Response to Reviewer Comments for "Urban aerosol chemistry at a land-water transition site during summer – Part 2: Aerosol pH and liquid water content" by Michael A. Battaglia Jr. et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-368-RC1, 2021"

We thank the Referee for their additional comments. We have addressed each comment with the Referee comments in bold and our reply in plain text immediately below.

In the revised manuscript, the authors provided additional details and explanation to the discussion, which are generally acceptable. However, I still have some concerns about their data presentation and interpretation.

The QA/QC information the authors provided is not based on their own data, but from another citation. Without the calculation results associated with their own data, it is very hard to judge the uncertainty of pH calculations associated with their ionic compounds measurement. The author seems to claim one-unit difference is sufficient small in the response to major comment 1. However, their pH fluctuation range is also around 1 unit. I still insist that the authors should perform separate QA/QC calculations to quantify the uncertainties on pH calculation.

We understand the Referee's concern here. We agree with the Referee that quantifying the uncertainty of aerosol pH calculations from thermodynamic equilibrium models represents an important priority in this research field. However, at present, challenges in the direct measurement of aerosol pH limit the extent to which uncertainty can be quantified in the pH calculations because uncertainty does not just derive from uncertainty in the measured ionic components. Pye et al. (2020) discuss this point extensively (see Sections 4 and 7). It is why the thermodynamic calculations of aerosol pH in Figures 3 – 7 of Pye et al. (2020) do not have error bars. Doing so would require a direct measurement of aerosol pH to quantify the model error. This is not a challenge that is unique to our study: for example, Zheng et al. (Science, 2020) do not present their thermodynamic predictions of pH with uncertainties, either (see their Figures 4b, 4c, S5, S6, S13b, and S14). We are following the standard convention described by Pye et al. (2020), and references therein.

Figure 4 &5: even though the authors provide some explanations in the caption, it is still not clear about their logic to group data points through such approach. How the bins were chosen and why process through such approach?

We have defined bins that are approximately evenly spaced across the range of observed values (in this case, temperature, aerosol liquid water, and total NH_3 concentration) with sufficient bin numbers and spacing so that trends in the mean values are apparent. We agree with the Referee that there is some level of subjectivity in the choice of bins in Figures 4 and 5. That is why we updated the figures so that all individual observations are plotted along with the bin means. We feel that the individual observations demonstrate that our primary conclusions are not sensitive to small changes in the bin definitions. However, all data in Figures 4 and 5 are publicly available (see data availability statement) so one could test different bin boundaries, if desired.

References

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Zheng, G., Su, H., Wang, S., Andreae, M. O., Pöschl, U., & Cheng, Y. (2020). Multiphase buffer theory explains contrasts in atmospheric aerosol acidity. *Science*, *369*(6509), 1374. https://doi.org/10.1126/science.aba3719