

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

Anonymous Referee #1

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This paper utilizes unique data sets to predict aerosol pH in the more polluted regions of China. Overall, the paper is a significant contribution since little is known about aerosol pH in these regions and even less on size resolved pH. However, in my view, the analysis is somewhat limited. The authors have an interesting data set that could be more fully utilized to assess the pH predictions, partitioning of inorganic species and understand aerosol pH from a more fundamental standpoint.

A suite of important inorganic gases was measured with the MARGA, but they are not significantly discussed in the paper. This is a major oversight. For example, in the comparison of the model to measurements the particle data are shown, but no gas data. For the MOUDI, no gas data is available so the pH is estimated by an iteration method, why not use the MARGA data, which includes gases, to test the sensitivity of pH to this approach?

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Greater utilization of the gas data could also help the authors understand fundamentally what is driving pH and the sensitivities to various parameters. This could include the use of S curves, as done extensively by Guo et al, to go beyond just simple variation of one variable at a time. Eg, why in the sensitivity analysis do changes in HNO₃ not affect pH, but changes in NH₃ do? These, and possibly other, more detailed analysis would reduce the sense that the authors simply run the thermodynamic model and plotted results.

pH is calculated under the assumption of a completely deliquesced particle with no phase separation, all the way down to very low RH, ie, to 30%. These assumptions at low RH need to be justified. Eg, the predicted and measured partitioning of NH₃/NH₄⁺, HNO₃/NO₃⁻, HCl/Cl⁻ etc (ie include analysis of the gases) could be assessed as a function of pH and see if changes occur at lower RH. Discussion of phase separation in the literature under various conditions (RH, T, O/C) etc should be discussed.

Finally, the paper needs editing for proper English usage and grammar.

Specific Comments Line 37 change specials to species

Line 202 and following, it is not just lack of NH₃ data that can affect predicted pH, what about HNO₃, HCl, etc?

Line 205, how much did the pH change when the iteration approach is used? Or, were the predicted gas species concentrations reasonable relative to what was measured during the MARGA study period.

Line 229 is superfluous, it is well known that low pH means high acidity.

Line 235 to 238: this paragraph seems out of place.

Fig 3 caption, what does transverse direction mean on a polar plot?

Line 246 change souther to southern.

Line 276-286. From Fig 4 it does not appear that pH and sulfate diurnal trends are

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always the same (actually inverse), as stated. Looks like a stronger inverse trend with liquid water. More quantitative analysis is needed to support the statements made in this section.

Line 327, provide a physical explanation for the U shape dependency of H^+ on NO_3^-

Line 330-331: Is it really true that there is a straightforward relationship between NH_3 and H^+ over broad NH_3 concentration ranges? I.e., will increases in NH_3 always lead to higher dissolved NH_3 ? Technically it may be true, but the relationship may be highly nonlinear under certain conditions. This statement seems too broad.

Line 335, this is an obvious statement based on Eq (1). In fact much of the discussion throughout relating pH, H^+ and LWC is obvious from Eq (1).

Line 358 and on regarding changes in pH with NO_3^- . The authors discuss the trends they observe in the sensitivity analysis and NO_3^-/SO_4^{2-} , but never provide an explanation. By just reporting of results, the value of this work is greatly limited, despite the what could be done with this unique data set.

Lines 380 and on regarding TA and TS. Most of these statements are technically incorrect (although, from a broad perspective they may have a grain of truth to them). The authors data show that the pH is far from neutral despite it being NH_3 rich. This analysis largely continues misconceptions of how aerosol composition depends on interactions between SO_4^{2-} , NH_3 , NH_4^+ , HNO_3 , NO_3^- and LWC. Eg, is HNO_3 only taken up once sulfate is so-called neutralized; maybe this can be tested with the data (there should be no NO_3^- and then a sudden jump in NO_3^- when $[TA]/2[TS]$ is greater than 1. Another example, why does pH vary, even for this data set, if NH_3 is always in great excess? It is suggested that the authors look at S curves (partitioning of say NH_3 and/or HNO_3 vs pH) instead of the analysis currently being used.

Line 419 to 421. The loss of buffering capacity of the coarse mode mineral dust during winter pollution events is very interesting and has direct implications for predictions of

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NO_2 + SO_2 oxidation pathways proposed by Wang et al 2016 and Cheng et al 2016. It is suggested that this finding be noted more prominently, maybe even included in the Abstract. However, this period does not seem to be shown in the plots?

The use of the word synthetically throughout the paper is confusing, it is suggested that it not be used since its meaning is unclear.

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