Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2022-139

Deming Xia (Referee)

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-RC2, 2022

α-dicarbonyls play important roles in the secondary organic aerosol (SOA) and brown carbon (BrC) formation in the atmosphere, but their sources remain unclear. Ji et al. selected GL as a model compound, and systematically investigated its aqueous-phase oligomerization mechanisms, kinetics, and contributions to SOA and BrC formations using quantum chemical calculations and some modeling methods. The simulation results were supported by previous experiments and can provide useful data for assessment on the role of α-dicarbonyls in SOA and BrC formations. The manuscript is well written and can be easily understood. The following minor aspects may be useful to further improve its quality:

- LINE 94 and 97: Why is the 6-311+G(3df,3pd) basis set more "*flexible*" than the /6-311G(d,p) basis set?
- LINE 97: What is the "M06-2X//M06-2X" level? Is it means that M06-2X/6-311+G(3df,3pd)//M06-2X/6-311G(d) level?
- LINES 98 and 123: Please add citation of the methods CCSD(T) and NBO.
- LINES 153 and 200: The sub-title "3.2" is the same as "3.3". I guess that the "3.3" should be written as: Oligomerization mechanisms *with* methylamine/ammonia.
- LINES 265 ~ 293: Some contents here belong to "Methods" rather than "Results and Discussion".
- LINES 278 ~ 279: The sentence is hard to understand.
- LINES 315 ~ 316: It is relatively arbitrary to judge the importance of heterogenous loss of GL in urban regions only based on the simulated lifetime. Urban regions may have more competing loss pathways of GL, compared with other regions. Therefore, more discussion should be supplemented to support the view "a more important role of heterogeneous reaction of GL in urban air quality compared with other conditions".
- TABLE 1: The k_{rate}(total) and *GR* in rural regions calculated by the authors are higher than the experimental ones by one order of magnitude (1.85×10^{-3} vs 1.90×10^{-4} and 0.57 vs 0.032). Please add more discussion.
- Figure 5: Please explain the â , â ı, and â ¢ in the caption.
- Variables are recommended to be expressed in *italics*.