

Review of the revised manuscript of Beardsley et al.

The authors have addressed most of my comments adequately. I recommend accepting manuscript after minor revisions.

1. The units in the manuscript are still confusing. The authors claimed that  $[H^+]$  has the unit of mol/L (p1 line13), but  $[SO_4^{2-}]$  has the unit of  $\mu\text{mol}/\text{m}^3$  in figure 4. Also, the authors cite Xu et al. (2015) in p20 line 27 stating that "...ambient isoprene SOA formation in the SE U.S. to be most highly correlated with  $[SO_4^{2-}]$ , and insensitive to  $[H^+]$  and LWC". It is important to note that in Xu et al. (2015),  $SO_4$  and LWC have unit of  $\mu\text{g}/\text{m}^3$  air, but  $H^+$  has the unit of mol/L. Thus, I suggest the authors clearly and carefully discuss the units throughout the manuscript and possibly use different symbols for mol/L and  $\mu\text{mol}/\text{m}^3$

2. Figure 4: Is the  $[SO_4^{2-}]$  reported in this figure measured by PILS-IC? I am surprised that the  $[SO_4^{2-}]$  is so constant. If it is truly from measurement, it suggests that all organosulfate decomposes in the PILS. This is also surprising to me considering that the organosulfate is generally low volatile and the steam in PILS is only  $\sim 100^\circ\text{C}$ .

3. p13 line 21-24. The authors argue that the large contribution from  $OM_{AR}$  to  $OM_T$  (i.e.,  $>65\%$ ) in nucleation experiment is consistent with previous studies by Surratt et al. (2006) and Nguyen et al. (2010). However, I don't agree with the authors. Surratt et al. (2006) showed that oligomers comprise only  $\sim 22-34\%$  of the high- $NO_x$  SOA mass. For the  $NO_x$ -free experiments in Surratt et al. (2006), organic peroxides (not all are oligomers) account for 61% of total OA formed by nucleation. In Nguyen et al. (2010), no additional  $NO_x$  was injected, which is different from the experimental conditions in the manuscript. Also, as noted by Nguyen et al. (2010), the fraction of oligomers generally increases with increasing initial precursor concentrations. This suggests that the oligomers are likely formed by the  $RO_2+RO_2$  reactions in the gas phase considering the high amount of initial VOC concentrations in the experiments.

4. p21, line 2. The authors cite Lewandowski et al. (2015) to support the importance of  $[H^+]$  (p33142 line 15). However, as pointed in my previous comments, sulfate ( $\mu\text{g}/\text{m}^3$ ) correlates perfectly with  $[H^+]_{\text{air}}$  ( $\text{nmol}/\text{m}^3$ ) in Lewandowski et al. (2015), so that it is difficult to argue if the yield enhancement is due to  $[H^+]$  or sulfate or LWC, etc...I suggest to remove this sentence.

5. p12 line 2. The calculation of  $[\text{SO}_4^{2-}]_{\text{free}}$  is still confusing. Firstly, how do the authors define “sulfate not associated with ammonium”? In  $\text{NH}_4\text{HSO}_4$ , is sulfate associated with ammonium? Secondly, the  $\text{SO}_4$  in ammonium sulfate can still act as a nucleophile to form organosulfate. Thus, the  $[\text{SO}_4^{2-}]_{\text{free}}$  should not be zero in ammonium sulfate.