

Anonymous Referee #2

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.

Response:

Thanks for your important comments, which are very useful to make our paper more rigorous. Please see our point-by-point responses to the comments and the revised manuscript for details. The order of the Figures or Tables in Response is the same as the corresponding Figure or Table appears in the main text and supplemental materials.

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 a 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?

Response:

The data set of MOUDI was obtained during 2013 and 2015, which was not synchronous with the online ion data (obtained in 2016 and 2017). There was no observation of gas precursors during the periods of MOUDI sampling, hence an iteration method used in Fang et al. (2017) and Guo et al. (2016) was applied in this work. As a brief summary, the predicted NH₃, HNO₃, and HCl concentrations from the *i*-1 run were applied to the *i*th iteration, until the gas concentrations converged. Based on these iterative gas phase concentrations, each MOUDI stage's measured aerosol ion concentrations and estimated gas concentrations, as well as the averaged RH and T during each group sampling time, were input the ISORROPIA-II to determine pH for each stage. The particles at each size bin were assumed to be internally mixed.

The comparisons of iterative and predicted NH₃, HNO₃, and HCl as well as measured and predicted

NO_3^- , NH_4^+ , Cl^- , $\epsilon(\text{NH}_4^+)$, $\epsilon(\text{NO}_3^-)$, and $\epsilon(\text{Cl}^-)$ for data from MOUDI samples were showed in Figure 3. The previous study showed that coarse mode particles are very difficult to reach equilibrium with the gaseous precursors due to kinetic limitations (Dassios et al., 1999; Cruz et al., 2000). Assuming coarse mode particles in equilibrium with the gas phase could result in a large bias between measured and predicted NO_3^- and NH_4^+ in coarse mode particles (Fang et al, 2017). We also find that in this work, it can be clearly seen that assuming coarse mode particles in equilibrium with the gas phase could overpredict NO_3^- and Cl^- and underestimate NH_4^+ in the coarse mode (the blue scatters), which could subsequently underestimate the coarse mode aerosol pH. Compared with the coarse mode particles, the measured and predicted NO_3^- , NH_4^+ , and Cl^- agreed very well in fine mode particles. Considering the kinetic limitations and nonideal gas-particle partitioning in coarse mode particles, the aerosol pH in coarse mode was determined by ignoring the gas phase.

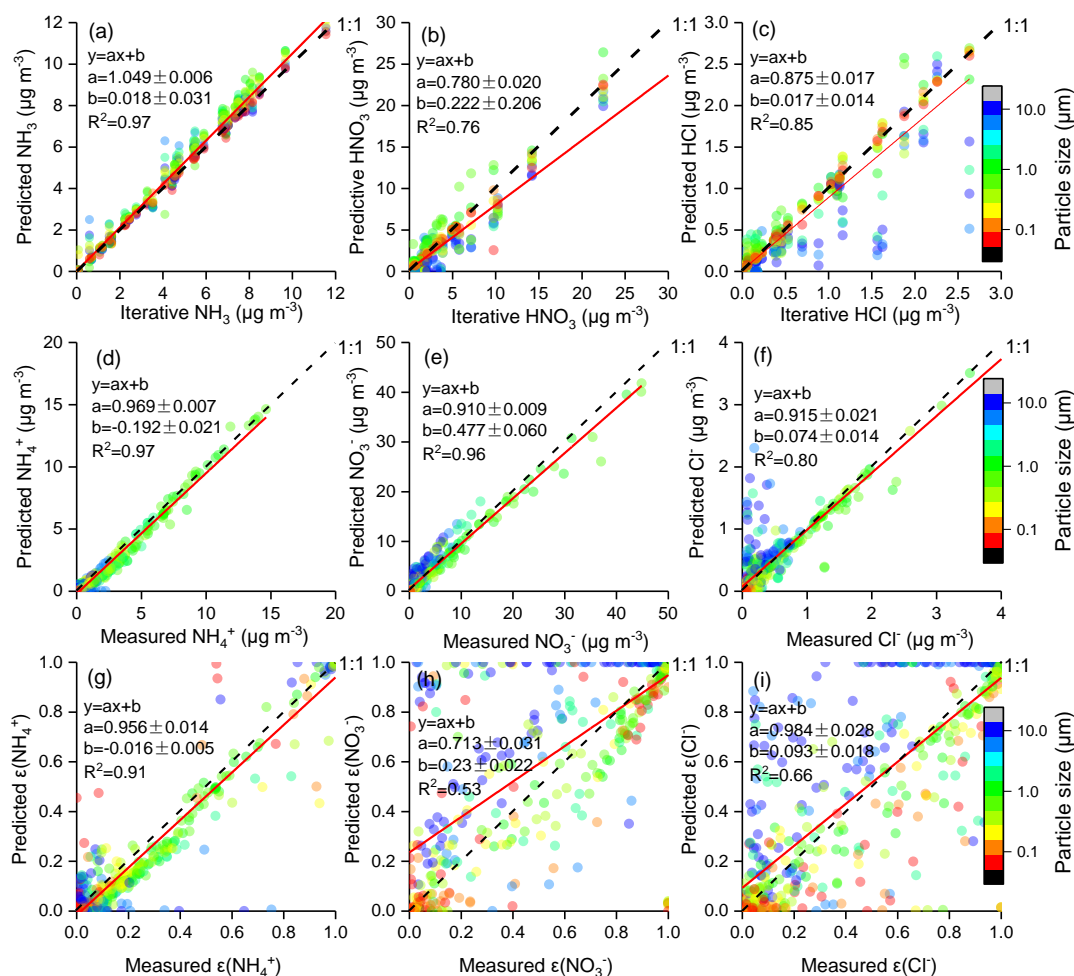


Figure 3. The comparisons of iterative and predicted NH_3 , HNO_3 , HCl as well as measured and predicted NO_3^- , NH_4^+ , Cl^- , $\epsilon(\text{NH}_4^+)$, $\epsilon(\text{NO}_3^-)$, $\epsilon(\text{Cl}^-)$ for data from MOUDI samples, which all

colored by particle size.

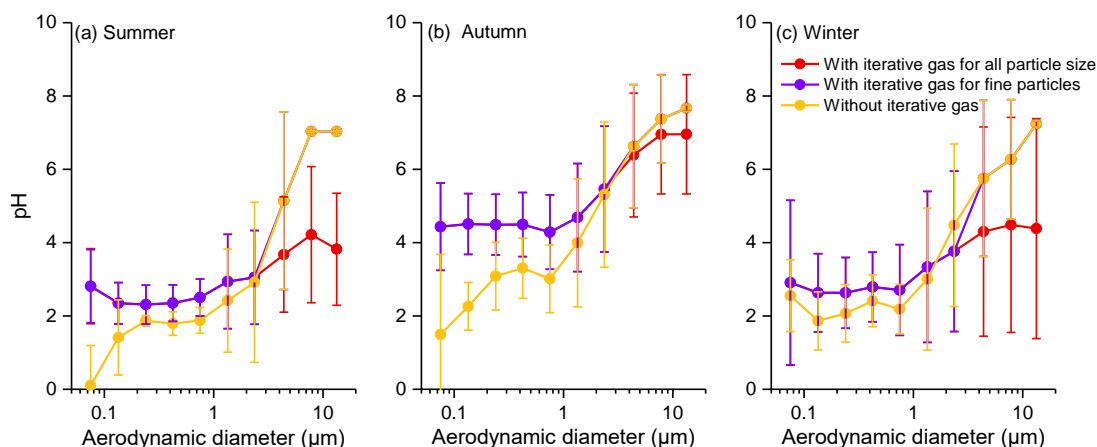


Figure R1 The averaged size-resolved aerosol pH in three seasons predicted with three assumptions: (1) predicted with no iterative gases, (2) predicted assuming lack of equilibrium with gas phase for coarse mode particles, (3) predicted assuming all particles in equilibrium with the gas phase.

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.

Response:

Thanks for your important advice. The detailed introduction of the method about aerosol pH driving factor analysis has been put in section 2.5. In the real ambient air, the thermodynamic process of the aerosol is complicated, it is not easy to tell the effect of one certain factor on the aerosol pH. The ALWC, H_{air} , and aerosol pH are all the output of ISORROPIA-II. They reflect an objective state of particles. Considering the relative independence between input parameters, it is reasonable to discuss the influence of input variables on output parameters with the results of ISORROPIA-II. Thus, in this paper, we focus on the sensitivity analysis of single-factor variation, which can reflect the variation tendency of aerosol pH caused by the change of each variable.

a. Did the authors consider restricting the output values used to calculate sensitivities (e.g.

Table 2) to space actually probed in the ambient? For example, ALWC output from the simulation varying RH spans 0-140 $\mu\text{g}/\text{m}^3$ while most other input parameters did not result in this range of ALWC values. Was 140 $\mu\text{g}/\text{m}^3$ 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?

Response: All data used in the sensitivity analysis were based on the actual observation, not randomly generated simulation data, which helps us capture a more real impact. When the RH was considered as a variable, ALWC output spans 0-140 $\mu\text{g}/\text{m}^3$, this mainly attributed to the vital impact of RH on ALWC, especially when the RH was higher than 80% owing to the exponential increase of ALWC with the RH. Whereas in other simulated cases, the averaged RH was generally within 50% ~ 75%, hence the output ALWC was relatively low. In summer and autumn, the actual ALWC was even more than 140 $\mu\text{g}/\text{m}^3$ when both aerosol components and RH were high.

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 a full range of inputs?

Response: The average value and variation range for each variable in all four seasons were listed in Table S1 and Figure S7. The aerosol pH1 is the value by averaging all individual pH predictions from each input variable, for example, the average aerosol pH was 3.74 ± 0.47 when the SO_4^{2-} was regarded as an input variable while other input parameters were fixed with the average value. The aerosol pH2 is the value by using average inputs for all input parameters. In theory, pH1 and pH2 cannot be the same, otherwise, the effect of the variables on aerosol pH will not be reflected.

Table S1 The average value and range for each variable in all four seasons, as well as the two average aerosol pH types. The aerosol pH1 is the value by averaging all individual pH predictions from each continuous input variable, for example, the average aerosol pH was 3.74 ± 0.47 when the SO_4^{2-} was regarded as a continuous input variable

while other input parameters were fixed with the average value. The aerosol pH2 is the value by using average inputs

for all input parameters. The unit of chemical components is $\mu\text{g m}^{-3}$.

Spring	SO_4^{2-}	NH_4^+	NO_3^-	Cl^-	RH, %	T, °C	Ca	Na	K	Mg
Average input	8.4	25.7	13.5	1.1	52	20.9	1.29	0.20	0.34	0.3
Variable range	3.0~41.4	0.1~33.9	0.4~77.6	0.03~6.27	30~92	10.0~33.3	0.1~3.0			
pH1	3.74±0.47 3.69±0.19 3.65±0.53 3.81±0.09 3.79±0.05 3.81±0.27 3.73±0.16									
pH2	3.82									
Winter	SO_4^{2-}	NH_4^+	NO_3^-	Cl^-	RH, %	T, °C	Ca	Na	K	Mg
Averaged	7.3	12.2	14.3	3.0	52	2.7	0.2	0.40	1.0	0.2
Ranges	2.0~34.6	1.3~46.7	0.8~49.3*	0.02~25.2	30~94	-8.7~16.2	0.01~0.7			
pH1	4.32±1.21 3.86±1.04 4.27±0.48 4.27±0.16 4.39±0.18 4.36±0.29 4.36±0.04									
pH2	4.36									
Summer	SO_4^{2-}	NH_4^+	NO_3^-	Cl^-	RH, %	T, °C	Ca	Na	K	Mg
Averaged	8.6	26.8	10.2	0.6	74	26.1	0.5	0.60	0.2	0.1
Ranges	0.6~40.1	1.2~69.6	0.3~59.8	0.1~2.8	30~97	14.2~38.1	0.02~2.9			
pH1	3.43±0.27 3.31±0.32 3.31±0.12 4.38±0.03 3.40±0.27 3.37±0.20 3.38±0.06									
pH2	3.38									
Autumn	SO_4^{2-}	NH_4^+	NO_3^-	Cl^-	RH, %	T, °C	Ca	Na	K	Mg
Averaged	9.3	27.8	20.3	1.0	72	16.4	0.4	0.3	0.2	0.1
Ranges	0.3~54.7	3.2~67.5	0.2~90.5	0.06~5.17	30~97	-1.1~33.3	0.02~2.3			
pH1	3.85±0.23 3.60±0.58 3.70±0.12 3.84±0.04 3.94±0.10 3.84±0.29 3.84±0.03									
pH2	3.84									

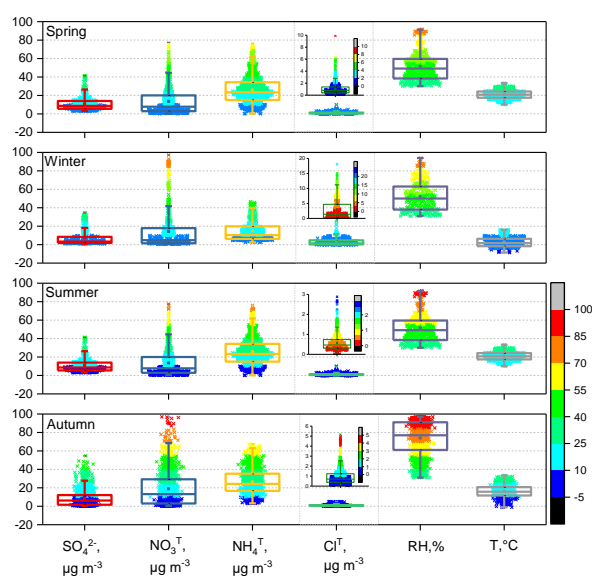


Figure S7. The distribution of each input variable for sensitivity analysis in four seasons

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

Response: Units to the quantities in table 2 were missed in the manuscript, in the revised manuscript, we replace the deviation by relative standard deviation as the evaluation target, hence the unit is unified to %.

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

Response: The relationships between input variables and aerosol pH are not simply linear. The method in this work based on the overall accurate relationship between variables rather than the permutation and combination in the mathematical sense, the latter may subversively change the relationship between variables and does not conform to the actual physical laws. Moreover, the predicted aerosol pH in the sensitivity analysis was realistic, which confirms that the method we used was reasonable.

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

Response: The Monte Carlo method is a good way to evaluate the uncertainty of the predicted aerosol pH and to determine if the input parameters are appropriate. However, as mentioned above, all input variables came from the actual observation to make sure the relationships between variables could conform to the actual physical laws. Moreover, the sensitivity analysis in this work focused on the variation tendency of aerosol pH rather the absolute aerosol pH value.

3. Instead of classifying PM_{2.5} into clean (0-75 ug/m³), polluted, and heavily polluted (>150 ug/m³), it may be illustrative to consider PM_{2.5} 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 PM_{2.5} concentrations as continuous, you may be able to better determine the association of pH with PM_{2.5}. 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.

Response: Thanks for your suggestion. Firstly, three groups for PM_{2.5} were classified by hourly PM_{2.5} mass concentration, not daily average PM_{2.5} mass concentration. Secondly, the differences in pH between clean, polluted, and heavily polluted conditions were indeed not significant, the conclusion in the manuscript was just taken from the average value of pH. More deep analysis has been added in the revised manuscript.

Table 1 showed that as the air quality deteriorates, all aerosol components, as well as ALWC and H_{air}⁺, increased, but the differences in pH between clean, polluted, and heavily polluted conditions are not statistically significant. The relationship between PM_{2.5} and aerosol pH was shown in Figure S8, the aerosol pH under clean condition spanned 2~7 while the aerosol pH under polluted and heavily polluted conditions mostly concentrated in 3~5. Time series of mass fraction of NO₃⁻, SO₄²⁻, NH₄⁺, Cl⁻, and crustal ions (Mg²⁺ and Ca²⁺) in total ions, as well as pH in all four seasons, were showed in Figure 4. It can be seen that on clean days, high aerosol pH (>6) was generally companied by high mass fraction of crustal ions, while the relatively low aerosol pH (<3) was companied by high mass fraction of SO₄²⁻ and low mass fraction of crustal ion, which was most obvious in summer (large part of aerosol pH with RH≤30% were excluded in spring and winter). On polluted and heavily polluted days, the aerosol chemical composition was similar, mainly dominated by NO₃⁻, hence the differences of aerosol pH on polluted and heavily polluted days were small. Compared with the mass concentration of PM_{2.5}, the different aerosol chemical compositions may be the essence that drives aerosol acidity. The impact of aerosol compositions on aerosol pH is discussed in Section 3.4.

Table 2. The averaged ambient temperature and $\epsilon(\text{NH}_4^+)$, $\epsilon(\text{NO}_3^-)$, $\epsilon(\text{Cl}^-)$ at different ambient RH levels in four seasons.

	RH	T, °C	$\epsilon(\text{NH}_4^+)$	$\epsilon(\text{NO}_3^-)$	$\epsilon(\text{Cl}^-)$
	≤ 30 %	24.8 ± 3.7	0.17±0.14	0.84±0.12	0.67±0.24
Spring	30~60 %	20.6 ± 3.8	0.25±0.14	0.91±0.06	0.82±0.16
	>60 %	15.8 ± 2.7	0.28±0.12	0.96±0.03	0.96±0.06

	$\leq 30\%$	5.4 ± 5.3	0.31 ± 0.13	0.78 ± 0.12	0.89 ± 0.14
Winter	30~60 %	1.0 ± 3.6	0.50 ± 0.21	0.89 ± 0.10	0.97 ± 0.03
	>60 %	-1.9 ± 2.1	0.60 ± 0.20	0.96 ± 0.03	0.99 ± 0.01
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	$\leq 30\%$	35.6 ± 0.4	0.06 ± 0.02	0.35 ± 0.20	0.39 ± 0.17
Summer	30~60 %	29.6 ± 4.2	0.17 ± 0.11	0.65 ± 0.23	0.43 ± 0.16
	>60 %	25.2 ± 3.8	0.26 ± 0.12	0.90 ± 0.12	0.71 ± 0.15
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	$\leq 30\%$	21.7 ± 7.5	0.07 ± 0.06	0.49 ± 0.25	0.45 ± 0.21
Autumn	30~60 %	20.8 ± 6.3	0.21 ± 0.14	0.82 ± 0.19	0.67 ± 0.21
	>60 %	14.9 ± 5.7	0.30 ± 0.19	0.92 ± 0.10	0.86 ± 0.13
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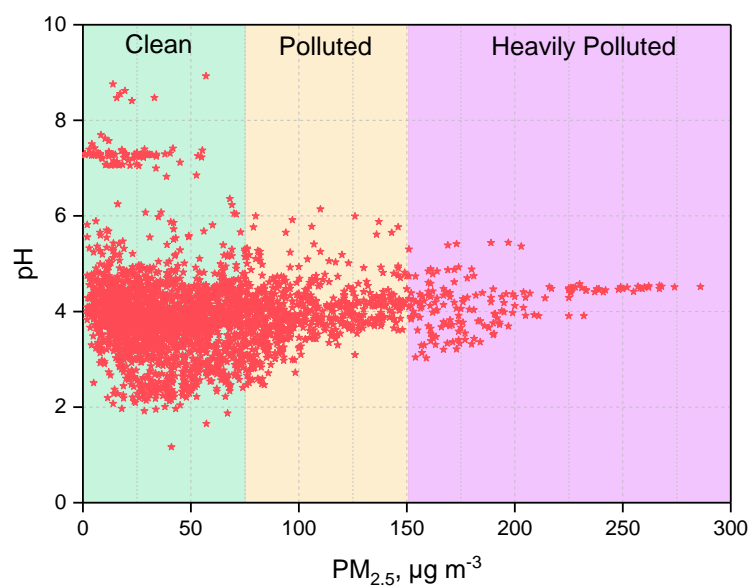


Figure S8. The relationship between $PM_{2.5}$ mass concentration and aerosol pH, the dots with $RH \leq 30\%$ were excluded.

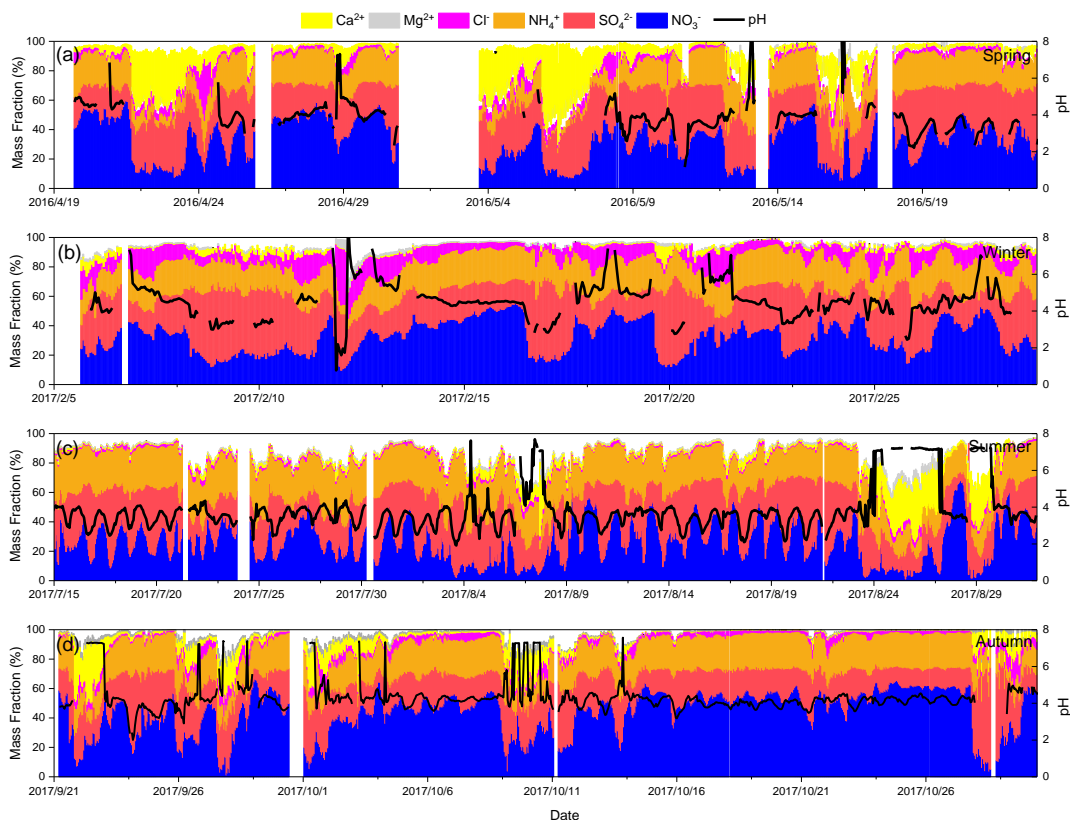


Figure 4. Time series of mass fraction of NO_3^- , SO_4^{2-} , NH_4^+ , Cl^- , crustal ions (Mg^{2+} , Ca^{2+}) in total ions as well as aerosol pH in all four seasons.

4. 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?

Response: Thanks for your suggestion. The data set of MOUDI was obtained during 2013 and 2015, whereas the online ion data was obtained in 2016 and 2017. (1) The sensitivity analysis in this work aimed at the $\text{PM}_{2.5}$ (*ie* fine particles) since the $\text{PM}_{2.5}$ components in four seasons were available and has a high temporal resolution (1h). In addition, the data set has a wide range, covering different levels of haze events, making it suitable for sensitivity analysis. The MOUDI data were only utilized to determine the size-resolved aerosol pH. (2) In this work, the coarse mode aerosol acidity was generally neutral, which mainly attributed to the higher mass concentration of mineral materials in the coarse mode. The sensitivity analysis in this work showed that the aerosol pH increased approximately linearly with the elevated Ca^{2+} in $\text{PM}_{2.5}$ (Figure S18). However, the impact of Ca^{2+}

has a limited impact on fine mode aerosol pH due to its low mass concentration in PM_{2.5}. Our previous paper showed that the mineral materials such as Ca²⁺ and Mg²⁺ mainly concentrated in the coarse mode (Figure R2, same data set with this work, Zhao et al, 2017; Su et al., 2018). We did some supplementary simulations under extreme cases that Ca²⁺ and Mg²⁺ are removed from the input files. The results showed that the presence of Ca²⁺ and Mg²⁺ in coarse mode has a crucial effect on aerosol pH (Figure S22), the difference of aerosol pH (with and without Ca²⁺ and Mg²⁺) for particles larger than 1 μm increased with the increasing particle size. The aerosol pH in coarse mode decreased by 4~6.5 unit when the Ca²⁺ and Mg²⁺ are removed.

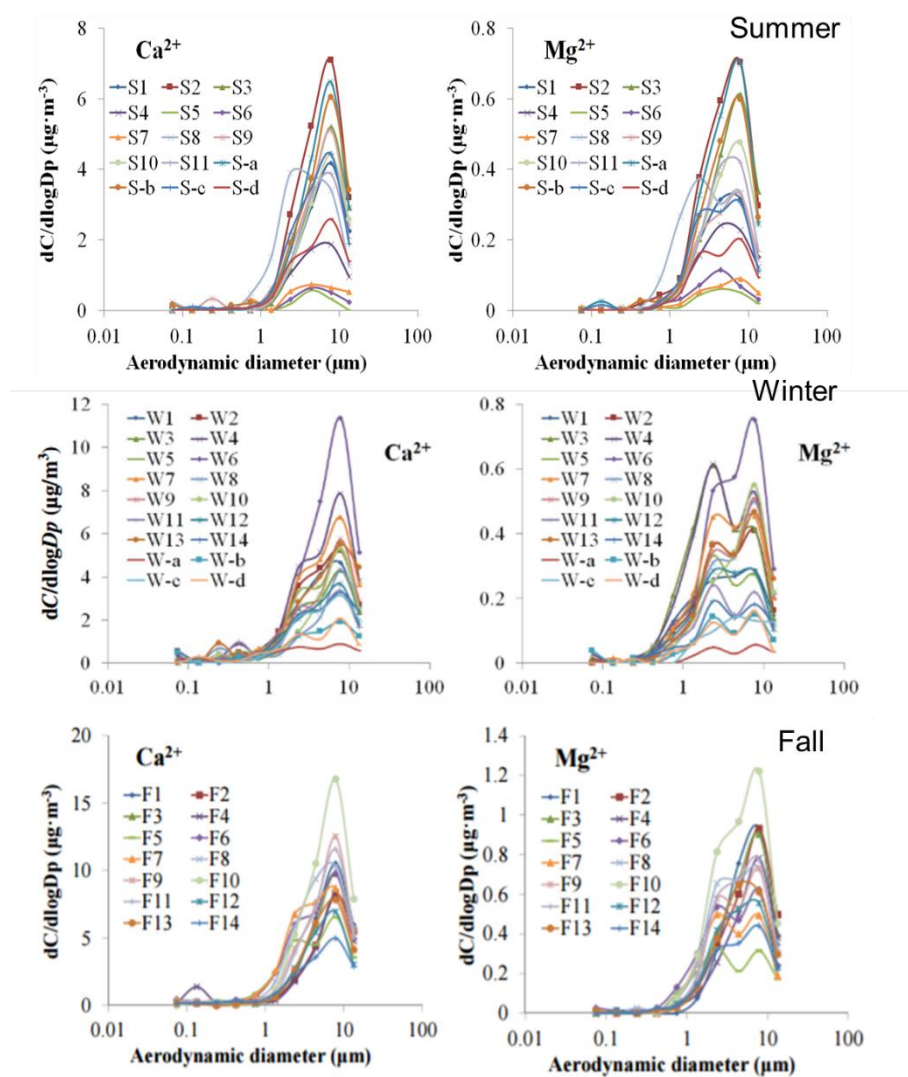


Figure R2. Size distributions of the mass concentration for Ca²⁺ and Mg²⁺ in summer, winter, and fall. (Zhao et al, 2017; Su et al., 2018)

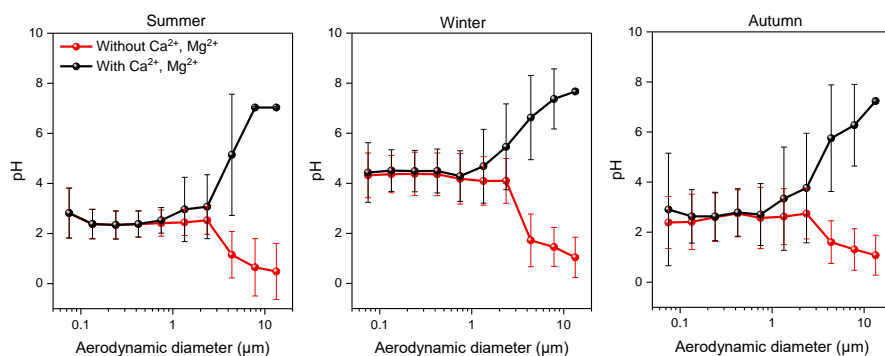


Figure S22. Size distributions of the aerosol pH with and without Ca^{2+} and Mg^{2+} in summer, winter, and autumn.

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.

Response: Thank you for your good advice, “usually” has been changed to “frequently” in the revised manuscript

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.

Response: Thank you for your correction, here we want to express that the simple ion balance cannot predict the hydronium ion concentration in the aerosol liquid water accurately. In the revised manuscript, this statement has been reworded.

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

Response: The sentences about this assumption were deleted in the revised manuscript.

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?

Response: Thank you for your question. This statement about nitrate is mainly aimed at the aerosol composition in China. Many studies in China showed that the fine mode nitrate generally exceeds

coarse nitrate except for the dust days. In Beijing, the fine mode ($\leq 2.5 \mu\text{m}$) nitrate concentration at different polluted level was 3~5 times higher than that in coarse mode (2.5~10 μm) (Meier et al., 2009; Tian et al., 2014; Sun et al., 2014), and the same size distribution was found in southern cities of China on non-dust days (Pan et al., 2009; Wang et al., 2015; Ding et al., 2017). However, in dust days, the PM_{10} concentration was much higher than that of $\text{PM}_{2.5}$, resulting in the elevated nitrates in coarse mode (Pan et al., 2009; Wang et al., 2015). In the revised manuscript, the statement was qualified.

5. Near line 155 and Figure 1: Spring shows a fairly persistent difference in the concentration of PM_{10} vs $\text{PM}_{2.5}$. Two dust episodes are mentioned. With the exception of these two episodes, do you have a sense of what is contributing to the PM_{10} - $\text{PM}_{2.5}$ material? Late September also indicates an episode in which PM_{10} is elevated compared to $\text{PM}_{2.5}$.

Response: The $\text{PM}_{2.5-10}$ was generally regarded as coarse particles. On clean days, the crustal materials could account for more than 30% of the total $\text{PM}_{2.5-10}$. During the dust events, crustal materials could account for more than 60% of the coarse particles (Xu, 2010). However, during the severe haze events, SO_4^{2-} , NO_3^- , NH_4^+ , OM, and EC also substantially accumulated in the coarse mode (Pan et al., 2009; Tian et al., 2014).

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.

Response: Thank you for your good suggestion. Surely part of organic species in particles such as water-soluble secondary organic carbon is hygroscopic, especially in ultrafine particles. In the revised manuscript, the threshold of 1 has been removed and a statement about the potential error incurred by ignoring ALWCo has been added as below.

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

Response: This paragraph has been deleted in the revised manuscript.

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)?

Response: The diurnal variation in sulfate was complex, especially during the severe haze episodes, where the rapid increase in mass concentration was mainly due to the accumulation induced by the unfavorable meteorological condition. Figure R2(a) and R2(b) showed that for most of the time, the mass fraction of SO_4^{2-} in total ions has little variation when SO_4^{2-} mass concentration increased largely, which could be regarded as the contribution of meteorology. However, at some moments in the nighttime (gray shadow in the figure), both mass concentration and mass fraction of SO_4^{2-} showed a significant increase, which mainly attributed to the secondary reaction of SO_2 . Overall, the mean SO_4^{2-} fraction in total ions at night in winter was slightly higher than that in daytime (Figure R2(c)), but differences are not statistically significant. Hence the diurnal variation in sulfate was more driven by meteorology.

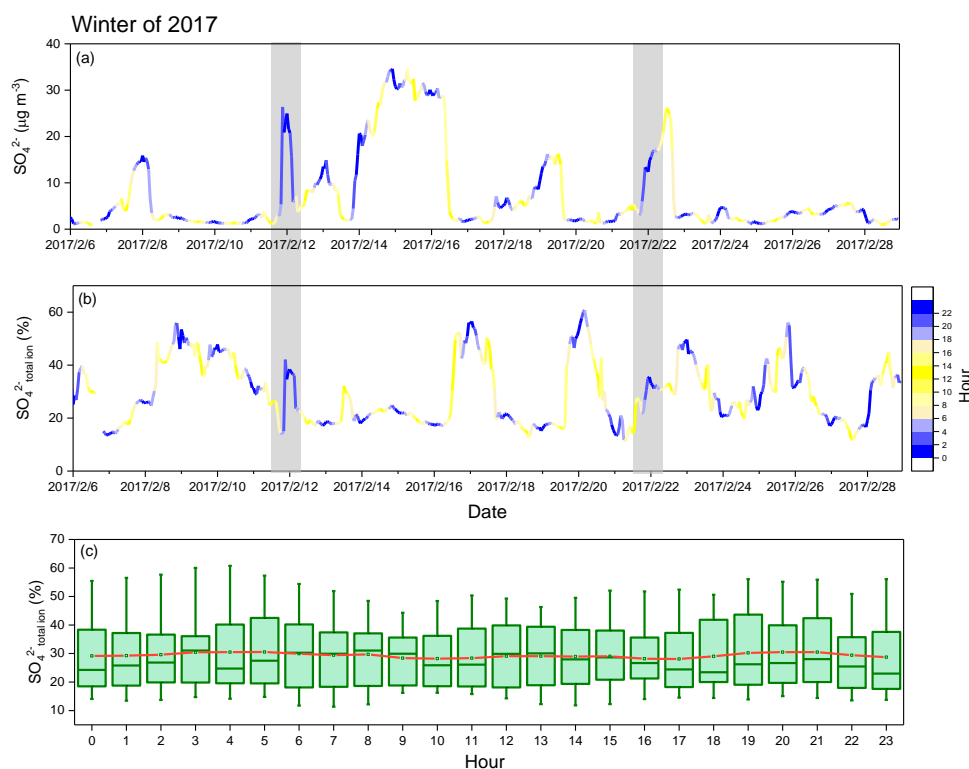


Figure R2. Time series of SO_4^{2-} mass concentration (a) and SO_4^{2-} mass fraction in total ions (b) as well as the diurnal variation of SO_4^{2-} mass fraction in total ions in winter.

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

Response: Thank you for your question. According to the record of literature (Guo et al., 2015), H_{air}^+ diurnal variation was less significant while the ALWC diurnal variation was significant, hence the diurnal pattern in pH was mainly driven by particle water dilution. However, in this work, we find that both H_{air}^+ and ALWC had significant diurnal variation, and the aerosol acidity variation agreed well with sulfate. In the North China Plain, the $PM_{2.5}$ mass concentration has a wide variation range and the average value was high. For example, in winter, the $PM_{2.5}$ mass concentration in Beijing was several to dozens times higher than that in the US, which means there are more seeds in the limited water vapor, hence the dilution of aerosol liquid water to H_{air}^+ doesn't work at all, the diurnal variation of aerosol components was more important. Therefore, we think both the higher concentrations and the greater variability in concentrations have important effects on the difference between the US and Beijing.

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.

Response: The statements here indeed have some problems. In the revised manuscript, we try to give the impact of NH_4^T on aerosol pH with another explanation. Elevated NH_4^T could reduce H_{air}^+ exponentially and slightly increase ALWC when the other input parameters were held constant, leading to the decrease of aerosol pH. As the NH_4^T increases, H_{air}^+ are consumed swiftly during the dissolution of NH_3 as well as the further reaction with SO_4^{2-} , NO_3^- , and Cl^- . And the elevated NH_4^T increases the $\epsilon(NO_3^-)$ and $\epsilon(Cl^-)$ when NO_3^T and Cl^T were fixed (Figure 10), which means the elevated NH_4^T alter the gas-particle partition and shifts more NO_3^T and Cl^T into particle phase, and the deliquescence of additional nitrate and chloride increased ALWC slightly.

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

ammonia.

Response: We are afraid that the reviewer misunderstood what we meant. Here we want to deliver that although the ammonia in the atmosphere is excessive, the other conditions are limited, the ALWC is one of them. Compared to the liquid water content in clouds and precipitation, ALWC is much lower, hence the dilution of aerosol liquid water to H_{air}^+ is much weaker. In the revised manuscript, we reword Line 376-388 to more clearly express our point.

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

Response: Thanks for your careful check, the caption to Table 2 has been clarified as below:

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

Response: Color scale in figures has been unified.

14. Figure 5, 6, 7, caption. These figures appear to be the sensitivity of ALWC, H^+ , and pH to chemical components. Reword caption.

Response: Thanks for your careful check, captions to Figure 5, 6, 7 (7-9 in the revised manuscript) have been clarified as below:

Figure 7. Sensitivities of H_{air}^+ to SO_4^{2-} , NO_3^T , NH_4^T , Cl^T , as well as meteorological parameters (RH, T) in summer and winter.

Figure 8. Sensitivities of ALWC to SO_4^{2-} , NO_3^T , NH_4^T , Cl^T , as well as meteorological parameters (RH, T) in summer and winter.

Figure 9. Sensitivities of aerosol pH to SO_4^{2-} , NO_3^T , NH_4^T , Cl^T , as well as meteorological parameters (RH, T) in summer and winter.

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

Response: In this work, the water-soluble ions of $PM_{2.5}$ samples and MOUDI samples were not collected synchronously. Water-soluble ions (SO_4^{2-} , NO_3^- , Cl^- , NH_4^+ , Na^+ , K^+ , Mg^{2+} , Ca^{2+}) of $PM_{2.5}$ and trace gases (HCl, HNO_3 , HNO_2 , SO_2 , NH_3) in the ambient air were measured by an online analyzer (MARGA) at hourly temporal resolution during the spring (April and May in 2016), winter (February in 2017), summer (July and August in 2017) and autumn (September and October in 2017). While the size-resolved sampling was conducted during July 12-18, 2013; January 13-19,

2014; July 3-5, 2014; October 9-20, 2014; and January 26-28, 2015. Compared to the real-time PM_{2.5} sampling, MOUDI sampling time is short, which is not conducive to analyze the variation tendency of aerosol composition and acidity in time. MOUDI samples were mainly used to analyze the change of aerosol composition and acidity in different particle size.

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

Response: The English in the manuscript has been improved by an English native speaker.

References

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