

## Comment on acp-2021-626

Anonymous Referee #3

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Referee comment on "Measurement report: Molecular characteristics of cloud water in southern China and insights into aqueous-phase processes from Fourier transform ion cyclotron resonance mass spectrometry" by Wei Sun et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-626-RC3>, 2021

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This work provides a comprehensive analysis of the compounds in cloud water as well as the interstitial PM2.5, and characterize the distribution of different groups of species by using FT-ICR-MS; daytime and nighttime comparison was also made. Based on such analysis, aqueous-phase processing and the reactions involved were inferred to enhance our understanding of the aerosol chemistry. The paper is fairly well written and provide useful information and knowledge regarding the cloud water organics, this reviewer however has a series of comments to be addressed first before its acceptance

- Line 51: as you stated here, "chromatographic and spectroscopic techniques only determined ~20% of all kinds of organics", then what does the FT-ICR-MS perform? Even though it has a super high mass resolution, is it being able to determine all existing species? If not, organics with what functionalities are preferred to be detected? What is the fraction of determined species to the total? How does this bias affect your interpretation? I think this issue should be clarified in your manuscript.
- Section 2.2: Even though instrumental details are included in the supplement, I think some key information, for example, the mass resolution, and how to remove background organics, etc can be briefly described here, as well as the IC and TOC/TN analysis.
- As described in (2), background organics or impurities during sample storage and treatment, might be detected as FT-ICR-MS is highly sensitive and has ultrahigh resolution. The number of molecules in cloud samples seem to be much higher than those in PM2.5 samples, I am wondering how do these excess compounds come from except from possible aqueous-phase processing?
- Line 170-179: This reviewer thinks that cloud cycling might need to be considered, as the interstitial PM2.5 sampled here may contain aqueous oxidation products inside cloud droplets as cloud droplets in reality cycle per few minutes. Therefore a high O/C value might be observed in PM2.5 samples rather than cloud water. Whether or not aqueous processing could enhance the oxidation degree of organics depends on the ageing time. In a short time scale, the organic oxidation degree could increase and the more oxidized species may fragment into low oxygenated ones given enough time.
- Line 286-292: Similar for OS, typically OS can be produced more efficiently in aerosol

water rather than liquid water, yet no statistical difference are observed here, as there are repeated cycling between cloud water and interstitial PM2.5.

#### Specific comments

- Line 25-26, do you mean CHON and CHO-containing species? It is not clear. The last sentence in Line 26 is not a full sentence.
- Line 28: A recent paper by Wang et al (2021, 118:e2022179118) demonstrates that aqueous-phase oxidation of aromatic species could be a source of SOA, this might be a supporting evidence that "CHON with aromatic structures are abundant in cloud water"
- Line 69: Consider to add citation<sup>1/4</sup> Ye et al., Atmos Environ 2020;223:117240, which determines the organic acids produced from aqueous-phase oxidation of a certain precursor.
- Line 144: RA means relative abundance, this reviewer somehow think the authors can directly use "relative abundance", it is easier to understand than RA<sup>1/4</sup>