

We thank the reviewers for careful reading and helpful comments that improve the quality of our manuscript. Reviewer comments have been copied followed by our responses in bold.

Anonymous Referee #2)

General Comments

This work by Lim et al. describes a model for SOA formation from methyl glyoxal through multiphase processing. The authors developed explicit aqueous-phase OH oxidation mechanisms for acetic acid and methyl glyoxal. They validate these mechanisms against laboratory oxidation experiments, including a correction factor for H₂O₂ absorption of light. The mechanisms are then incorporated in their explicit glyoxal SOA model, and use it to simulate SOA formation under cloud water and aerosol water conditions. Since the mechanisms are explicit, they are able to determine product yield and which products dominate under which concentration conditions. They find that at low precursor concentrations, organic acids, specifically oxalic and pyruvic acid, dominate the products. At higher precursor concentrations, radical-initiated oligomer products dominate. This work presents a nice increase in our molecular-level understanding of aqueous-phase SOA formation. I think that this paper is suitable for publication in ACP after addressing the minor comments below.

Specific Comments

R2C1) Page 7, line 12: Is there a reason the authors allowed the path length to vary while keeping the literature extinction coefficient, rather than keeping the known path length and allowing the extinction coefficient (which might have larger relative error bars than the path length) to vary?

Response: We used a fixed path length that is not very different from the physical measurement of the mean path length for fluid in the reaction vessel. The value we used enables us to use literature values of extinction coefficients. We must include extinction coefficient values for all (potential) light absorbing compounds (e.g., methylglyoxal, pyruvic acid, oligomers) in our model. The approach we used requires only one (reasonable) fitted parameter. For this reason, we take the approach of using the literature values of light absorbing compounds and a constant but fitted path length.

R2C2) Page 8, line 30: Can the authors give examples of precursors and conditions that are relevant for batch reactions vs. CSTR reactions?

Response: Yes. Thank you to both reviewers that suggested this. This suggestion has improved the paper. Please see our response to R3C3.

R2C3) Page 10, line 8: Is there a reason CSTR reactions do not go above 10⁻⁴ M initial glyoxal or methyl glyoxal concentration?

Response: There are technical challenges associated with conducting the CSTR runs at higher concentrations and the range of concentrations modeled provided enough comparison points with the batch assumptions to be confident in the conclusions.

R2C4) Page 15, line 16: Even if the authors did not identify the compounds responsible for light absorption, did they observe a color change in their solutions?

Response: There was no visible color change. There was also no visible color from hydrogen peroxide, but it still clearly absorbs light.

R2C5) Page 18, line 8: Can the authors quantify “substantial”, especially as from glyoxal, from methyl glyoxal, and from other compounds?

Response: We add the following at the end of the line:

(Although uncertainties are large, recent modeling studies (Carton et al., 2008; Fu et al., 2008; Liu et al., 2012) suggest that the magnitude of aqSOA is comparable to SOA formed via gas-particle partitioning of semivolatile organic products from gas-phase oxidation of VOCs)

R2C6) Can the authors comment on how the product chemical composition and distribution might change if chemistry with inorganics is allowed, as is likely in real aerosols with often contain quantities of ammonium sulfate and/or ammonium nitrate?

Response: At cloud conditions, glyoxal (or methylglyoxal) + OH is the major reaction. Our previous (and in press) photooxidation experiments and rate calculations suggest that ammonium, sulfate and nitrate do not affect glyoxal + OH reactions, and do not form sulfur or nitrogen containing organics at cloud-relevant concentrations. However, during droplet evaporation and in wet aerosols, others have shown that organics can react with ammonium, sulfate as well as OH radicals. It is not clear whether inorganic-organic compounds formed via non-radical reactions interfere with or enhance glyoxal (or methylglyoxal) + OH reactions.

We have made this more clear in the text. Please see our response to R4C2.

Technical Corrections

R2C7) Page 3, line 18: b in “based” should not be capitalized.

Response: We correct this.

R2C8) Page 4, line 9: Perhaps use “unit mass resolution electrospray ionization. . .”

Response: We accept this and have made this change.

R2C9) Page 4, line 14: Reference for the triiodide method?

Response: We now reference Banerjee et al., Anal. Chem., 1964.

R2C10) Page 6, line 21: Do the authors mean “Specifically” rather than “Specially”?

Response: We changed this.

R2C11) Page 11, line 11: “. . . faster than abstraction. . .” rather than “. . . faster than Abstraction. . .”

Response: We corrected this.

R2C12) Page 11, line 25: I believe a tertiary carbon means that it is covalently bonded to three other carbon atoms (which do not exist in either glyoxal or methyl glyoxal). Perhaps the authors could change the wording to “triply substituted carbon” or something similar to indicate they mean carbons bound to three non-H atoms?

Response: We now change “tertiary carbon” to “triply substituted carbon (i.e., carbon bound to three non-H atoms).”

R2C13) Page 13, line 9: “. . . to our knowledge. . .” rather than “. . . in our knowledge. . .”

Response: We corrected this.

R2C14) Page 15, line 2: “. . . slower rate than the literature value. . .” rather than “. . . slower rate than the literature value. . .”

Response: We correct this.

R2C15) Page 15, line 23: “Further work is needed to investigate this hypothesis.” rather than the current concluding sentence.

Response: We correct this.

R2C16) Page 17, line 26: OH radicals rather than OH radical

Response: We correct this.