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Reply on RC1

Yuemeng Ji et al.

Author 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-AC1, 2022

Comments: This theoretical work conducted a comprehensive examination on the essential aqueous-phase oligomerization pathways of glyoxal without and with amine/ammonia and reveals their rate-limiting steps during isomeric processes. The detailed thermodynamic and kinetic characterization of the heterogenous reactions of glyoxal in this study provided valuable insights into the formation mechanisms formation of secondary organic aerosol (SOA) and brown carbon (BrC) starting from small alpha-dicarbonyls in the atmosphere. The evidences are sufficient and the mechanisms are elucidated well. There are minor issues with the manuscript:

Response: Thank you for the review and feedback. We have made careful revisions on the original manuscript according to your kind and helpful comments. The changed sentences have been marked as red color in the revised manuscript.

Question 1) Lines 277-278: A little confuse about the statement here: If γ_{GL} used was only suitable for urban condition, estimating other atmospheric conditions should be unreasonable, right? Also, does it mean that there are no available γ_{GL} values for remote and rural conditions from literatures like Liggio (2005)'s study? BTW, "are not" should be "is not".

Response: We are sorry that our expression led to the reviewer's confusion. The γGL used in our study is more suitable for the urban condition than other conditions, because it was determined at very low relative humidity which is closer to urban condition relative to other conditions (J. Geophys. Res., 2005, 110(D10): D10304). Hence, it suggests that the rate estimated under urban condition is more suitable than those under remote and rural conditions. In addition, there are no available γGL values, which are explicitly obtained under remote and rural conditions in the previous studies, like Liggio et al. (J. Geophys. Res., 2005, 110(D10): D10304). Hence, for comparative analysis, the same γGL value is used to estimate the rates under three different conditions. According to the reviewer's helpful comment, to make the expression clearer, the corresponding sentences were modified as following: "**The krate value under urban condition almost agrees with that of the experimental data and is slightly larger than those of the experimental data under other conditions (Liggio et al., 2005a). The lower values under remote and rural conditions are explained by the γGL used here, which is more suitable for the urban condition (Liggio et al., 2005a).**" (Please see lines

283-285)

Question 2) Lines 294-303: Did the authors identify these primary reaction pathways based on their branching ratios?

Response: These primary reaction pathways were determined not only based on branching ratios but also the reaction energies, activation energies, and rate constants.

Question 3) Line 320: please also estimate the mean tau or its ranges for gas-phase processes.

Response: According to the reviewer's suggestion, we added the mean τ in the revised manuscript, which is determined by Liggio et al. (J. Geophys. Res., 2005, 110(D10): D10304).

"**Especially, the τ value under urban condition is significantly shorter than the total gas-phase loss (125 min) (Liggio et al., 2005a)**." (Please see lines 326-327)

Question 4) Line 28: The full name of SOA is missed.

Response: Thanks to the reviewer's kind reminder. The full name of SOA was supplemented on line 28 of the text.

Question 5) Line 30: "particle" -> "particles"

Response: They were modified as suggested.

Question 6) Line 34: "particle-phase and aqueous-phase" -> "particle- and aqueous-phase"

Response: They were modified as suggested. (Please see lines 31 and 34-35)

Question 7) Figure 5: Please describe each circle ring in the caption.

Response: According to the reviewer's suggestion, the following sentences were added to describe each circle ring in the caption of Figure 5 as "**The inside circle ring represents the ion-mediated initial reaction of GL to yield DL, TL and 1st-CBs; The middle circle ring corresponds to the formation of RODs and 2nd-CBs; The outer circle ring denotes the formation and propagation of ROTs from the association reactions of 2nd-CBs with DL/TL.**" (Please see lines 559-562)