Response to Editorial Comment on the manuscript:

egusphere-2024-3197

Atmospheric processing and aerosol aging responsible for observed increase in absorptivity of long-range transported smoke over the southeast Atlantic.

EC: Editor Comment RC: Reviewer Comment AR: Author Response

EC: Public justification (visible to the public if the article is accepted and published): I want to take this chance to say thanks to the reviewers for the valuable comments. I also appreciate the authors being serious about the comments. After reading all of the materials, I have some additional suggestions for this manuscript:

EC: 1. This manuscript is generally lengthy, especially the method part already reaches line 372. Though the text helps to demonstrate the broad background knowledge of the authors, it does not help to keep readers focused on the important scientific findings. Here are several specific suggestions for your consideration:

1) Delete the text regarding the introduction of the structure of the paper,

AR: We have complied with your recommendation and have removed the paragraph accordingly.

2) The first few sentences introducing the measurement modes of the 4STAR. You are not using the other two modes at all. Start directly from "This study…",

AR: We appreciate your suggestion and have revised the section. Section 2.3 now reads:

2.3 Airborne measurements

This study uses sky radiance measurements from the airborne 4STAR instrument (Dunagan et al., 2013) aboard NASA P-3B aircraft, enabling AERONET-like observations in remote regions beyond ground-based network coverage. Its frequent co-deployment with in-situ instrumentation offers a more comprehensive characterization of aerosol properties. This analysis focuses on observations made in the sky-scanning mode (Pistone et al., 2019), using both ALM and principal plane (PPL) scans. Given that most ORACLES flights occurred near solar noon, limiting the angular range of ALM scans, PPL scans were selectively included if they met specific quality control (QC) criteria. Here, we processed 4STAR sky scans using the QC criteria from Mitchell et al. (2023). These criteria were adapted from Pistone et al. (2019) for four-wavelength 4STAR retrieval of ORACLES 2016-2018 and serve as a proxy for AERONET level 1.5 aerosol inversion standards. The criteria are: (1) AOD (400 nm) > 0.2, (2) altitude difference < 50 m, (3) sky error < 10%, (4) minimum scattering angle < 6°, (5) maximum scattering angle > 50°, (6) mean scattering angle difference < 3° (between 3.5 and 30°), (7) maximum scattering angle difference < 10° (between 3.5 and 30°), (8) roll standard deviation < 3°, (9) passes retrieval boundary test - ensuring that the retrieval is within limits of parameter space, and (10) maximum altitude < 3000 m. A summary of the valid QC'd 4STAR retrievals of SSA, AOD, EAE (Mitchell et al., 2023) in the ORACLES dataset (Oracles, 2020) for all three deployments is given in Table 1.

3) The Second paragraph introducing the WRF-CAM5 is a good summary of the model, but I don't think it is necessary here unless you have done something unique to the model. As a method part, the first and last paragraphs are enough.

AR: We appreciate your suggestion and the section now reads:

2.4 WRF-CAM5: Concept and Configuration

WRF-CAM5 is an adaptation of the WRF-Chemistry (WRF-Chem) model (Grell et al., 2005), which integrates the physics and aerosol packages of the global CAM5 (Ma et al., 2014; Zhang et al., 2015a), making it suitable for studying multi-scale atmospheric processes and evaluating aerosol and physics parameterizations in global climate models (Wang et al., 2018). WRF-CAM5 has been widely applied to investigate air quality and climate interactions in Asia and the United States (Campbell et al., 2017; Wang et al., 2018; Zhang et al., 2015b) and has shown good skill in capturing smoke concentration, aerosol properties, and the vertical distribution of BBA in the SEA region (Doherty et al., 2022; Chang et al., 2023). In this study, the model is configured at 36km horizontal resolution with 74 vertical layers over the spatial domain 41°S-14°N, 34°W-51°E, initialized every five days using the National Center for Environmental Prediction (NCEP) Final Operational Global Analysis (NCEP FNL) and Copernicus Atmosphere Monitoring Service (CAMS) reanalysis datasets as detailed in Shinozuka et al. (2020a) and Doherty et al. (2022), with daily smoke emissions from the Quick-Fire Emissions Dataset version 2 (QFED2) (Darmenov and Da Silva, 2015).

4) There is a long introduction on why to separate the BL contribution from TC. The sentence "Dang et al. (2022) showed that BBA sampled during ORACLES dominate… " is good enough, and all of the text before can be potentially removed.

AR: We have revised the introduction to section 2.7 on why we separate the BL contribution from TC. In line with your recommendation, we have removed portions of the text while retaining a few sentences to briefly describe the transport of BBA by the AEJ-S from the continent to the maritime atmosphere over the SEA. The revised section now reads:

2.7 Separating Boundary Layer (BL) contributions from Total Column (TC) observations

As sea surface temperatures increase, the boundary layer (BL) deepens offshore before transitioning to a cumulus regime (Zhang and Zuidema, 2019, 2021; Ryoo et al., 2021). Continental BBA are lofted above 6 km and advected above the cloud layer by free troposphere (FT) winds (Ryoo et al., 2021), particularly during strong south African Easterly Jet (AEJ-S) episodes (Adebiyi and Zuidema, 2016). Although generally elevated, subsidence (Wilcox, 2010) and low-level easterlies (Diamond et al., 2018) can entrain BBA into the BL between June and August, altering aerosol properties (Dobracki et al., 2025).

Dang et al. (2022) showed that BBA sampled during ORACLES dominate the FT while sea salt aerosols may often dominate the BL over the SEA with a fraction of BBA mixed with sea salt aerosols in the BL. Therefore, our goal of investigating the evolution of BBA from TC observations is complicated by the potential contribution of non-BBA aerosols from the MBL. To address this, and given that AERONET and 4STAR provide columnar retrievals above the observation altitude, we employed a two-pronged approach, detailed in Section 2.7.1 and 2.7.2, to isolate the FT aerosol from the columnar observations. First, we applied a model-derived ratio to partition aerosol loading in the FT and BL over the SEA. Subsequently, we implemented a size thresholding technique to exclude contributions from larger particles, ensuring our analysis remains focused on BBA properties.

EC: 2. RC2 5 and text in the revised manuscript: There are several problems in this revised paragraph.

1) Why emphasize flaming here? Smoldering is more efficient in producing rBC and brown carbon.

2) Organic coating is the condensation of organic vapors on the existing particles, while nucleation produces new particles. Don't mix them.

3) Many lab experiments have shown that the biomass burning BrC has a lifetime of hours to 2 or 3 days. This bleaching process might be the main reason for the increase in SSA during transport.

4) Increasing OA does not mean increasing SSA. It's true only when the OA is not absorbing or less absorbing.

5) During 6-8 days, aging probably already causes organic loss in the particles. Therefore, the lensing effect is weaker, which increases SSA. But due to the loss of less absorbing species (compared to black carbon), the mass absorption efficiency increases and size decreases, both lead to decreased SSA. The authors need to carefully change the text.

AR: We thank you for your insights. We have revised the paragraph as below:

The changes in SSA presented in this study are primarily associated with chemical and physical processes in the atmosphere (Dobracki et al., 2023). Fresh aerosols over the continent exhibit a low SSA, which we attribute to a high proportion of rBC compared to other aerosol components. This interpretation agrees with previous findings further offshore (Denjean et al., 2020b; Wu et al., 2020; Dobracki et al., 2023) and is characteristic of emissions from flaming grassland fires. As these aerosols age in the atmosphere, they accumulate an organic coating, a process that begins rapidly within hours and continues for the first few days (Hodshire et al., 2019; Sedlacek et al., 2022), increasing the contribution of OA to the total mass of the aging particles. Unlike field measurements over the Atlantic Ocean that are relevant for more aged aerosols, our results show a progressive increase in SSA during the first 6 days of aging. After 6-8 days, however, the SSA starts to decrease, consistent with in-situ observations over the maritime atmosphere. We hypothesize that this decrease in SSA is due to increased absorption per particle from lensing effects, consistent with Taylor et al. (2020). Moreover, heterogeneous oxidation may drive the repartition of aerosol mass back to the gas phase, reducing the overall OA:BC mass ratio. In addition to compositional changes driven by the accumulation and/or evaporation of organic coatings, there is likely a shift in fine-mode particle size, potentially resulting from the same chemical and physical processes that also influence the optical properties.

3. The abstract must be one single paragraph.

AR: The abstract is now combined into a single paragraph.

4. The authors used indentation for starting some new paragraphs, but not to all. Please be constant.

AR: Thank you for your suggestion. We have revised the manuscript to consistently apply indentations for new paragraphs.