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Comment on acp-2021-724

Anonymous Referee #1

Referee comment on "Biomass burning and marine aerosol processing over the southeast Atlantic Ocean: a TEM single-particle analysis" by Caroline Dang et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-724-RC1>, 2021

Dang et al. ACPD

This manuscript presents an interesting data set on the composition of aerosol sampled during ORACLES and CLARIFY downwind of Africa over the Atlantic Ocean during burning season in 2017 and 2018. Single particle elemental analysis conducted with TEM-EDX provides information about the mixing state (internal versus external), shape, viscosity, and volatility of the collected particles that can provide insight into photochemical, aqueous and heterogenous processing during transport which is not available from techniques measuring bulk aerosol.

The authors suggest that the new data presented in this paper provide important complement to previously published AMS data from the 2 campaigns that will address 3 key questions: 1) do the aerosols in the 2 campaigns differ as a function of the age of the smoke plumes, 2) what are the differences between aerosol in FT and MBL, and 3) what processes caused any differences found while investigating the first 2 questions? Authors also state that determining the mixing state of the aerosol is a motivating question, but I assert that the mixing state is information that may contribute to answering the other questions rather than being of high intrinsic interest by itself. The evolution of smoke advected over the ocean, and how that evolution may be modified by mixing with seasalt aerosol, as well as how seasalt aerosol may be modified by mixing with smoke, have climatic relevance so I was looking forward to hearing the answers to the 3 questions the authors stated they were addressing.

Unfortunately, bulk of the paper presents the single particle composition in a large number of ways that are not very well connected to each other, and rarely clearly connected back to the motivating questions. The fact that the conclusions of this manuscript include nothing related to the fact that CLARIFY sampled older smoke than ORACLES, and almost nothing about differences between aerosol in the FT and MBL (just that there was less

depletion of Cl in fresh seasalt aerosol in the MBL compared to in the FT) is illustrative. Also, more than a few interesting (sometimes puzzling) observations are pointed out almost in passing, with little attempt at understanding what they may suggest about aerosol processing.

In the following I work through the paper noting sections that could/should be expanded to perhaps develop one or more compelling arguments. Along the way I will also note sections or statements that are not as clear as they might be.

In 3rd paragraph of the introduction the summary of prior work on the extent of coating on BC during CLARIFY (very thick) compared to ORACLES (less thick with some evidence for loss of coating with age) is interesting since the smoke sampled in CLARIFY was significantly older and might therefore have less coating. I assumed this was setting up a major line of investigation based on the TEM-EDX results, but really never found it.

In second paragraph of section 2.3 it might be helpful to say something about where most of the fires were. Alternatively, modifications to Fig 1 or supplemental Fig 1 might be a way to convey this information (more on that later).

Second paragraph of section 2.4, were there SP2 instruments on both aircraft? If not, which had one and which did not?

Lines 202-203 state that Table 1 includes BC mass/total PM1 but I do not find that. Rather it includes BC mass and the number of BC particles/cm³.

Second paragraph of section 3.1 introducing Fig 1. Lot of questions about the trajectories: where were the fires, how far above the fire(s) did given parcel pass (i.e., many trajectories quite high, hence may not have entrained much smoke)? It might be easier for the reader to figure some of this out if there were 4 panels, separating BL and FT samples, with fire hot spots included on the maps.

Line 216 Table 1 shows a filter collected 8/30/17 and 2 on 9/30/18 so "except for two" should be except for three.

Section 3.2.1 The finding that the organic aerosol in CLARIFY was more volatile (hence lost more quickly when hit by TEM beam) is interesting, and a noteworthy finding. But it is not clear to me that the AMS f44 fully supports this. While there are a few ORACLES samples with high f44 most of the samples in both campaigns cluster from 0.18 to 0.24 (per right panel in Fig 2). Please explain how the combination of f43 and f44 is, or may not be, consistent with your inference based on single particle analysis.

The third paragraph in this section is also interesting, using TEM images to assess viscosity and volatility. But there are very few particles shown in Fig 3. Would be much stronger with a numerical summary of how many particles in ORACLES looked like the round one on left compared to the one in the middle, etc.

The loss of tarballs with age is an important finding.

Section 3.2.2 I find the first sentence to be a little confusing, by including internally and externally mixed variants of K-salts. If you counted a particle that was mostly OA, with a little K (or BC with small K crystal attached) as OA (or BC) instead of K would the number fraction of K-salts be much smaller?

More important point. Table 2 shows pretty big difference between BL and FT in all three columns, but the sign of all differences is opposite between the two campaigns. Given motivating question 2 (and 3) nearly requires that the authors at least try to explain this.

Section 3.2.3 I am not convinced that it is a good decision to ignore ORACLES in this section. Kind of seems the point of entire paper is to compare and contrast the 2 campaigns, in the framework of looking at old versus very old smoke (and how both kinds of smoke interact with seasalt).

Line 301. The atomic ratio of Na:Cl in SS is 0.84. Seasalt is not halite.

Displacement of Cl from seasalt by acids is well established (based on many studies), so need not be emphasized so much here. The Cl rich particles are probably new, hence more interesting.

Lines 348-349 Not convinced that Na in the FT (mixed with BBA) requires mixing BBA into MBL and then modified aerosol back out. Could just mix seasalt into FT and have it interact with BBA there.

On the other hand, the Gold1 filter does support BBA mixing into the MBL (but is this really surprising?)

359-366 As noted above, the Cl rich particles are interesting. But are they important (e.g., what impact on radiative forcing, good CCM)?

Section 3.3 last paragraph? Any evidence that the change in the response of K-salts to the electron beam translates to an atmospherically relevant change (like the decreasing viscosity of the sulfur/organic particles in the same samples)

Section 3.4 end of second paragraph. Dilution of particles sourced from fires is not unexpected as transport distance increases. Does the similar fraction of the S/K particles in FT and BL during CLARIFY suggest more mixing between the FT and BL (compared to ORACLES). Is there a meteorological reason this might happen (like more convection, as suggested elsewhere based on transition in cloud field)

lines 502-505 Case for more S bearing particles in the FT during ORACLES being due to cloud processing is not really supported by Fig 8a and lines 420-425 where it is clear that it was in the MBL that ORACLES samples had more cloud influence.

Lines 507-519 This is a jumpy paragraph that is hard to glean anything from. Not clear that collocation of K, Cl and S requires gas phase HCl condensing on BBA as seasalt contains all 3 of these elements.

Section 4 (Conclusions) See opening comment regarding lack of connection to motivating questions.

Lines 528-530 Given the loss of OA in the TEM, the large fraction of internally mixed K and BC is artificially inflated, perhaps by a lot in CLARIFY samples.

Line 539-540 First sentence of next paragraph has no real point

Lines 553-554 see comment above on lines 507-519