Review of "Significant contrasts in aerosol acidity between China and the Unites States" by B. Zhang et al.

This manuscript examines differences in aerosol pH between China and the US using both model simulations and network observations. The analysis investigates differences in aerosol pH between the two locations, primarily focusing on composition and concentration differences. Aerosol pH is an important topic and this work is timely and original. It is certainly appropriate for ACP and will be of interest to a broad scientific community. The organization is mostly fine, though some of the discussion is unnecessary (see comments below), and the writing is good. I do have some concerns that need to be addressed before I can recommend the manuscript for publication. My specific comments are below:

I think that the performance of the model is greatly overstated, as summarized in lines 235-237. As the manuscript states, model predictions of aerosol pH are frequently evaluated using comparison of modeled and measured species partitioning (NH_3 and HNO_3 are the most common species used). Figures S3, S4, and S5 show some significant problems predicting key species and parameters (especially εNH_4^+ and εNO_3^-), such that the pH predictions are also questionable in many times/locations. I think that these differences are mostly minimized in the manuscript, or not discussed accurately (e.g., Section 3.1.2). While some of the underlying differences are identified (e.g., the need for better NH3 emissions in China), the uncertainty in the pH predictions are not acknowledged. I think that the acceptable threshold for pH predictions should be much tighter than +/- 2 pH units (as line 225 – 228 seems to indicate). To address this concern, the manuscript needs to be more transparent and detailed in the discussion of the difficulties predicting both εNH_4^+ and εNO_3^-, and how this translates to uncertainty in the pH predictions.

This is no fault of the authors, but a significant paper was recently published that must be discussed (Zheng et al., Science 369, 1374–1377 (2020)), especially because the present manuscript presents several contrasting findings compared to Zheng et al. Specifically, Zheng et al. characteries differences in pH between China and the US, and the reasons for these changes. They find that the two most important factors are temperature and ALW. The present manuscript does account for ALW differences because composition and concentration both affect ALW; however, their analysis does not acknowledge the importance of temperature differences at all. Also, they discuss all of the differences as if composition has the biggest effect (e.g., adding NH3 neutralizes the acidic species…), when it may be the effect on ALW that is the most important factor, at least according to Zheng et al. Other studies have also identified the importance of temperature in driving pH differences (Battaglia et al., 2017; Tao and Murphy, 2019). Further, Zheng et al. conclude that NVCs make a very small contribution (on the order of ~5%) to the pH difference between the two regions, which seems to contradict the present study. So, the present manuscript needs to add significant discussion to address similarities and differences between their study and Zheng et al. They should also more broadly discuss other factors that are known to influence pH, such as temperature.

Finally, there is quite a bit of space (both figures and discussion) dedicated to analyses that don't seem to add much to the manuscript. For example, Line 350 describes the process for segregating the data into different groups to further examine the effects of TNH3. This was a good idea, however, the results (shown in Fig. 8) don't add any new insight to our current understand of aerosol pH. I would say the same is true for Fig. 11 and the associated discussion, and for the analysis of the TNO3/TSO4 molar ratios. I would strongly suggest moving these figures and discussion to the Supporting Information, especially in light of the added discussion and possible analyses needed to address the above comments.

Technical Corrections

Line 51: delete the period appearing in the middle of the sentence

Line 62-63: what are "large-scale" measurements?

Line 93: I question the stated accuracy of the AMoN NH3 measurements – especially given the variability between duplicate samples reported by the network.

Line 97: "Its" should not be capitalized

Line 101 – 103: provide the criteria for identifying outliers, and the number of outliers excluded from the respective datasets

Line 158-159: specify if sulfate was also adjusted?

Line 241-241: need to acknowledge that most of the pH predictions over China cannot be evaluated due to limitations in observational data.

Line 251-252: these correlations are weak, so the description of a "significant positive correlation" is misleading.

Line 325: "this could be due to higher biases in H+ concentration by ISORROPIA in ammonia poor conditions" – I don't follow this explanation?

Line 457: awkward as written

References

Battaglia, M. A., Douglas, S., and Hennigan, C. J.: Effect of the Urban Heat Island on Aerosol pH, Environ. Sci. Technol., 51, 13095–13103, https://doi.org/10.1021/acs.est.7b02786, 2017.

Tao, Y. and Murphy, J. G.: The sensitivity of $PM_{2.5}$ acidity to meteorological parameters and chemical composition changes: 10-year records from six Canadian monitoring sites, Atmos. Chem. Phys., 19, 9309–9320, https://doi.org/10.5194/acp-19-9309-2019, 2019.

Zheng, G., Su, H., Wang, S., Andreae, M.O., Pöschl, U., Cheng, Y., Multiphase buffer theory explains contrasts in atmospheric aerosol acidity, *Science*, 369 (1374-1377), 2020.