Review of "Secondary organic aerosol formation in biomass-burning plumes: Theoretical analysis of lab studies and ambient plumes" by Bian et al.

General Comments

This paper presents a modeling analysis of SOA formation in aging biomass burning (BB) plumes. The study presents several new angles on this topic, namely a) the effects of vapor wall losses on SOA quantified in chamber studies and b) the effects of plume dilution (as related to fire size and meteorological stability) on SOA production. Both of these effects appear to have significant impacts on the interpretation of chamber data and ambient BB plume evolution. The scope of the work is clearly appropriate for *ACP*, the results are novel and will be of interest to many in the atmospheric chemistry community. Overall, the manuscript is well organized, the writing is good, and the presentation is clear. Thus, I recommend the paper for publication in *ACP* after the below comments are addressed.

Specific Comments

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. 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.

2. As stated by the authors (line 316), $OAER_{chem}$ 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 $OAER_{chem}$, but there are many more. I found the motivation for $OAER_{chem}$ 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 $OAER_{inert}$, but there would seem to be equal (if not greater) limitations of $OAER_{chem}$ simply introduced by different models or the choice of model parameters.

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.

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?

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.

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.).

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?

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).

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.

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

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

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

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

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

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

8. Line 328: "…simulations <u>are</u> shown…"

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

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

References

Akagi, S. K., et al.: Evolution of trace gases and particles emitted by a chaparral fire in California, Atmos. Chem. Phys., 12, 1397-1421, 2012.

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.

Cubison, M. J., et al.: Effects of aging on organic aerosol from open biomass burning smoke in aircraft and laboratory studies, Atmos. Chem. Phys., 11, 12049–12064, doi:10.5194/acp-11-12049-2011, 2011.

Grieshop, A. P., et al.: Laboratory investigation of photo chemical oxidation of organic aerosol from wood fires 1: measurement and simulation of organic aerosol evolution, Atmos. Chem. Phys., 9, 1263-1277, doi:10.5194/acp-9-1263-2009, 2009

Hennigan, C. J., et al.: Chemical and physical transformations of organic aerosol from the photooxidation of open biomass burning emissions in an environmental chamber, Atmos. Chem. Phys., 11, 7669-7686, 2011