

Response to the comments of Anonymous Referee #2

This paper demonstrates that interaction with water, especially in the presence of iron, leads to production of OH from secondary organic aerosol (SOA). The authors explain that this is reasonable because organic peroxides are a major component of secondary organic aerosol, and OH can be formed by decomposition of organic peroxides in water.

The science is sound and the results are important because they help to explain how condensed phase oxidation can lead to the aging of SOA in the atmosphere (both fragmentation and functionalization of compounds). This can change the O:C ratio and the mass of organic aerosol.

Response:

We thank the referee for review and positive comments. The point-by point responses are given below.

1. The authors discuss several reasons why the formation of OH radicals in wet particles might be important, including in the oxidation of sulfur dioxide to sulfuric acid. However, they miss one: water-soluble gases, such as aldehydes, can be taken up by wet particles and if there is sufficient OH present they can react to form low volatility products and (e.g., organic acids and oligomers). Thus SOA can form from gaseous precursors through aqueous phase reactions in wet aerosols, if sufficient oxidant is present (see work by Ervens, Turpin, Monod, and others). McNeill and Ervens both argue that this chemistry will be OH limited. Thus the availability of OH radicals in the condensed phase will aid that chemistry.

Response: Thanks for raising this important point. We will add a discussion on SOA formed through aqueous phase chemistry into the manuscript as follows:

Water-soluble gases such as aldehydes taken up by deliquesced particles may undergo reactions under presence of OH radicals to form low volatility products, including organic acids, peroxides, peroxyhemiacetals, and oligomers (Lim et al., 2010; Ervens et al., 2011; Liu et al., 2012; Ervens, 2015; Lim and Turpin, 2015; McNeill, 2015). Thus, the formed OH radicals would promote chemical aging of SOA especially in the presence of iron ions (e.g., SOA coated mineral dust particles) (Chu et al., 2014) and may also induce aqueous-phase oxidation of sulfur dioxide forming sulfuric acid (Harris et al., 2013).

2. The authors might also find the following recent ACP reference about organic peroxides and OH helpful: Lim, Y. B., and B. J. Turpin. "Laboratory evidence of organic peroxide and

peroxyhemiacetal formation in the aqueous phase and implications for aqueous OH." Atmospheric Chemistry and Physics 15.22 (2015): 12867-12877.

And this one about formation of oligomers from OH oxidation in wet aerosols (relevant to page on page 30027): Lim, Y. B., Tan, Y., Perri, M. J., Seitzinger, S. P., and Turpin, B. J.: Aqueous chemistry and its role in secondary organic aerosol (SOA) formation, Atmos. Chem. Phys., 10, 10521– 10539, doi: 10.5194/acp-10-10521-2010, 2010.

Response: Yes, these reference papers are closely related to this work. We will include them in the revised manuscript. Please see the response above.

3. Figure 2 and first paragraph of results. The reader would benefit from more complete explanation of what is shown in Figure 2 and what it means.

Response: As explained in the third paragraph of the results and discussion session, Figure 2 shows EPR spectra of various samples of water extracts of SOA by α -pinene, β -pinene, limonene, and isoprene as well as field fine particles. The four peaks at specified positions are characteristics for OH radicals and the formation of OH radicals can be enhanced by Fe^{2+} . We have revised section 3 by adding more descriptions.

4. Bigger fonts are needed in the figures. The meaning of the shading in Figure 7 should be explained in the figure caption. Figure 8 – implications should include reactive uptake and SOA formation from oxidation of water-soluble gases.

Response: The fonts in most figures were enlarged and more description on the meaning of the shading in Figure 7 was added to the text. Regarding Fig. 8: this study has indeed important implications for reactive uptake and SOA formation from oxidation of water-soluble gases. However, we would like to keep Fig. 8 as simple as possible.