RC3: Review of: Combining POLDER-3 satellite observations and WRF-Chem numerical simulations to derive biomass-burins aerosol properties over the Southeast Atlantic region. Authors: A. Simeon, F. Waquet, J-C Pere, F. Ducos, F. Thieuleux, F. Peers, S. Turquety, and I. Chiapello.

RC3: Recommendation: Accept with Minor Revisions

RC3: In this study, the authors focus on biomass burning (BB) particle plumes transported above clouds over the Southeast Atlantic (SEA) region, off the southwest coast of Africa. They employ simulations from a regional model (WRF-Chem) coupled with meteorological reanalyzes data and aerosol retrievals from POLDER in clear sky (POLDER/GRASP) and cloudy scenes (POLDER-3/AERO-AC), to better characterize the physico-chemical and absorption properties of aerosols.

RC3: Other reviewers have asked relevant questions, so I will just add some additional comments below:

RC3: Page 6, line 155: What is the temporal resolution of the WRF-Chem configuration?

AC: Thank you for this question. The temporal resolution of the WRF-Chem configuration is hourly. We added the following sentence at line 155, page 6: "The temporal resolution of the WRF-Chem numerical simulations is hourly.". This point is now clarified in the revised manuscript.

Are the vertical levels evenly distributed from surface to 50hPa or the vertical resolution is finer near the surface? Could you clarify in the text?

AC: Thank you, this point is now clarified in the revised manuscript. We adopted the hybrid sigma-pressure vertical coordinate which is terrain-following near the surface with a finer resolution than at the fixed top pressure (flat). We modified the sentence at lines 155-156, page 6, as follows: "The atmospheric layer is divided into 50 vertical levels using the hybrid sigma-pressure vertical coordinate. The vertical levels are terrain-following near the surface with a finer resolution than at the upper pressure level set at 50 hPa.".

Line 157: Whilst I agree that the first half of July is representative for the whole month, this short period is not representative for the entire biomass-burning season. As Adebiyi et al, 2015 and Deaconu et al, 2019 showed, September and October months are characterized by different meteorological conditions and larger amounts of BBA transported over the SEAO. Also, the BBA are lifted at higher altitudes, limiting the contact aerosol-clouds. How do you justify choosing this period to study?

AC: This is an interesting remark. We chose to study this period because the aerosol absorption is the strongest (very low SSA). If we can go low enough for this period, our approach is more likely to be able (in theory) to reproduce the higher SSA observed in August and September. The SSA cycle is shown in Fig. 1 in Peers et al. (2016) for the fire season 2006 over the SEAO (5°N-30°S, 20°E-20°W). Furthermore, Figure A (not shown in the article) shows the monthly variabilities of spectral ACAOD (left) and ACSSA (right) retrieved by POLDER-3/AERO-AC above clouds in its native resolution (6 x 6 km^2), during the fire season 2008, and averaged over the Southeast Atlantic (SEA) region (20°W-30°E, 39.9°S-10°N).

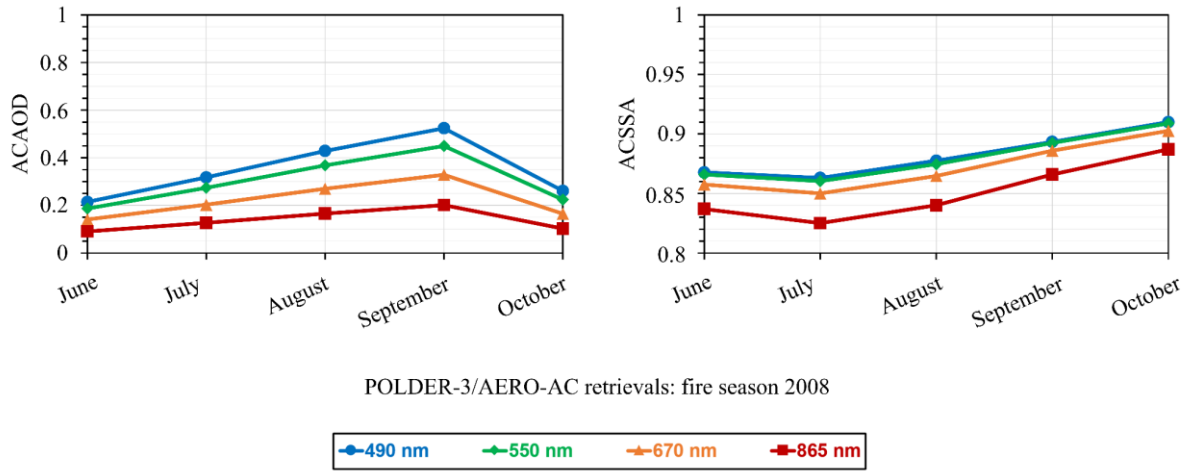

Figure A: Monthly variabilities of aerosol optical depth (ACAOD, left) and single scattering albedo (ACSSA, right) retrieved by POLDER-3/AERO-AC above clouds at a 6 x 6 km^2 spatial resolution from blue to near infrared over the Southeast Atlantic region (20°W-30°E, 39.9°S-10°N) during the fire season 2008.

Figure A indicates that the lowest values of ACSSA, corresponding to the strongest absorptions, are obtained for the month of July 2008. For example, the value of POLDER-3/AERO-AC ACSSA at 865 nm (red curve, right) decreases from 0.837 in June to 0.825 in July and then increases to 0.887 in October.

What was the computational cost of the 30 days (with 15 days spin-up) simulation and could you apply the optimized model configuration over September/October 2008 to check the consistency over this period?

AC: The computational cost of the 30 days (with 15 days spin-up) simulation was less than 1 week. It depended on the number of available and used processors. As suggested, we applied the WRF-Chem optimized configuration until the end of September 2008 to test its ability to represent the absorption cycle observed during the fire season of 2008. Figure B shows the temporal evolutions of the ACAOD (left) and ACSSA (right) retrieved by POLDER-3/AERO-AC (solid line curves) and simulated with the WRF-Chem optimized configuration (dotted curves) above clouds at 490, 550 and 865 nm (blue, green and red curves, respectively), during the July-August-September 2008 quarter.

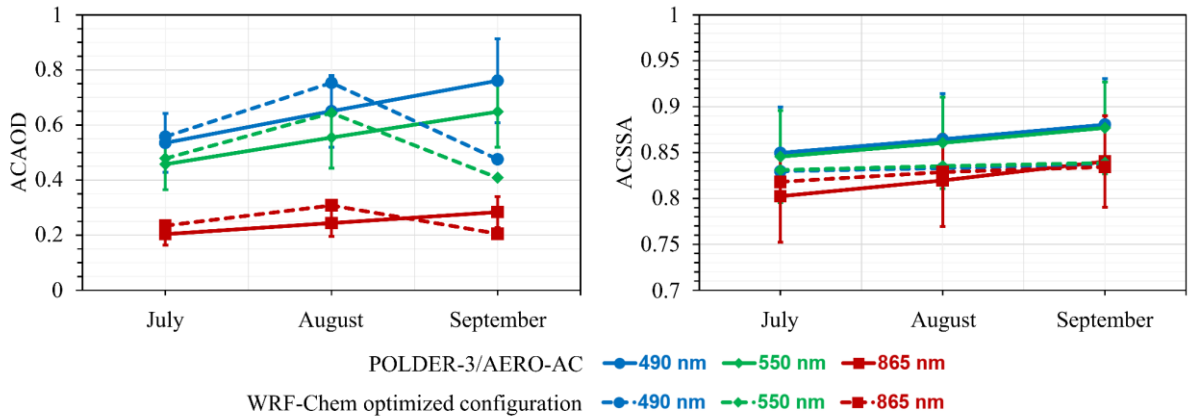

Figure B: Temporal evolutions of the ACAOD (left) and the ACSSA (right) retrieved by POLDER-3/AERO-AC (solid line curves) and simulated with the WRF-Chem optimized configuration (BCx2, OC/2.5 with 2.5% of BrOC, dotted curves) above clouds over the Southeast Atlantic Ocean at 490, 550 and 865 nm (blue, green and red curves, respectively). The optical properties are averaged geographically within the coordinate study area of (0°E-15°E, 15°S-5°S) and monthly over the July-September 2008 period. The vertical error bars correspond to the uncertainties associated with POLDER-3 data with an accuracy of ±20 % on the aerosol optical depth and ±0.05 on the single scattering albedo over the entire spectral domain.

On the one hand, Figure B shows that the WRF-Chem optimized configuration reproduces in part at least the increase in ACAOD retrieved by POLDER-3/AERO-AC between July and August, but does not reproduce the variability observed between August and September 2008. For September 2008, the WRF-Chem optimized configuration fails to adequately simulate the aerosol concentration peak which is typically observed at this time of year over the Southeast Atlantic region. Indeed, the WRF-Chem optimized configuration underestimates the amounts of aerosols transported over the marine stratocumulus clouds with an average bias of -0.24 for ACAOD at 550 nm compared to the POLDER-3/AERO-AC retrievals in September 2008. This underestimation could be related to uncertainties about the evolution of biomass burning emissions during the season in the inventory used (APIFLAMEv1, Turquety et al. 2014). Recently, a numerical sensitivity study was conducted by Pan et al. (2019) by testing six global biomass burning emissions inventories (excluding APIFLAMEv1) in the NASA's GEOS-Chem chemistry-transport model. In particular, this modelling study focused on the South African continent in September 2008. The results of the study showed an underestimation of the simulated AODs over land for these six inventories, with a maximum bias of 0.23 compared to MODIS-Aqua data at 550 nm and at least 50% compared to data from the AERONET site in Mongu, Zambia. This discrepancy between modelling and observations could not be explained by different synoptic conditions because the meteorology and aerosol mechanisms (transport and removal) were identical for these six numerical experiments. The authors of this study therefore suggest that estimates of the amounts of aerosols emitted by biomass burning should be revised high, in particular by updating the emission factors according to the season and the conditions of activity of the fires.

On the other hand, Figure B shows that the WRF-Chem optimized configuration struggles to represent very satisfyingly the seasonal cycle of ACSSA retrieved by POLDER-3/AERO-AC, which is characterized by a decrease in aerosol absorption (increase in ACSSA values) during the advance of the fire season. This result suggests that the chemical composition of the biomass burning aerosol plumes, especially the carbonaceous aerosols, changes during the dry season, likely in relation to a change in the type of fuel burned and in combustion conditions (Zuidema et al. 2016b; Pan et al. 2019). It is therefore necessary to study the aerosols physico-chemical

and optical properties on a month-by-month basis in order to better understand and constrain the biomass burning aerosols absorption cycle over the South-East Atlantic region in WRF-Chem.

RC3: Figures 1 and 2: From these figures looks like you could have chosen a different study area for the clear-sky cases, say between 5° and 15° N, that would have covered more of the biomass-burning emissions (correlated with the PM2.5 in Fig.1) and also more clear-sky days (Fig.2). Why did you choose this particular box?

AC: This is an interesting question. We chose this particular box (12°E-30°E, 10°S-0°S) because the aerosol load is maximum (see Fig. 7, left, in the revised manuscript), and the uncertainties related to the retrievals of the aerosol optical properties by POLDER-3/GRASP are the lowest.

RC3: Page 10, Fig2b: You are showing number of observation days of clear and cloudy scenes. It would be useful to plot also the data with coincident aerosol retrievals (e.g., number of observation days used in the study) for clear and cloudy skies.

AC: Thank you for this suggestion. In fact, an inversion is performed for each of these days.

RC3: Page 16, line 395: What is the scale of the biases between the AOD in clear-sky POLDER/GRASP compared to WRF-Chem reference configuration? From Fig. 7, it looks like the model is still strongly underestimating the AOD over land. Since the range of uncertainties for BB emission inventories reported in literature in 2 to 4, why did you choose to scale the APIFLAMEv1 with a factor of 1.5?

AC: This is an interesting question. The scale of the biases between the AOD in clear-sky POLDER/GRASP compared to WRF-Chem reference configuration is 2.24. We first applied a multiplying factor of 2 to the APIFLAMEv1 BB emissions inventory and the result was not concluding. Over land (12°E-30°E, 10°S-0°S), simulated AOD values ($AOD_{mod,565} = 0.98$) were in much better agreement with those retrieved by POLDER-3/GRASP ($AOD_{obs,565} = 1.10$) with a moderate underestimation of 0.12 on a geographical average. Nevertheless, above the ocean, simulated AOD values ($AOD_{mod,565} = 0.86$) were overestimated compared to POLDER-3/GRASP retrievals ($AOD_{obs,565} = 0.61$), with a positive bias of 0.25 on a geographical average. This could be explained by a higher emitted and then transported aerosol load. This could also be interpreted as a numerical diffusion problem of a dense biomass burning aerosol plume, which causes a spread of the load between the continent and the ocean due to the width of the mesh of WRF-Chem (as a reminder, a grid of 30x30 km^2). Finally, the adjustment factor of 1.5 was chosen as being a good compromise, even if small biases were still present. This point is now clarified in the revised manuscript.

RC3: Page 17, line 408: Deaconu et al., 2019 showed that oxygen pressure method underestimates the cloud height compared to the CALIOP retrieval, by about 2-300 m for low clouds. Therefore, the WRF-Chem underestimation of cloud top could not be as high as 500 m. Could you have used the CTH from CALIOP instead of POLDER for the model optimization (or scale up the POLDER CTH using CALIOP retrievals)?

AC: This is an interesting suggestion. We applied the empirical relationship proposed by Deaconu et al. (2019) to correct the altitude of the cloud top height retrieved by POLDER-3. The advantage of the corrected POLDER-3 data is the much greater spatial coverage than that of the CALIOP lidar, which allows to obtain better statistics. This point is now clarified in the revised manuscript.

RC3: In the beginning of the paper, you mention having simulated only half of July as representative for the entire month. In Fig.2 you mention '01-15 July period' and everywhere else you mention only 'July 2008', which leaves the wrong impression the data (satellite and/or model) are averaged over the entire month. Please clarify in the captions and in the text where necessary.

AC: This point has been now clarified in the revised manuscript. The simulation of the first half of July 2008 only concerns the numerical experiments. The assessments of the WRF-Chem initial and reference configurations were done for the whole month. We modified the text at lines 157-160, page 6, as follows: "We have simulated the first half of July 2008 (plus 15 days spin-up) for the numerical experiments to reduce numerical costs. The computation time was about three days for this time period and for one numerical simulation against about six days for the whole month (plus 15 days spin-up). Furthermore, the first half of July 2008 appears to be representative of July 2008. Indeed, the Above-Cloud Aerosol Optical Depth (ACAOD) and the Above-Cloud Single Scattering Albedo (ACSSA) retrieved by POLDER-3/AERO-AC at 550 nm are respectively 0.45 and 0.85 over the first half of July 2008 and 0.46 and 0.85 over July 2008 on average over our combined studied areas (black frames in Figure 2).". We added the following sentence at line 304, page 11: "It is worth noting that the assessment conducted in the first step is done for the whole month of July 2008.".

RC3: Minor corrections:

RC3: Page 10, line 279: '…to evaluate the aerosol extinction simulated with WRF-Chem at 550, as well as…'

RC3: Page 16, line 360: '…This bias could be due to…'

RC3: Page 17, line 399: '…besides the simulated aerosol loads…'

RC3: Page 18, line 431: '…WRF-Chem configuration simulates well the …'

line 447: '…depolarization ratio method…'

RC3: Sometimes the phrases are too long, and there are missing commas or linking words that could make reading more elegant and easier.

AC: We appreciate the suggested corrections and took them into account. We also tried to reduce the sentences as much as possible.

AC: References:

Deaconu, Lucia T., Nicolas Ferlay, Fabien Waquet, Fanny Peers, François Thieuleux, and Philippe Goloub. 2019. « Satellite Inference of Water Vapour and Above-Cloud Aerosol Combined Effect on Radiative Budget and Cloud-Top Processes in the Southeastern Atlantic Ocean ». Atmospheric Chemistry and Physics 19 (17): 11613-34. https://doi.org/10.5194/acp-19-11613-2019.

Pan, Xiaohua, Charles Ichoku, Mian Chin, Huisheng Bian, Anton Darmenov, Peter Colarco, Luke Ellison, et al. 2019. 'Six Global Biomass Burning Emission Datasets: Inter-Comparison and Application in One Global Aerosol Model'. Preprint. Aerosols/Atmospheric Modelling/Troposphere/Physics (physical properties and processes). https://doi.org/10.5194/acp-2019-475.

Peers, F., N. Bellouin, F. Waquet, F. Ducos, P. Goloub, J. Mollard, G. Myhre, et al. 2016. 'Comparison of Aerosol Optical Properties above Clouds between POLDER and AeroCom Models over the South East Atlantic Ocean during the Fire Season'. Geophysical Research Letters 43 (8): 3991–4000. https://doi.org/10.1002/2016GL068222.

Turquety, S, Laurent Menut, B Bessagnet, Alessandro Anav, Nicolas Viovy, Fabienne Maignan, and M Wooster. 2014. APIFLAME v1.0: High-Resolution Fire Emission Model and Application to the Euro-Mediterranean Region. Vol. 7. https://doi.org/10.5194/gmd-7-587-2014.

Zuidema, Paquita, Jens Redemann, James Haywood, Robert Wood, Stuart Piketh, Martin Hipondoka, and Paola Formenti. 2016b. 'Smoke and Clouds above the Southeast Atlantic: Upcoming Field Campaigns Probe Absorbing Aerosol's Impact on Climate'. Bulletin of the American Meteorological Society 97 (7): 1131–35. https://doi.org/10.1175/BAMS-D-15-00082.1.