

Interactive comment on “Molecular insights on aging and aqueous phase processing from ambient biomass burning emissions-influenced Po Valley fog and aerosol” by Matthew Brege et al.

Anonymous Referee #2

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This manuscript presents molecular-level analyses of fresh versus aged fog and samples influenced by biomass burning. The authors aim to explore the potential importance of aqueous phase processing on alteration of organic matter chemical compositions. The authors reported that aged aerosols and fresh fog samples show similarity in composition, indicating the possibility of aged aerosols that served as fog nuclei.

One of my major concerns for this manuscript is that the authors attributed the CHON and CHOS compounds exclusively to organonitrates and organosulfates based on FT-ICR MS analyses, but this is not supported by NMR spectra! This seems to be a major finding but it was not discussed in great detail. It looks to me that other types of organic

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nitrogen and organosulfur compounds may contribute to formation of detected CHON and CHOS that need further investigations.

In addition, nitro groups (R-NO₂) and nitrooxy groups (R-ONO₂) are different. They have distinct formation processes and physiochemical properties as well (e.g., lifetime against hydrolysis). The authors need to be clear when discussing their findings in context of literature.

For the results and discussion, the current form of manuscript is a bit lengthy and repetitive when reporting the FT-ICR MS data. A more concise presentation will greatly improve the readers' reading experience.

Also, reactions in aerosol liquid water content and in fog should be discussed separately. Based on the results presented (with only 1 sample in each category), the aqueous processing does alter the chemical compositions of organic matter, but the pathways are rather inconclusive.

Overall, this is still a nice case study that provides useful information. Below I provide a few more specific comments for the authors' consideration and clarification.

Specific Comments:

1) Page 4, lines 7-9: The aerosol filter extracts were filtered with 0.45 μm PTFE membrane, while the fog water was filtered through 47 mm quartz fiber filters. What is the pore size of 47 mm quartz fiber filters? Why did the authors use two different filtering methods here? Since the FT-ICR MS analysis is very sensitive, potential artifacts (even trace amounts) during sample preparation should be avoided.

2) Page 7, line 6: Does “SOA-like” mean oxygenated/or functionalized/or fragmented? It is not clear here.

3) Page 10, lines 25-35: Since aged aerosols could act as fog nuclei, scavenging of organosulfates resided in aged aerosols into fog might have contributed to the observed organosulfates in fresh fog water. Based on the data presented, I don't really

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see direct evidence here showing that aqueous processing leads to CHOS production. Similarly, on Page 13 lines 12-17: the authors concluded that the current data provide strong evidence of aqueous processing that dominates the production of S-containing organic matter. I would tone down this statement.

4) Page 12, line 32: “hygroscopic” is a better term to describe aged/oxygenated organics that contribute to droplet formation.

5) Page 13, lines 8-9: it is confusing when the authors stated “some evidence of dimerization” here. This was not presented in “results and discussion” but suddenly mentioned in summary.

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