We thank both reviewers for their time and comments and address their comments below. Line numbers refer to the track changed version on the manuscript.

Referee 1

In my review of the original version of this manuscript I noted that the wide variety of particles that were sampled and described was kind of interesting, but there was no coherent narrative establishing progressive evolution of aged BBA nor how the changes during aging might be optically or biogeochemically important. This revised version is perhaps even less coherent. Nearly every particle type now has multiple hypotheses advanced in explanation, with nearly no attempt to develop arguments favoring one theory over the other.

It is now not clear whether the authors are confident that the TEM EDX results can distinguish between SSA and BBA after processing, nor whether the unusual Cl rich particles are secondary or primary. The lack of tarballs in the older smoke sampled in these studies compared to abundant tarballs in SAFARI remains interesting, but suggesting "a removal process, potentially through deep precipitation" that selectively scrubs tarballs but not the rest of BBA is not at all helpful.

Thank you for your comments. We address the evolution of BBA, TEM-EDX, describe optical property implications, Cl-rich particles, and tar balls below.

We think there is a narrative of evolution of BBA described in the paper, and summarize here. A major theme of the paper is the interaction between sea salt aerosol with BB plumes and biomass burning aerosol with marine air. Our findings are that BB aerosol are affected by the marine boundary layer and cloud processing as they age, and that organic becomes increasingly volatile with aging. Na and/or Cl can be mixed with the black carbon, potassium salts and organic from biomass burning plumes through aqueous processing or secondary processing. BB plumes and the higher levels of NO_x and SO_x can affect sea salt aging through more rapid Cl depletion. There is evidence of BL and FT top-of-cloud mixing as the BB plume advects west. TEM EDX results can distinguish between SSA and BBA, primarily because SSA will have a predominance of Na and/or Cl, and BBA aerosol will have more K in the form of potassium salts and/or the presence of BC. TEM is useful in that in can detect small changes in individual particle composition and processing on a single particle level, for example increased Na mixing with BC, so while there can seem to be a merging of particle types- we still can distinguish source based on the main element and their prevalence in individual particles. Caveats throughout the paper (ie, Cl and Na elements have been found in biomass burning particles) are for context, but if a particle is predominantly Na or Cl then we deduct that it is from a marine source; further we use ancillary data such as CO, altitude at sampling and backtrajectories to inform our assessments.

The unusual Cl rich particles were found on filters Gold 14, 15, and 18. While Gold 14 was collected above-cloud and EDX spectra showed strong Cl and N peaks, Gold 15 was collected below-cloud with C, Cl and small amount of K and Si. Gold 18 had Ca and Mg mixed with the Cl. Different mechanisms were proposed for the three filters based on elemental composition. We cannot assess whether they are secondary or primary, given that these types of particles have not been often observed in field observations, but put forth different potential pathways for their formation such as HCl uptake onto

liquid particles or gas-to-aerosol partitioning, and Ca and Mg distributed in a sol-gel particle for the Cl particles containing Ca and Mg based on prior studies (lines 426 to 462).

As for climate implications, radiative effects due to inorganic mixing with BC and changes in in hygroscopicity are noted in lines 604-607. Further we note tar ball effects on models in lines 620 to 627: "Tar ball incorporation in BB models (Jacobson 2014) have been hindered due to lack of data, as tar balls can only be definitively detected with time-intensive single particle electron microscopy. While other work shows that TBs are a significant fraction of BB aerosol, with some showing that tar balls outnumber BC by a factor of 10 (Hand et al. 2005; China et al., 2013), our analysis shows a lack of tar balls in the aged BB plumes, consistent with Posfai et al. (2003) who also reported a dearth of tar balls in aged plumes as a puzzling phenomenon. Since tar balls are a light-absorbing particle, the absorption from aged plumes is dominated by non- tar ball components like BC, brown carbon, dust; the absence of tar balls in these aged plumes can help constrain models on radiative forcing in the region."

As the processes for tar ball removal are not clear, we have changed the line to : "This suggests a removal process, and while there are many unknowns regarding loss processes for tar balls, precipitation near the coast or heterogeneous, photolytically-driven processes which may affect the solubility or volatility of tarballs as they are advected west over the ocean may contribute to their removal. Posfai et al. (2003) also reported a dearth of tarballs when sampling in the haze layers representing aged BB plumes, without a clear explanation for their absence." (lines 313-317)

Referee 2

I believe the paper is much improved in terms of the overall presentation, discussion and analysis of results, and the summary of the conclusions and significance. I have only a couple minor comments. Line numbers below refer to the new manuscript without tracked changes.

Page 17, lines 597-600: "This is important as it suggests that the salts formed in the fire via evaporation and recondensation drive the mixing of the carbon aerosol as the secondary inorganic condenses, and that the organic fraction is separate. This is consistent with findings regarding emissions of BC and K-salts and other salts in the flaming phase of a fire, while organic emissions occur during the pyrolysis or smoldering phases (Haslett et al., 2018)."

The discussion quoted above is interesting and provides useful insight on BBA mixing state. However, I would point out that the work of (Haslett et al. 2018) and related work of (Fawaz et al. 2021) utilized uniform sections of woody biomass combusted under tightly controlled conditions, which can differ from field conditions where temperature gradients may exist within the fire and fuel can be highly variable (in terms of leafy vs woody and/or wet vs dry). This can lead to some degree of spatial or temporal variability in fire emissions and may impact the very near-source mixing of fresh BBA. I don't believe that the quoted passage needs to be altered, but I do suggest the authors consider whether to add a caveat to this discussion.

We have added a caveat that field conditions may also affect the near- source mixing for fresh BBA, aside from the smoldering and flaming phases. (lines 609-611)

 Page 17, lines 606-609: “While the SAFARI campaign and other recent biomass burning campaigns found tar balls, our TEM analysis did not find tar balls other than on filters RF10 and RF11, which were aged for approximately 1 and 2 days, respectively. This finding implies a reduction in tar balls in aged African BB plumes.”

Would this paragraph be an appropriate place to discuss potential implications of tar ball loss with aging, for example regarding the evolution of optical properties of African BB plumes? While a major area of tar ball research has been their optical properties, I realize there are still many unknowns regarding the specifics of tar ball optical properties as well as transformation or loss processes, however the direct TEM observations in the present work give the authors a unique position to comment.

Yes, thank, you, we discuss the implications in the reply to Referee 1.

Fawaz, M., Avery, A., Onasch, T.B., Williams, L.R., and Bond, T.C. (2021). Technical note: Pyrolysis principles explain time-resolved organic aerosol release from biomass burning. *Atmos. Chem. Phys.* 21 (20):15605–15618. doi:10.5194/acp-21-15605-2021.
Haslett, S.L., Thomas, J.C., Morgan, W.T., Hadden, R., Liu, D., Allan, J.D., Williams, P.I., Keita, S., Liousse, C., and Coe, H. (2018). Highly controlled, reproducible measurements of aerosol emissions from combustion of a common African biofuel source. *Atmos. Chem. Phys.* 18 (1):385–403. doi:10.5194/acp-18-385-2018.