In this work the authors report OH radical observations in aqueous solutions of secondary organic aerosol in the absence of radiation. They postulate that organic hydroperoxides (ROOH) can decompose in water and form OH radicals. The formation rate of OH was enhanced in the presence of Fe2+, following a Fenton-like mechanism to catalyze radical production. The manuscript is well written, and the experiments were carefully performed. I have a few minor comments, and recommend publication in ACP after these concerns are addressed.

Major comments:

- How is the concentration of OH quantified? Since the major finding of this work is OH radical formation, it seems that the quantitation method should be clearly described and justified. It seems to me that the OH is estimated by fitting the EPR data (Supplement Figure S1). What is the sensitivity of the fitting to quantification? What are the detection limits, limit of detection, limit of quantification etc.? Are there standards for quantification? It is acceptable if no standards are available (since OH is very difficult to measure), but perhaps there are other methods (e.g. tracer method used by the Abbatt group in Toronto) for comparison? Given that OH is so difficult to measure, the authors should provide more evidence that their quantitation method is sound.

- It was noted in the manuscript that the pH of SOA solution was in the range of of 4.8 – 6.4. In Fenton reaction, Fe2+ reacts with hydrogen peroxide to yield OH radical:

$Fe2+ + H2O2 \rightarrow Fe3+ + OH. + OH-$

Can OH radical or OH ion generation be quenched under acidic condition? e.g.

$2Fe2+ + H2O2 + 2H+ \rightarrow 2Fe3+ + 2\ H2O$

What would be the pH effect for OH production from SOA? Will OH production be underestimated or overestimated in this paper, especially when the particle is inhaled into the lung where pH is around 7? (Richard and Francis, J Clin Invest. 1969)

- The observation that ROOH (not HOOH or ROOR) decomposition leads to OH formation is interesting. Any speculations as to why ROOH preferentially decomposes? Also, the R group seems to be also important, since b-pinene forms more OH than a-pinene (and they are very similar). Also, why was isoprene SOA not studied? Isoprene SOA is known to contain a large amount of ROOH (Surratt et al., 2006) and is likely more important than terpene SOA.

- The addition of Fe2+ leads to elevated OH concentrations, but the concentration of Fe2+ seems high relative to SOA for ambient particles. The mass ratios in aerosols are more likely to be around 0.01 to 0.1 (if we assume 10 ng/m3 of Fe and 1 ug/m3 of SOA). Perhaps in cloud droplets, the ratios would be higher due to higher water content and therefore more soluble iron? I suggest adding some reference of ratio of organic to Fe in cloud droplets to clarify.

- Pg. 30027 line 7-11: The authors should be careful about making the hypothesis that the OH formation revealed in this study is analogous to autoxidation reactions. From the cited studies, autoxidation reactions occur as a result of intramolecular reaction in the gas phase. These are favorable in the gas phase because the lifetimes of radicals are longer. The same type of mechanism is unlikely to happen in the condensed phase since the lifetimes of radicals are much shorter due to extensive quenching by other radicals. On the other hand, propagation of radicals is likely promoted by the higher concentrations. The authors should clarify that the hypothesized mechanisms are not similar.

- The link to oxidative stress in the lung lining fluid is very interesting. What is the formation rate of OH compared to clearance rate by the mucus in the airway?

Minor/Technical comments:

- Pg. 30026 line 20: “illustrated”

- Fig. 4: why is there an abrupt change in OH concentration when Fe2+/SOA reaches a certain ratio for a-pinene and b-pinene, but not for limonene?

- Pg. 30020 line 5: RH for naphthalene experiments is 30%. What about the other experiments?

- Pg. 30020 line 9: the typical size is 50-400 nm. Is that by number? What is the volume mode or median diameter?

- Pg. 30020 line 15: I assume 0-20 ppb of ozone is what was measured after the ozone denuders. A clarification is needed.

- Acknowledgements usually include funding information.