

Atmos. Chem. Phys. Discuss., referee comment RC2
<https://doi.org/10.5194/acp-2021-626-RC2>, 2021
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Comment on acp-2021-626

Anonymous Referee #2

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-RC2>, 2021

General Comments:

The manuscript by Sun et al. presents the mass spectral characteristic of cloud water samples throughout a long-lasting cloud event by FT-ICR-MS, and attempts to shed light on the potential influences of in-cloud aqueous phase reactions, which are currently uncertain for the formation of SOA. They show that CHON with aromatic structures are the most abundant type in cloud water, suggesting their enhanced formation in cloud. Their results also indicate distinctly differences between day and night, which is most probably attributed to diurnal differences in aqueous chemistry.

Such observation could provide valuable cloud chemistry data for the community, and has the potential to be published after considering my comments. The major weakness is the limited dataset, thus the authors have to clearly indicate in the discussion of the diurnal difference of cloud chemistry between day and night, since there could be other factors contributing to such difference.

Specific Comments:

-Introduction: Overall it is OK, but it would be better to include the aqueous formation mechanisms related to CHON and CHOS.

- Lines 172, "the current understanding that aqueous-phase reactions generally increase the degree of oxidation (Ervens et al., 2011)." Please also include the reasons to this understanding. Does such aqueous reactions refer to in cloud processing?

- Lines 182, "The O/C ratios and OSC of CHO collected during the daytime is slightly lower than the nighttime...". What about the influence of primary emission? Since the samples collected during the daytime and nighttime may originally presents different characteristics without oxidation.

- Lines 195, is there any result of aromaticity related to traffic emission or other sources, in addition to coal combustion and biomass burning? Since the present OA molecular does not correspond to these sources, i.e., coal combustion and biomass burning as discussed.

- Lines 251, It is an interesting result that coal combustion contributes to S-containing formulas in cloud water more significantly compared with CHO and CHON. Is there any other evidence to support the demonstration, such as the correlation between CHOS with the concentration of so₂ or sulfate?

- Lines 278, "For CHO, the most abundant C₁₇H₂₆O₄ in cloud water is not detected in the PM_{2.5} samples, suggesting a formation by the in cloud aqueous-phase reactions, although the contribution from BVOCs cannot be ruled out." Reasons should be discussed for such a contradiction.