Review of "Role of ammonia on fine-particle pH in agricultural regions of China: Comparison between urban and rural sites" by S. Wang et al.

General Comments:

This manuscript presents simultaneous measurements of inorganic aerosol composition and key gas-phase species (NH_3, HCl, HNO_3) at two urban and three rural sites in Henan Province, China. The measurements occurred during the winter and capture one of the well-documented winter haze episodes. The focus of the manuscript is modeling aerosol pH at the urban-rural sites, and performing sensitivity tests to characterize the factors that most control pH during such polluted conditions. This is a novel and valuable data set that can add important insight to our understanding of aerosol pH. The topic is appropriate for *ACP* and will be of interest to a broad audience. The manuscript organization and figure quality are generally good, however certain elements of the presentation – namely the English usage – require improvements throughout to bring it up to publication quality. Several analyses and/or results require clarification, and a number of key references are missing. These comments are detailed below.

Specific Comments:

Missing from the manuscript is a discussion of measurement uncertainty and statistical significance of the various analyses. For example, throughout the manuscript (line 27-78, 194-195, 203-204, 228-229, 250-254, 260-261) concentrations or values at the five sites are compared and ranked. In some cases, the differences appear to be quite small, and are not likely statistically significant if uncertainty and measurement variability are taken into account.

Figures 5, 6, and 7 and the associated discussion in Section 3.3 need clarification. It is not clear how the different sensitivity analyses were performed (What was held constant? What was varied? Which conditions were used for the base simulations?). In some cases, the interpretations also require clarification: e.g., the authors use "%RSD" – how is this actually defined? It seems in the text that they interpret %RSD as actual pH values instead of a percentage, but this could just reinforce the point that this discussion needs to be improved. On the point of RSD, the description in the text does not seem consistent with what is plotted in Fig. 5 or Fig. 7.

As stated above, the manuscript requires editing for grammar, English usage, and punctuation. Since this is an issue throughout the manuscript, specific areas for improvement are not identified in 'Technical Corrections'.

A number of key references are missing. These need to be cited and the discussion enhanced to include the context they provide. To Sections 3.3 and 3.4 add discussion of Weber et al. (2016), Vasilakos et al. (2018), and Nenes et al. (2019). On the meteorological effects on pH, add discussion of Battaglia et al. (2017) and Tao and Murphy (2019). For recent discussions of aerosol pH importance, definitions, and reported ambient levels, add discussion of Pye et al. (2019). Line 67 refers to several other studies that have examined aerosol pH in agricultural regions of China – the present results should be contrasted with these prior studies.

The discussion of Fig. 3 (line 208 - 218) needs revision. For Figures 3d, 3e, and 3f, the authors discuss the strong anticorrelated relationships, but why are these separated by case 1, 2, and 3? This is the same location and the measurements are all within a few week span, so the differences in slope and intercept are curious. Discussion of the physical meaning should be provided (e.g., what explanation is there for the greater sensitivity of H^+ to pH in case 1?). It would probably be more instructive to combine these into one plot. For Figures 3g, 3h, and 3i, what explanation do the authors have for a linear relationship between H^+ and TWSII on a semi-log plot? For a given TWSII concentration, the H^+ level appears to vary by several orders of magnitude, which seems to agree with Guo et al. (2015), Hennigan et al. (2015) and Murphy et al. (2017).

The results in Figure S2 need much more discussion. This is not just limited to the HCl and HNO_3 results, though more explanation should be provided. Typically, the model predictions of pH are validated by the predictions of NH_3/NH_4^+, HCl/Cl^-, and HNO_3/NO_3^- partitioning in lieu of direct pH measurements. In addition to the problems with HCl and HNO_3 at all sites, it looks like there are systematic differences in NHx partitioning between the model and measurements at the U-ZZ and R-PY sites. Why is that and what does this mean for the associated pH predictions at these sites?

Technical Corrections:

A map of the five sites should definitely be included – either in the main manuscript or supplemental.

Line 152: ‘distraction’ is not the right term here.

Fig. S5 needs improvement: the scale is not evident from the figures, nor is the relative locations and proximity of the five different sites.

Line 238-239: I don’t see where sensitivity of pH to crustal species was analyzed?

Line 262-263: yes, but these are presumably correlated?

Line 270-271: this comment is misguided - see Weber et al (2016) for more context and explanation.

Line 286: suggest removing ‘obviously’.

Lines 291-292: what is the physical meaning of these equations?

Line 298-299: this sentence is confusing – I suggest re-writing.

Line 306: I’m not sure the evidence supports this statement. What about meteorology? Also, differences in local vs. regional emissions would need to be accounted for.

Line 333: consider changing ‘promote’ to ‘perturb’?

References:

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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.

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