General Comments

In their manuscript "Biomass burning and marine aerosol processing over the southeast Atlantic Ocean: A TEM single particle analysis," the authors Dang et al. present a series of analyses on aged biomassburning and marine aerosols collected during the CLARIFY and ORACLES field campaigns sampling biomass-burning aerosol and biomass-burning impacted marine air masses originating from central Africa. The paper focuses on the transmission electron microscopy analysis of collected particles and the insights that can be gained from this analysis. The analysis is wide-ranging, the paper is overall well written, interesting results are presented, and the geographic area and emission timescale covered by the field-collected samples address a significant current measurement gap. However, there are several sections of the data presentation and discussion where I feel that the authors need to include more detail, either to support the conclusions that are drawn, to cover other possible explanations for the observations, or to address the results of prior work. I believe the manuscript will be suitable for publication in *Atmospheric Chemistry and Physics* after the questions and issues below are addressed.

 The authors discuss the atmospheric processing of BBA in detail, but I'm not certain I agree with all aspects of the discussion and conclusions. The authors conclude with "Due to the considerable processing of organic aerosol and the noted effects of the MBL on BBA, it appears that aqueous processing, photolysis, and interaction with the MBL are the key drivers in physical and chemical properties such as mixing state and elemental composition of very aged BBA, rather than source" (page 16, lines 584-586).

The conclusion that fuel type is not a major factor in affecting composition and morphology of aged BBA appears to be based on observations that BBA becomes mixed with other inorganic salts through atmospheric and cloud processing and therefore the initial BBA composition is of lesser significance. This conclusion may be reasonable for BBA originating in the currently studied geographic region but does not seem generally applicable to BBA where interaction with marine air masses will not necessarily occur in the days following emission. Additionally, as the authors note, fuel type has a substantial influence on the relative and absolute prevalence of salt and chloride phases in BBA (e.g., Jahn et al. 2021; Goldberger et al. 2019; Liu et al. 2017; Levin et al. 2010) and salt phases will influence the ability of particles to uptake water and act as CCN (e.g., Gomez et al. 2018; Semeniuk et al. 2007; Pósfai et al. 2003). The present work suggests that cloud and/or aqueous processing can significantly impact BBA aging, so would the ability of fresh BBA to uptake water and act as CCN (which is strongly influenced by inorganic salt content and therefore fuel type) not be a potentially major driver in affecting how atmospheric aging proceeds?

The processing of organic aerosol is discussed in terms of organic ratios (Org_{tot}, BC, f44, f43) derived from AMS (and SP2?) measurements and from TEM observations on particle viscosity and volatility. The description of f44 and f43 in section 2.4 does not appear to be consistent with the cited work (Ng et al. 2011). Ng et al. stated that "m/z 44 is thought to be mostly due to acids... or esters" (not diacids or organic peroxides, page 5 line 190) and that "m/z 43 is predominantly due to non-acid oxygenates" (not acids, page 5 line 187). The authors mention derivation of f60 in section 2.4 but do not utilize this organic fraction during later analysis. Recent work (Hodshire et al. 2019) has highlighted the usage of the ratio f60:f44 to analyze BB plume aging in a number of studies: could such a comparison be useful in the present work as well? The authors seem to focus more on photolysis and fragmentation (in addition to aqueous processing) as ways that BBOA evolves during atmospheric aging and less on oxidation of

organics (in either the gas or particle phase), mentioning oxidation specifically only once (page 16, line 574). Oxidation increases the AMS oxidized mass fractions (f44 and f43), leads to oxidative fragmentation, and can change molecular photoabsorption cross sections. Photoxidation of laboratory-generated BBA has also been observed to alter BBOA volatilization behavior (Jahn et al. 2021) and physical properties (Jahl et al. 2021). I think the authors should address in more detail the role that oxidation may play in altering BBOA, in addition to photolysis, evaporation/condensation, and aqueous processing.

- 2. Can the authors provide more detail on sample collection? Was any size segregation performed during particle sampling? From the TEM images in the manuscript, it looks like many particles are in the submicron range; is this characteristic of all the particles that are included in the tabulated data and statistics? How long were individual filter sample collection times? Is it possible that particle morphologies or compositions were altered during collection, and if so could this affect aspects of the discussion on particle mixing state? For example: could particle impaction alter particle morphology due to, e.g., the spreading of organic material around other particle constituents or the lacey carbon framework; or could inorganic material undergo chemical reactions following collection as the aircraft passes into new airmasses?
- 3. I think the authors need to include more detail when describing the composition of different particle populations, specifically what's shown in Table 3 and Figure 11. Table 3 lists average ratios of Na:Cl and Na:S across CLARIFY filters but it's difficult to tell how much heterogeneity exists within the filter populations as a whole. Relatedly, Figure 11 illustrates the extent to which S, Na, K, and Cl are co-located within the ORACLE and CLARIFY samples but doesn't communicate anything about the elemental ratios, which makes it difficult to draw conclusions on particle composition, mixing state, and population-level compositional heterogeneity. I suggest the authors construct a series of ternary diagrams (either for the main text or SI) to more clearly show the ratios of these elements within particles, which will better allow the reader to understand and visualize the composition, mixing state, and heterogeneity of each filter's particle population. I think this would also make it easier for a reader to follow the discussion in section 3.0—Results and evaluate the conclusions in section 4—Conclusions.

Specific comments

Page 3, lines 84-85: I realize that this summary of the conclusions of Posfai et al (2003) is modeled after what's stated in the abstract of that work, but I don't believe that this is the most complete way to represent the conclusions of that work. Posfai et al. wrote "Even if the carbonaceous part of organic particles were water insoluble, the inorganic K-salt inclusions should make the mixed particle hydrophilic. Thus, organic particles with inorganic inclusions are likely responsible for the high cloud-nucleating potential of biomass smoke particles." Posfai et al. discussed the high degree of mixing between inorganics and both organics and soot, and in the abstract emphasized that observations on internal mixing informed their hypothesis on the cloud nucleus constituents of smoke. Thus, I think the authors of the present study should also emphasize that inorganic material was theorized to be important for BBA hygroscopic behavior in Posfai et al. Related to this, I would like to suggest another reference relevant for this discussion: the related work of Semeniuk et al. (2007), who analyzed the hygroscopic behavior of SAFARI BBA through TEM and concluded that inorganic particle components were the main driver behind particle water uptake and hygroscopic behavior.

Page 3, line 110: "very" is used to describe the degree of aging here and elsewhere; please be more quantitative when describing the degree of aging, either highlighting time since emission, time in cloud, and/or estimated radical exposure levels (or the ranges of these quantities, if more than one sample is being discussed).

Page 7, lines 242-244: what time period does each data point in Figure 2 (right) represent? What CO values would be considered typical of BB-influenced air vs relatively pristine marine air?

Page 7, line 249: I'm not sure what "groups" refers to in this sentence: individual particles? Different samples? Please clarify.

Page 8, lines 266-275: the observations and discussion on tar balls here is interesting. Can the authors comment on any potential reasons (differences in fuel or fire conditions, differences in other parameters measured during sampling, etc.) for why tar balls may have been observed in these two fresh samples but not others? Additionally, can the authors comment on any potential removal or transformation mechanisms?

Page 8, lines 285-288: do the authors intend to describe the mechanism by which BC is coated with inorganics? Prior work (Jahn et al. 2020; Li et al. 2003; Gaudichet et al. 1995) has described the formation and prevalence of inorganic phases in BBA as resulting (in part) from the volatilization-deposition of inorganic elements present within the biomass fuel, which can theoretically deposit to any particles generated in the BB plume.

Page 8, lines 288-289: was BC mixed with both organics and salt a mixing state that was observed at all?

Page 9, line 304: Prior electron microscopy work has indicated that there can be significant deviation from the nominal Na:Cl atomic ratio in sea spray aerosol, which is ~0.86:1 (not 1:1) and which also varies as a function of particle size (Krueger et al. 2003, see Figure 7). I suggest that the authors potentially consider additional metrics or ratios to use to examine the aging of sea spray aerosol, or at least discuss the natural variability that can be present.

Page 9, line 305: please provide a reference for the sea spray aerosol processing timeline. Is this timeline mostly universal for sea spray aerosol or does it vary by region or with influencing air masses?

Page 9, line 306: please provide citation(s) for the statement on SO_2 oxidation, as there are several mechanisms by which this can occur.

Page 9, lines 307-308: Cl would be volatilized as a gaseous molecule, not in its atomic form. Cl gases can also partition to existing Na-free aerosol and do not necessarily need to be involved in the process by which new particles form.

Page 9, line 308-309: please provide citations for the statement on determining SSA aging, as (I assume) this methodology has been described and used in prior work.

Page 9, line 320: I don't know how significantly this will affect the overall sea spray aging discussion, but I think it's worth acknowledging that prior work has observed significant variation (<1-13%) in the atomic % of S in fresh sea spray particles (Ault et al. 2013).

Page 9, line 324: specify the Na:Cl ratio on Gold 23.

Page 9, line 335: explain the rationale behind the 120 ppbv CO cutoff: is this based on other measurements from these field campaigns or prior literature?

Page 10, lines 345-346: are the high amounts of NOx based on measurements during the current campaigns or prior literature? Please specify and provide citations.

Page 10, lines 347-348: could the BC-NaNO₃ particle originate from biomass burning rather than sea spray? It is difficult to tell the size of the Na region(s) in Figure 6e. Please also specify in the text here that the proper TEM image is Figure 6e.

Pages 10-11: I have several questions related to filters Gold 14, 15, and 18 and the discussion here.

- Gold 8 was noted to have several Cl-rich particles and was described as having other markers for mixing between the boundary layer and free troposphere; could the Cl-rich particles on Gold 8 be related to those on Gold 14, 15, or 18 in any way?
- The composition of particles on Gold 14 (N and Cl) suggests that these may be composed of NH4Cl, and the discussion of the HNO3-NH3-NH4NO3 system seems to imply that the authors consider this possibility; do the authors think it's likely that the observed N-Cl particles are NH4Cl? If so, please state this directly.
- Based on the EDX data, the identity of the Cl⁻ counterion appears to be unclear in most of the Gold 15 particles; is this correct? Do the authors have any hypotheses regarding the Cl phase? Is it possible this is NH4Cl but the N signal is relatively weaker compared to Gold 14 and is therefore not visible?
- The composition of particles on Gold 18 (Mg/Ca and Cl) seems like it could suggest particles of a marine origin, as particles of similar composition (relatively enriched in Mg or Ca) and morphology appear to have been observed in prior work (Kirpes et al. 2018; Prather et al. 2013). Is the elemental composition of these particles homogeneous across each whole particle? If these particles originate from sea spray without additional processing, would that change the discussion in this section?
- Gold 18 has CO levels below 120 ppbv listed in Table 4 (119 ppbv), does this mean that this sample was not influenced at all by BB air as would be suggested based on the previously mentioned 120 ppbv cutoff? 120 vs 119 ppbv is obviously a small difference, but it illustrates the difficulties of using a sharp cutoff value.
- The authors propose that new particle formation occurred before/during flight C042, but this does not appear to be a period where new particle formation was discussed in the work of Wu et al. (2020) cited earlier in this section. Can the authors provide similar evidence (SMPS size distributions) for the assertion in the present work as was used in Wu et al. (2020)? Looking at Figure 7 it appears to me that very little time was spent in the altitude range 2250-2800 m and from Table 1 it appears that there is no AMS or SP2 data for this time period either; given the lack of data, is it reasonable to speculate on the composition and formation mechanisms of these particles? Or, are there alternative explanations to the one given in the manuscript?
- I disagree with the characterization of ammonium nitrate as semivolatile: ammonia and nitrate (as nitric acid) are volatile/semivolatile, but ammonium nitrate salt is not semivolatile.
- With the reported RH measurements, the authors seem to be implying that (at least some) aerosols are expected to be deliquesced; if this is the case, please state so directly. Prior work (Jahn et al. 2021; Semeniuk et al. 2007) has also observed BBA to take up water in this RH range.

- The work of Gunthe et al. (2021) concluded that high levels of ammonia were necessary to drive HCl partitioning to the aerosol phase. Are there any measurements during this or similar time periods that point towards what could potentially drive HCl to condense and react? I realize this may be beyond the scope of the current work, but can thermodynamic modelling (e.g., Pye et al. 2020) during this or similar periods be used to constrain the likelihood and conditions under which HCl could partition to the aerosol phase (e.g., how much HCl would need to condense to generate the observed particles, and is this amount reasonable given ambient sources and concentrations)?
- The inorganic/crustal elements observed on filters Gold 14, 15, and 18 (Si, K, Ca, Mg) have all been observed in BBA during prior work (Li et al. 2003); if the authors believe these signatures are from SSA and not BBA please make that clear.
- The authors posit (page 11, lines 389-391) that condensation of CI-species gives rise to the particles observed on filters Gold 14, 15, and 18. Prior work examining sea spray aerosol suggests that sea spray composition is heterogeneous and can vary episodically depending on the aerosol generation mechanism and other conditions (Ault et al. 2013; Prather et al. 2013; Krueger et al. 2003). Is there a possibility that some of the particles on Gold 14, 15, and 18 are primary?

Page 12, line 420: my understanding of cloud microphysics is somewhat limited; is cloud processing intensity a generally used metric? If so, please include a citation, and if not, please explain the rationale. My understanding of droplet coalescence is that the overall size distribution is relevant for predicting coalescent collisions and that clouds with high aerosol loading can have high cloud water content but a low rate of coalescence.

Page 12, line 432: please provide a citation (e.g., a recent review article) that provides some background on cloud droplet coalescence processes.

Page 13, Figure 10/discussion before section 3.4: over what area is the elemental wt% measured in each particle? If possible, please note the area over which EDX was performed, as in Figure 9. Do the EDX wt% values listed include all non-C elements? For the particles shown in the top panel, do the authors have before/after EDX data to inform what phases volatilized from the particle during analysis? The authors write that these are K-phases, but it also looks like crystalline material remains after EDX on RF5_Filter1_47. Can the authors clarify what they mean by "degradation of the K-salt during cloud processing"? Is this chemical reaction to form mixed K-phases, some degree of amorphization originating from hydration/dissolution and recrystallization, or something else? Some salt phases (some nitrates and sulfates, for example) have also been observed to be unstable upon electron beam exposure following exposure to water vapor and/or acid gases without aqueous processing (e.g., Jahn et al. 2021; Hoffman et al. 2004), offering an alternative explanation for the behavior observed here.

Pages 13-14, section 3.4: I found the discussion in this section a little difficult to follow. The paragraphs list a lot of percentages that are difficult to mentally keep track of and I ended up rereading this section a couple times. I would consider reorganizing some of this material to start a paragraph/section with a conclusion statement and then go through the numbers that support it; for example, starting the paragraph at page 13, line 476 with the idea that BBA is more diluted on CLARIFY than ORACLES filters and then explaining the numbers that support this.

Page 14, line 504: consider emphasizing that aqueous formation pathways for sulfate in cloud water are generally predicted to be faster than-gas phase formation pathways.

Page 14, line 512: In a preceding paragraph the authors suggested a marine influence for the Cl in CLARIFY aerosol, and in the preceding section (3.2) and later in this paragraph the authors discuss the mixing/coalescence of sea spray and BB particles and the fact that sea spray aerosol may contain some amount of K. Therefore, I'm wondering why co-located K/S+Cl implies Cl condensation to BBA in CLARIFY aerosol rather than mixing of sea spray and BBA or simply the presence of sea spray aerosol? Or is the condensation of Cl implied solely for the filters described in Table 4?

Page 15, lines 528-529: I don't say this to lessen the relevance and importance of the present work, but Posfai et al. (2003) did observe a large degree of internal mixing between BC and salts.

Page 15, lines 539-540: consider rephrasing this sentence, as it implies (to me) that CLARIFY/ORACLES weren't focused on BBA and that SAFARI didn't attempt to examine the noted particle constituents (which they did).

Page 15, line 547 and page 16, line 558: consider combining these paragraphs, as they appear to discuss the same phenomenon.

Technical Comments

References to Dobracki et al. (2021), Sedlacek et al. (2021), and Che et al. (2021) are included in numerous locations throughout the manuscript but are not included in the list of references. Even if these manuscripts are not published, a bibliographic entry is needed for each with the author list, working title, and preparation status.

The word "collocate" is used in various places in the manuscript; I believe the authors intend to use the word co-locate (alternatively colocate), as collocate is a separate word

Page 2, line 53: "BBA aerosol" is redundant

Page 4, line 125: include the product ID# for the holey (lacey?) TEM grids and nuclepore filters

Page 5, line 206 (and potentially elsewhere): "above cloud samples" \rightarrow "above-cloud samples"

Page 10, line 369: add that Wu et al. (2020) is also based on the CLARIFY campaign to emphasize the relevance to the discussion here

Page 10, line 372: add the CPC to the instrumentation listed in the Methods section

Page 12, line 417: there's an extra "spent"

Page 13, line 459: "about" \rightarrow "amount"

General Table and Figure comments: please try to increase the size of axis labels, legends, and in-figure labels, as these were overall difficult to read. I also urge the authors to use a, b, c... labelling for figure panels to enable clearer references to specific panels within the manuscript. As many of the TEM images in tables are too small to see clearly, consider including larger images in a supplementary file.

Page 25, Figure 5: three numbers are present in the ratio above the left-most TEM image but the order and identity of the third number isn't listed

Page 26, Table 3 caption: "forum" → "from"

Page 30, Figure 11: The ORACLES and CLARIFY labels within the figure are switched from the caption

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