

Review of “Molecular distribution and stable carbon isotopic compositions of dicarboxylic acids and related SOA from biogenic sources in the summertime atmosphere of Mt. Tai in the North China Plain” by Meng et al.

This manuscript reported the concentrations and isotopic compositions of dicarboxylic acids in the summertime of Mt. Tai. The bulk of the results presented focuses on the following aspects: (1) comparing the concentrations of species and the ratios of different dicarboxylic acids between day and night; (2) correlating the species concentrations with other parameters (T, O₃, RH, pH, etc); (3) the $\delta^{13}\text{C}$ of the dicarboxylic acids. The authors concluded that the dicarboxylic acids in Mt. Tai, a forested area with high elevation, mainly originate from local photochemical oxidation of biogenic emissions. Given the surrounding in Mt. Tai and sampling time, this conclusion is not surprising. Overall, the data analysis is routine and the conclusion is generally supported by experimental evidence. I recommend publication after major revisions.

Major Comments

1. My major concern is the lack of innovation of this study. As noted in the abstract, a few studies have performed similar measurements and analysis in the same site. The novelty of this manuscript as claimed by the authors is that the measurements were performed in summertime for the first time. It is not well justified that why it is important to conduct the measurements in summer. Also, considering the surroundings of measurement site and sampling time (i.e., summer), isn't the conclusion largely expected? It is not clear what are the scientific questions that authors aimed to address.
2. Another major comment is regarding the comparison between daytime and nighttime. Due to the high elevation, the sampling site is in the residual layer at night. At night, the residual layer is isolated from surface emissions. The dicarboxylic acids and other species at night are mainly carried over from late afternoon formation. Also, the late afternoon mixing ratios of BVOCs, NO_x and O₃ govern overnight chemistry within the residual layer. Please refer to Edwards et al. (2017). Assuming no further oxidation at night, the species concentrations are expected to be similar to that in the late afternoon. Thus, the day (8am-8pm) vs night (8pm-night day 8am) comparison may

merely reflect the difference in species concentration between noon and 6pm (when sampling site is above the boundary layer).

This study is an excellent opportunity to study the residual layer chemistry. However, in current most of discussions (Line 184, Line 223, Line 298, etc), the format is like “something is higher in daytime than nighttime, suggesting stronger photochemistry or stronger biogenic activity in the day”. The authors should clearly state the physical model behind the day vs. night comparison and better explain the observations (including the NH_4NO_3 concentration in Line 184). The authors are encouraged to look in depth into the day vs. night comparison.

3. Line 193-194. Why is LWC higher in daytime than nighttime?

4. Line 243-248. The discussions on the higher concentration of dicarbonyls at night is confusing. Firstly, dicarbonyls are produced from photooxidation of isoprene and other VOCs, not aqueous phase reaction. Secondly, how does the “opposite pattern” suggest “aerosol aqueous phase oxidation”? Thirdly, I believe “impressed” is a typo. In light of my previous comments, the authors should explore more reasons in terms of this day vs night comparison.

5. Line 268-269. Please provide evidence that C2/C4 and C3/C4 can be regarded as indicators of photochemical aging. Also, the measured C2/C3 and C2/C4 ratios are actually very close to that from vehicle exhausts. This is contradictory to the conclusion that dicarboxylic acids are mainly from biogenic emissions.

6. Line 279-282. This is one of many cases that the authors need to better explain the link between evidence and conclusion. Why would the “correlation between C2/C4 and temperature at night” suggest “local photochemical oxidation”?

7. Line 360-362. The role of nighttime chemistry is not justified at all. The lack of correlation between BSOA tracers and O_3 at night could be attributed to that most of BSOA tracers at night are carried over from later afternoon product and the O_3 at night may be quickly titrated by residual NO. Again, this comment falls in the scope of day vs. night comparison.

Minor Comments

1. Line 61. “water” is uncountable noun. Replace “waters” with “water”.

2. Line 132. What type is the GC column?

3. Line 304. The change in topic is too quick. Rephrase the sentence to improve the transition.

4. Line 360. As mentioned above in the text, it is not only from O₃ oxidation, also OH oxidation.
5. Line 377. Replace “linger” with “linear”.

Reference

Edwards, P. M., Aikin, K. C., Dube, W. P., Fry, J. L., Gilman, J. B., de Gouw, J. A., Graus, M. G., Hanisco, T. F., Holloway, J., Hübler, G., Kaiser, J., Keutsch, F. N., Lerner, B. M., Neuman, J. A., Parrish, D. D., Peischl, J., Pollack, I. B., Ravishankara, A. R., Roberts, J. M., Ryerson, T. B., Trainer, M., Veres, P. R., Wolfe, G. M., Warneke, C., and Brown, S. S.: Transition from high- to low-NO_x control of night-time oxidation in the southeastern US, *Nat Geosci*, 10, 490, 10.1038/ngeo2976, 2017.