Reviewers Comments:

Reviewer #1 (Remarks to the Author):

The authors present a comprehensive seven-year observational study of low molecular weight organic acids (LMWOAs) in cloud water at Whiteface Mountain, providing valuable insights into their seasonal patterns, sources, contributions to ion balance and cloud droplet acidity. They found formate and acetate were the dominant monocarboxylic acids, showing seasonal variations driven by biogenic emissions. In contrast, oxalate concentrations were elevated in smoke-impacted clouds. A growing proportion of samples exhibited a surplus of ammonium over sulfate and nitrate—a trend previously linked to unmeasured organic acids. This hypothesis is supported here by the observed positive correlation between surplus ammonium and LMWOA concentrations. Furthermore, the correlation between oxalate and DOC, which strengthened under higher ozone levels, points to an in-cloud secondary production pathway for oxalate that is enhanced under high oxidant conditions. The work is well-executed, the data are robust, and the conclusions are largely supported by the findings. The topic is timely and addresses a critical gap in our understanding of evolving cloud water chemistry in the high-wildfire-activity environment. However, several points could be strengthened to enhance the clarity, impact, and broader implications of the study. Specific comments are listed below.

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Line 310-315 and Figure 6: The positive correlation between surplus NH_4^+ and LMWOA sum in samples with surplus NH_4^+ is a key finding. The authors note that for DOC-rich samples in 2023-2024, the relationship is less clear, possibly due to reactions forming unmeasured nitrogen-containing organics. This is a reasonable hypothesis. Could the authors provide a brief quantitative estimate or citation regarding the potential impact of such sink reactions? For instance, referencing approximate reaction rate constants or yields from previous studies would strengthen this discussion.

Section 2.4.4 “contribution of LMWOAs to cloud droplet acidity”: The analysis of MAF and MAF* clearly demonstrates the growing importance of unmeasured anions. The authors state that including LMWOAs explains 5-40% of the missing acidity annually. To make this range more interpretable, could the authors add a brief discussion linking the yearly variability in this percentage to specific environmental conditions? For example, is the lower explanatory power (5%) associated with years of high wildfire smoke influence (like 2021, 2023), where higher molecular weight organics might dominate, as suggested later in the smoke section?

Line 365-370 and Section 2.4.5 (Smoke Influence): The comparison between "Aged" and "Fresh" smoke events is insightful. The hypothesis that aging leads to oligomerization, increasing DOC while decreasing LMWOAs, is plausible. To further support this, could the authors comment on the potential changes in the average oxidation state (e.g., O:C ratio) or double bond equivalent implied by this transformation, even if not directly measured? Referencing previous work (e.g., Cook et al., 2017, cited in the manuscript) that observed such trends in smoke could strengthen the argument.

Line 455-460 and the Conclusion: The study rightly concludes that a substantial fraction of cloud droplet acidity and anions remains unaccounted for, pointing to unmeasured organic species. This has significant implications for atmospheric modeling and chemistry. To elevate the impact, the authors should more explicitly state the priority classes of compounds that future research should target based on their findings. For instance, should the focus be on di-/tricarboxylic acids beyond oxalate, organosulfates/nitrates, or humic-like substances (HULIS)? A specific recommendation would guide future analytical efforts.

Line 85-110 (Site Description) and Generalizability: Whiteface Mountain is an excellent remote, forested site. The authors briefly note its relevance for studying long-

range transport. In the conclusion or discussion, a short statement on the expected similarities or differences in LMWOA dynamics at other key site types (e.g., coastal, arid, or heavily polluted regions) would help readers assess the broader applicability of the trends observed here. This is particularly relevant given the global increase in wildfire smoke and changing emissions.

Minor Comment:

Line 150-155 (Equations 3-4): The assumption that other cations are negligible is standard but worth a quick justification. A reference to typical cation concentrations in remote cloud water (or a citation to the site's prior work like Lawrence et al., 2023) would suffice.