

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2021-3

Anonymous Referee #1

Referee comment on "Contrasting effects of secondary organic aerosol formations on organic aerosol hygroscopicity" by Ye Kuang et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-3-RC2>, 2021

The manuscript of Kuang et al.: "Contrasting effects of secondary organic aerosol formations on organic aerosol hygroscopicity" shows that the oxidation state of the SOA does not always correlate to their degree of the hygroscopicity and they propose that the hygroscopicity is controlled by additional factors along with the oxygenation degree of the OA. To show that, the authors use the data set of a field campaign conducted in China (Pearls River Delta) using a suite of online and high-resolution instrumentation. This paper is well written and I recommend publication after the authors address comments below.

Major:

Lines 166-171: The authors state that they did not perform a calibration at the sampling state. They used an RIE=4 for ammonium taken from the last calibration. This could be a serious issue for ammonium concentration resulting in artifacts (lower or higher ammonium concentration). How well the ammonium concentration correlated with filter measurements (mentioned on line 178) using a RIE=4? Please provide the R2. Also please explain how stable is this parameter for the specific instrument. How much does the ammonium concentration changes if you use a RIE of 3.0, 3.5 and 4.5? Please provide the corresponding NH₄ mass concentrations and its % contribution to the PM₁ for the above 3 cases.

Lines 235-243, Table 1 and equation 2: How it is possible to have (NH₄)HSO₄ and

(NH₄)₂SO₄ and NH₄NO₃ at the same time? Ammonia will first neutralize all available sulfate and it will bring the aerosol in the form of (NH₄)₂SO₄. Then, whatever ammonia exists in the atmosphere will react with HNO₃ to form NH₄NO₃. The co-existence of (NH₄)HSO₄ and NH₄NO₃ is not compatible according to thermodynamic laws. (Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Seinfeld and Pandis, Wiley, 3rd edition, 2016). The mass balance should be recalculated. After that, please discuss the potential compounds present in the atmosphere during the campaign.

(NH₄)HSO₄ should be deleted from equation (2) and (3) and the calculations should be done again. How much the results (kOA) do change with this correction?

Lines 293-294: Is this NH₄NO₃ formation during the night due to the lower temperature during the night? The dissociation constant K_p(T) of the ammonia-nitric acid system (NH₃(g) + HNO₃(g) ⇌ NH₄NO₃(s)) is a function of the temperature and it is very sensitive to the temperature changes. As the temperature decreases K_p(T) also decreases, and the equilibrium shifts towards the aerosol phase increasing the NH₄NO₃ mass concentration (Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Seinfeld and Pandis, Wiley, 3rd edition, 2016). It seems that this is the case taking account Figures 2a and 2c. Please discuss.

Lines 307-310: This explanation is not sufficient stated. If this is the case (i.e., N₂O₅ production during the night, then NO₂ should be reduced, but Figure 2f shows that NO₂ increases during the night. Please make sure that you explain the phenomena correctly.

Line 310: "Nitrate concentrations increased quickly since 16:00:". This is contradictory to Figure 2c, where nitrate decreases from 09:00 to 16:00 and then starts increasing, reaching its maximum at 03:00.

Line 312: "nighttime heterogenous formation of nitrate, " please check if this is the case.

Line 337: "On average, $\delta^{15}\text{N}$ increased slowly during the nighttime". From Figure 3c it seems that kOA is rather stable during the night and it increases after 03:00 (a.m.).

Lines 357-359: "However, $\delta^{15}\text{N}$ was also negatively correlated with LOOA 357 (Fig.4d), whose mass concentration increase rapidly after sunrise and are likely secondary due to local photochemistry with potential precursors such as isoprene and anthropogenic VOCs." How do you support this? Could you discuss any results from the gas-phase?

Minor:

Please use past tense throughout the whole manuscript. There are parts that the present tense alternates with past tense (e.g., section 2.2).

Line 62: Please replace "evolvment" with "evolution".

Lines 69-71: Please add here that the volatility could be another factor that affects the hygroscopicity of the SOA.

Lines 71-72: Please cite here the following 4 papers:

- Kuwata, M., Kondo, Y., Mochida, M., Takegawa, N., and Kawamura, K.: Dependence of CCN activity of less volatile particles on the amount of coating observed in Tokyo, *J. Geophys. Res.*, 112, D11207, doi:10.1029/2006JD007758, 2007.
- Asa-Awuku, A., Engelhart, G. J., Lee, B. H., Pandis, S. N., and Nenes, A.: Relating CCN activity, volatility, and droplet growth kinetics β -caryophyllene secondary organic aerosol, *Atmos. Chem. Phys.*, 9, 795–812, 2009.
- Frosch, M., Bilde, M., Nenes, A., Praplan, A. P., Jurányi, Z., Dommen, J., Gysel, M., Weingartner, E., and Baltensperger, U.: CCN activity and volatility of β -caryophyllene secondary organic aerosol, *Atmos. Chem. Phys.*, 13, 2283–2297, 2013.
- Kostenidou, E., Karnezi, E., Hite Jr., J. R., Bougiatioti, A., Cerully, K., Xu, L., Ng, N. L., Nenes, A., and Pandis, S. N.: Organic aerosol in the summertime southeastern United States: components and their link to volatility distribution, oxidation state and hygroscopicity, *Atmos. Chem. Phys.*, 18, 5799–5819, 2018.

Lines 106-110: Please rewrite this part. It should be not mentioned any sections, but rather describe and explain what it will follow in the next.

Line 116: Please replace "locates" with "was located".

Line 117: Please add "the" before "megacity".

Line 130: Please add "the" before "physical".

Line 132: Please add "the" before "aerosol".

Line 134: Please replace "of" with "the".

Line 135: Please add "the" before "aerosol".

Lines 137-139: Please rephrase this sentence.

Line 139: Please replace "Two" with "Two sensors".

Line 146: Please add "a" before "flow".

Line 146: Please add "the" before "sampling".

Line 149: Please add "the" before "particle".

Line 152: Please add "the" before "size-resolved".

Line 153: Please delete "basically".

Lines 161-162: Please rephrase this sentence.

Line 183: "As a wildly used source analysis method" I am not sure what do you mean here.

Lines 186-188: Please rephrase this sentence.

Lines 190-199: This is not the right place for this paragraph. It should be moved to the Results section.

Lines 206-207: Please rephrase this sentence.

Lines 223-226: Please rephrase this sentence.

Line 250: Please define better the parameters κ and ϵ .

Lines 255-256: Please rephrase this sentence.

Line 352: Please provide the corresponding literature.

Line 356: "It was generally thought that secondary aerosol formation would result...", This phrase is not well connected to the previous sentences.

Line 359: What do you mean by average O/C"? Each factor derived from PMF analysis has a constant O/C ratio.

Lines 360-363: This sentence is quite big and complicate. Please rephrase and simplify.

Line 394: Please replace "?" with ".".