Authors' response to comments made by anonymous reviewer #2:

Summary

Wang et al. presents a comprehensive investigation of how ammonia (NH3) emissions affect size-resolved aerosol composition and acidity on a global scale. Using the EMAC atmospheric chemistry-climate model with three different ammonia emission schemes, the authors analyze the complex interactions between ammonium, sulfate and nitrate in different sizes, geographic regions, and chemical environments. Research advances our understanding of atmospheric aerosol dynamics and has significant implications for air quality management and climate modeling. I recommend publication after addressing the following comments.

We thank the reviewer for the thoughtful and positive comments on our manuscript. We appreciate the constructive feedback provided which helped us improve the clarity and quality of the manuscript. Below is a point-by-point response (in black) to all the points raised (in red).

Specific Comments:

1. Research questions and modeling approach lack clarity

The Introduction provides a comprehensive overview of aerosol emission trends and how $PM_{2.5}$ components respond to different clean air policies. It also notes that these responses vary depending on particle size ranges. However, the progress in modeling this size-resolved response is not clearly presented. The intended model for use in this work and its suitability are not well explained, and the scientific question lacks clarity. The authors are encouraged to clearly articulate the rationale for selecting the model, i.e., why EMAC is appropriate for this question?

We thank the reviewer for this valuable feedback. We recognize the importance of clearly articulating both the scientific motivation and the rationale behind our modeling approach. In response, we have thoroughly revised the manuscript to clarify the research objectives and the suitability of the EMAC model for addressing our questions. Specifically, we have updated the final paragraph of the Introduction to clearly state the scientific questions driving this study, including the size-resolved response of aerosol composition and acidity to changes in precursor emissions. Furthermore, we justify the selection of the EMAC model in the introduction and model description (i.e., section 2), emphasizing its state-of-the-art capability to simulate global-scale aerosol–chemistry–climate interactions with size resolution, and its integration with the ISORROPIA-II thermodynamic module for aerosol pH estimation. ISORROPIA-II is a widely used thermodynamic model well-suited for simulating aerosol pH, as it efficiently handles size-resolved inorganic aerosol systems under varying humidity conditions. While it simplifies some aspects (e.g., assuming unity for certain activity coefficients), it has been extensively validated and remains a practical choice for large-scale pH simulations. These revisions aim to provide a more coherent narrative linking the research context, modeling framework, and study objectives.

2. *The role of organic aerosols in affecting aerosol pH*

Although the main goal of this work is to study the size-resolved SNA and pH response to different ammonium emission inventories, it would be beneficial to include some discussions on the role of organics in influencing these outcomes. As significant components of aerosol particles with diverse hygroscopic properties, organic aerosols can absorb water and impact both aerosol liquid water content and pH. Including a discussion on how organics might alter the size-resolved response would strengthen the analysis.

For example, the reported 104% increase in NH_4^+ in response to an 18% rise in ammonia emissions could not solely from interactions with sulfate and nitrate, but may partly result from reactions between ammonia and organic acids (e.g., forming ammonium oxalate). These processes can influence pH, especially in the 0–1 μm range. Neglecting the role of organics risks overattributing observed effects to SNA alone.

We appreciate the reviewer's suggestion to further discuss the role of organic aerosols in influencing aerosol pH. While the primary focus of this study is on the size-resolved response of SNA aerosols to changes in ammonia emissions, we agree that organics can also play a role, particularly through their contribution to aerosol liquid water content (ALWC) and, to a lesser extent, their influence on hydrogen ion activity. In our model, the effect of water-soluble organic aerosols on ALWC is accounted for via the GMXe module, which includes both inorganic and organic contributions. Organic aerosol formation is simulated using the ORACLE module, and the associated water uptake is calculated assuming a κ-hygroscopicity value of 0.14 for all organic components (Tsimpidi et al., 2014). This influences the total aerosol water content used in pH calculations. However, our model in the present set-up does not account for chemical interactions between ammonia and organic acids (e.g., formation of ammonium oxalate), and it treats the inorganic and organic aerosol phases independently. Consequently, while organics can indirectly affect pH through water uptake, changes in NH_3 emissions do not influence organic aerosol formation or the associated water content in our simulations. Therefore, the reported increase in NH_4^+ is attributed solely to interactions with inorganic aerosol components. Although the effect of organics on hydrogen ion activity coefficients is not explicitly included, previous studies have shown that water-soluble organic aerosols exert only a minor influence on aerosol pH. For example, Pye et al. (2018) estimated that organic-associated hydrogen ions increase $PM_{2.5}$ pH by only ~0.1 units, while Vasilakos et al. (2018) found that organics induce pH deviations of less than 2% across a range of compounds and environmental conditions. These findings are consistent with other studies (Battaglia Jr et al., 2019; Pye et al., 2020; Guo et al., 2015; Liu et al., 2017), supporting the limited role of organics in modulating aerosol acidity. We have added this clarification to Section 2.2.1, along with a discussion of the model's limitations in representing ammonia–organic interactions.

Minor comments:

1. *Line 170: The cases are not clearly defined in the texts that describe Table 1. What are noNH_3 and Meta cases? You can briefly introduce why you conduct these two cases here. Is Top-Dep case using the Top-down scheme?*

Thank you for pointing this out. In the noNH_3 case, all ammonia emissions are turned off. The Meta case is identical to the base case, except that the ISORROPIA model is run in metastable mode. The Top-Dep

case applies the top-down emission scheme. We have added a brief explanation of these simulation cases in lines 176–180 to clarify their purpose and setup.

2. *Line 207: The symbols and Italic fonts used in the texts and equations throughout the paper, such as $E_{NH3,mod}$ do not follow standard scientific writing conventions. For guidance, you may refer to this document.:https://iupac.org/wpcontent/uploads/2016/01/ICTNS-On-the-use-of-italic-and-roman-fonts-for-symbols-in-scientific-text.pdf*

We sincerely appreciate the reviewer's guidance on the proper use of italic and roman fonts for scientific symbols. Following the IUPAC recommendations, we have systematically revised the formatting of all symbols throughout the text and equations to ensure consistency with standard conventions.

3. *Line 217: Remove the dot after number 74.*

Done.

4. *Line 230: Using lighter background colors in Figure 1(a) would improve clarity and make the hotspots easier to distinguish.*

Thank you for the suggestion. We have redrawn Figure 1(a) using a lighter background color to enhance visual clarity and improve the distinction of hotspot regions.

5. *Line 235 and 245: Since these are comparative descriptions rather than time series trends, I'll avoid using "increase" and instead opt for terms like "overestimate" or "biases".*

Thank you for the helpful suggestion. We have revised the wording accordingly.

6. *Line 295: references for IPCC(2023)?*

We have added the appropriate reference for IPCC in the reference list.

7. *Line 351: Typos in this paragraph. Change SO_4^- to SO_4^{2-} .*

Corrected.

8. *line 505: References for the statement: "high-latitude marine aerosols are more acidic ..."?*

High-latitude marine aerosols are generally more acidic than those over remote ocean regions, primarily due to the long-range transport of anthropogenic pollutants such as H_2SO_4 and HNO_3 from continental sources. This enhanced acidity has been observed and simulated in several studies. For example, Karydis

et al. (2021) conducted a global modeling study showing that aerosol pH in high-latitude marine regions is significantly influenced by anthropogenic outflow, particularly from Europe and North America. Similarly, Myhre et al. (2013) discuss how anthropogenic aerosols, including sulfate and nitrate, are transported to remote marine environments, altering their chemical composition and acidity. We have updated the text in the revised manuscript to include these references.

9. *Line 538: Table 9?*

We have corrected the table number accordingly.

10. *Line 539: It would be better to provide more context for the motivation of conducting the noNH_3 case earlier in the text-when introducing the cases in Table 1—rather than introducing it abruptly here.*

Thank you for the suggestion. We have added a brief explanation of the noNH_3 case in lines 176–180 to improve the flow of the manuscript.

11. *Line 556: Since the effects of different ammonia emission scheme are a crucial aspect of this research, and the title is "The Influence of Ammonia Emissions...," it would be more appropriate to move the sentences discussing the importance of the ammonia emission inventory and its effects earlier.*

We appreciate this thoughtful suggestion. After careful consideration, we have decided to retain the current structure to preserve the logical flow of the manuscript. Our approach begins with the model and observational datasets (Sections 2 and 3), followed by evaluation of the base case (Section 4), then analysis of global and regional aerosol chemical regimes (Sections 5 and 6), and finally the emission sensitivity analysis (Section 7). Reordering Section 7 earlier could disrupt this flow, as the interpretation of the emission scenario results depends on understanding the performance and limitations of the base case.

12. *Line 605: The figure captions for Figure 7 and Figure 8 are almost the same. You can change the caption of figure 8 to "The same as figure 7, but for the difference between Top-Dep case and base case.*

Thank you for the suggestion. We have revised the caption for Figure 8 accordingly.

References

Battaglia Jr, M. A., Weber, R. J., Nenes, A., and Hennigan, C. J.: Effects of water-soluble organic carbon on aerosol pH, Atmos. Chem. Phys., 19, 14607-14620, 10.5194/acp-19-14607-2019, 2019.

Guo, H., Xu, L., Bougiatioti, A., Cerully, K. M., Capps, S. L., Hite Jr, 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.

Karydis, V. A., Tsimpidi, A. P., Pozzer, A., and Lelieveld, J.: How alkaline compounds control atmospheric aerosol particle acidity, Atmos. Chem. Phys., 21, 14983-15001, 10.5194/acp-21-14983-2021, 2021.

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, Geophysical Research Letters, 44, 5213-5221, https://doi.org/10.1002/2017GL073210, 2017.

Myhre, G., Samset, B. H., Schulz, M., Balkanski, Y., Bauer, S., Berntsen, T. K., Bian, H., Bellouin, N., Chin, M., Diehl, T., Easter, R. C., Feichter, J., Ghan, S. J., Hauglustaine, D., Iversen, T., Kinne, S., Kirkevåg, A., Lamarque, J. F., Lin, G., Liu, X., Lund, M. T., Luo, G., Ma, X., van Noije, T., Penner, J. E., Rasch, P. J., Ruiz, A., Seland, Ø., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K., Wang, P., Wang, Z., Xu, L., Yu, H., Yu, F., Yoon, J. H., Zhang, K., Zhang, H., and Zhou, C.: Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations, Atmos. Chem. Phys., 13, 1853-1877, 10.5194/acp-13-1853-2013, 2013.

Vasilakos, P., Russell, A., Weber, R., and Nenes, A.: Understanding nitrate formation in a world with less sulfate, Atmos. Chem. Phys., 18, 12765-12775, 10.5194/acp-18-12765-2018, 2018.