

Interactive comment on “In-cloud processes of methacrolein under simulated conditions – Part 2: Formation of Secondary Organic Aerosol” by I. El Haddad et al.

Anonymous Referee #1

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This manuscript describes the formation of high molecular weight products and secondary organic aerosol from the aqueous OH radical oxidation of methacrolein and water evaporation. This paper provides important evidence supporting the formation of secondary organic aerosol through aqueous reactions in clouds, fogs and aerosol water.

The experimental work described has been carefully conducted and the interpretation of results is well considered. There are a few points that are important to make to prevent mis-use of the results.

1. The statement "This provides, for the first time to our knowledge, strong experimental

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evidence that cloud processes can act as important contributors to SOA formation in the troposphere." is stated too broadly to be true. As stated, Loeffler certainly could be said to be the first. Sorooshian papers from ICARTT and MACE (and Crahan before them) provide atmospheric measurements that could also claim this. Liggio, Volkamer, and Hennigan provide key evidence for these types of processes in aerosol water. The modeling papers by Fu and Carlton could also claim this.

2. This reviewer encourages the authors to examine their mixed standard in the presence of H₂O₂ and separately in the presence of UV to determine whether any quantified products are effected by these during experiments intended to measure reactions with OH.

3. Concentrations of methacrolein and OH: The authors should comment on how these concentrations compare to those likely to be found in cloud water or in aerosol water.

4. Generation of aerosols by atomization and water evaporation: 1) is there a way to ensure that the nebulizer does not induce additional formation of high molecular weight products? 2) It would be nice to know how atomization of the measured mix of quantified products compares to atomization of the reaction vessel solution (i.e., differences in SOA mass/size obtained should indicate how much of the mass was in high molecular weight products. 3) It was very important that NaCl was also atomized to understand just how large the losses are.

5. The authors should make the reader aware that the size distributions obtained depend on how the atomizer is operated. Only the change in the size distribution, not the absolute size distribution is meaningful.

6. How much variation is seen in the size and mass obtained from the atomizer given no difference in the solution? Certainly losses in the atomization process are very large.

7. Can the authors say anything about how much of the SOA formed comes from quan-

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tified species vs high MW products? The authors should make the reader aware that the importance of oligomer products will probably differ with precursor concentration.

8. Everyone wants to present yields when they do this kind of work. But it is not clear that the yields obtained are appropriate to the atmosphere. The yields obtained depend not only on reaction time, but also will be different if the OH radical concentration or organic precursor concentration is different. Yields might also differ with temperature or RH. It is very important that this is made clear. How do the conditions in the reaction vessel compare to the real atmosphere? What should a modeler consider before putting these yields into a regional or global model?

The results do provide experimental evidence that cloud processes of methacrolein can produce significant SOA - (if reactions were conducted at cloud relevant concentrations). If they are produced instead at concentrations relevant to aerosol water, this statement should be altered.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 6425, 2009.