

## ***Interactive comment on “Chemical insights, explicit chemistry and yields of secondary organic aerosol from methylglyoxal and glyoxal” by Y. B. Lim et al.***

**Anonymous Referee #3**

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### General Comments

This paper describes a detailed model of the aqueous-phase oxidation of glyoxal and methylglyoxal. Main improvements over a previous glyoxal oxidation model have to do with simulating laboratory experiment conditions better, namely the attenuation of the photolysis beam by HOOH absorption. In addition, tetraperoxy compound decomposition rates were allowed to vary by an order of magnitude (staying within published limits) in order to better fit the data. However, in order to make the simulations match the data, two additional adjustments were made. First, glyoxylic acid photolysis rates and product ratios were adjusted away from published measurements. Second, the

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absorption of unknown product molecules was added in. There is reasonable justification for at least the latter adjustment. Most of these adjustments were only needed for the highest concentration runs, in any case. The model, once validated in this way, was used to calculate overall SOA yields for glyoxal and methylglyoxal oxidation in the aqueous phase. A variety of concentrations were used, in order to produce data relevant to both atmospheric clouds and aqueous aerosol particles. The model predicts mass-based aerosol yields near 100% for the aqueous phase oxidation of glyoxal and methylglyoxal, despite the potential for evaporation of some of the semivolatile products. However, the product gas/particle partitioning values used in the model come from atmospheric measurements, so all is believable. This paper represents an advance in our understanding of SOA formation in the aqueous phase, especially from methylglyoxal.

### Specific Comments

To avoid confusion the authors should restate the sentence in the abstract: “Oligomerization of unreacted aldehydes during droplet evaporation could enhance yields,” The statement is true inasmuch as these accretion reactions are another competing pathway to SOA formation, and could increase SOA mass; however, as they do not depend on OH oxidation, their SOA-forming contribution cannot be numerically added to the measured yields of this study in a meaningful way.

p. 8 line 16: The paragraph should make clear that this is how aqueous SOA can form in the model. There are certainly other possible mechanisms in the atmosphere.

p. 8 last sentence: Can the authors give some examples of atmospheric conditions / precursors present when a given assumption might be appropriate? Readers will not know what to make of this statement otherwise.

p. 11 line 27: Could the authors list specifically which runs of the model allowed radical – radical reactions to occur, and which runs had them turned off? If the radical – radical reactions are turned off, what is the main sink for radicals in the model?

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p. 15 line 22: The work of Barbara Noziere et al. on aldol condensation reactions forming brown carbon in aerosol particles could be appropriately cited here.

p. 16 line 8: An explanation / justification is needed for the assumption that all dissolved, water-soluble organics in an aerosol particle will have the same ability to form oligomers as glyoxal. As mentioned on p. 11, the authors showed in a recent publication that acetic acid, for example, cannot form oligomers under these conditions.

Fig 1: Is an ROO + NO pathway relevant in the aqueous phase?

Fig 8: For comparison, this figure should show a model result when using literature values for pyruvic acid photolysis. Since this is the most questionable adjustment made to the model, readers should be able to see how important its effect is in achieving decent fits to experimental data.

#### Technical Corrections

p. 15 last line: The initial phrase of this sentence is confusing. Do the authors mean "For the batch reactor runs ..."?

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 4687, 2013.