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Nitrate-dominated $PM_{2.5}$ and elevation of particle pH observed in urban Beijing during the winter of 2017

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Supplementary Materials

Table.S1 Literature list of the history data exhibition

Label	Sample time	Data Type	Reference
1999	1999-12-01 to 2000-02-01	Averaged	He et al., 2001
2011	2011-12-01 to 2012-01-30	Averaged	Cheng et al., 2013
2013	2013-01-5 to 2013-01-24	Pollution	Huang et al., 2014a
2015-p	2015-01-21 to 2015-02-04	Pollution.PM_1	Wang et al., 2016
2015-a	2015-01-01 to 2015-02-28 2015-12-01 to 2015-12-31	Averaged	Liu et al., 2019
2016	2016-12-15 to 2017-01-14	Averaged	Shao et al., 2018
2017-a	2017-12-15 to 2017-02-25	Averaged	This Study
2017-p	2017-12-15 to 2017-02-25	Pollution	This Study

Filter Comparisons

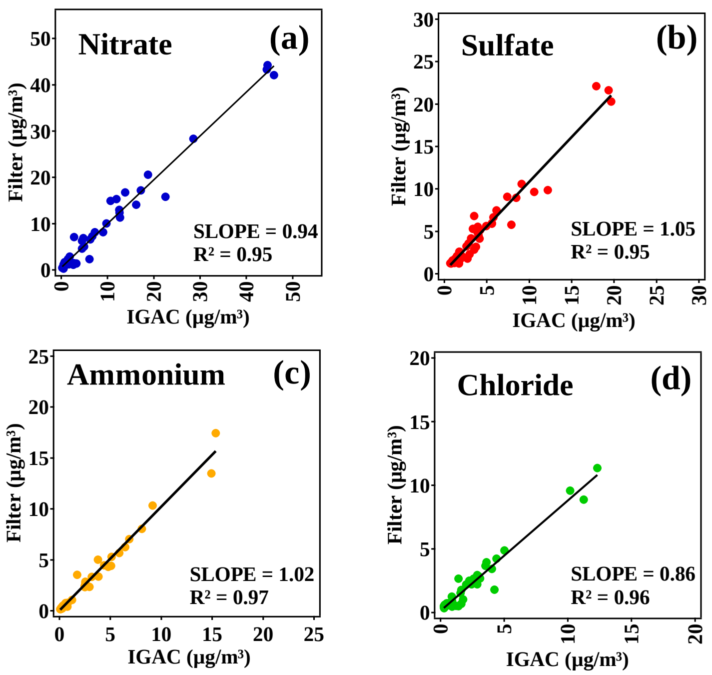

Fig.S1 Comparison of major ions in $PM_{2.5}$ between filter sampling and IGAC measurements during the campaign.

Effect of crustal ions on particle pH prediction

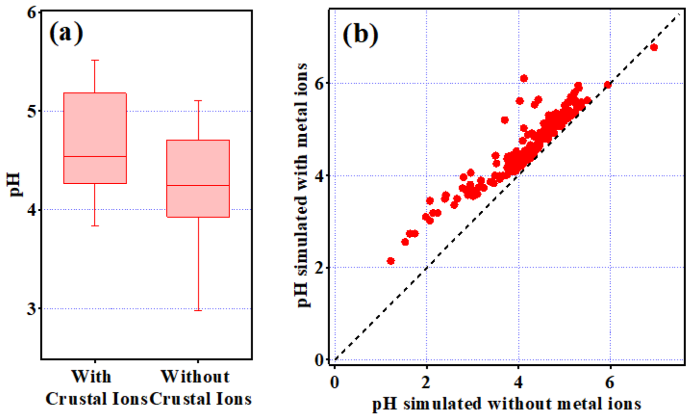

Fig.S2 Comparison of particle pH simulation with/without Crustal Ions: (a) pH level comparison and (b) Scatter Plot of the pH simulated with/without crustal ions.

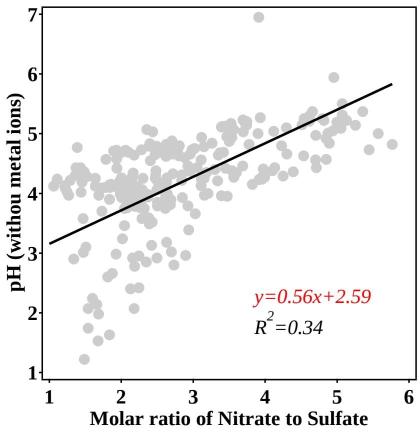

Fig.S3 Scatter plot of pH simulated without crustal ions and the Ratio $_{\text{N-to-S}}$

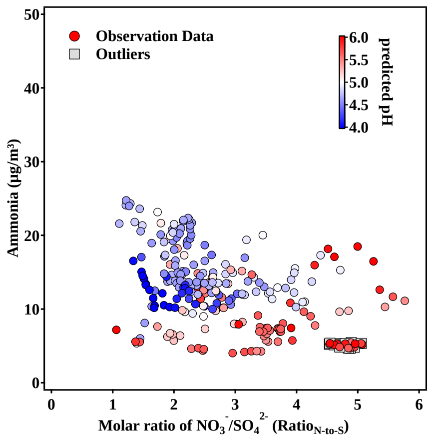

FIG. S4 Scatter plot of NH_3 vs. the Ratio$_{\text{N-to-S}}$ colored with predicted pH. Only the data corresponding to the pollution categories and with sufficient aerosol liquid water (above 5 μg m^{-3}) is chosen and the data during Chinese New Year is excluded. Note that the grey frame depicts the outliers which have lower F_{NH4} and lower Ammonia concentration.

Sensitivity result of controlled sulfate transferring

In order to provide more evidence that nitrate-dominated particles could increase particle pH compared to sulfate-dominated particles, a sensitivity test by replacing the sulfate with equivalent nitrate (e.g. 1 mole of sulfate would be transferred to 2 moles of nitrate) was conducted with an increment of 10% of sulfate replaced (Fig.S5). The test results were limited to the same criteria as presented in the main article (ALWC more than 5μg/m^3 and during the pollution period). A forward model run with metastable assumption was kept the same as the simulations in the manuscript. Comparison between sulfate-transferred results vs. original results shows significant increase of pH values due to the replacement of sulfateby nitrate. The elevation was less noticeable when the transfer fraction was around below 20%. Then, the increase outstands when the transfer fraction gets higher. When the fraction ranges in 30% to 80%, the predicted pH increase showed a double-end pattern: the data with high RatioN-to-S (above 3) and high pH (between 5.6 to 6.4) as well as lower RatioN-to-S (around 1.5) and lower pH (below 4) exhibit greater elevation of pH. As the transfer fraction exceeds 80%, the pH increase was suppressed, i.e. there is no further elevation of pH. While the overall pH simulated with transferred input was higher than original data, the "double-end" phenomenon disappeared as these two clusters' pH decreased compare to the transferring fraction of 80%.

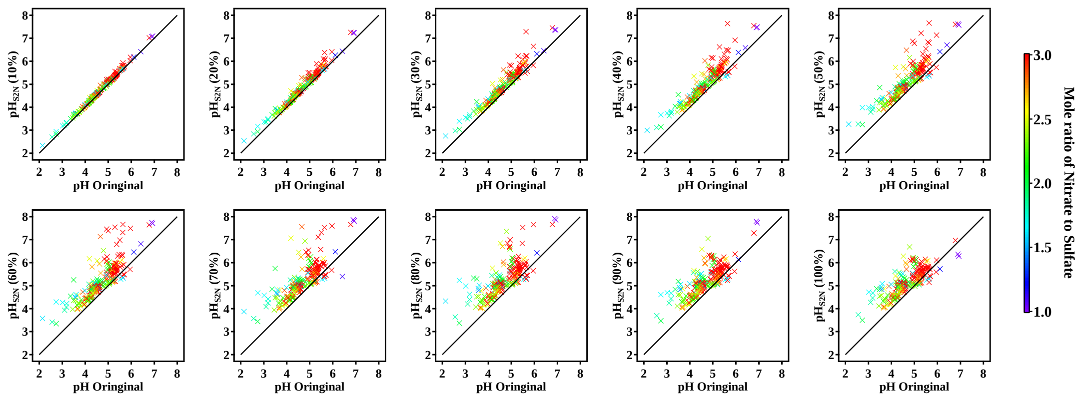

FIG.S5 Scatter plot of sensitivity test results versus original simulated pH using observation data. Data points are colored by the ratio of nitrate to sulfate. Black lines in each scatter plot are 1:1 line as a reference. The transfer fraction was labeled on Y axis with the abbreviation of "S2N XX%"

Correlation between ALWC, Temperature and Ratio$_{N\text{-to-}S}$

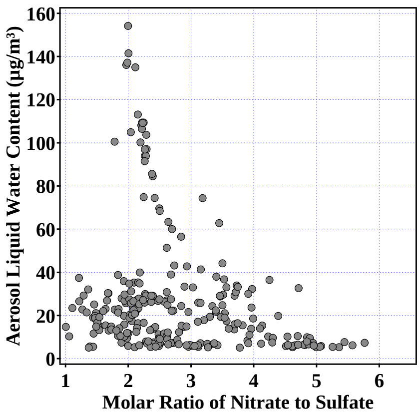

FIG. S6 Scatter plot of ALWC vs. the Ratio$_{N\text{-to-}S}$. Only the data corresponding to the pollution categories and with sufficient aerosol liquid water (above 5 μg m^{-3}) is chosen and the data during Chinese New Year is excluded.

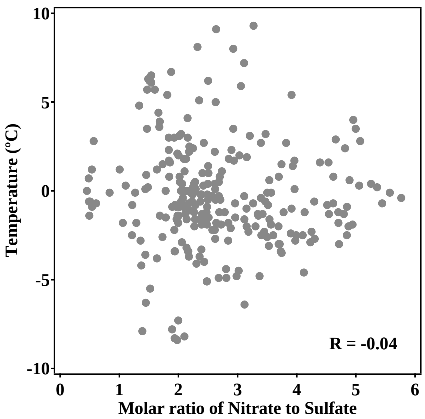

FIG. S7 Scatter plot of Temperature vs. the Ratio$_{N\text{-to-}S}$. Only the data corresponding to the pollution categories and with sufficient aerosol liquid water (above 5 μg m^{-3}) is chosen and the data during Chinese New Year is excluded.

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