Atmospheric
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Discussions

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Interactive comment on "Aerosol pH and its influencing factors in Beijing" *by* Jing Ding et al.

Anonymous Referee #2

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This paper presents observations and analysis of the inorganic aerosol system in Beijing for 2017. The pH values are realistic; however, more analysis to verify the methods would make for a stronger paper.

Major comments:

1. Clarify the methodology in terms of how pH was calculated. How was the pH in different size ranges modeled and combined? Even if that appears in other work (as indicated in the text), a quick summary of the method would be useful. Line 209 indicates pH (for the coarse mode?) was determined by ignoring the gas phase and running ISORROPIA in forward mode with zero gas. How was this assumption verified? Figure 2 shows a comparison of total species modeled vs predicted, but that doesn't give a sense of how the size dependent predictions worked. Line 438 indicates that NH3, HNO3, and HCl were determined through iteration when MOUDI data was used.

Was that just for the fine mode particles?

2. Driving factor analysis: The driving factors for pH were obtained by holding all composition, RH, and T parameters at average values and then varying one of the input values (line 291 and thereafter-consider putting some of this method in section 2). A larger change in ALWC, H+air, or pH due to varying one input was interpreted as that input having a major influence on pH. The authors do note that this method will not capture the effect of simultaneous changes in more than one factor.

a. Did the authors consider restricting the output values used to calculate sensitivities (e.g. Table 2) to the space actually probed in the ambient? For example, ALWC output from the simulation varying RH spans 0-140 ug/m3 while most other input parameters did not result in this range of ALWC values. Was 140 ug/m3 ALWC predicted for any of the actual atmospheric conditions? What space is actually probed in the ambient atmosphere in terms of ALWC, H+air, and pH compared to what is probed in the simulated data holding all but one parameter constant?

b. How can the method be evaluated? Does using average inputs result in the same predicted pH that would be obtained by averaging all individual pH predictions from individual inputs? Could the average pH and input be indicated on each panel of Figure 5 to 7? How evenly distributed over the input range are the various inputs? Would it be more appropriate to focus on the interquartile range instead of full range of inputs?

c. Are there units to the quantities in table 2?

d. How would a multiple linear regression analysis differ from the technique of varying one quantity at a time?

e. Could a Monte Carlo method or other technique be used to make sure atmospherically relevant combinations of inputs are being used?

2. Instead of classifying PM2.5 into clean (0-75 ug/m3), polluted, and heavily polluted (>150 ug/m3), it may be illustrative to consider PM2.5 in a continuum. 0-75 ug/m3 on

a daily average is not very clean as it includes concentrations that exceed air quality standards. In addition, by considering PM2.5 concentrations as continuous, you may be able to better determine the association of pH with PM2.5. Consider that the pH for the three classifications is reported with a range/uncertainty that indicates the differences in pH between clean, polluted, and heavily polluted conditions are not statistically significant (values on line 262 overlap). However, if considered as a continuous variable, a regression with confidence interval could be provided and might provide a more robust analysis of the association.

3. Better connect the size resolved measurements with the rest of the text. To what degree did the presence of coarse material drive ambient pH? Do figures 5-7 and the analysis regarding drives of pH only consider fine mode pH?

Minor comments:

1. Line 49. Instead of stating that aerosol acidity is "usually estimated" by the charge balance, I would indicate "sometimes" or "frequently," but not usually as many studies do use a thermodynamic model.

2. Line 52-55 wording indicates ion balance fails because acidity is estimated by aerosol water extract. This doesn't follow well as ion balance (e.g. difference between number of charge equivalent anions and cations) doesn't require extraction.

3. Line 95: may want to indicate models "often" assume internal mixtures (but that is not a requirement).

4. Line 98-99: For this statement indicating nitrate is mainly in the fine mode, does that need to be qualified by indicating a location or time of year? Does fine mode nitrate generally exceed coarse nitrate?

5. Near line 155 and Figure 1: Spring shows a fairly persistent difference in the concentration of PM10 vs PM2.5. Two dust episodes are mentioned. With the exception of these two episodes, do you have a sense of what is contributing to the PM10-PM2.5

material? Late September also indicates an episode in which PM10 is elevated compared to PM2.5.

6. Line 190 indicates water uptake onto hydrophilic organics can be ignored unless the fraction of particle water due to organics is near 1 (100%). Water due to uptake on organics is presumably important even when it is not the sole contributor to particulate water. The threshold of 1 should be removed and perhaps a statement about the potential error incurred by ignoring ALWCo should be added.

7. Text on lines 235-238 seems misplaced or unnecessary.

8. Line 277 highlights sulfate as a driving factor for pH. Sulfate peaked at night during the winter (Figure 4) when photochemical activity is lower. To what degree is the diurnal variation in sulfate driven by chemistry vs meteorology (e.g. planetary boundary layer depth)?

9. Line 284: Is the key difference between the US and Beijing more driven by the higher concentrations or the greater variability in concentrations?

10. Line 384:386 represents a simplified description of ammonia partitioning in which ammonia acts first to neutralize sulfate and then any leftover ammonia can react with nitrate to make ammonium nitrate. Perhaps the authors do not mean this so simply. Reword to reflect the semivolatile nature of ammonia and nitrate.

11. Line 388: Do the authors mean that aerosol would be fully neutralized except for the fact that ammoia is taken up into clouds and precipitation? Reword to reflect the buffering nature of ammonia.

12. Caption to table 2: This table appears to be the sensitivity of acidity, ALWC, and H+air to chemical components (not the other way around). Please clarify caption.

13. Figure 3: use a common color scale for all panels.

14. Figure 5, 6, 7, caption. These figures appear to be the sensitivity of ALWC, H+air,

and pH to chemical components. Reword caption.

15. Line 136: Have you looked at trends from 2013, 2015, and 2017 datasets you have collected?

16. Additional improvements in terms of editing would be useful.