Response to Reviewer

Guo et al. analyze gaseous sulfuric acid in urban Beijing, and derive three proxies (the OH-CS based, UVB-CS based, and UVB-$PM_{2.5}$ based proxies) based on its production and loss pathways. These proxies are able to estimating sulfuric acid concentration well between 10:00 and14:00. This study also shows that the UVB-CS based and UVB-$PM_{2.5}$ based proxies can reproduce daytime sulfuric acid concentration at the Hyytiälä boreal forest site in Finland. This suggests that the three steady-state based proxies are likely applicable to other atmospheric sites. Since sulfuric acid is a key precursor for cluster formation and subsequent growth, the sulfuric acid datasets provided here are highly valuable. The proposed proxies can estimate the overall sulfuric acid concentrations at other sites, thereby to some extent facilitating aerosol-related researches.

The previous two referees raised four main comments. First, since OH radical in this study is obtained from model, the reliability of model should be illustrated. Second, the description of the modeling section does not match the overall style of the manuscript and needs refinement. Third, it should be better if how well these proxies perform during new particle formation periods can be explained. Finally, the referee wonder what are the conditions when estimated sulfuric acid concentration deviate significantly from the measured one in Figure 5. The authors responded to these main and additional minor comments by supplementing data, updating figures, revising the text, and providing detailed explanations, and revised the manuscript and supplement accordingly. Overall, the responses are well reasoned and well supported. I think this article can be published after the following questions being explained.

We thank the reviewer for the constructive comments and suggestions. We have carefully revised our manuscript and supplement accordingly. The point-to-point response to the comments is given below. And the comments, our replies, and the corresponding changes in the manuscript and supplementary information are in black, blue, and green, respectively.

1. The three sulfuric acid proxies presented in this study originate from the OH-CS based proxy, mainly focusing on their performance during 10:00–14:00. How does the OH-CS based proxy perform during other daytime periods?
Response: Thanks for your comment.

Figure S15 shows the performance of the OH-CS based proxy during 08:00–10:00 and 14:00–16:00, as well as 06:00–08:00 and 16:00–18:00. As the time window moves further from noon, the proxy increasingly underestimates the sulfuric acid concentration. Moreover, as the time window shifts away from noon, the relationship between proxy and measured sulfuric acid becomes increasingly nonlinear. This suggests that in the early morning or at nightfall, sulfuric acid sources other than OH+SO_2 pathway cannot be neglected.

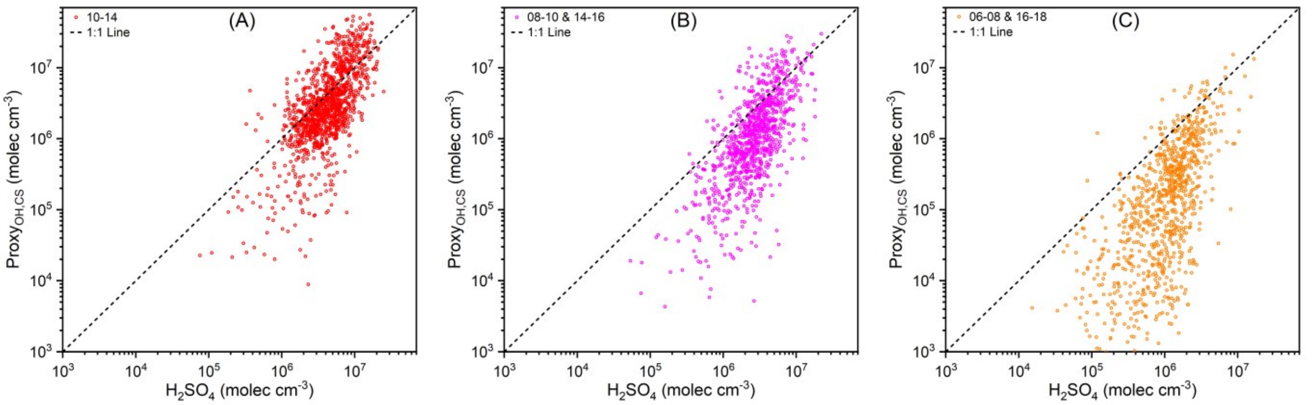

Figure S15. OH-CS based proxy vs. measured sulfuric acid during daytime in 2019 for (A) 10:00–14:00, (B) 08:00–10:00 and 14:00–16:00, as well as (C) 06:00–08:00 and 16:00–18:00. The black dashed lines are 1:1 lines.

To clarify this consideration, we added Figure S15 in the revised supplement and the following explanation in the revised manuscript:

"It should be pointed out that, as the time window moves further from noon, the OH-CS based proxy increasingly underestimates the sulfuric acid concentration. And as the time window shifts away from noon, the relationship between proxy and measured sulfuric acid becomes increasingly nonlinear. This implies that in the early morning or at nightfall, sulfuric acid sources other than OH+SO_2 pathway cannot be neglected (Figure S15)." (Line 404–408, Page 15–16)

2. The manuscript notes that the k values in the CS = k · $PM_{2.5}^{2/3}$ equation vary across different sites, which then leads to different pre-factors in the UVB-$PM_{2.5}$ based proxy expressed by Eq. (18). The k value for Beijing is stated to be 1.68 times that of the Hyytiälä site. What are the differences of the k values at other sites compared with Beijing?

Response: Thank you very much for your comment.

We use the dataset from one Nanjing site (Nie et al., 2022) to illustrate (Figure S16). At this site, the coefficient k in the CS = k · $PM_{2.5}^{2/3}$ equation is 1.62×10^{-3} s^{-1} $\mu g^{-2/3}$ m^2, which falls between those of Beijing (2.67×10^{-3} s^{-1} $\mu g^{-2/3}$ m^2) and Hyytiälä (1.59×10^{-3} s^{-1} $\mu g^{-2/3}$ m^2), and is closer to that of Hyytiälä. This implies that although the k values vary among sites, this difference remains modest.

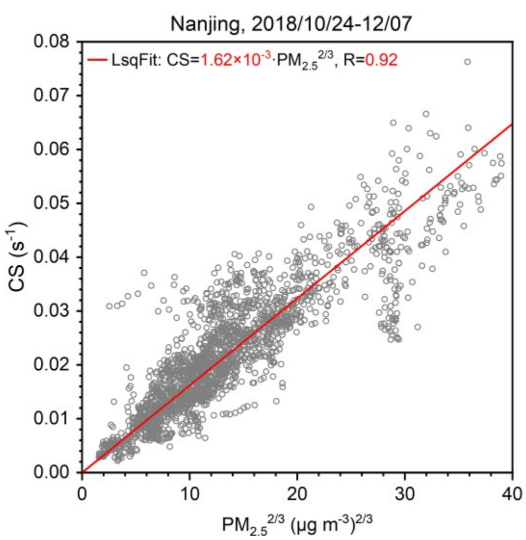

Figure S16. Relationship between condensation sink (CS) and $PM_{2.5}^{2/3}$ for Nanjing site (Nie et al., 2022) from 24th October to 7th December, 2018.

To clarify this consideration, we added Figure S16 in the revised supplement and the following explanation in the revised manuscript:

"Moreover, this coefficient k for one Nanjing site is 1.62×10^{-3} s^{-1} $\mu g^{-2/3}$ m^2 (Figure S16), which falls between those of Beijing and Hyytiälä. This implies that although the values of k vary among sites, this difference remains modest." (Line 498–500, Page 19)

In addition, we are sorry that some units of coefficient k were written incorrectly in the manuscript. We have corrected these units in the revised version.

"… where the values of k are 2.67×10^{-3} s^{-1} $\mu g^{-2/3}$ m^2 and 1.59×10^{-3} s^{-1} $\mu g^{-2/3}$ m^2 for Beijing …" (Line 494, Page 19)

3. The manuscript mentions that sulfuric acid may originate from traffic emissions. Do the datasets presented here provide any indication that sulfuric acid is emitted from such sources?

Response: During the COVID lockdown period, people were kept home and traffic emissions were dramatically reduced (Huang et al., 2021;Zhang et al., 2020;Yan et al., 2022). If traffic was a major sulfuric acid source, difference should emerge between the pre-lockdown and lockdown periods. Figure R1 shows the relationship between the OH-CS based proxy and measured sulfuric acid for the entire year of 2019, pre-lockdown and lockdown periods. It is clear that as in other periods, data points during the lockdown period also fall on the 1:1 line. This suggests that the dominant production and loss mechanisms of sulfuric acid remained unchanged during the lockdown period, and that traffic exerted only a minor influence on its formation. To further demonstrate that, the diurnal variations of sulfuric acid for pre-lockdown and lockdown periods, as well as weekdays and weekends in 2019 are plotted in Figure R1 (B) and (C). During morning rush hour, the diurnal curves of pre-lockdown and lockdown periods overlap, and there is also no discernible difference between weekdays and weekends. This also indicates that traffic emissions were unlikely to be a major sulfuric acid source in urban Beijing.

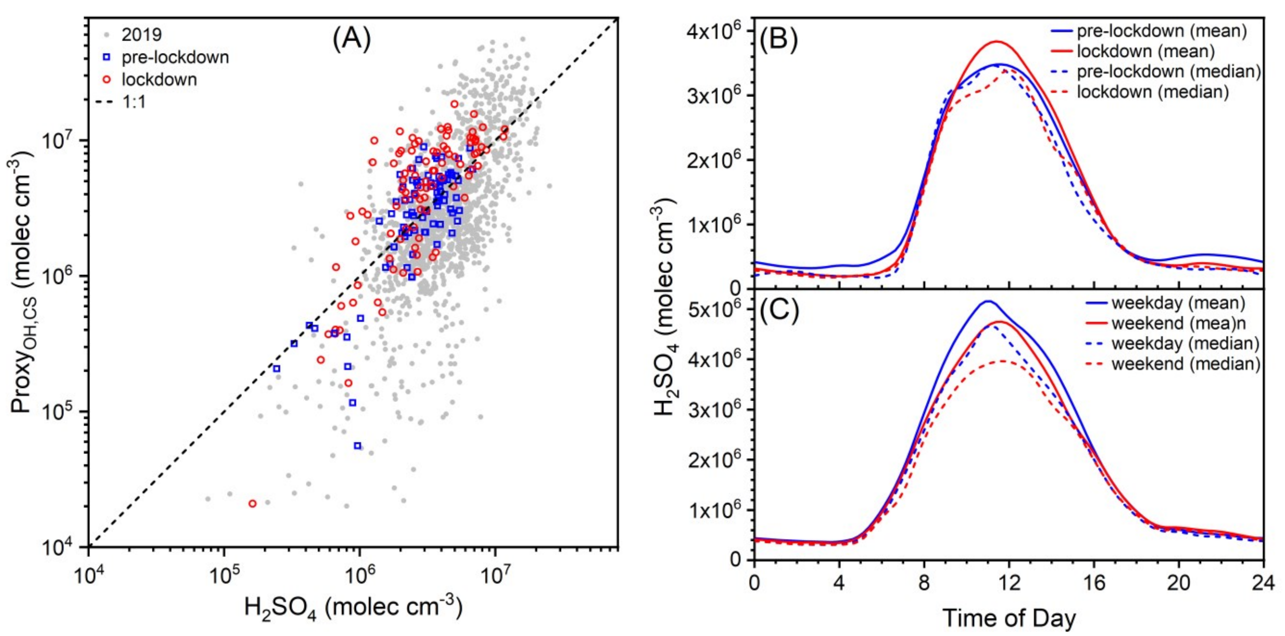

Figure R1. (A) OH-CS based proxy vs. measured sulfuric acid at daytime (10:00-14:00) during different periods. Gray circle, blue hollow square, and red hollow circle are data points of the entire year of 2019, pre-lockdown (1st to 22nd January, 2020), and lockdown (23rd January to 19th February, 2020) periods, respectively. The black dashed line is 1:1 line. (B) Median and mean diurnal variations of sulfuric acid for pre-lockdown and lockdown periods. (C) Median and mean diurnal variations of sulfuric acid for weekdays and weekends in 2019.

After careful consideration, we decided not to include the above discussion in the manuscript. There are two main reasons. First, this study focuses on daytime sulfuric acid and derives three proxies to estimate its concentration during 10:00–14:00. In these proxies, the source of sulfuric acid is the oxidation of SO_2 via OH radical. However, traffic emissions are not the main source of daytime sulfuric acid, and thus lie outside the core scope of this research. An extensive discussion of this topic would make the content somewhat scattered. Second, a comprehensive analysis of traffic emissions requires additional data and supporting evidence. For instance, emission indicators such as benzene (Yang et al., 2021) should be needed. However, no long-term measurement of such indicator was available during the study period. The traffic emission analysis based on Figure R1 remains superficial. Consequently, Figure R1 and its accompanying explanation were not included in the manuscript.

REFERENCES

Huang, X., Ding, A., Gao, J., Zheng, B., Zhou, D., Qi, X., Tang, R., Wang, J., Ren, C., Nie, W., Chi, X., Xu, Z., Chen, L., Li, Y., Che, F., Pang, N., Wang, H., Tong, D., Qin, W., Cheng, W., Liu, W., Fu, Q., Liu, B., Chai, F., Davis, S. J., Zhang, Q., and He, K.: Enhanced secondary pollution offset reduction of primary emissions during COVID-19 lockdown in China, Natl Sci Rev, 8, nwaa137, 10.1093/nsr/nwaa137, 2021.

Nie, W., Yan, C., Huang, D. D., Wang, Z., Liu, Y., Qiao, X., Guo, Y., Tian, L., Zheng, P., Xu, Z., Li, Y., Xu, Z., Qi, X., Sun, P., Wang, J., Zheng, F., Li, X., Yin, R., Dallenbach, K. R., Bianchi, F., Petäjä, T., Zhang, Y., Wang, M., Schervish, M., Wang, S., Qiao, L., Wang, Q., Zhou, M., Wang, H., Yu, C., Yao, D., Guo, H., Ye, P., Lee, S., Li, Y. J., Liu, Y., Chi, X., Kerminen, V.-M., Ehn, M., Donahue, N. M., Wang, T., Huang, C., Kulmala, M., Worsnop, D., Jiang, J., and Ding, A.: Secondary organic aerosol formed by condensing anthropogenic vapours over China's megacities, Nature Geoscience, 15, 255-261, 10.1038/s41561-022-00922-5, 2022.

Yan, C., Shen, Y., Stolzenburg, D., Dada, L., Qi, X., Hakala, S., Sundström, A. M., Guo, Y., Lipponen, A., Kokkonen, T., Kontkanen, J., Cai, R., Cai, J., Chan, T., Chen, L., Chu, B., Deng, C., Du, W., Fan, X., He, X. C., Kangasluoma, J., Kujansuu, J., Kurppa, M., Li, C., Li, Y., Lin, Z., Liu, Y., Liu, Y., Lu, Y., Nie, W., Pulliainen, J., Qiao, X., Wang, Y., Wen, Y., Wu, Y., Yang, G., Yao, L., Yin, R., Zhang, G., Zhang, S., Zheng, F., Zhou, Y., Arola, A., Tamminen, J., Paasonen, P., Sun, Y., Wang, L., Donahue, N. M., Liu, Y., Bianchi, F., Daellenbach, K. R., Worsnop, D. R., Kerminen, V. M., Petäjä, T., Ding, A., Jiang, J., and Kulmala, M.: The effect of COVID-19 restrictions on atmospheric new particle formation in Beijing, Atmos. Chem. Phys. Discuss., 2022, 1-24, 10.5194/acp-2021-1079, 2022.

Yang, L., Nie, W., Liu, Y., Xu, Z., Xiao, M., Qi, X., Li, Y., Wang, R., Zou, J., Paasonen, P., Yan, C., Xu, Z., Wang, J., Zhou, C., Yuan, J., Sun, J., Chi, X., Kerminen, V.-M., Kulmala, M., and Ding, A.: Toward building a physical proxy for gas-phase sulfuric acid concentration based on its budget analysis in polluted Yangtze River Delta, East China, Environmental Science & Technology, 55, 6665-6676, 10.1021/acs.est.1c00738, 2021.

Zhang, R., Zhang, Y., Lin, H., Feng, X., Fu, T.-M., and Wang, Y.: NOx Emission Reduction and Recovery during COVID-19 in East China, Atmosphere, 11, 10.3390/atmos11040433, 2020.