerosols, as illustrated in Fig. B (see below). The first scenario corresponds to the initial GOCART AFWA desert dust aerosol emissions scheme (DUST x1, initial configuration in orange curves). The second corresponds to the modified GOCART AFWA desert dust aerosol emissions scheme (DUST x0.5, partially constrained configuration in blue curves). The last scenario corresponds to the removal of desert dust aerosol in WRF-Chem (no DUST, yellow curves).
Figure B: Spectral dependencies of the single scattering albedo of aerosols simulated by three WRF-Chem experiments (colored curves) over land in clear sky (12°E-30°E, 10°S-0°S, dashed curves) and above clouds of the Southeastern Atlantic Ocean (0°E-15°E, 15°S-5°S, dashed curves), and retrieved by POLDER-3 (GRASP in dashed black curve, AERO-AC in solid black curve) on average for July 2008. The black vertical error bars correspond to the uncertainties associated with the POLDER-3 data with an accuracy of ±0.05 (per pixel estimates) over the whole spectral domain. The first WRF-Chem scenario corresponds to the initial GOCART AFWA desert dust aerosol emission scheme (DUST x1, initial configuration, orange curves). The second WRF-Chem scenario corresponds to the modified GOCART AFWA desert dust aerosol emission scheme (DUST x0.5, partially constrained configuration, blue curves). The last scenario of WRF-Chem corresponds to the removal of desert dust aerosols in the model (no DUST, yellow curves).

Figure B compares the spectral values of SSA resulting from these three WRF-Chem experiments (colored curves) with the retrievals from POLDER-3/GRASP (solid black curve) over land in clear sky (12°E-30°E, 10°S-0°S) and from POLDER-3/AERO-AC (dashed black curve) above clouds over the SEAO (0°E-15°E, 15°S-5°S), on average for July 2008. The three sets of simulations show that the effect of desert dust aerosols is slightly visible in the near infrared (1,000 nm) with a small decrease in absorption with increasing desert dust aerosols concentration: the SSA values increase of approximately 0.005 between the desert dust aerosols-free scenario (no DUST, yellow curves) and the WRF-Chem partially constrained configuration (DUST x0.5, blue curves), both in clear and cloudy atmospheres. In the ultraviolet (300-400 nm), the high absorption of desert dust aerosols is absent: the SSA spectral values only decrease by about 0.001 and 0.003 between the WRF-Chem partially constrained configuration (DUST x0.5, blue curves) and the desert dust aerosols-free scenario (no DUST, yellow curves), respectively over land in clear sky and above clouds over the SEAO. We can conclude from these WRF-Chem experiments that the presence of desert dust aerosols area has little influence on the SSA modeling in our study. Therefore, we kept desert dust aerosols to obtain simulated aerosol chemical composition and optical properties the more realistic as possible.

RC2: p. 25 line 568: “an increased of the aerosol absorption’ => ‘increased aerosol absorption’

AC: Thanks, we took into account this correction in the revised manuscript.

AC: Thanks, we took into account your corrections at line 403, page 17, lines 562 and 572, page 25 and line 668, page 29 in the revised manuscript.

RC2: p. 28, line 643: is there any evidence for more desert dust becoming incorporated into the aerosol layer as it moves westward other than from the WRF-Chem model? Any observations of this?

AC: Thank you for this question. Please see Fig. A at the beginning and its associated discussion, and the Deaconu et al. (2019) paper.

RC2: p. 29 line 661: remove “that”, ‘Located” “the geographic coordinates area”

AC: Thanks, we took into account your corrections. Besides, we removed “of geographic coordinates” everywhere in the revised manuscript.

RC2: p. 30 line 692: is the Leahy 2007 representative of July? Eck 2013, Zuidema 2018 both show a strong seasonal evolution to the SSA so good to make sure it’s about the same time frame.

AC: Thank you for this question. The mean SSA of 0.85±0.02 (550 nm) obtained during the SAFARI-2000 airborne campaign is not representative of July (Leahy et al. 2007). We modified the sentence at line 691, page 30, as follows: “In particular, the simulated mean SSA is consistent with the mean value of 0.85±0.02 observed during SAFARI-2000, although not fully representative of the same period of the BB season (Leahy et al. 2007).”

For instance, Peers et al. (2016) also showed a strong seasonal evolution of the SSA over the SEAO with a higher aerosol absorption in July (see their Fig. 1).

RC2: p. 33 references: the formatting of the references is not consistent, check to make sure they fit the ACP format. It also seems like there are more references than are used?

AC: Thank you for this information. We used Zotero to automatically create the bibliography.

RC2: Additional reference suggestions:


Kacarab, M., et al, 2020: Biomass Burning Aerosol as a Modulator of Droplet Number in the Southeast Atlantic Region. Atmos. Chem. Phys., 20, p. 3029-3040, doi:10.5194/acp-20-3029-2020 - this focuses on the month of August, so slightly later than July, nevertheless supports the idea that in the early part of the BB season, much aerosol is also present in the boundary layer, and, indicates the microphysical implication.


Shinozuka, Y., P. E. Saide, G. A. Ferrada, et al. 2020: Modeling the smoky troposphere of the southeast Atlantic: a comparison to ORACLES airborne observations from September of 2016. Atmos. Chem. Phys., 20, p.11,491-11,526, doi:10.5194/acp-20-11491-2020 - could be interesting (in a future study) to see how the new WRF-Chem representation does within this same comparison, the data are accessible in an easy meta format. Also useful for more context on where WRF-Chem falls in the pantheon of aerosol model representations. The downward ‘slumping’ of the aerosol layer shown in Fig. 9 is also shown to be common to many other aerosol modelling efforts.


AC: References:


