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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "The effects of isoprene and NO$_X$ on secondary organic aerosols formed through reversible and irreversible uptake to aerosol water" *by* Marwa M. H. El-Sayed et al.

Anonymous Referee #2

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General comments: This work examines aqueous SOA, both reversible (able to evaporate upon drying) and irreversible, in the Eastern US using measurements of water soluble compounds in both the gas and particle phase. Additional measurements (isoprene, NOx) are used to infer that the reversible SOA is a result of isoprene epoxydiol (IEPOX) uptake to an aqueous medium. This paper examines an important issue with implications for what controls IEPOX SOA formation. However, to further their conclusions it would be good to demonstrate stronger connections between isoprene and the reversible SOA since no chemical identity beyond WSOCp (particulate WSOC) and WSOCp,dry (dried WSOC) is known for the organic aerosol. The major pieces of evidence for IEPOX being the precursor to reversible SOA come from NOx and isoprene

concentrations and time lag analysis. The WSOCp peaks 9 hours after isoprene (consistent with IEPOX being 2nd generation plus an additional lag), and the reversible SOA is highest when NOx/Isoprene is lowest which is consistent with our understanding of IEPOX formation in the gas-phase. However, formation of IEPOX may not be the limiting factor for IEPOX SOA formation (sulfate and its influence on particle surface area/volume as well as acidity may be responsible). In addition, other aspects of the ambient atmosphere are changing in addition to NOx and isoprene as a function of season. Two areas that could be furthered include:

1. Can mass closure be reached in terms of how much isoprene is present and the amount of WSOCp and WSOCg?

a. E.g. Page 6 line 25: Do you get mass closure if you assume 5 ppbC of isoprene reacted forms 2 ugC/m3 IEPOX?

b. Is the reversible IEPOX SOA just dissolved IEPOX or is it a reversibly formed reaction product? Are the levels of reversible IEPOX SOA consistent with dissolved IEPOX? Sareen et al. 2017 indicate dissolved IEPOX alone is a very small concentration (especially compared to IEPOX SOA from AMS PMF analysis).

2. Were other proxies for chemistry besides NOx/Isoprene examined?

a. Page 7, near line 10: Is the diurnal variation in sulfate involved in IEPOX SOA?

b. Figure 7 shows seasonality in the WSOCp,dry/WSOCp ratio consistent with changes in NOx/Isoprene. What else changes as a function of season that could also explain the ratio? Oxidants? How is ALW changing? If the horizontal axis was sulfate or SOx divided by isoprene would it show the same behavior?

Other Specific Comments:

3. Page 1: Lines 23-24 indicate that the trend towards lower NOx/Isoprene ratios may mean more IEPOX SOA in the future. Given the dependence of IEPOX SOA on sulfate, wouldn't we expect this pathway to decrease with de-

creasing sulfate levels in the future as demonstrated by Marais et al. 2017 ERL (http://iopscience.iop.org/article/10.1088/1748-9326/aa69c8/meta)?

4. Page 1: Line 29 indicates isoprene is the dominant SOA precursor in summer. I would define dominant as responsible for >= 50% of SOA. Hu et al. 2015 ACP (https://doi.org/10.5194/acp-15-11807-2015) indicate isoprene (or IEPOX) is responsible for 17% to 36% of Southeast US SOA. So while it is important, it is not dominant.

5. Page 2: Lines 21-23: I would characterize both Marais et al. 2016 and Pye et al. 2013 as irreversible IEPOX uptake since both use a reactive uptake formulation. The major difference between Marais et al. and Pye et al. is the Henry's law coefficient which leads to different amounts of IEPOX SOA. They also simulated different years. Budisulistiorini et al. 2017 has shown that reversible (simpleGAMMA, McNeill et al. 2012) and irreversible (CMAQ, Pye et al. 2013) models of IEPOX uptake can agree when the parameters going into them are identical (for $\sim$6 hours of processing time).

6. Page 3: Line 17-18: which instruments may not measure reversible SOA?

7. Page 4: Near line 30: Can you clarify the relationship between WSOCp and aqSOA? What fraction of WSOCp is aqSOA? How was aqSOA identified?

8. Page 8: Line 35: How much higher is the fraction of reversible aqSOA? Insert value

9. Page 8: Line 36: For the range of 0-60%, what is the typical value (mean, median, or similar)?

10. Page 9: Line 18-22: I am unclear as to whether or not the work of Wong et al., 2015 is atmospherically relevant if their experiments did not produce SOA from IEPOX. D'Ambro et al. 2017 ES&T (http://pubs.acs.org/doi/abs/10.1021/acs.est.7b00460) demonstrates that IEPOX is the atmospherically relevant pathway to isoprene SOA and laboratory experiments with unrealistic concentrations may be activating pathways that are not important in the atmosphere.

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