

Atmos. Chem. Phys. Discuss., referee comment RC2  
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## Comment on acp-2022-313

Anonymous Referee #2

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Referee comment on "Long-term monitoring of cloud water chemistry at Whiteface Mountain: the emergence of a new chemical regime" by Christopher E. Lawrence et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-313-RC2>, 2022

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The work by Lawrence et al. builds on a dataset of cloud water information for White Face Mountain in the northeastern U.S. by extending it to more recent years and adding trends in organic species. The trends in the different cloud water components (sulfate, nitrate, total organic carbon) are discussed and differences explored. The authors are careful to consider criteria for including samples in the analysis which is shown to significantly affect the fraction of samples considered "valid" in more recent years. This paper will likely be a great community resource of data and has highlighted the growing importance of organics in cloud water conditions.

I share the first reviewers concerns about the use of cations in the analysis and whether they were in ionic forms or not. Cations are not only from dust but are present in trace amounts from many sources as shown in the work of Reff et al. (2009) et al.

Can more information be provided on the trends in cloud water itself? Based on the criteria on Page 5 for cloud collection, has the fraction of the year when samples are collected changed over time? This could help link changes in cloud water composition to changes in general atmospheric state. How often are cloud samples being dumped due to bottles being too full? Given the role of valid/invalid samples in the analysis, some commentary on the actual sample coverage would be useful.

#### Minor comments:

- The code availability is excellent. Consider creating a persistent identifier (doi) for the github code as well. Several free services are available.
- Is there any information on the seasonality and likely WSOC parent hydrocarbons important for the unmeasured anions? Are they likely biogenic or anthropogenic?
- Figure 5: Could measurement uncertainty be propagated to the cation/anion ratio?
- Is cloud water S always in the form of inorganic sulfate ( $\text{SO}_4^{2-}$ )? Are hydroxymethanesulfonate or isoprene organosulfates ever included in the cloud water sulfate concentrations?
- What is the likely source of WSOC and insoluble OC in this data set? Are they likely changing over time. Figure 3 plots with both WSOC and TOC trends are very helpful.
- Could some bounding analysis be performed on the amount of organic acids needed to reconcile the data and how that fits with the measured WSOC and likely abundance of organic acids?

#### Reference:

Reff, A., Bhawe, P. V., Simon, H., Pace, T. G., Pouliot, G. A., Mobley, J. D., and Houyoux, M.: Emissions Inventory of PM<sub>2.5</sub> Trace Elements across the United States, *Environ. Sci. Technol.*, 43, 5790-5796, 10.1021/es802930x, 2009.