The authors thank the editor and reviewers to review our manuscript and particularly for the valuable comments and suggestions that are very helpful in improving the manuscript. We provide below point-by-point responses to the reviewers' comments. We also have made most of the changes suggested by the reviewers in the revised manuscript.

Reviewer: 1

This study investigated the key factors to aerosol pH variability based on machine learning with SHAP. Especially, they found the regime-dependent influence of RH due to interactions with other factors, which is less investigated before. In general, this study is based on promising new method, and found interesting results. However, the breadth and depth of this study needs to be improved before it can be accepted.

Response: We sincerely thank the reviewer for the positive assessment and constructive suggestions. To address the concerns about breadth and depth, we applied the same analytical framework to other cities for comparative analysis. In the revised manuscript, we have strengthened the discussion by explicitly acknowledging the site-specific nature of the regime-dependent behavior and thresholds, and by incorporating more in-depth explanations for the observed interactions. Detailed responses to these two aspects are provided below.

(1) How will the threshold of the regimes vary with sites? There're many public available datasets for aerosol acidity calculations (e.g., https://doi.org/10.5194/acp-19-9309-2019), and such investigations should be easy with established methods.

Response: We thank the reviewer for this important comment. Following the reviewer's suggestion, we applied the same analytical framework to the eastern Canadian datasets from Tao and Murphy (2019) (https://doi.org/10.5194/acp-19-9309-2019). The results revealed that the specific interactions and regime thresholds identified in Beijing were not fully reproduced at the Canadian sites. For example, RH at the Canadian sites is predominantly higher than 50%, and the interaction between RH and temperature is less pronounced than in Beijing, with a

lower threshold separating temperature regimes (around 10 °C). The threshold value for RH–PM_1 SHAP interaction (approximately 2.5 μg m^{-3}) is also much lower than that in Beijing, while no obvious interaction between RH and the N/S mass ratio was observed (Fig. R1). These differences are likely associated with regional variations in chemical properties, emission patterns, and meteorological conditions.

Nevertheless, the primary objective of this study is to provide a quantitative and interpretable framework for understanding how multiple environmental factors jointly contribute to aerosol pH variation. This framework can be readily applied to other datasets to explore regime-dependent pH variations in diverse atmospheric environments. We appreciate the reviewer's suggestion and agree that a systematic multi-site and long-term analysis represents an important direction for future work to further understand the regime-dependent pH variations and underlying mechanisms.

To address this concern, we have explicitly acknowledged the site-specific nature of the regime-dependent behavior and thresholds, as well as the limitations of our study, in the revised manuscript. The relevant paragraph (lines 413–432) now reads as:

"The thresholds and interaction patterns identified here are based on observations at a single urban site in Beijing and may therefore reflect local meteorology and emission characteristics. Nevertheless, this approach provides a quantitative framework for identifying the nonlinear relationships between pH and complex coupled environmental parameters. In China's evolving atmosphere, meteorological factors, together with variable particle properties, may jointly modulate aerosol acidity. …..Extending this SHAP-based analytical framework to diverse atmospheric conditions would be a valuable direction for future research to further understand the regime-dependent pH variations across different environments".

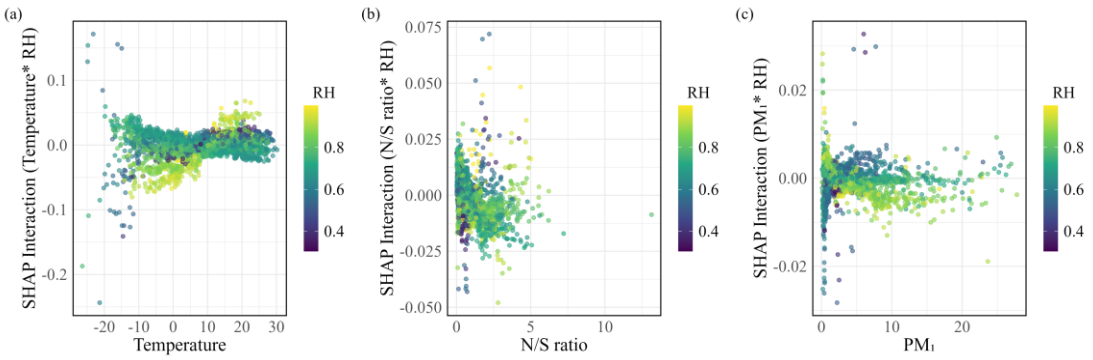

Fig. R1 SHAP interaction plots showing the joint contributions of RH with temperature (°C) (a), N/S mass ratio (b), and PM_1 mass loading (μg m^{-3}) (c) to aerosol pH predictions. The color scale represents RH, and the SHAP interaction values indicate the direction and magnitude of the interactive contributions to pH across different regimes. The SHAP analysis followed the same procedure as for the main Beijing dataset. Aerosol pH in this dataset was calculated using the E-AIM II model. As PM mass loading was not available, the sum of particulate Na^+, K^+, Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-}, NO_3^-, and NH_4^+ was used as a proxy for aerosol loading.

Reference:

Tao, Y. and Murphy, J. G.: The sensitivity of $PM_{2.5}$ acidity to meteorological parameters and chemical composition changes: 10-year records from six Canadian monitoring sites, Atmos. Chem. Phys., 19, 9309–9320, 10.5194/acp-19-9309-2019, 2019.

(2) Accordingly, more in-depth explanation of the regime-dependent interactions should be applied. Current explanations are too general, and the conclusions and patterns can hardly be confidentially applied to other scenarios. At least, whether and why it applies to all ammonia-Rich urban atmosphere, as outlined in the title, should be discussed and investigated in more detail.

Response: We thank the reviewer for this constructive suggestion. We agree that more in-depth interpretation of the regime-dependent interactions is essential. In response, we have expanded the discussion by considering gas–particle partitioning of semi-volatile species, heterogeneous reactions of gaseous precursors and secondary formation of inorganic species. These processes jointly modulate the competition between H^+ accumulation and aerosol liquid water dilution—

two key factors regulating aerosol pH—and likely underlie the identified regime-dependent interactions. We also acknowledge that the interaction patterns are based on a single urban site and may therefore reflect local meteorological and emission characteristics. Nevertheless, our primary contribution is a quantitative and interpretable analytical framework that can be readily applied to other scenarios, as also discussed in our response to Comment (1). These clarifications have been incorporated into the revised title, abstract, and conclusion.

In the revised manuscript, the discussion of the regime-dependent interactions now reads as follows (lines 295–373):

"This regime-dependent association behavior identified by SHAP model is consistent with the coupled influence of temperature and RH on gas–particle partitioning, aerosol water uptake, and secondary inorganic formation, which together may modulate the balance between H^+ input and dilution—two key factors related to different pH variations (Ding et al., 2019; Tao et al., 2025). According to previous studies, under low-temperature conditions, ……. Elevated RH could not only enhance aerosol water uptake but also facilitate heterogeneous reactions of gaseous precursors to form secondary inorganic aerosols, accompanied by H^+ accumulation likely primary from the dissociation of HNO_3 upon aqueous uptake and the multiphase oxidation of SO_2 (Wang et al., 2016; Liu et al., 2017). ……In contrast, at higher temperatures, elevated RH still enhances aerosol water uptake. However, enhanced volatilization of semi-volatile species shifts gas–particle partitioning towards the gas phase, likely reducing the availability of these precursors for aqueous-phase reactions and thereby limiting H^+ generation in the particle phase. As a result, the increased aerosol water at high RH predominantly exerts a dilution effect (Ding et al., 2019; Tao et al., 2025). Consistently, ……

Strong statistical interactions were also observed between RH and chemical parameters,……These patterns are consistent with the complex multiphase chemical processes among different regimes (Wang et al., 2016; Xie et al., 2020; Wang et al., 2025). Under cleaner conditions with low PM_1 loading, elevated RH is expected to substantially enhance ALWC. At the same time, secondary inorganic aerosol formation might be limited by the low availability of precursor gases. As a result, the additional aerosol water may act mainly through dilution,

which is likely associated with higher pH. In contrast, under high-PM_1 conditions, the abundant particle surface area and elevated ionic strength may create favorable media for multiphase reactions (Mekic et al., 2021). The enhanced water uptake at elevated RH likely facilitates heterogeneous/aqueous-phase formation of secondary inorganic aerosols (Wang et al., 2025), thereby promoting additional acidity production that may offset or outweigh the water dilution effect (Fig.5). Regarding the RH–N/S interaction, ……This pattern likely suggests that under this regime, the efficient aqueous medium is likely more favorable for sulfate production (e.g., through multiphase oxidation of SO_2 and NOx) (Wang et al., 2016). This process likely introduces additional acidity to the particle phase and is likely associated with a decrease in pH. ……".

To more accurately reflect the scope of this study, the title has been revised as: "Quantitative insights into regime-dependent aerosol pH variability in ammonia-rich urban Beijing from explainable machine learning".

A corresponding clarification has also been added to the abstract:

"……Here, we integrate machine learning with interpretable model analyses, field observations, and thermodynamic modeling to quantitatively assess the relative contributions and associations of key factors to aerosol pH variability in the ammonia-rich urban atmosphere of Beijing".

The limitations of our study are explicitly stated in the revised conclusion (as detailed in our response to Comment 1): "The thresholds and interaction patterns identified here are based on observations at a single urban site in Beijing……would be a valuable direction for future research to further understand the regime-dependent pH variations across different environments".

Reference:

Ding, J., Zhao, P., Su, J., Dong, Q., Du, X., and Zhang, Y.: Aerosol pH and its driving factors in Beijing, Atmos. Chem. Phys., 19, 7939-7954, 10.5194/acp-19-7939-2019, 2019.

Liu, M., Song, Y., Zhou, T., Xu, Z., Yan, C., Zheng, M., Wu, Z., Hu, M., Wu, Y., and Zhu, T.: Fine particle pH during severe haze episodes in northern China, Geophys. Res. Lett., 44,

5213-5221, 10.1002/2017gl073210, 2017.

Mekic, M. and Gligorovski, S.: Ionic strength effects on heterogeneous and multiphase chemistry: Clouds versus aerosol particles, Atmos. Environ., 244, 117911, 10.1016/j.atmosenv.2020.117911, 2021.

Tao, J., Wu, Y., Deng, Y., Hu, B., and Zhou, Z.: Thermodynamic regulation of aerosol pH by temperature and humidity: The role of gas-particle partitioning of semi-volatile inorganic species in a subtropical region, Environ. Res., 286, 122727, 10.1016/j.envres.2025.122727, 2025.

Wang, G., Zhang, R., Gomez, M. E., Yang, L., Levy Zamora, M., Hu, M., Lin, Y., Peng, J., Guo, S., Meng, J., Li, J., Cheng, C., Hu, T., Ren, Y., Wang, Y., Gao, J., Cao, J., An, Z., Zhou, W., Li, G., Wang, J., Tian, P., Marrero-Ortiz, W., Secrest, J., Du, Z., Zheng, J., Shang, D., Zeng, L., Shao, M., Wang, W., Huang, Y., Wang, Y., Zhu, Y., Li, Y., Hu, J., Pan, B., Cai, L., Cheng, Y., Ji, Y., Zhang, F., Rosenfeld, D., Liss, P. S., Duce, R. A., Kolb, C. E., and Molina, M. J.: Persistent sulfate formation from London Fog to Chinese haze, Proc Natl Acad Sci U S A, 113, 13630-13635, 10.1073/pnas.1616540113, 2016.

Wang, J., Wu, Z., Qiu, T., Man, R., Zong, T., Qiu, Y., Fang, W., Chen, S., Liang, D., Song, M., Ahn, J., Lee, J., and Hu, M.: Impacts of aerosol acidity and liquid water content on secondary inorganic aerosol pollution in East Asian megacities: Beijing and Seoul, Atmos. Environ., 361, 10.1016/j.atmosenv.2025.121479, 2025.

Xie, Y., Wang, G., Wang, X., Chen, J., Chen, Y., Tang, G., Wang, L., Ge, S., Xue, G., Wang, Y., and Gao, J.: Nitrate-dominated PM2.5 and elevation of particle pH observed in urban Beijing during the winter of 2017, Atmos. Chem. Phys., 20, 5019-5033, 10.5194/acp-20-5019-2020, 2020.

Reviewer: 2

I appreciate the authors' efforts in addressing the important issue of aerosol pH variability in an ammonia-rich urban atmosphere. The manuscript combines field observations, thermodynamic modeling, and an explainable machine-learning framework to examine how meteorological and chemical factors are associated with aerosol pH. The topic is relevant, and the regime-dependent RH effect is potentially interesting. However, I think the manuscript requires substantial revision before the conclusions can be fully supported.

Response: We sincerely thank the reviewer for the positive and constructive assessment of our work. We fully agree that substantial revisions are necessary to strengthen the conclusions. Below we provide point-by-point responses to all comments and have made corresponding revisions accordingly. We greatly appreciate the reviewer's valuable suggestions, which have significantly improved the quality of our manuscript.

Overall comment

The central concern is that the scientific role of the XGBoost-SHAP framework is not sufficiently justified. In this study, aerosol pH is not directly measured but calculated using ISORROPIA II, and the ML model is subsequently trained to reproduce this ISORROPIA-derived pH using variables that are either direct inputs to the thermodynamic calculation or closely related derived indicators, including Excess NHx, N/S ratio, PM1 mass loading, temperature, and RH. As a result, the SHAP analysis mainly characterizes how the thermodynamic-model-derived pH responds to these variables under the observed covariance structure of the dataset. This can still be useful, but its added value beyond conventional thermodynamic sensitivity analysis needs to be clearly articulated. In particular, the authors should clarify whether the main contribution of the XGBoost-SHAP approach is to diagnose nonlinear pH responses under real-world co-varying conditions, rather than to identify independent causal mechanisms. The manuscript should therefore more carefully distinguish thermodynamic-model behavior, statistical associations in the observational dataset, and actual

atmospheric chemical processes. Some current interpretations appear to overstate SHAP-based associations as mechanistic evidence, and these should be revised accordingly.

Response： We sincerely thank the reviewer for this critical comment. We fully agree that the scientific role of the XGBoost-SHAP framework—particularly its added value beyond conventional thermodynamic sensitivity analysis—needs to be more clearly articulated. Conventional thermodynamic sensitivity analyses typically vary one input parameter at a time while holding others at average conditions, or prescribe two-variable gradients to examine pairwise effects (Guo et al., 2017; Ding et al., 2019; Xie et al., 2020; Wei et al., 2023). These approaches are valuable for understanding thermodynamic model behavior under idealized, decoupled conditions, but they cannot systematically quantify how multiple factors contribute to pH variation under the complex, co-varying conditions of the real atmosphere—where temperature, RH, chemical composition, and aerosol loading vary simultaneously and interact nonlinearly. The added value of the XGBoost-SHAP framework lies in its ability to diagnose nonlinear and interactive pH responses from observational data in a sample-level, data-driven manner. Specifically, it characterizes sample-level heterogeneity, quantifies the relative importance of different factors, and identifies regime-dependent behaviors, providing complementary insights into pH variation under real-world co-varying conditions.

We clarify that the XGBoost–SHAP approach serves primarily as a statistical and interpretable tool. Its main contribution is to diagnose nonlinear pH responses under real-world co-varying conditions and to provide a data-driven decomposition of predictor contributions to pH, rather than to identify independent causal mechanisms. Accordingly, following the reviewer's suggestion, we have more carefully distinguished thermodynamic-model behavior, statistical associations, and atmospheric chemical processes throughout the revised manuscript. Causal language that may have overstated SHAP-based associations as mechanistic evidence has been reframed to present these results as statistical associations consistent with known atmospheric chemistry.

In the revised manuscript, a clarification of the XGBoost-SHAP framework has been added in Section 2.3 (lines 182-191):

"Unlike conventional thermodynamic sensitivity analyses that typically vary one factor at a time (or examine pairwise effects) under fixed conditions (Guo et al., 2017; Ding et al., 2019; Xie et al., 2019; Wei et al., 2023), the XGBoost-SHAP framework diagnoses nonlinear and interactive pH responses from observational data in a sample-level, data-driven manner. It characterizes sample-level heterogeneity, quantifies the relative importance of different factors, and identifies regime-dependent behaviors, providing complementary insights into pH variation under the complex, co-varying conditions of the real atmosphere. We emphasize that the XGBoost–SHAP approach serves primarily as a statistical and interpretable tool, providing a data-driven decomposition of how each predictor contributes to pH variations, rather than identifying independent causal mechanisms".

In addition, causal language in SHAP-result interpretation has been carefully revised throughout the manuscript. For example:

Line 295: "This regime-dependent behavior can be explained by ……" has been reframed to "This regime-dependent association behavior identified by SHAP model is consistent with……".

Line 309: "That means……" has been reframed to "This likely suggests that……".

Line 311: "……resulting in increased aerosol acidity……" has been reframed to "which might be associated with increased aerosol acidity".

Lines 322-324: "Temperature therefore modulates ……, producing ……"has been reframed to "Temperature therefore likely modulates ……, which may be associated with……".

Lines 340-341: "……, such that dilution by aerosol water likely outweighed, resulting in elevated pH……" has been reframed to "……, suggesting that dilution by aerosol water likely outweighed H^+ input, which might be associated with elevated pH……".

Line 346: "These might be related to……" has been reframed to "These patterns are consistent with……".

Line 368: "That means under this regime, ……" has been reframed to "This likely suggests that……".

Line 371: "……and leads to a decrease in pH again……" has been reframed to "……and is likely associated with the decrease in pH".

Line 382: “RH generally showed negative contribution to pH in winter……” has been reframed to “RH generally showed negative association with pH in winter……”.

Line 393: “……reinforcing the plausibility of the interaction patterns identified here” has been reframed to “……supporting the consistency of the interaction patterns identified here”.

The figure caption for Figure 3: “SHAP interaction plots illustrating the joint effects of RH with temperature (℃) (a), N/S mass ratio (b), and PM_1 mass loading (μg m^{-3}) (c) on aerosol pH prediction. The color scale represents RH, and the SHAP interaction values indicate the direction and magnitude of RH modulation on predicted pH under different regimes” has been reframed to “SHAP interaction plots showing the joint contributions of RH with temperature (℃) (a), N/S ratio (b), and PM_1 mass loading (μg m^{-3}) (c) to aerosol pH predictions. The color scale represents RH, and the SHAP interaction values indicate the direction and magnitude of the interactive contributions to pH across different regimes”.

References:

Ding, J., Zhao, P., Su, J., Dong, Q., Du, X., and Zhang, Y.: Aerosol pH and its driving factors in Beijing, Atmos. Chem. Phys., 19, 7939-7954, 10.5194/acp-19-7939-2019, 2019.

Guo, H., Liu, J., Froyd, K. D., Roberts, J. M., Veres, P. R., Hayes, P. L., Jimenez, J. L., Nenes, A., and Weber, R. J.: Fine particle pH and gas–particle phase partitioning of inorganic species in Pasadena, California, during the 2010 CalNex campaign, Atmos. Chem. Phys., 17, 5703-5719, 10.5194/acp-17-5703-2017, 2017.

Wei, Y., Wang, S., Jiang, N., Zhang, R., and Hao, Q.: Comparative multi-model study of PM(2.5) acidity trend changes in ammonia-rich regions in winter: Based on a new ammonia concentration assessment method, J. Hazard. Mater., 458, 131970, 10.1016/j.jhazmat.2023.131970, 2023.

Xie, Y., Wang, G., Wang, X., Chen, J., Chen, Y., Tang, G., Wang, L., Ge, S., Xue, G., Wang, Y., and Gao, J.: Nitrate-dominated PM2.5 and elevation of particle pH observed in urban Beijing during the winter of 2017, Atmos. Chem. Phys., 20, 5019-5033, 10.5194/acp-20-5019-2020, 2020.

Specific comments

1. The model validation strategy should be strengthened. The current random training/testing split may overestimate model performance for continuous time-series observations, because temporally adjacent samples, similar pollution episodes, and data from the same season may be included in both training and testing sets. I suggest adding blocked or time-aware validation, such as leave-one-month-out cross validation, season-based cross-validation, or training in one season and testing in another. This would better show whether the model captures generalizable relationships rather than temporally correlated patterns.

Response: We thank the reviewer for this constructive suggestion. We agree that the conventional random training/testing split may overestimate model performance for continuous time-series observations. Following the reviewer's suggestion, we adopted leave-one-month-out cross-validation, which preserves temporal order and prevents data leakage between adjacent samples. As shown in Table R1, model performance decreased under this time-aware validation compared with our primary model—an expected outcome given that the model is tested on temporally independent data, consistent with previous studies (Huang et al., 2019; Chen et al., 2021). Nevertheless, the average leave-one-month-out R^2 of 0.86 ± 0.13 (range: 0.63–0.98) suggests that the model maintains good predictive performance across independent time periods, supporting its ability to capture generalizable relationships rather than temporally correlated patterns.

In the revised manuscript (Section 2.3, lines 161-166), we have added the leave-one-month-out cross-validation as follows:

"To evaluate the temporal generalizability of the model, we additionally performed leave-one-month-out cross-validation, in which each month was iteratively held out as the test set while the remaining months were used for training (Bergmeir and Benítez, 2012; Chen et al., 2021). This validation yielded an average R^2 of 0.86 ± 0.13 (Table S1), suggesting that the model maintains good predictive performance across independent time periods and captures generalizable relationships rather than temporally correlated patterns".

Table R1 has been added to the revised supporting information as Table S1.

Table R1 Monthly performance metrics (R^2, RMSE, MAE) of the XGBoost model from leave-one-month-out cross-validation.

Month	R^2	RMSE	MAE
Dec-14	0.64	0.23	0.15
Jan-15	0.63	0.19	0.16
Feb-15	0.68	0.23	0.17
Mar-15	0.90	0.21	0.15
Apr-15	0.94	0.17	0.12
May-15	0.97	0.13	0.08
Jun-15	0.98	0.10	0.07
Jul-15	0.97	0.10	0.06
Aug-15	0.96	0.13	0.10
Sep-15	0.94	0.19	0.11
Oct-15	0.88	0.19	0.12
Nov-15	0.90	0.11	0.08
Dec-15	0.74	0.19	0.15
Average	**0.86±0.13**	**0.17±0.05**	**0.12±0.04**

References:

Bergmeir, C. and Benítez, J. M.: On the use of cross-validation for time series predictor evaluation, Inform. Sciences, 191, 192–213, 10.1016/j.ins.2011.12.028, 2012.

Chen, C.-C., Wang, Y.-R., Yeh, H.-Y., Lin, T.-H., Huang, C.-S., and Wu, C.-F.: Estimating monthly $PM_{2.5}$ concentrations from satellite remote sensing data, meteorological variables, and land use data using ensemble statistical modeling and a random forest approach, Environ. Pollut., 291, 118114, 10.1016/j.envpol.2021.118114, 2021.

Huang, C.-S., Lin, T.-H., Hung, H., Kuo, C.-P., Ho, C.-C., Guo, Y.-L., Chen, K.-C., and Wu,

C.-F.: Incorporating satellite-derived data with annual and monthly land use regression models for estimating spatial distribution of air pollution, Environ. Modell. Softw., 114, 181–187, 10.1016/j.envsoft.2019.01.017, 2019.

2. The very high model performance should be interpreted cautiously. Since the target pH is calculated by ISORROPIA II and the predictors are closely related to the thermodynamic inputs, a high R2 is expected. The manuscript should avoid presenting the high R2 as strong evidence that the ML framework independently captures atmospheric processes.

Response: We thank the reviewer for this critical comment. We fully agree that the high prediction performance (e.g., R^2) should be interpreted as the ML model successfully capturing the nonlinear relationships between the predictors and pH as derived from the thermodynamic simulations, serving as a reliable basis for the subsequent SHAP-based interpretation—rather than as evidence that the ML framework independently captures atmospheric processes.

To more accurately clarify this, the relevant sentence in the revised manuscript (Section 2.3, lines 156–161) now reads as:

"Model performance was evaluated using the root mean square error (RMSE, 0.12), mean absolute error (MAE, 0.08), and coefficient of determination (R^2 = 0.98; Fig. S2). This high performance suggests that the model has adequately learned the dominant relationships embedded in the thermodynamic behavior and providing a reliable basis for the subsequent SHAP-based interpretation".

3. The dependence among input features should be evaluated, and the robustness of the SHAP results to feature selection should be tested. Excess NHx, N/S ratio, and PM1 mass loading are not independent variables. Excess NHx is derived from ammonia/ammonium and acidic species; N/S ratio is derived from nitrate and sulfate; and PM1 mass loading is partly composed of the same inorganic species involved in the pH calculation. These correlations may influence SHAP importance rankings and interaction values. The authors should provide a correlation matrix and test whether the main conclusions remain robust when alternative feature sets are used, for

example by removing PM1 mass loading, replacing N/S ratio with nitrate and sulfate separately, or excluding strongly derived variables.

Response: We thank the reviewer for this constructive suggestion. We agree that Excess NHx, N/S ratio, and PM_1 mass loading are not strictly independent from a chemical perspective, and such potential correlations may influence SHAP importance rankings and interaction values. To quantitatively assess feature dependence, we calculated the correlation coefficients (R^2) among all input features. As shown in Fig. R2, the correlations among all variable pairs are weak ($R^2 < 0.2$), indicating that multicollinearity is negligible and that the features can be treated as relatively independent predictors in the XGBoost-SHAP analysis.

In addition, to evaluate the robustness of the SHAP results to feature selection, we performed tests using alternative feature sets, as suggested by the reviewer. Specifically, we (1) excluded PM_1 mass loading, and (2) replaced N/S ratio with NO_3^- and SO_4^{2-} as separate features. For each alternative configuration, we retrained the model and recalculated SHAP values. Model performance remained high for both configurations, as expected given the thermodynamic consistency between the target pH and input features (as discussed in response to Comment 2). Quantitatively, removing PM_1 led to a slight decrease in performance, while replacing N/S ratio with NO_3^- and SO_4^{2-} led to a slight increase, likely due to better alignment with the ISORROPIA-II inputs (Table R2).

Regarding SHAP interpretations (Fig. R3 and R4), the key conclusions remained robust across all alternative configurations:

(1) When PM_1 was removed, the SHAP importance rankings of the remaining features were consistent with the primary model—temperature remained the dominant factor, followed by Excess NHx, N/S ratio, and RH. The regime-dependent RH–temperature (threshold ~15 °C) and RH–N/S ratio (threshold ~1.25) interactions were also consistently reproduced.

(2) When N/S ratio was replaced with NO_3^- and SO_4^{2-}, the importance rankings of other features (Temperature, Excess NHx, and PM_1) remained consistent. Nitrate showed a positive contribution to pH while sulfate showed a negative contribution, consistent with the positive SHAP contribution of N/S ratio in the primary model (i.e., higher N/S ratio corresponds to

higher nitrate and lower sulfate). Furthermore, the regime-dependent RH–temperature and RH–PM_1 interactions were consistently reproduced. In addition, we observed opposite regime-dependent patterns for RH vs. nitrate and RH vs. sulfate: higher RH was associated with lower pH under low sulfate/high nitrate conditions, and with higher pH under high sulfate/low nitrate conditions. This is consistent with the RH–N/S ratio interaction pattern observed in the primary model, where adopting N/S ratio likely captures the coupled associations with nitrate and sulfate.

These tests indicate that our main SHAP results and conclusions are robust to the choice of feature sets. We acknowledge that the derived variables (Excess NHx, N/S ratio, and PM_1) were chosen as concise proxies for neutralization capacity, nitrate/sulfate dominance, and pollution level, respectively, providing a more generalized perspective on how aerosol chemical properties are associated with pH rather than focusing on absolute concentrations of individual species.

In the revised manuscript (Section 2.3, lines 145–151), we have added the correlation matrix and clarified the rationale for using these derived variables as input features. The paragraph now reads as:

"The input features included chemical indicators such as excess ammonia (Excess NHx, calculated as described in Text S2), nitrate-to-sulfate mass ratio (N/S ratio), and PM_1 mass loading, together with meteorological parameters (temperature and RH). Aerosol pH calculated using ISORROPIA II served as the target variable. The derived variables (Excess NHx, N/S ratio, and PM_1) were chosen as proxies for neutralization capacity, nitrate/sulfate dominance, and pollution level, respectively, providing a more generalized perspective on how aerosol chemical properties are associated with pH rather than focusing on absolute concentrations of individual species. The correlation matrix (Fig. S3) showed weak correlations among all feature pairs ($R^2 < 0.2$), indicating that multicollinearity is negligible and that the features can be treated as relatively independent predictors in the XGBoost-SHAP analysis".

Fig. R2 has been added to the revised supporting information as Fig. S3.

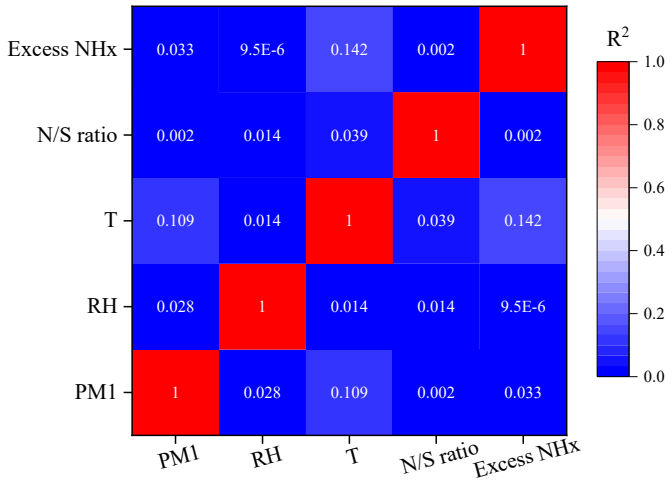

Fig. R2 Correlation matrix of input features. The color scale represents the correlation coefficient (R^2).

Table R2 Comparison of model performance (R^2, RMSE, MAE) for the primary feature set and two alternative configurations.

Performance	R^2	RMSE	MAE
Primary feature set (our study)	0.98	0.12	0.08
Removing PM_1	0.98	0.14	0.09
Replacing N/S ratio with NO_3^- and SO_4^{2-} as separate features	0.99	0.12	0.07

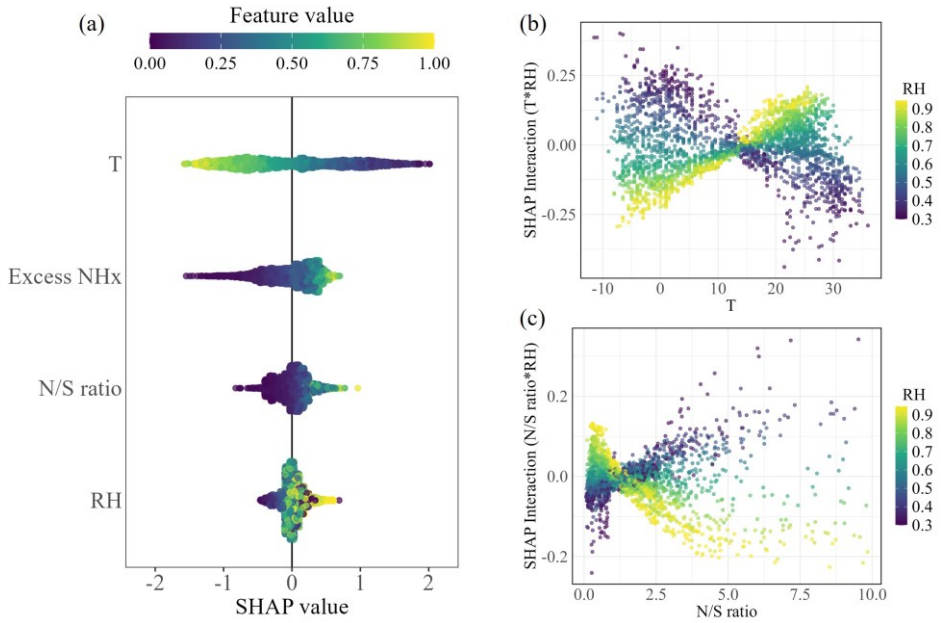

Fig. R3 Global SHAP feature importance rankings (a) and SHAP interaction plots between RH and temperature (T, °C) (b), RH and N/S ratio (c), for the alternative feature set excluding PM_1 mass loading.

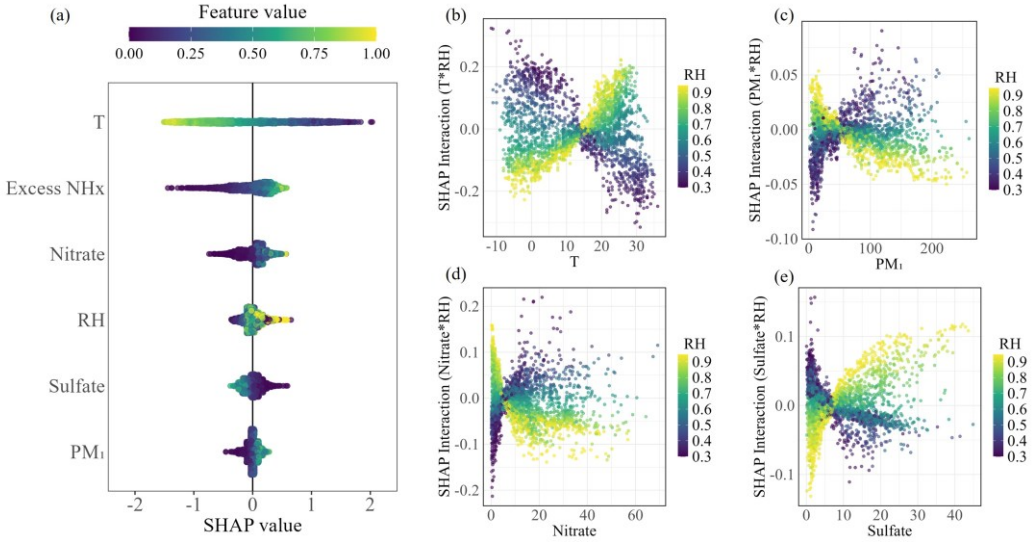

Fig. R4 Global SHAP feature importance rankings (a) and SHAP interaction plots for RH versus temperature (T, °C) (b), RH versus PM_1 (μg m^{-3}) (c), RH versus nitrate (μg m^{-3}) (d), and RH versus sulfate (μg m^{-3}) (e), for the alternative feature set replacing N/S ratio with nitrate and sulfate as separate features.

4. The regime thresholds should be supported more quantitatively. The manuscript identifies transition values around 15 °C, N/S ≈ 1.25, and PM1 ≈ 50 μg m-3. These thresholds appear to be inferred from SHAP interaction plots, but their statistical robustness is unclear. The authors should test whether these values remain stable under different random seeds, train/test splits, bootstrapping, or smoothing methods. If not, they should be described as empirical transition ranges rather than fixed thresholds.

Response: We thank the reviewer for this important suggestion. We agree that the thresholds identified from SHAP interaction plots should be statistically supported. To evaluate their robustness, we retrained the model under different random seeds (1234, 5678, 9012) and train/test split ratio (0.6, 0.7, 0.8). The resulting threshold values remained stable around T≈15 °C, N/S ≈ 1.25, and PM_1 ≈ 50 μg m^{-3} (Fig. R5, R6), suggesting the stability of these approximate thresholds. In addition, we performed bootstrap resampling (n = 100) on the training data, retraining the model and recalculating SHAP interaction values for each bootstrap sample. As shown in Fig. R7, the 100 bootstrap trend lines (gray) consistently clustered around the primary model trend (red) for all three interactions (RH–T, RH–N/S, and RH–PM_1), suggesting overall model stability across different data subsamples. We note that the bootstrap analysis was designed to evaluate overall model stability rather than to pinpoint exact threshold locations, which were more directly assessed through the sensitivity tests under different random seeds and split ratios described above. We acknowledge that no smoothing was applied to the SHAP interaction plots in our analysis. The thresholds were identified visually from the SHAP interaction plots, where the SHAP interaction values crossed zero and the direction of the RH–pH relationship reversed. Given the inherent uncertainty in this visual identification, we describe these values as approximate thresholds (e.g., T ≈ 15 °C, N/S ≈ 1.25, and PM_1 ≈ 50 μg m^{-3}) rather than fixed exact values in the manuscript.

In the revised manuscript, the validation of the statistical robustness of these thresholds has been added as follows (lines 281–285):

"The thresholds were identified from inflection points in the SHAP interaction plots, where the SHAP interaction values crossed zero and the direction of the RH–pH relationship reversed.

The robustness of these approximate thresholds was supported by sensitivity tests under different random seeds and train/test split ratios (Fig. S10 and S11)".

The threshold values have been consistently stated as approximate values throughout the revised manuscript.

Fig. R5 and R6 have been added to the revised supporting information as Figs. S10 and S11, respectively.

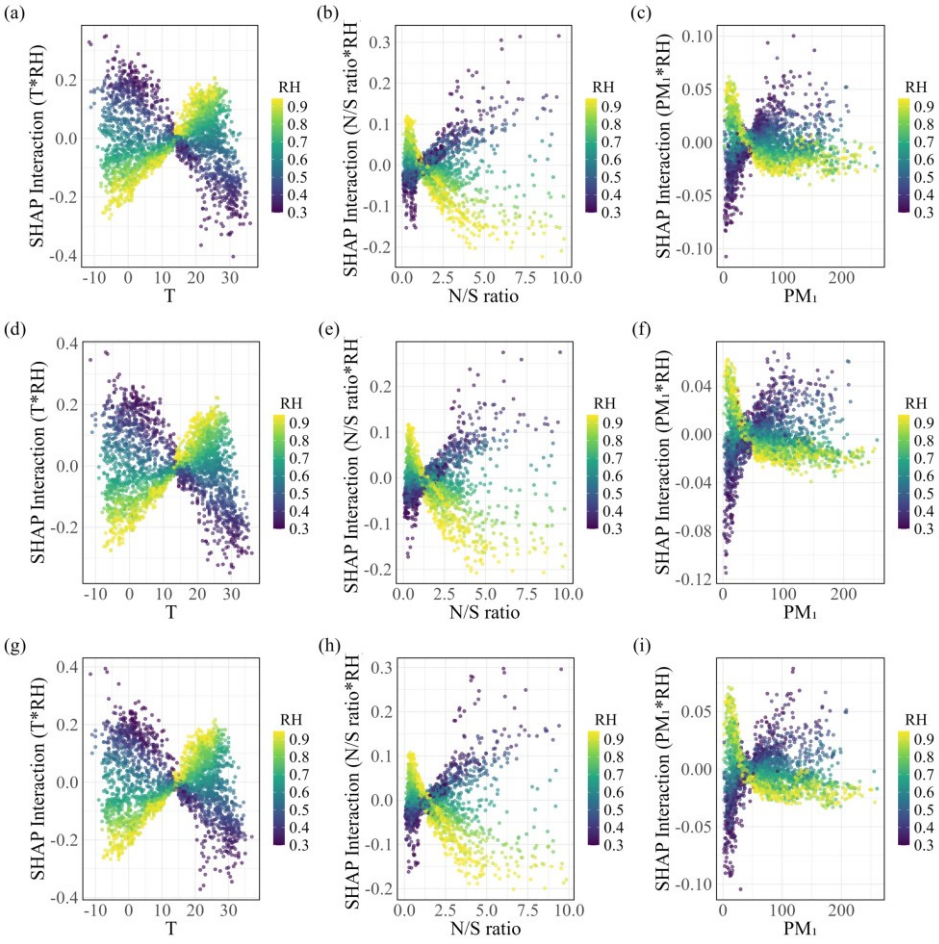

Fig. R5 SHAP interaction plots for RH versus temperature (T, °C), N/S mass ratio, and PM_1 mass loading (μg m^{-3}) under different random seeds of 1234 (a, b, c), 5678 (d, e, f), and 9012 (g, h, i). The color scale represents RH, and the SHAP interaction values indicate the direction and magnitude of the interactive contributions to pH across different regimes.

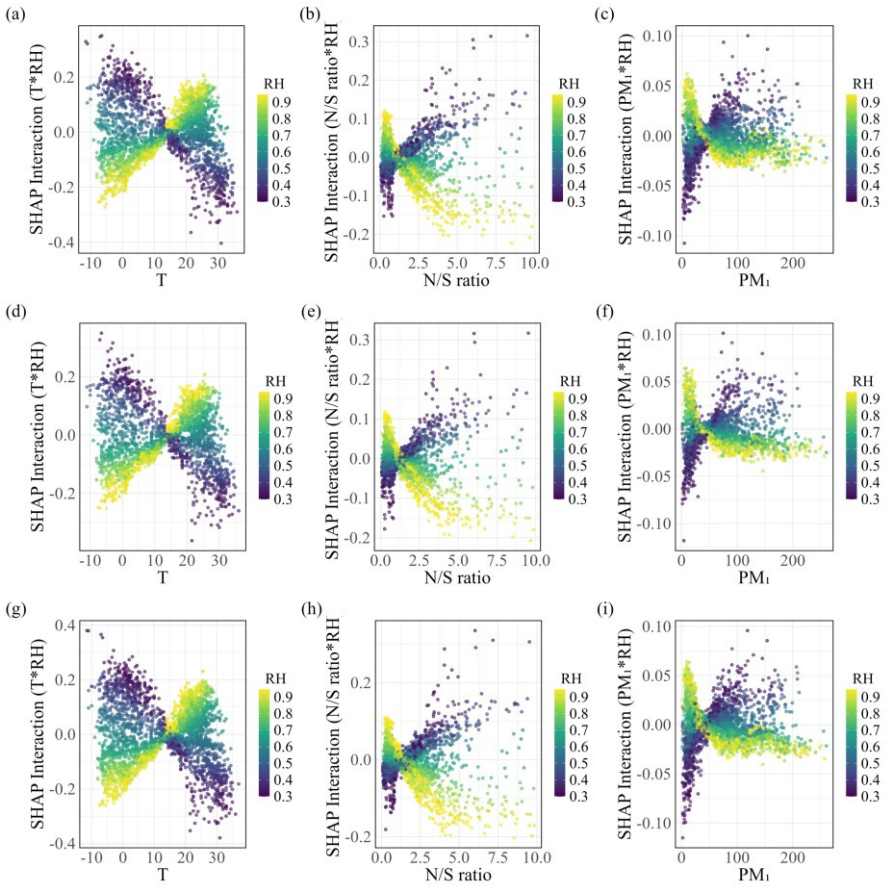

Fig. R6 SHAP interaction plots for RH versus temperature (T, °C), N/S mass ratio, and PM_1 mass loading (μg m^{-3}) under different train/test split ratio of 0.7 (a, b, c), 0.6 (d, e, f), and 0.8 (g, h, i). The color scale represents RH, and the SHAP interaction values indicate the direction and magnitude of the interactive contributions to pH across different regimes.

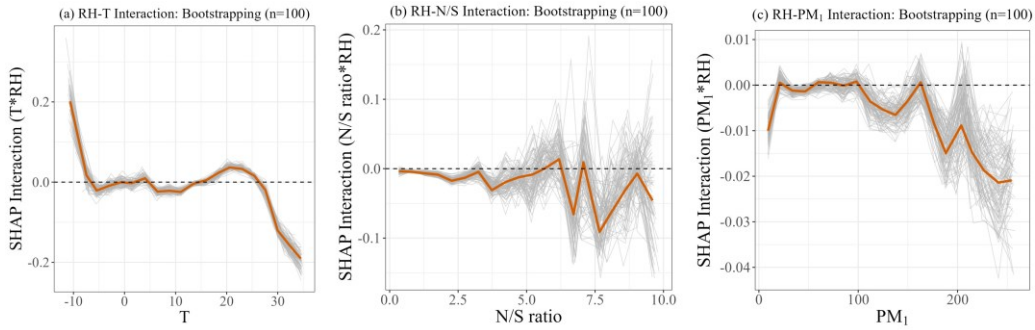

Fig. R7 Bootstrap resampling (n=100) of SHAP interaction patterns for RH versus temperature (T, °C) (a), RH versus N/S ratio (b), and RH versus PM_1 mass loading (μg m^{-3}) (c), Gray lines represent the mean trends from individual bootstrap samples, and red lines represent the mean trend from the primary model.

5. The uncertainty in the pH calculation should be expanded. The assumptions of γH+ = 1, neglect of organic aerosol liquid water, and exclusion of alkaline cations should be discussed more carefully. Since all ML results depend on the ISORROPIA-derived pH, these assumptions may affect the SHAP interpretation.

Response: We thank the reviewer for this valuable suggestion. We agree that the uncertainty in pH calculation should be further clarified. In our study, aerosol pH was estimated using ISORROPIA-II, in which γ_{H+} is set to unity as a simplifying assumption to facilitate computational efficiency in large-scale applications (Fountoukis and Nenes, 2007; Hennigan et al., 2015). This treatment is inherent to the ISORROPIA-II model and has been followed in numerous ISORROPIA-based pH studies (Guo et al., 2015; Song et al., 2018).

Regarding organic aerosol liquid water, given the limited influence of $ALWC_o$ on pH calculation reported in previous studies (Guo et al., 2015; Zhou et al., 2022) and the lack of organic hygroscopicity (κ_{org}) measurements in this study, we primarily used $ALWC_i$ for pH calculation. To assess this assumption, we estimated $ALWC_o$ using a global mean κ_{org} of 0.12 (Pöhlker et al., 2023). The estimated $ALWC_o$ averaged 11.2 ± 18.1 μg m^{-3}, substantially lower than $ALWC_i$ (43.2 ± 75.5 μg m^{-3}). Including $ALWC_o$ changed the pH from 3.82 ± 0.96 to 3.96 ± 0.93 (~4% change; R^2 = 0.99; Fig. R8), suggesting that the influence of $ALWC_o$ on our pH estimates is limited.

Furthermore, we acknowledge that alkaline cations (e.g., Ca^{2+}, Mg^{2+}, Na^+, K^+) were not included in our calculations due to data unavailability, which may lead to a systematic underestimation of aerosol pH, as these species can contribute to acid neutralization (Guo et al., 2017).

In the revised manuscript, the potential uncertainties associated with these assumptions in the ISORROPIA II model have been further clarified and discussed. The revised paragraph in Section 2.2 (lines 102-125) now reads as:

"$$pH = -\mathrm{Log}_{10}\gamma_{H^+}H^+_{aq} = -\mathrm{Log}_{10}\frac{1000\gamma_{H^+}H^+_{air}}{ALWC_i+ALWC_o} \cong -\mathrm{Log}_{10}\frac{1000\gamma_{H^+}H^+_{air}}{ALWC_i}$$ (1)

where γ_{H^+} is the activity coefficient of hydronium ion (assumed to be unity, following the default ISORROPIA-II setting, Fountoukis and Nenes, 2007), H^+_{aq} (mol L^{-1}) is the concentration of hydronium ion in particle liquid water, H^+_{air} (μg m^{-3}) is the mass concentration of hydronium ion in air, and $ALWC_i$ and $ALWC_o$ (μg m^{-3}) is the water concentration of bulk particle related to inorganic and organic species, respectively. Given the limited influence of $ALWC_o$ on pH calculation reported in previous studies (Guo et al., 2015; Zhou et al., 2022) and the lack of organic hygroscopicity (κ_{org}) measurements in this study, we primarily used $ALWC_i$ for pH calculation. To further assess this assumption, we estimated $ALWC_o$ using a global mean κ_{org} of 0.12 (Pöhlker et al., 2023), following the equation (Guo et al., 2015):

$$ALWC_o = \frac{m_{org}\rho_w}{\rho_{org}} \frac{\kappa_{org}}{(\frac{1}{RH}-1)} \quad (2)$$

where m_{org} is the mass concentration of organic aerosol, ρ_w is the density of water (ρ_w = 1.0 g cm^{-3}), ρ_{org} is the density of organics (ρ_{org} = 1.4 g cm^{-3}; Guo et al., 2015).

The estimated $ALWC_o$ averaged 11.2 ± 18.1 μg m^{-3}, substantially lower than $ALWC_i$ (43.2 ± 75.5 μg m^{-3}). When $ALWC_o$ was included in the pH calculations, the resulting pH values showed a strong correlation with those calculated without $ALWC_o$ (R^2 = 0.99; Fig. S1), with the pH increasing slightly from 3.82 ± 0.96 to 3.96 ± 0.93 (~4% change). This suggests that the influence of $ALWC_o$ on the estimated pH in our dataset is relatively limited. It should also be noted that alkaline cations (e.g., Ca^{2+}, Mg^{2+}, Na^+, K^+) were not included in our simulations due to the lack of measurements, which may lead to a systematic underestimation of aerosol pH (Guo et al., 2017)".

Fig. R8 has been added to the revised supporting information as Fig. S1.

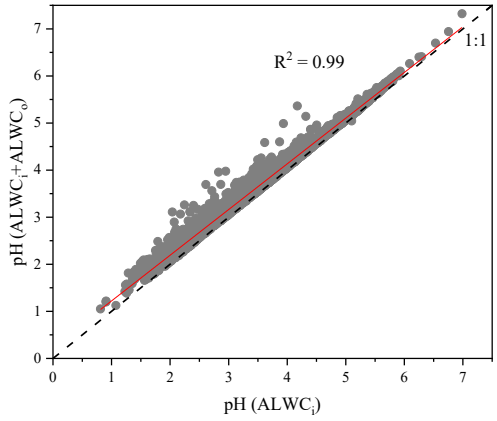

Fig. R8 Comparison of pH calculated with and without organic aerosol water ($ALWC_o$).

References:

Fountoukis, C. and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K^+–Ca^{2+}–Mg^{2+}–NH_4^+–Na^+–SO_4^{2-}–NO_3^-–Cl^-–H_2O aerosols, Atmos. Chem. Phys., 7, 4639-4659, 2007.

Guo, H., Xu, L., Bougiatioti, A., Cerully, K. M., Capps, S. L., Hite, J. R., Carlton, A. G., Lee, S. H., Bergin, M. H., Ng, N. L., Nenes, A., and Weber, R. J.: Fine-particle water and pH in the southeastern United States, Atmos. Chem. Phys., 15, 5211-5228, 10.5194/acp-15-5211-2015, 2015.

Guo, H., Liu, J., Froyd, K. D., Roberts, J. M., Veres, P. R., Hayes, P. L., Jimenez, J. L., Nenes, A., and Weber, R. J.: Fine particle pH and gas–particle phase partitioning of inorganic species in Pasadena, California, during the 2010 CalNex campaign, Atmos. Chem. Phys., 17, 5703-5719, 10.5194/acp-17-5703-2017, 2017.

Hennigan, C. J., Izumi, J., Sullivan, A. P., Weber, R. J., and Nenes, A.: A critical evaluation of proxy methods used to estimate the acidity of atmospheric particles, Atmos. Chem. Phys., 15, 2775-2790, 10.5194/acp-15-2775-2015, 2015.

Pöhlker, M. L., Pöhlker, C., Quaas, J., Mülmenstädt, J., Pozzer, A., Andreae, M. O., Artaxo, P., Block, K., Coe, H., Ervens, B., Gallimore, P., Gaston, C. J., Gunthe, S. S., Henning, S., Herrmann, H., Krüger, O. O., McFiggans, G., Poulain, L., Raj, S. S., Reyes-Villegas, E., Royer, H. M., Walter, D., Wang, Y., and Pöschl, U.: Global organic and inorganic aerosol

hygroscopicity and its effect on radiative forcing, Nat. Commun., 14, 6139, 10.1038/s41467-023-41695-8, 2023.

Song, S., Gao, M., Xu, W., Shao, J., Shi, G., Wang, S., Wang, Y., Sun, Y., and McElroy, M. B.: Fine-particle pH for Beijing winter haze as inferred from different thermodynamic equilibrium models, Atmos. Chem. Phys., 18, 7423-7438, 10.5194/acp-18-7423-2018, 2018.

Zhou, M., Zheng, G., Wang, H., Qiao, L., Zhu, S., Huang, D., An, J., Lou, S., Tao, S., Wang, Q., Yan, R., Ma, Y., Chen, C., Cheng, Y., Su, H., and Huang, C.: Long-term trends and drivers of aerosol pH in eastern China, Atmos. Chem. Phys., 22, 13833-13844, 10.5194/acp-22-13833-2022, 2022.

6. The upper end of the calculated pH range needs further explanation. The manuscript reports pH values up to 6.7, which is close to neutral and unusually high for fine-mode urban aerosols. The authors should examine the high-pH cases, for example pH > 5 or pH > 6, and clarify under what conditions they occur. In particular, they should show whether these values are associated with very low ALWC, low inorganic mass, extremely ammonia-rich conditions, or other situations where thermodynamic pH predictions may be uncertain. It would also be useful to evaluate whether the NH3/NH4+ partitioning remains reliable for these high-pH samples, rather than only reporting overall model performance.

Response: We thank the reviewer for this critical and constructive comment. We have further examined the aerosol pH values in our study. As shown in Fig. R9, the majority of samples exhibited pH values below 5. Specifically, approximately 12% of the data points showed pH > 5, and only 5 samples (less than 1% of the dataset) had pH values exceeding 6. These relatively higher pH values (e.g., pH > 5) were predominantly observed in wintertime, consistent with previous wintertime studies in Beijing that reported pH values frequently exceeding 5 and occasionally above 6, likely reflecting the ammonia-rich and nitrate-dominated chemical regime (Ding et al., 2019; Xie et al., 2020). We agree with the reviewer that pH predictions under very low ALWC conditions can carry large uncertainties. In our analysis, we had already

excluded data points with RH < 30% to avoid unrealistic pH estimates arising from misestimated ALWC at low RH, following the practice of previous studies (Liu et al., 2017; Ding et al., 2019; Xie et al., 2020). To further evaluate the thermodynamic predictions for high-pH samples, we examined the NH_3/NH_4^+ partitioning for pH > 5 samples against those with pH < 5 (Fig. R10). As shown in the figure, samples with pH > 5 fall along a similar trend as the rest of the dataset, with no obvious systematic deviation from the overall relationship. This indicates that the thermodynamic model performs consistently across the observed pH range.

To more accurately describe the pH range in our campaign, we have updated the text in the revised manuscript as follows (lines 205-210):

"Time series of meteorological and chemical measurements, together with ISORROPIA-derived pH and ALWC, are shown in Fig. 1. The average aerosol pH value was 3.8 ± 0.96, with the samples predominantly falling within the range of 1.5–6.0 (Fig. S5). This range is comparable to previous observations in urban Beijing (Ding et al., 2019; Xie et al., 2020), and displayed pronounced seasonal and synoptic variability."

Fig. R9 has been added to the revised supporting information as Fig. S5.

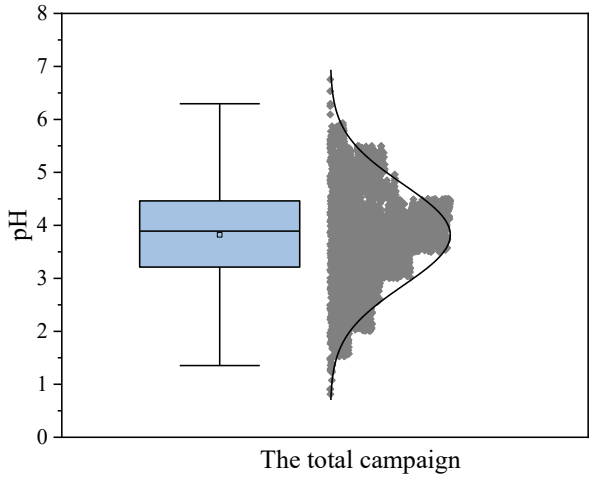

Fig. R9 Distribution of aerosol pH in this study. The box spans the 25th–75th percentiles, with whiskers extending to the 10th and 90th percentiles. The line and square inside the box indicate the median and mean, respectively.

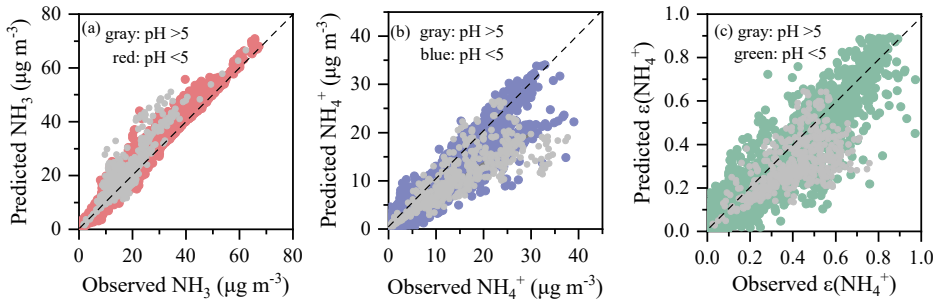

Fig. R10 Comparisons of predicted versus observed NH_3 (a), NH_4^+ (b), and $\varepsilon(NH_4^+)$ (c) for samples with pH > 5 and pH < 5. $\varepsilon(NH_4^+)$ is calculated as the molar ratio of $NH_4^+/(NH_4^+ + NH_3)$.

References:

Ding, J., Zhao, P., Su, J., Dong, Q., Du, X., and Zhang, Y.: Aerosol pH and its driving factors in Beijing, Atmos. Chem. Phys., 19, 7939-7954, 10.5194/acp-19-7939-2019, 2019.

Liu, M., Song, Y., Zhou, T., Xu, Z., Yan, C., Zheng, M., Wu, Z., Hu, M., Wu, Y., and Zhu, T.: Fine particle pH during severe haze episodes in northern China, Geophys. Res. Lett., 44, 5213-5221, 10.1002/2017gl073210, 2017.

Xie, Y., Wang, G., Wang, X., Chen, J., Chen, Y., Tang, G., Wang, L., Ge, S., Xue, G., Wang, Y., and Gao, J.: Nitrate-dominated PM2.5 and elevation of particle pH observed in urban Beijing during the winter of 2017, Atmos. Chem. Phys., 20, 5019-5033, 10.5194/acp-20-5019-2020, 2020.

7. The causal language in the discussion should be moderated. The RH-related pH reversal is interesting, but interpretations involving secondary inorganic aerosol formation, heterogeneous reactions, aqueous-phase chemistry, and H+ accumulation are not directly resolved by the XGBoost-SHAP model. These explanations should be framed as plausible mechanisms or patterns consistent with known chemistry, rather than as direct evidence from the ML analysis.

Response: We thank the reviewer for this important and constructive comment. We fully agree that the XGBoost-SHAP analysis identifies statistical associations and feature importance patterns rather than directly resolving causal physicochemical mechanisms. Accordingly, we have carefully revised the discussion throughout the manuscript to moderate the causal

language. The relevant interpretations involving secondary inorganic aerosol formation, heterogeneous reactions, aqueous-phase chemistry, and H+ accumulation are now framed as plausible mechanisms or patterns consistent with known atmospheric chemistry.

For example:

Line 295: "This regime-dependent behavior can be explained by ……" has been reframed to "This regime-dependent association behavior identified by SHAP model is consistent with……".

Line 309: "That means……" has been reframed to "This likely suggests that……".

Line 311: "……resulting in increased aerosol acidity……" has been reframed to "which might be associated with increased aerosol acidity".

Lines 322-324: "Temperature therefore modulates ……, producing ……"has been reframed to "Temperature therefore likely modulates ……, which may be associated with……".

Lines 340-341: "……, such that dilution by aerosol water likely outweighed, resulting in elevated pH……" has been reframed to "……, suggesting that dilution by aerosol water likely outweighed H^+ input, which might be associated with elevated pH……".

Line 346: "These might be related to……" has been reframed to "These patterns are consistent with……".

Line 368: "That means under this regime, ……" has been reframed to "This likely suggests that……".

Line 371: "……and leads to a decrease in pH again……" has been reframed to "……and is likely associated with the decrease in pH".

Line 382: "RH generally showed negative contribution to pH in winter……" has been reframed to "RH generally showed negative association with pH in winter……".

Line 393: "……reinforcing the plausibility of the interaction patterns identified here" has been reframed to "……supporting the consistency of the interaction patterns identified here".

The figure caption for Figure 3: "SHAP interaction plots illustrating the joint effects of RH with temperature (°C) (a), N/S mass ratio (b), and PM_1 mass loading (μg m^{-3}) (c) on aerosol pH prediction. The color scale represents RH, and the SHAP interaction values indicate the direction and magnitude of RH modulation on predicted pH under different regimes" has been

reframed to "SHAP interaction plots showing the joint contributions of RH with temperature (°C) (a), N/S ratio (b), and PM_1 mass loading (μg m^{-3}) (c) to aerosol pH predictions. The color scale represents RH, and the SHAP interaction values indicate the direction and magnitude of the interactive contributions to pH across different regimes".

8. The manuscript should be more cautious in generalizing the results. The analysis is based on one urban site in Beijing during 2014-2015. The identified thresholds and interaction patterns may reflect local meteorology, emissions, and aerosol composition. Statements about broader predictive capability or future scenarios should therefore be toned down unless supported by additional datasets or validation.

Response: We thank the reviewer for this important caution. We fully agree that our analysis is based on observations from a single urban site in Beijing during 2014–2015, and that the identified thresholds and interaction patterns may reflect local meteorological conditions, emission characteristics, and aerosol composition, as acknowledged in the conclusion. Following the reviewer's suggestion, we have further toned down statements about broader predictive capability or future scenarios. The revised conclusion now emphasizes the SHAP framework as a diagnostic approach for understanding local pH variations, and frames the extension to other environments as a recommended future direction rather than an established capability. The revised paragraph (lines 413–432) now reads as:

"The thresholds and interaction patterns identified here are based on observations at a single urban site in Beijing and may therefore reflect local meteorology and emission characteristics. Nevertheless, this approach provides a quantitative framework for examining the nonlinear relationships between pH and complex coupled environmental parameters. In China's evolving atmosphere, meteorological factors, together with variable particle properties, may jointly modulate aerosol acidity. Distinct environment conditions not considered in this study, such as coastal regions influenced by sea salt (Wang et al., 2022; Xu et al., 2025) and dust-affected areas enriched in alkaline metals (Shi et al., 2019; Huang et al., 2020), likely also exert important roles in pH variation. Extending this SHAP-based analytical framework to diverse

atmospheric conditions would be a valuable direction for future research to further understand the regime-dependent pH variations across different environments".

9. Minor issues:

(1) The sentence in the Abstract, "their behavior differs across environmental regimes and remain incompletely understood," is grammatically inconsistent. It should be revised to "their behavior differs across environmental regimes and remains incompletely understood," or rewritten.

Response: Thank you for this correction. We have revised the sentence to "their behavior differs across environmental regimes and remains incompletely understood" in the Abstract.

(2) The word "systematical" should be changed to "systematic."

Response: Corrected. "Systematical" has been changed to "systematic". The revised sentence now reads: "RH-related interactions are generally stronger and more systematic than those of other variable pairs……".

(3) The phrase "That means" is too informal for a scientific manuscript and should be replaced with "This indicates that" or "This suggests that."

Response: We agree with this suggestion and have replaced "That means" with "This suggests that" in the relevant sections as follows:

Line 309: "This likely suggests that under low-temperature conditions, enhanced inorganic aerosol formation……".

Line 368: "This likely suggests that under this regime, the efficient aqueous medium is likely more favorable for sulfate production……".

(4) Equation (2) should be checked carefully. The notation for the sample index and feature index is confusing, and the summation should clearly refer to feature contributions.

Response: We thank the reviewer for pointing this out. We have revised the equation accordingly, using x to denote the sample and j as the feature index in the summation. The revised equation now reads as:

$$f(x_i)=\phi_0(f,x)+\sum_{j=1}^{M}\phi_j(f,x_i) \qquad (3)$$

where $f(x_i)$ is the predicted value for sample (x_i) with M features, $\phi_0(f, x)$ is the base value (expected output), and $\phi_j(f, x_i)$ is the SHAP value of feature j (Lundberg et al., 2020; Hou et al., 2022).

(5) The terminology for the nitrate-to-sulfate ratio should be standardized throughout the manuscript. For example, "NtoSratio" in the figures should be changed to "N/S ratio" to match the text.

Response: Corrected. We have standardized the terminology for the nitrate-to-sulfate ratio as "N/S ratio" throughout the revised manuscript. "NtoSratio" in the figures has been changed to "N/S ratio" to match the text.

(6) Figures 2 and 3 are difficult to read because the fonts and data points are too small. The c-ICE panels in Fig. 2 are particularly dense. The authors should enlarge the figures, improve labels, or move some panels to the Supplement.

Response: We thank the reviewer for this constructive suggestion. To improve readability, we have enlarged the fonts and data points in both figures and moved some c-ICE panels to the Supplement. The revised figures are shown in Figs. R11–R13. Fig. R11 has been revised as Fig. 2 and Fig. R13 as Fig. 3 in the manuscript. Fig. R12 has been added to the Supporting Information as Fig. S7.

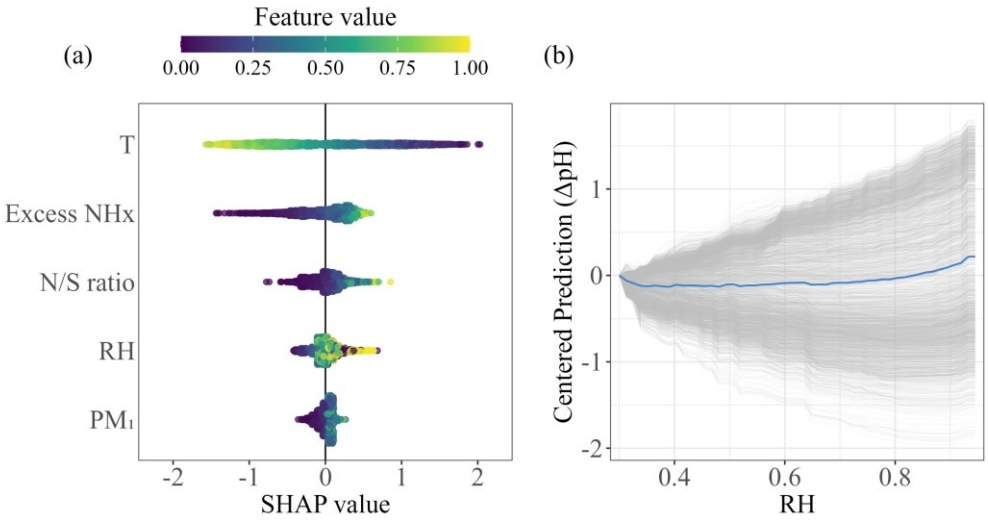

Fig. R11 Global SHAP summary plot (a) showing the relative importance of temperature (T, ℃), RH, Excess NHx (μg m^{-3}), N/S ratio, and PM_1 mass loading (μg m^{-3}) to aerosol pH. Panel (b) shows c-ICE curves of RH, with gray lines representing sample-specific effects and the blue line showing the averaged response.

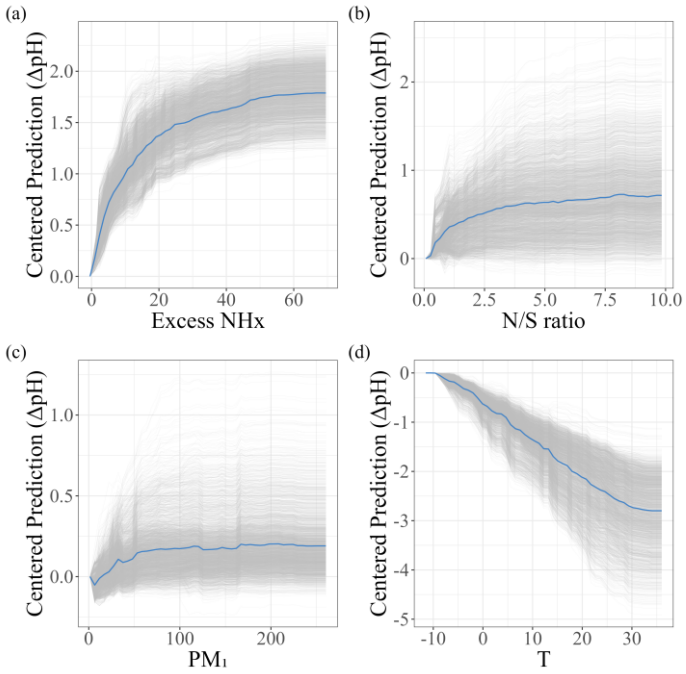

Fig. R12 c-ICE curves of (a) Excess NHx (μg m^{-3}), (b) N/S ratio, (c) PM_1 mass loading (μg m^{-3}), and (d) temperature (℃). Gray lines represent sample-specific effects on aerosol pH, and the blue line shows the averaged response.

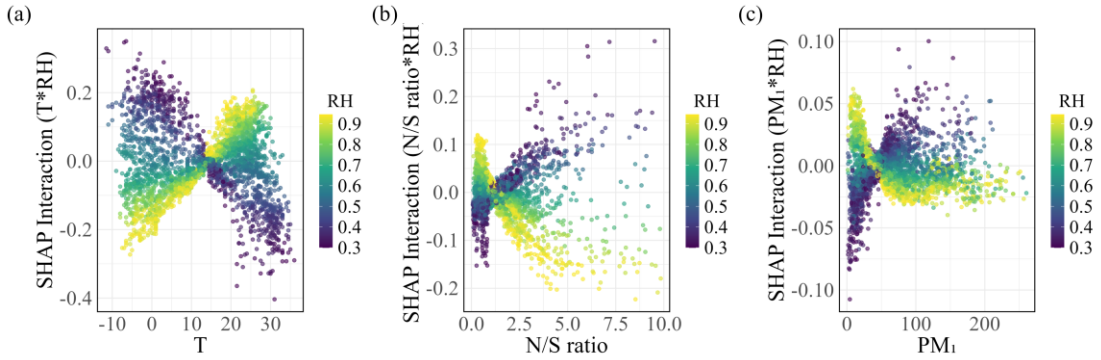

Fig. R13 SHAP interaction plots showing the joint contributions of RH with temperature (°C) (a), N/S ratio (b), and PM_1 mass loading (μg m^{-3}) (c) to aerosol pH predictions. The color scale represents RH, and the SHAP interaction values indicate the direction and magnitude of the interactive contributions to pH across different regimes.