Review of "Role of ammonia on fine-particle pH in agricultural regions of China: Comparison between urban and rural sites" for *Atmospheric Chemistry and Physics*

General Comments

Wang et al. analyzed a novel dataset of inorganic aerosol constituents and their gaseous precursors as well as organic aerosol constituents in a province in central China with heavy agricultural activity. They used ISORROPIA, an inorganic thermodynamic aerosol model, and previously published methods to estimate aerosol pH for the observed particles. The measurements were made with state of the art instruments and the modeling was carefully evaluated. They positioned their work in the broader context of aerosol acidity work very well.

The analysis sought to elucidate differences between the pH and the pH sensitivity to aerosol constituents between urban and rural sites. In this aspect of the work, more care could be taken. First, a map with the key emissions and measurement locations marked would help readers unfamiliar with the area characterize the results. Secondly, the differences between the urban and rural sites appears to be fairly random and small. The authors draw conclusions about the rankings between sites or the degree of difference in the aerosol pH sensitivities based on these measurements. Without understanding the uncertainties in the measurements and the accuracy of calibrations between the five instruments, their conclusions seem poorly supported. With the addition of estimated uncertainty on the measurements as well as evidence of the calibration of the instruments against a standard, the authors would be better able to support their conclusions if they remain valid. Finally, relative standard deviation as a representation of sensitivity of aerosol pH to total ammonia is unfamiliar to me. This metric seems dependent on the range of values selected for the parameters of interest (e.g., total sulfate, temperature) and is difficult to interpret. Please consider using a unit-based sensitivity analysis approach that would indicate the influence of each parameter without respect to the observed range of each influential parameter.

Accordingly, I would consider this work publishable with major revisions. A few more specific comments are included below that may be relevant if the major revisions yield publishable results.

Specific Comments

A. Text

Line	Comment
29-31	These two ideas are not independent. Please choose to state one or the other.
41	Typicallly, the phase is included in the subscript (e.g., "$NH_{3(g)}/NH_{4(aq)}^+$").
42	"element" should be "elements".
102	"electron" is unclear here.

174   Case 2 seems to have different meteorological episodes within it. Consider splitting the lower RH and corresponding lower concentration days into a fourth episode.

209-10   This conclusion is simply based on Equation 1. Please consider removing the log-linear dependence from Figure 2 and this sentence. This correlation exists because of the calculation.

213-5   This statement makes it difficult to see that Case 3 is being compared with Case 1. Consider making the single sentence into two sentences.\

221   Perhaps the back trajectories in the SI indicate the degree of influence from transport, but it may be helpful to include a summary of the influences in the description of the cases if this information will be referenced in the discussion.

264   "figure" should be "Figure".

289   "liner" should be "linear".

B. Figures and Tables

Fig 1 & 2   Please add uncertainty shading or bars to the inorganic constituent measurements and to ammonia. If you could please propagate that uncertainty to aerosol pH, the aerosol pH difference across sites could be evaluated.

Fig 5   This figure is very difficult to understand. What were the conditions across which modeling was conducted at each site? Did those ranges that are plotted as contour colors extend beyond the ranges plotted as a bar? More information would be required in the caption or an associated table in the SI to make this figure helpful.

Fig 6   Why is excess NHx not just $NH_{3(g)}$ as predicted from ISORROPIA? As calculated, excess NHx ignores nonideality as well as bisulfate formation.

Fig 7   How was $SO_{4(aq)}^{2-}$ specified when running ISORROPIA? If the authors mean total sulfate, please correct the figure caption and axis label. Although all of the sulfate remains in the aerosol phase in ISORROPIA, it does not all become $SO_{4(aq)}^{2-}$ necessarily.