

Comment on acp-2022-139

Anonymous Referee #3

Referee comment on "Elucidating the critical oligomeric steps in secondary organic aerosol and brown carbon formation" by Yuemeng Ji et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-139-RC3>, 2022

This work systematically investigated the aqueous-phase reaction mechanism and kinetics of glyoxal (GL) using quantum chemical and kinetic rate calculations. The critical oligomeric steps in secondary organic aerosol (SOA) and brown carbon (BrC) formation were elucidated. The authors also revealed that the loss rate of aqueous-phase reaction of GL is higher than that of photolysis and photo-oxidation. This paper is well organized and clearly written. And the outcomes are very helpful to understand the role of the aqueous-phase chemistry of GL in SOA and BrC formation. I recommend publication after the minor comments are addressed.

Detailed comments:

- Line 28: "produce (SOA) particle mass" is a vague expression. It should be replaced with "produce secondary organic aerosol (SOA) particles".
- Line 60: "have" should be "has".
- Line 87: MG in "MG+MA/AM reaction systems was not mentioned before. Please clarify.
- Methods: How does the authors deal with the influence of liquid-phase environment? It should be included in the Methods.
- Line 122: The term of "the natural charge population analysis" should be replaced with "the natural population analysis"?
- Line 147: The results show that "the k values ...are $4.14 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$ and their half-lives ($t_{1/2}$) are lower than $\sim 10^{-4} \text{ s}$ ". How are the half-lives calculated or has the author considered the real atmospheric concentration of hydrogen ion in aerosol?
- Line 156: The authors states "the $\text{C}\alpha\text{C}\beta\text{O}(\text{H})$ bond of 1st-CB1 after protonation is elongated by 0.05 \AA ". Please clarify the reference, that is, what is the $\text{C}\alpha\text{C}\beta\text{O}(\text{H})$ bond longer than?
- Line 188-189: "Current results reveal that cyclic oligomers are difficult to be formed from the CBs with the positive charge center close to O(H) atom". The authors should point out which CBs in Figure 1b are difficult to form cyclic oligomers.
- According to the data in Figure 4a, the corresponding ΔG_r value in line 217 is -0 kcal mol^{-1} .

- Line 237: " G_r " should be " ΔG_r ".
- Line 263: "ammonia salts" should be "ammonium salts".
- Line 315: "conditions" should be "condition".