We thank reviewer 1 for their thoughtful and helpful review. Our response is below.

1. *I worry that the study overestimates the effects of dilution on OA concentrations. Specifically, Fig. 6d predicts significant evaporation of OA for the two lowest-intensity fires (approximately 40% reduction in OA mass over the first ~30 min). These predictions do not seem consistent, qualitatively or quantitatively, with any ambient observations that I am aware of. For example, see Fig. 7 of Cubison et al. (2011), which compiles results for BB plume aging over similar timescales. Even in Akagi et al. (2012), where a net decrease in OA was observed, the ambient observations are qualitatively quite dissimilar from the predictions in this paper.*

Response: Our simulations suggest the patterns of OA evolution are sensitive to the fire sizes. The burn area for Williams prescribed fire in the study of Akagi et al. (2012) was 81 hectare (i.e. 0.81 km^2). This may be more comparable with our simulation for the fire size of 1 km^2. May et al. (2015) showed that OA reduction was approximately 50% during the plume evolution, which is more similar to our smaller simulated fires. We added the following in the main text from line 68: "…production or even a net loss (Akagi et al., 2012: May et al., 2015). OA loss in first hour after emission was approximately 50% in the study of May et al. (2015), OA consists of …"

The current results would seem to predict that BB emissions at night would undergo even more dramatic decreases in OA, since they would likely be far more impacted by dilution than chemical SOA production (even assuming nitrate radical chemistry). I'm not sure if nighttime BB plume evolution has ever been observed, and perhaps some of the differences noted above are due to fire intensity, but I would push the authors to evaluate their predictions of dilution/evaporation further.

7. *Similarly, it may be outside the scope of this study, but can the authors use their results to make conclusions about the evolution of BB emissions at night?*

We combined our response to address above two points about night-time evolution: It may be difficult to generalize about day/night differences due to various aspects being different between day and night on average. In general, nighttime plumes may have (1) less dispersion in the boundary layer due to more-stable air, (2) different chemistry, and (3) lower emission fluxes as peak fire intensities are typical during the day (this may affect fire size too; Zhang and Kondragunta, 2008; Wooster and Lagoudakis, 2009). It's unclear how the convolution of these differences might impact the plumes, and it probably varies between cases.

We added the text from line 554: "For nighttime OA evolution, it may be difficult to generalize about day/night differences due to various aspects being different between day and night on average. In general, nighttime plumes may have (1) less dispersion in

the boundary layer due to more-stable air, (2) different chemistry, and (3) lower emission fluxes as peak fire intensities are typical during the day (this may affect fire size too; Zhang and Kondragunta, 2008; Wooster and Lagoudakis, 2009). It's unclear how the convolution of these differences might impact the plumes, and it probably varies between cases."

2. *As stated by the authors (line 316), OAERchem cannot really be evaluated against observations. It is completely dependent upon parameters that can vary quite a bit across different models. This study demonstrates a few of the model parameters that influence OAERchem, but there are many more. I found the motivation for OAERchem to be quite confusing (lines 309-317). I encourage the authors to more clearly describe what it is that they hope to show with this quantity, and how it can be used in practice (beyond the current study). For example, they point to some valid limitations of OAERinert, but there would seem to be equal (if not greater) limitations of OAERchem simply introduced by different models or the choice of model parameters.*

Response: $OAER_{chem}$ can certainly vary across models due to different assumptions. We introduced $OAER_{chem}$ to isolate the effect of SOA formation to give an alternate metric to $OAER_{inert}$, which is the convolution of evaporation and SOA formation. While $OAER_{chem}$ cannot be evaluated against measurements, it does tell us what the isolated impact of SOA formation is *for the choice of model parameters used in the simulation*. We have modified the motivation of $OAER_{chem}$ from line 309: "To isolate the impact of SOA formation alone on our simulations, we introduce the chemistry OA mass enhancement ratio ($OAER_{chem}$) to give an alternate metric of $OAER_{inert}$ (which is the convolution of both evaporation and SOA formation). We define $OAER_{chem}$ as the ratio of predicted…"

3. *In Section 3.4, the authors should add some discussion to prior studies that make similar observations: e.g., Capes et al. (2008) observed significant increases in O:C ratios of the organic aerosol, but a small decrease in the normalized OA mass concentrations; Hennigan et al. (2011) present similar observations through their "aged POA" analysis.*

Response: We have added text in the lines 552-555. "Papers analyzing field observations have suggested this possibility. Capes et al. (2008) and Cubison et al. (2011) observed significant increases in O:C ratios of the organic aerosol with aging, but a small decrease in the normalized OA mass concentrations; Akagi et al. (2012) observed the decrease of OA with aging and attributed this to the processes of particle evaporation. Similarly, Jolleys et al. (2015) observed increasing O:C elemental ratio with aging but lowering normalized OA concentrations in the smoke plumes, and they attributed this to the combination of dilution and chemical processing. May et al. (2015)

also suggested the competition between dilution-driven evaporation and SOA formation during the plume transport may be occurring in their observed plumes, as they found approximately 50% reduction of OA after several hours of aging with increasing in the O:C ratio. Additionally, the lab study of Hennigan et al. (2011) also showed increased O:C ratios in experiments with decreasing OA concentrations. Our modeling result is consistent with the findings from these observational studies reporting increased oxygenation with time for the OA even with observed decreases in the relative amount of OA (or a relative constant or lower $OAER_{inert}$).”

4. *In the treatment of vapor wall loss, does the model allow for the reversible partitioning of vapors from the walls back to the gas phase as a compound is oxidized? Vapor wall loss is described as an equilibrium process (line 96), which implies that it is reversible – if this is/is not treated – how does this impact the current predictions?*

Response: The vapor wall loss is treated as a reversible partitioning process. Previous studies (Bian et al., 2015, Zhang et al, 2015) suggested two variables could influence vapor wall loss: the effective saturation of vapor with respect to the wall ($C_w/M_w\gamma_w$) and the accommodation coefficient for vapor into the wall (α_w). We performed the sensitivity tests on these two variables and showed that the simulations overlap with the measurement of Hennigan et al. (2011). However, as we stated in the manuscript, we are unable to determine which set of α_w, $C_w/M_w\gamma_w$, and chemistry assumptions best represent the actual processes occurring in the chamber, since different combinations of these values can reproduce the observed $OAER_{inert}$ range. However, if vapor wall loss is turned off, the amount of OA mass increases greatly over simulations with vapor wall loss on – regardless of what vapor-wall-loss and chemistry parameters are chosen. Therefore, the prediction of vapor wall loss has large uncertainties depending on the two variables, but this does not influence our main conclusion.

5. *This is more of a stylistic comment, but the writing in the first person is highly distracting. The terms "we" and "our" are used too extensively throughout the paper. I recommend changing to the third person voice, where possible.*

Our use of "we" and "our" is to keep our writing concise and direct, and to generally use the active voice. This link (https://cgi.duke.edu/web/sciwriting/index.php?action=passive_voice) provides a nice overview of the pros and cons of active and passive voice in scientific writing (but does not conclude that one must err to using one or the other). I (Jeff Pierce writing here) feel personally that the advantages of active voices outway disadvantages, and I personally find writing that avoids "we" and "our" harder to follow and more work to read. If you see me (Jeff again) at a conference or meeting, feel free to approach me about this if you don't mind losing your anonymity. I'm interested in learning about why you

feel "we" and "our" is distracting as I realize that not everyone has the same writing preferences, and it's good to try to write in a way that satisfies as broad an audience as possible.

6. *This is probably outside the scope of this study, but it is worth noting that other factors related to fire intensity may also contribute to different aging characteristics in BB plumes (e.g., in a high intensity fire, the smoke optical thickness may produce differences in photochemistry...the formation of pyrocumulus clouds could also dramatically impact chemistry...etc.).*

Fire intensity certainly influences OA evolution in the plume. We performed the test on the high and low emission mass flux (5×10^{-6} and 2×10^{-8} kg m^{-2}s^{-1}). For OA evolution for fire size of 1 km^2 under Atmospheric Class of D in Fig 7 and 8, high emission mass flux (i.e. large fire intensity) has lower OAER$_{inert}$ and OAER$_{chem}$, compared with low emission mass flux, suggesting that under the same dilution ratio, lower emission mass flux has slightly more-effective SOA formation. OA concentrations for lower emission mass flux quickly drop close or below the background non-volatile OA concentrations and further dilution does not lead to further evaporation. The evaporated organics are available for SOA formation. Both of OAER$_{inert}$ and OAER$_{chem}$ after 4 hrs were thus higher for low emission mass flux than high emission mass flux.

We added text to the paragraph after line 523: "Fire intensity also influences OA evolution in the plume through changes in emission fluxes. Compared OA evolution for fire size of 1 km^2 under Atmospheric Class of D in Fig 7 and 8, high emission mass flux (i.e. large fire intensity) has lower OAER$_{inert}$ and OAER$_{chem}$ than that of low emission mass flux, suggesting that under the same dilution ratio, lower emission mass flux has slightly more-effective SOA formation. OA concentrations for lower emission mass flux quickly drop close or below the background non-volatile OA concentrations and further dilution does not lead to further evaporation. The evaporated organics are available for SOA formation. Both of OAER$_{inert}$ and OAER$_{chem}$ after 4 hrs were thus higher for low emission mass flux than high emission mass flux." We also added the background OA concentration line in the Figures 6 to 8.

Mok et al. (2016) estimated that the reduced UV due to brown carbon and black carbon could slow down the photochemical rate as radicals OH, HO2 and RO2 was decreased in the plume by 17%, 15% and 14%, respectively. Also, cloud processing of smoke from biomass burning in the pyrocumulus clouds (and other clouds that the plume cycles through) could largely alter the smoke chemistry (Yokelson et al., 2003; Akagi et al., 2011). However, due to the limited information to constrain the chemical mechanism in our model, we only simulate gas-phase functionalization and do not include aerosol-phase or heterogeneous reactions, or cloud processing. We also added the text after

line 228: "We also do not include aerosol-phase or heterogeneous reactions, cloud processing, or effects of smoke on oxidant fields in our model, although these processes may affect the chemistry of plume (Yokelson et al., 2003; Akagi et al., 2011; Mok et al., 2016). The SOA mass yield $\alpha_{i,j}$ is assumed to be 1 for all reactions. We use this simple assumption of chemistry as a first test in our chamber and plume systems as we found that we did not have enough information to constrain gas-phase yields or additional chemistry mechanisms beyond this."

8. *Finally, the References need to be carefully checked – they are out of order, and some are not the correct form (e.g., ACPD article cited when the article has been published in ACP).*

Corrected.

Technical Corrections

1. *Delete Lines 141 – 149 ("We describe our aerosol microphysics model…presents our conclusions.") – the sections have clear headings so this is redundant.*

Done.

2. *Delete the sentence starting on line 320 – the section heading is just above this sentence.*

Done.

3. *Delete the sentence starting on line 344 – the section heading is just above this sentence.*

Done.

4. *Line 571: change "the" to "some"*

Done.

5. *Line 67: Grieshop et al. (2009) was a chamber study, not a field study.*

Deleted.

6. *Line 188: most chambers are rectangular or cubic – what is chamber radius?*

The chamber of Carnegie Mellon University was nearly cubic. We assume the chamber to be a sphere to allow for an analytical solution of turbulent wall-loss rates following Crump and Seinfeld (1981) and implemented in Pierce et al., (2008) on a similarly shaped chamber.

Changed to "…R is the radius of the chamber on the assumption that the chamber is a sphere…"

7. *Line 324: rewrite this sentence to be less awkward.*

Done.

8. *Line 328: "…simulations are shown…"*

Done.

9. *Line 438: is the term "perfect accommodation" technically preferred?*

Changed to "…A value of 1 represents no limitation on the vapor-wall loss rates due to this process…"

10. *Line 520: do the authors mean 'OA' instead of 'BC'?*

Corrected.

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Capes, G., et al.: Aging of biomass burning aerosols over West Africa: Aircraft measurements of chemical composition, microphysical properties, and emission ratios, J. Geophys. Res., 113, D00C15, 2008.

Crump and Seinfeld: Turbulent deposition and gravitational sedimentation of an aerosol in a vessel of arbitrary shape, J Aerosol Sci., 5, 405-415, 1981.

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