

## Response to Reviewers

### Anonymous Referee #4

This manuscript presented a study of Saharan dust based on airborne observations made over the Eastern Tropical Atlantic near the western African coast. The measurements were targeted to characterize dust microphysical, chemical and optical properties, including size distribution, particle shape, mass loading, composition, refractive indices, and SSA. This study contrasted the dust properties in Saharan Air Layer and the marine boundary layer. The authors highlighted several important findings which will advance the current understanding and benefit later modeling studies. The manuscript is logically organized and well written. It is noted that the authors provided meticulous details about the instrument, data reduction, and uncertainty analysis. This reviewer believes that this manuscript shall be published after the authors considering a few suggested minor changes, which will not alter the major finds of this study.

We are pleased that the reviewer finds our results important and beneficial to modelling studies, and that they consider our data processing and uncertainty analysis meticulous. We thank the reviewer for their detailed comments which we hope have led to a clearer manuscript. We have responded to each comment in turn below. In particular, we thank the reviewer for their point about the relationship between SSA,  $k$  and  $d_{\text{eff}}$ , and have tried to clarify our aims here in confirming whether  $k$  or  $d_{\text{eff}}$  is the main contributor to the variability in SSA. Full details are given below.

#### Minor Comments:

Page 4, Line 4: Please clarify that while light shadowing techniques are not impacted aerosol composition or Mie theory conversion issues, they still can be impacted by non-spherical particles.

Done

Page 5, Line 7: Change “Table 1” to “Table 2”

Done

Page 5, Line 8, please describe the AOD and clarify if the AOD is calculated over the dust layers. Please also make changes to the table caption so that it will be consistent with the text

The AOD is calculated across the entire aircraft profile, not just the dust. We have changed the text, the table does not need changing.

We added to Section 2.1, “The events sampled revealed accumulation mode AODs at 550 nm (see Section 2.3.1)...”

We added to Section 2.3.1, “Accumulation mode AODs are calculated from aircraft profiles by integrating the scattering and absorption measurements between the minimum aircraft altitude (typically around 30 m above sea level) to the top of the profile (typically around 6 km). Therefore AODs represent both SAL and MBL aerosol.”

Page 5, Line 26: Change “Figure 1b” to “Figure 1”

Changed

Page 6, Lines 18-19: The authors note that visually identifying and tracking dust plumes is subjective, difficult, and potentially error-prone. Would it be possible to instead obtain the underlying satellite data and apply an objective threshold?

As in the response to reviewer 3, we feel that we potentially over-emphasized the disadvantages of the SEVIRI method, and have changed the text accordingly.

Methods have been developed to automatically detect dust emission using the SEVIRI imagery, e.g. Ashpole and Washington (2012). To our knowledge none have been developed to track or back-track transported plumes. Applying a threshold to the underlying data may indeed have been possible, but would still be subject to variability due to underlying surface emissivity from surface temperature and surface type, altitude of dust in the atmosphere, obscuration by clouds.

Page 7, Line 5, please add a brief discussion on the choice using PSAP correction by Turnbull (2010) and difference between this correction and that by Virkkula, AS&T, 44:706-712, 2010

Turnbull (2010) reports on corrections necessary to the FAAM PSAP measurements based on the original work by Bond et al. (1999), and clarifications to this publication described in Ogren (2010), and clarifies any errors in calculations performed by Haywood and Osborne (2000).

Virkkula et al (2010) report corrections to the Virkkula et al (2005) publication, dealing with inconsistencies between a one and three-wavelength PSAP. The 2010 publication resolves the discrepancies. Since we employ a one-wavelength PSAP, not a three wavelength PSAP, we do not consider these publications in our corrections.

We have added the Bond et al. (1999) and Ogren (2010) references to the text.

Page 11, line 31, please provide a more quantitative criteria to define the word “dominant”

This has been changed to, “they were classified according to their dominant component type which made up the greatest oxide percentage.”

Page 13, Line 1: Figure 8b is unrelated to SSAs; perhaps Figure 13b was intended?

This sentence has been deleted.

Page 13, Line 8: Please restate the rationale to hold the real part of the refractive index at 1.53, in the context that in Section 3.5, the real part is found to be 1.47-1.49 based on the filter sample composition.

We also tested the sensitivity of our results to different real parts of the refractive index: 1.48 and 1.58. We found that effective diameter (resulting from varying the RI used to correct the PCASP and CDP data) changed by under 5%. For the optical properties calculated, scattering and absorption each changed by around 4%, but SSA changed by under 0.5% since typically both scattering and absorption changed in the same direction.

Also, for the reasons stated in Section 3.5, we believe the filters real refractive index results to be biased low.

We added, “Although a value of 1.53 for  $m^{550}$  is higher than that produced by the filter sample composition results (Section 3.5, 1.47-1.49) the filters result is likely biased low due to the reasons discussed in Section 3.5. We also performed a sensitivity test to using  $m^{550}$  of 1.48 and 1.58, and found that deff changed by up to 5% and SSA by under 1%.”

Page 13, Lines 14-20: This information might be better suited to a table, which could also include the actual refractive index used for each substance.

Although we appreciate this suggestion, since the spectral refractive index data is used rather than just a value at 550 nm, this would not fit into a table easily.

Page 14, Line 4: Change “Table 3” to “Table 4”

Done

Page 16, line 25. Please provide a brief discussion on how the “best-fit” compare to observed volume size distribution and number size distribution and Change “Table 4” to “Table 5”

Differences between the effective diameter calculated with the best-fit lognormal curves and the observed PSDs are between 10-15%. This relatively large error derives largely from the size range between diameters 3

to 20  $\mu\text{m}$  measured by the CDP. The fluctuating nature of the CDP PSD does not reconcile it to an easy fit with lognormal curves. This has been added to this paragraph.

Table reference changed.

Page 18, Line 2: Change "Table 5" to "Table 6"

Done

Page 18, Lines 8 and 11: These two statements regarding a potential decrease in  $d_{\text{max}}$  with height seem contradictory. Please clarify to make them consistent.

This has been changed to, "There is no clear trend of  $d_{\text{max}}$  decreasing with altitude."

Page 18, Line 14: There is no Figure 8c

Changed to 8b.

Page 19, Line 14: Please supply a reference for the dust density value.

The value comes from Tegen and Fung (1994), and is used by Woodward (2001) and Bellouin et al. (2011) as the default density value for dust aerosol in the Met Office Unified Model and also the radiation scheme within the model. References have been added.

Page 21, Line 15: Change "Table 6" to "Table 7"

done

Page 23, Line 26 (and Figure 13b): Some readers may wonder if finding a good agreement between the imaginary part of the refractive index and the SSA is expected, given the relationship between  $k$ , absorption, and SSA. The authors should consider the significance of confirming that the relationship exists in this case.

We are not trying to show or confirm the existence of a relationship between  $k$ , absorption and SSA. We acknowledge that this is an established relationship. However, the PSD can also impact the SSA. Therefore the SSA can be influenced by several factors, and our aim is to investigate which factors dominate the variability of the SSA. During Fennec, for example, the variability in the PSD was the dominant controller of the SSA, rather than  $k$ . Contrastingly, during AER-D we found that the PSD did not vary much, and therefore variability in  $k$  dominated the variability in SSA.

We have rewritten this paragraph to try and better explain this finding and its significance, and also in the conclusion. This paragraph now reads:

"Contrastingly to Figure 13a, Figure 13b shows that the SSA variability was strongly influenced by the variability in composition. This is the case for both accumulation mode observations of SSA, and for the full size distribution. It is not surprising that variability in  $k^{550}$  influences absorption and therefore SSA. However, the SSA can be influenced by several factors, including the PSD. Our aim is to investigate which factors influence the variability of the SSA. Therefore it is notable that there is so little variation in the PSD during AER-D that the composition (or  $k^{550}$ ) is the main factor contributing to the variability of the SSA. This finding is notably the opposite from that found during Fennec, where the size distribution was the dominant controller of optical properties. Liu et al. (2018) show that hematite content is important in the ICE-D/AER-D samples as a controlling factor on optical properties. Moosmuller et al. (2012) and Caponi et al. (2017) also show dependencies of refractive index on iron content. This is consistent with our findings that the calculated refractive index from the filter samples is strongly influenced by the iron content and its absorbing properties. It appears that over the Sahara, variations in the PSD (affected by dust age) have an important impact on SSA, while over the ocean the impacts of composition (perhaps either by chemical aging or by sampling dust from different sources) become more important."

Page 24, Line 20: Change "Table 5" to "Table 6"

Done

Figure 4: This figure suggests the flight b924 and b934 did not have extensive sampling in MBL, please make changes in text accordingly

We are not sure why the reviewer thought this. Figure 4 shows aircraft profile observations. Each flight included one SLR in the MBL. We have added information to Section 2.1 (Flight Patterns) to further explain the aircraft manoeuvres. See also the response to reviewer 3.

Figure 6: The blue shading was very faint on my screen. Perhaps a darker shade, or even hatching, could be used instead. Also, as there are no other parts to this figure, "Figure 6(a)" should be changed to "Figure 6" (as well as in the associated text).

'a' has been removed. The shading appears fine on our screen and print-offs. We will closely monitor the readability of this figure in proof-reading, where the original eps files will be used rather than a convert to .png contained in the MS Word document.