

This study provides a detailed characterization of the chemical, physical and optical properties of transported biomass burning aerosols (BBAs) in the Ascension Island marine boundary layer (MBL), utilizing datasets from a long-term in-situ measurement platform. The authors present temporal variations of BBAs properties within the Ascension Island MBL, which are associated with variations in source burning conditions and transport pathways. The low single scattering albedo (SSA) values observed in this study are linked to low organic (OA) to black carbon (BC) ratios, attributed to OA loss as a consequence of oxidation processes. This finding underscores the importance of improving the representation of OA loss mechanisms and, consequently, aerosol optical properties in future modeling studies. I recommend publishing this work after addressing the corrections listed below.

General comments

1. Marine background environments over the Ascension Island have been considered when presenting chemical compositions such as ΔSO_4 . However, the marine background environments should be given more consideration when presenting the BBAs results such as FrBC and SSA.
 - 1) In this manuscript, the FrBC in the BBA was “calculated by dividing the number concentrations of rBC-containing particles measured by the SP2 by the total particle number concentration measured by the CPC” (lines 211-212). In the MBL over Ascension Island, the FrBC during BB-impacted periods is not only related to BBAs, but also likely affected by marine emissions. The concentration levels between background marine aerosols and mixed BBAs, as well as the mixing state (internally or externally) between BBAs and marine aerosols will affect the FrBC values.
 - 2) Similar to FrBC, the SSA given in this manuscript is also likely affected by such as marine sulfates in the MBL.

The marine backgrounds may have small effects on these BBA properties, and the temporal trends shown in this manuscript may still depend on primarily difference in source conditions and transport processes. However, including the aforementioned details would provide a more comprehensive picture of BBAs in the MBL.
2. In Sect 4.1, 4.4 and 5.1, the term of “ $SO_4:rBC$ ” is used for discussions. However, based on descriptions in Sect 3, the term of $SO_4:rBC$ should be changed to $\Delta SO_4:rBC$ (refer to above-background ΔSO_4 mass concentrations). For example, in lines 804, 809, 816 etc.
3. In Sect. 4.1, the authors state that (lines 824-825) “high OA:rBC, large rBC coating-to-core mass ratios, and high $SO_4:rBC$ would suggest that the fires were inefficient, *while the large values of rBC_{gpd} and rBC_{mpd} and high FrBC would indicate that the fires were efficient*”. This assumes that efficient burning is related to larger BC core sizes. However, in the second paragraph of Sect. 4.2, the authors discuss that “the relationship between BC core size and source burning conditions are not sure from this study and previous studies”. These present conflicting clarifications to some extent. I suggest rephrasing this part.
4. In Sect. 4 (line 755-757), the authors state that “Lastly, we examine the dependence of SSA_{530} on the chemical properties of the BBA and explain why the BBA in the MBL has a lower SSA_{530} than that in the FT”, and further explanations follows in Sect. 4.5. I agree that the authors provide a detailed explanation on the relationship between SSA_{530} and OA:rBC, combining the LASIC MBL and ORACLES FT. However, more details should be provided regarding the discussion of low SSA_{530} in the Ascension Island MBL and the combination of different datasets:
 - 1) In line 998, the authors state that “The SSA_{530} values in the Ascension Island MBL even lower than those in the nearby FT by about 0.07”. However, it is not clearly demonstrated that what this “nearby FT” data is here. Does this “nearby FT data” refer to ORACLES FT? I suggest clarifying it clearer here.
 - 2) Following the above, the authors state that “This result is surprising, as mixing with clean marine air should increase SSA_{530} in the MBL all else equal (Wu et al., 2020)”.

In Barrett et al., (2022), LASIC ARM observed much less scatterings than CLARIFY FAAM during intercomparison flight legs, while the absorptions measurements were relatively consistent. This discrepancy in scattering measurements is attributed to the differences in the upper cut sizes of the impactors. LASIC scattering measurements are likely less affected by submicron coarse mode particles from marine sea sprays, due to its lower cut size (0.78 μm) compared to CLARIFY (1.0 μm). If the authors compare the LASIC-MBL/ORACLES-FT SSA trends to CLARIFY-MBL/FT SSA trends, the inconsistency in scattering measurements should be considered.

3) Is there any consistency or inconsistency in optical/chemical measurements between the LASIC and ORACLES platforms, if the authors combined two datasets to establish the correlation between SSA_{530} and OA:rBC? E.g.

5. The uncertainties of calculated values from different instruments, such as SSA, should be provided in this manuscript.
6. When getting to Ascension Island, the clouds tend to be decoupled from the surface mixed layer (e.g. Abel et al., 2020), and the upper part of the MBL is relatively separated to the lower surface layer. Under this condition, surface layer measurements from LASIC may not directly represent BBAs properties below the cloud. This should be taken into account when interpreting the implications of LASIC datasets.

Specific comments

1. Page 1, Line 23: The authors state that “OA to rBC mass ratios (OA:rBC) in the MBL between 2 and 5 contrast to higher values of 5 to 15 observed in the nearby FT”. I suggest adding the ages of BBAs for this MBL and “nearby FT”.
2. Page 7, Line 200: What is the supporting material (reference or else) for using $\text{BC} < 20 \text{ ng/m}^3$ as the criteria to calculate background SO_4 mass concentration?
3. Page 7, Table 2: Error with captions, is it ΔSO_4 or SO_4 ? And the same question for CO, is it clean period CO or ΔCO ?
4. Page 8, Lines 225-226: What is the supporting material (reference or else) for using $\text{BC} > 150 \text{ ng/m}^3$ as the threshold to define plume events?
5. Page 9: In Table 3, Regime 3 burning condition is described as “Inefficient and efficient”.
Page 10, line 292-293: Regime 3 is described as “Although fires are still highly active in grassland regions during this time, these values indicate *inefficient combustion*”.
Page 25: The reasons why Regime 3 had a mixture of inefficient and efficient combustion sources is explained in Sect 4.1.
The description of Regime 3 burning condition is not consistent in Sect. 3 and 4, which may confuse the readers. I suggest rephrasing this part.
6. Pages 11-13, Figures 3-5: There are errors with figure caption, (b) should be surface relative humidity, and (c) should be land use map.
7. Page 14, Table 4: the last line of SSA_{530} , “0.81 0.01”, a “ \pm ” is missing.
8. Page 16, Lines 499-502: The authors state that “the exception f44 values of P5 and P10... These differences possibly result from regime transitions and/or more complex fuel source mixtures”. Could you provide more explanations why more complex fuel source mixtures may lead to the exception f44?

9. Page 18, Line 557: May need a reference for Hoppel minimum.
10. Page 18, Line 567-568: “The average of the bimodal size distributions during BBA-laden times had an Aitken mode with a peak 200 cm^{-3} at a larger diameter near 45 nm”. It should be clear to specify, “at a larger diameter” is larger than pristine period or?
11. Page 18, Line 584: “A lower fraction of the BBA-laden days (4 of 12) than *Regimes 1 and 2*”.
12. Page 20, Line 628: “For example, the lowest SSA_{530} , 0.75 ± 0.01 , in P6, also had the highest FrBC”. The highest FrBC is in P9, rather than the P6. Please rephrase this.
13. Page 20, Line 653: There is no Table S1 in the supplement. Please add it if it is needed.
14. Page 23, Line 744-745: There is a redundant reference of “(Ryoo et al., 2022)” in one sentence.
15. Page 24, Table 5: Wu et al. (2020) provided the SSA values at green channel. I suggest replacing SSA_{658} with the blue channel SSA from Wu et al. (2020) in Table 5.
16. Page 26, Line 837: “(although still within instrument uncertainty)”. What is the uncertainty of “ rBC_{mpd} ”?
17. Page 26, Lines 847-849: The authors state that “However, large rBC_{gpd} and rBC_{mpd} values of 135 and 205 nm, respectively, were also observed during Regime 3, in late August and early September when burning conditions were less efficient ($\text{rBC}:\Delta\text{CO} = 0.008$)”. The values ($\text{rBC}:\Delta\text{CO} = 0.008$) for Regime 3 is not consistent with the average value (0.007) presented in Table 4.
18. Page 26, Lines 850-852: The authors state that “This contradicts this study’s findings that rBC_{gpd} and rBC_{mpd} values were smaller (125 and 195 nm) when fires were also less efficient ($\text{rBC}:\Delta\text{CO} = 0.009$)”. Are these BC core size and $\text{rBC}:\Delta\text{CO}$ values from Regime 1 or which single plume event? I suggest clarifying this clearer.
19. Page 27-28, Lines 900-901: The authors state that “f44 values were greater than 0.22 and OA:rBC values were less than 5 for BBA sampled in the FT and MBL near Ascension Island”. In Fig. 16, only the FT f44 values are presented. Is “MBL” near Ascension Island needed here?
20. Page 31, Line 1040: Error with the figure caption. a) SSA 530 nm versus “FrBC”.

References:

- Abel, S. J., Barrett, P. A., Zuidema, P., Zhang, J., Christensen, M., Peers, F., Taylor, J. W., Crawford, I., Bower, K. N., and Flynn, M.: Open cells exhibit weaker entrainment of free tropospheric biomass burning aerosol into the south-east Atlantic boundary layer, *Atmos. Chem. Phys.*, 20, 4059–4084, <https://doi.org/10.5194/acp-20-4059-2020>, 2020.
- Barrett, P. A., Abel, S. J., Coe, H., Crawford, I., Dobracki, A., Haywood, J. M., Howell, S., Jones, A., Langridge, J., McFarquhar, G., Nott, G., Price, H., Redemann, J., Shinozuka, Y., Szpek, K., Taylor, J., Wood, R., Wu, H., Zuidema, P., Bauguutte, S., Bennett, R., Bower, K., Chen, H., Cochrane, S. P., Cotterell, M., Davies, N., Delene, D., Flynn, C., Freedman, A., Freitag, S., Gupta, S., Noone, D., Onasch, T. B., Podolske, J., Poellot, M. R., Schmidt, S. K., Springston, S., III, A. J. S., Trembath, J., Vance, A., Zawadowicz, M., and Zhang, J.: Intercomparison of airborne and surface-based measurements during the CLARIFY, ORACLES and LASIC field experiments, *Atmos. Meas. Tech.*, 15, 6329–6371, <https://doi.org/10.5194/amt15-6329-2022>, 2022.
- Wu, H., Taylor, J. W., Szpek, K., Langridge, J. M., Williams, P. I., Flynn, M., Allan, J. D., Abel, S. J., Pitt, J., Cotterell, M. I., Fox, C., Davies, N. W., Haywood, J., and Coe, H.: Vertical variability of the properties of highly aged biomass burning aerosol transported over the southeast Atlantic during CLARIFY-2017, *Atmos. Chem. Phys.*, 20, 12697–12719, <https://doi.org/10.5194/acp-20-12697-2020>, 2020.