Point by Point Response to Review Comments

Daytime and nighttime aerosol soluble iron formation in clean and slightly-polluted moisture air in a coastal city in eastern China

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We thank the **Reviewer #1** for the detailed and constructive comments. We provide below point-by-point response to the comments. The reviewer's comments and the original contents of the manuscript are in **black**. The response text is in **blue**. Revisions in the manuscript are in **red**.

General comments:

This manuscript investigated the components of total and soluble iron in $PM_{2.5}$ in both daytime and nighttime in a coastal city, and tried to evaluate the effects of aqueous-phase and photochemical reactions on that. The topic is interesting and the discussion is comprehensive.

However, the language needs to be extensively improved throughout the manuscript, and the content could be more concise. Moreover, the references need to be double-checked. The original source should be cited as much as possible. For example, the original source of ISORROPIA II should be acknowledged as well.

General response:

In the revised manuscript, we have enhanced the linguistic expression, rectified grammatical inaccuracies, and rendered the descriptions more succinct. Moreover, we have conducted a thorough verification of the references, ensuring the inclusion of the original literature. Additionally, an acknowledgment has been extended to ISORROPIA II, as detailed below:

"*Acknowledgements. We gratefully acknowledge the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model, available at (https://www.ready.noaa.gov/HYSPLIT.php), and the Global Data Assimilation System (GDAS). Additionally, we acknowledge the use of ISORROPIA II, accessible at (https://www.epfl.ch/labs/lapi/models-and-software/isorropia/), developed by the Schools of Earth & Atmospheric Sciences and Chemical & Biomolecular Engineering at the Georgia Institute of*

Technology, for the calculation of aerosol pH and liquid water content."

Comment (1): In Table 1, aerosol pH values are with high standard deviations. Does it make the mean pH values less compatible? Are the data points actually quite overlapped?

Response:

As shown in Table 1, the standard deviations (SD) of pH value are approximately 0.8, indicating a considerable variability. This level of variability aligns with findings from a previous study, which documented similar SD values, exceeding 0.7 across various seasons (Ding et al., 2019). Ruan et al. (2022) observed SD values of 0.9 in clean air and 0.5 in heavily polluted air in Beijing, indicating that the observed SD levels of pH in our study are within expected ranges. In addition, the aerosol pH exhibited a pronounced difference between daytime and nighttime, as demonstrated by Figure R1. Although there is a substantial overlap in pH values between daytime and nighttime, this is anticipated due to the collection of aerosol samples under the same atmospheric conditions (either clean or slightly-polluted (SP) periods).

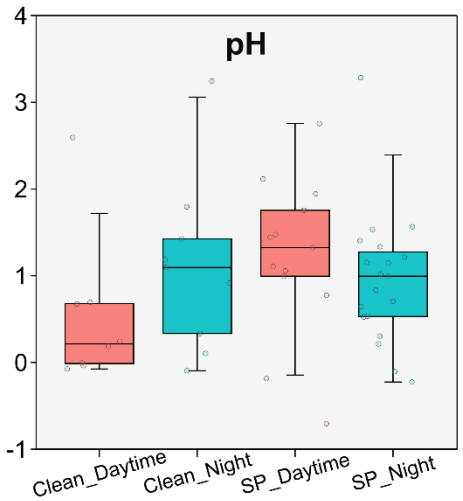

Figure R1: Aerosol pH during clean and slightly-polluted (SP) periods. Boxes and error bars represent the 10th, 25th, 50th, 75th, and 90th percentiles from bottom to top, respectively.

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.

Ruan, X., Zhao, C., Zaveri, R. A., He, P., Wang, X., Shao, J., and Geng, L.: Simulations of aerosol pH

in China using WRF-Chem (v4.0): sensitivities of aerosol pH and its temporal variations during haze episodes, Geosci. Model Dev., 15, 6143-6164, 10.5194/gmd-15-6143-2022, 2022.

Comment (2): Figure 3, the data points are for both clean and slightly-polluted periods. They are mixed together, not separately marked like in Figure 5, why?

Response:

This is because we want to show the overall situation and demonstrate that the influence of aqueous-phase promoted acid processes on Fe solubility (%Fe_S) was profound not only during clean and SP periods, but also the whole sampling period. Figure 5 contains only Clean and SP data, with limited data points, so different markers are used for differentiation. In contrast, Figure 3 contains the whole sampling dataset, with about 140 points, making it visually unappealing and impractical to distinguish Clean and SP periods by using different markers (Figure R2), and also not conducive to performing regression analysis on clean and SP data points separately.

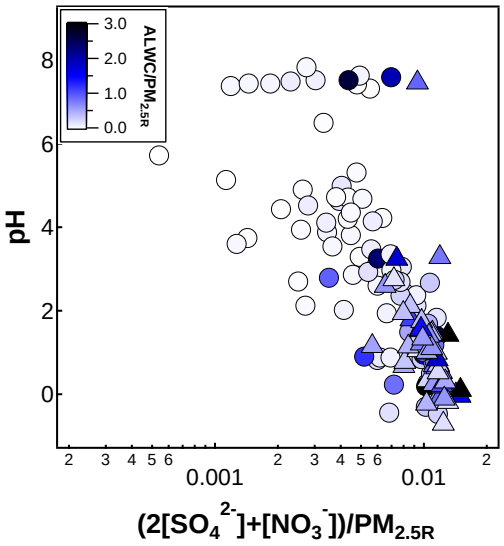

Figure R2: Relationship between aerosol pH and the normalized relative abundance of main acidic species ($2[SO_4^{2-}] + [NO_3^-]$) with respect to the reconstructed $PM_{2.5R}$. The data of clean and SP periods is marked by triangles.

Based on the above reasons, the dependence of pH on the ratio of $(2[SO_4^{2-}] + [NO_3^-])/PM_{2.5R}$ during clean and SP periods is shown in the subgraph at the bottom-left of Figure 3(a) show the robust influence of acid species on aerosol pH specifically during the clean and SP periods. In the revision,

we also added the dependence of pH on the ratio of ALWC/$PM_{2.5R}$ during clean and SP periods that is shown in the subgraph at the bottom-left of Figure 3(b). In terms of the relationship between $\%Fe_S$ and the ratio of $(2[SO_4^{2-}] + [NO_3^-])/PM_{2.5R}$ during clean and SP periods, it was provided by Figure S4 in the ***Supplementary Information.*** We also modified the color-coding variable of ALWC in Figure 3(c) into unit aerosol mass (i.e., ALWC/$PM_{2.5R}$) to ensure consistency with Figure 3(a) and 3(b).

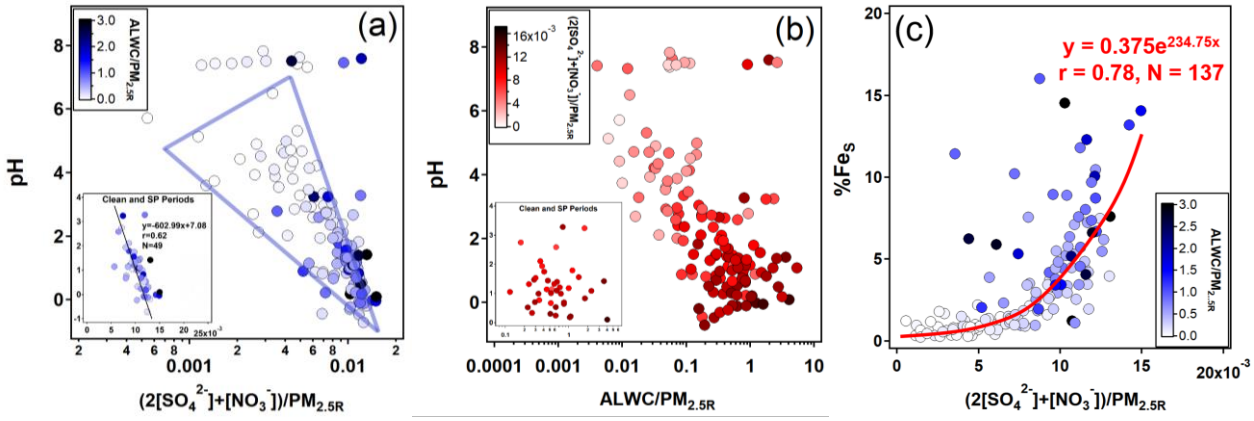

Figure 3: Relationships among aerosol pH, the normalized relative abundance of ALWC (unit: μg m^{-3}) and main acidic species (= $2[SO_4^{2-}] + [NO_3^-]$, unit: μmol m^{-3}) with respect to the reconstructed $PM_{2.5}$ ($PM_{2.5R}$, unit: μg m^{-3}), and $\%Fe_S$. The subgraph at the bottom-left of figures (a) and (b) show scatter plots during clean and SP periods with the linear regression line obtained by using the Igor Pro-based program developed by Wu and Yu (2018).

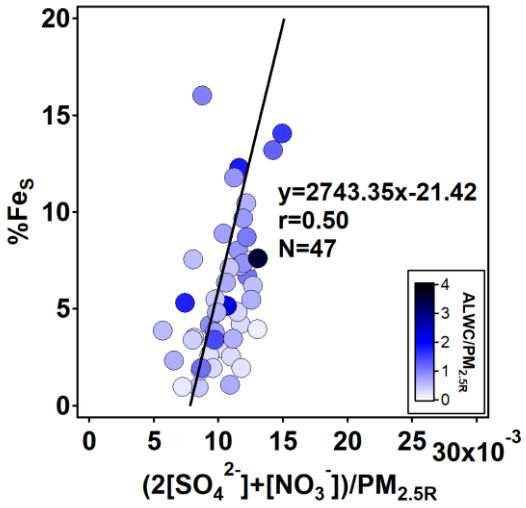

Figure S4: Same as Figure 3c in the manuscript but only for clean and SP periods.

Comment (3): Figure 6, color bar is missing.

Response:

We added color bars in Figure 6 and more descriptions in the figure caption. The revised figure is shown as follows:

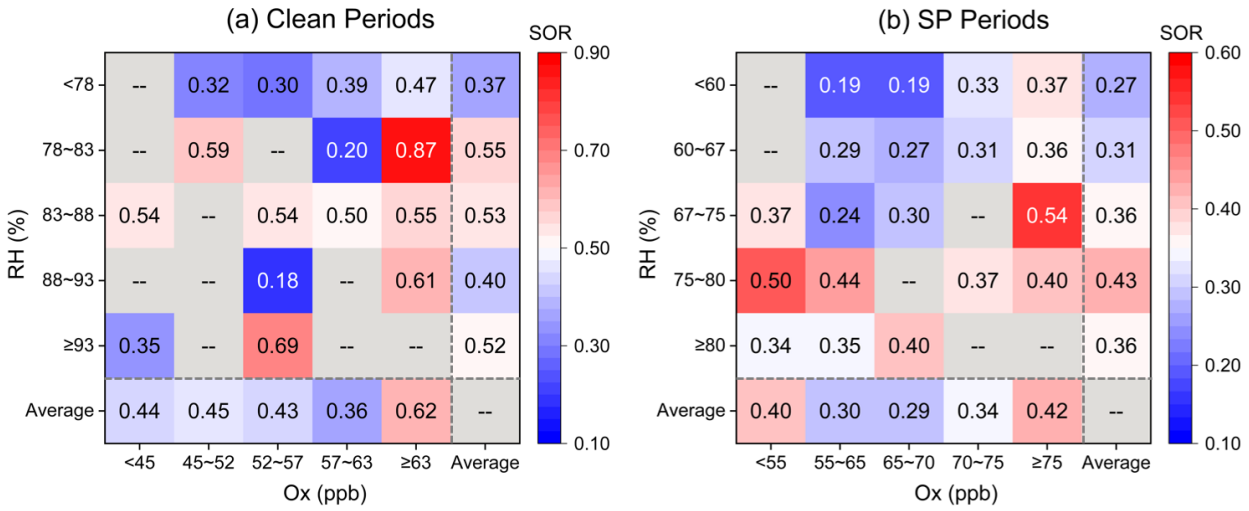

Figure 6: RH-O_x image plots colored by SOR during clean and SP periods. The last row and last column of the matrices represent the average value of SOR in the corresponding ranges of RH and O_x.