Response to Reviewer #1's Comments

In this manuscript, the authors focused on the atmospheric physiochemical characterizations of a transported dust event that passed through the city of Shanghai and then circled back again. However, it was written in poor English and lacking for scientific questions/gaps, which made it just like a data analysis note of various instruments of a super monitoring site based on three stages of dust event. In general, this manuscript has no innovations and valuable contributions for the science community, and it should be reorganized and discussed with other authors. As it presented in low quality, and I cannot accept it for the publication in present status, and it should be rejected.

We thank for the reviewer's comments and suggestions on this manuscript. Based on the specific comments, we have substantially reorganized the whole manuscript and made numerous changes. We have responded to all the comments point-by-point and made corresponding changes in the manuscript as highlighted in the track change mode. Please check the detailed responses to all the comments as below, and we hope to receive further comments based on this version.

Major questions:

1. The title of this manuscript is obscure and unmatched to the research contents.

The original title is not precise. In the revision, the title is changed as "Secondary aerosol formation under a special dust transport event: impacts from unusually enhanced ozone and dust backflows over the ocean".

2. The Introduction part had lengthy writing and lacking for logics among paragraphs. Each paragraph also lacks the topic sentence. The mainstream references are lacking in this part. As mentioned above, the key defect is lacking of research gaps and scientific questions.

Thanks for suggestion. We have removed all the redundant references and writings. We

have also updated more related studies. Some paragraphs have been reorganized, and we have made the goal of this study more clearly written in the last paragraph in the introduction.

Since there are numerous changes in the introduction section, please check the changes in **Line 59 - 191**.

3. The math formulas should be provided in the methodology part, such as Lines 292-293 and Lines 535-550.

Thanks for the suggestion. The formulas in the original Lines 292-293 have been moved to the methodology part. As for the method in Lines 535-550, it is based on the discussion of dust backflows, thus we prefer to state it in the main text.

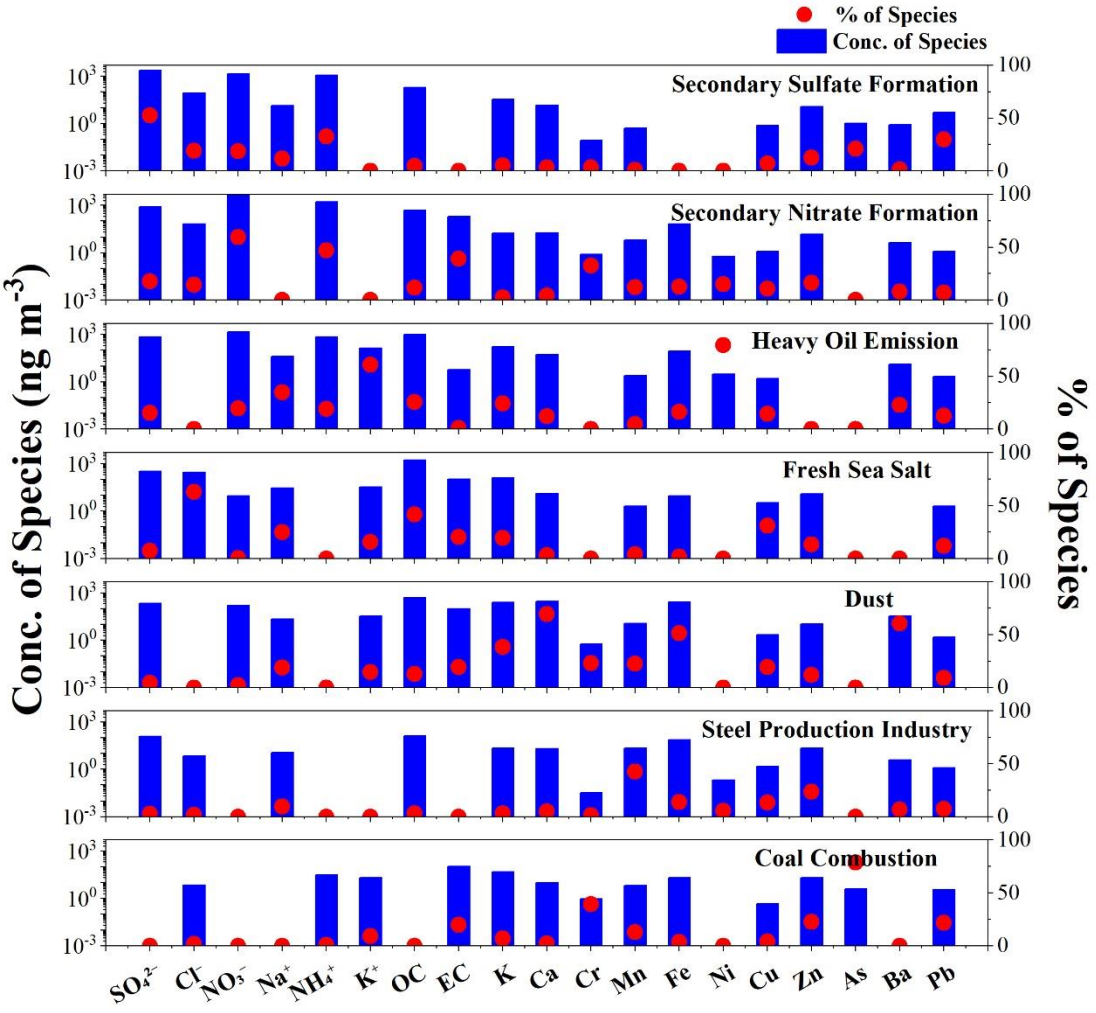
In the revision, we added a section about the uptake coefficient of NH_3 (γ_{NH3}) on particles in the methodology part. γ_{NH3} is used to deeply analyze the formation of secondary aerosol formation. Please check the changes in **Lines 263 - 277**.

4. The authors claimed that they develop "a simple method of relating the upstream and receptor simultaneous measurements", however, this method lacks for the basic scientific verifications, such as comparisons with other source apportionment methods (PMF, CMB, etc.).

Thanks for the suggestion and we do agree that this method should be compared with other methods. We have conducted PMF modeling and seven sources are identified as shown in the figure below. It can be seen sulfate and nitrate have very low loadings in the identified dust factor or sea salt factor. This means dust have contributed very little to the secondary aerosols based on the PMF results. This is expected as although PMF is a powerful source apportionment tool, it is not designed to apportion the contributions from secondary formation and long-range transport.

In this study, dust transport is frequent and the formation of sulfate and nitrate evidently occurred during the transport. However, it is clear that both the local formation and long-range transported sulfate and nitrate have been grouped into Factor 1 and Factor 2 of the PMF modeling. Thus, we need to develop other methods for the separation of

transported aerosols and secondary formed aerosols.

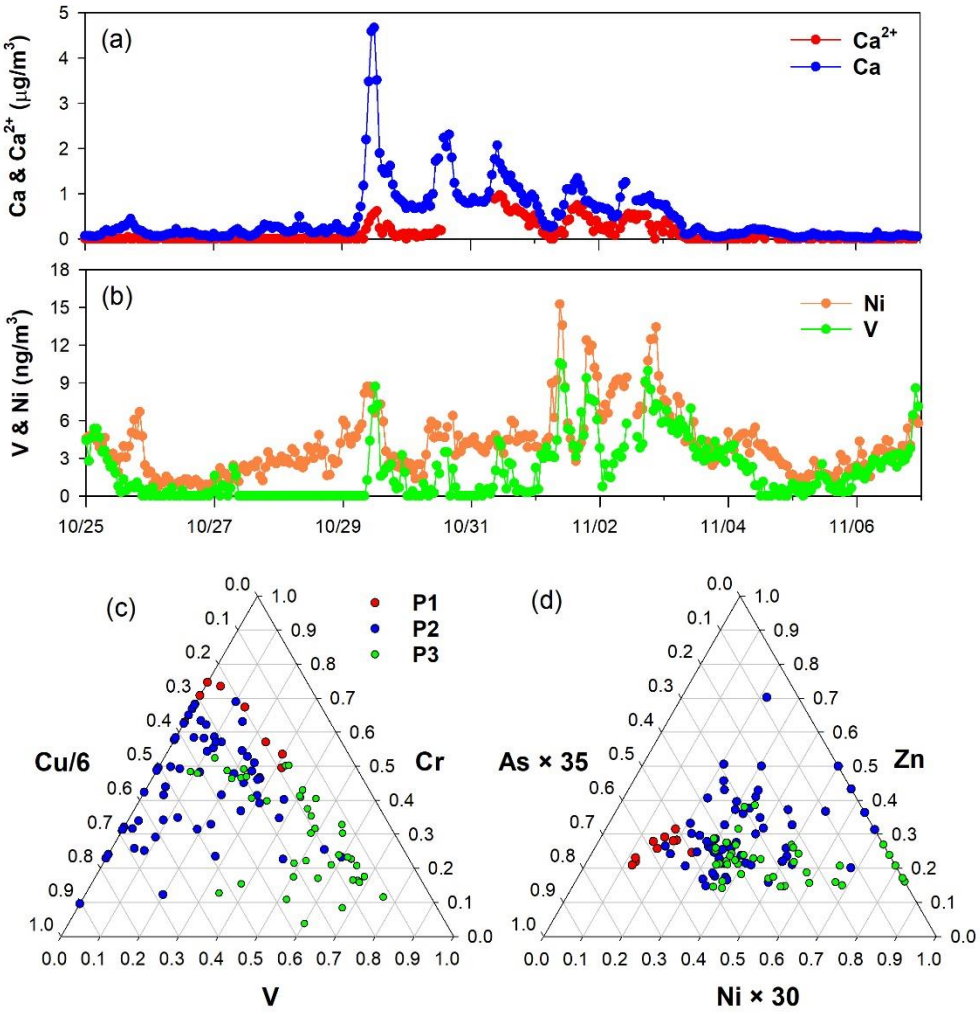


5. Too many figures in this manuscript, and the authors can only use the linear analysis in them, and this reflects that this manuscript is written by a student and lacking for the basic guidance.

Thanks for the suggestion and we quite agree with reviewer that linear analysis is overused in the original manuscript. In the revision, we have removed all figures from Figure 6 to Figure 12. The original writings in Section 3.3 & 3.4 have been almost deleted. We have fully reorganized the analysis and the major changes are summarized below.

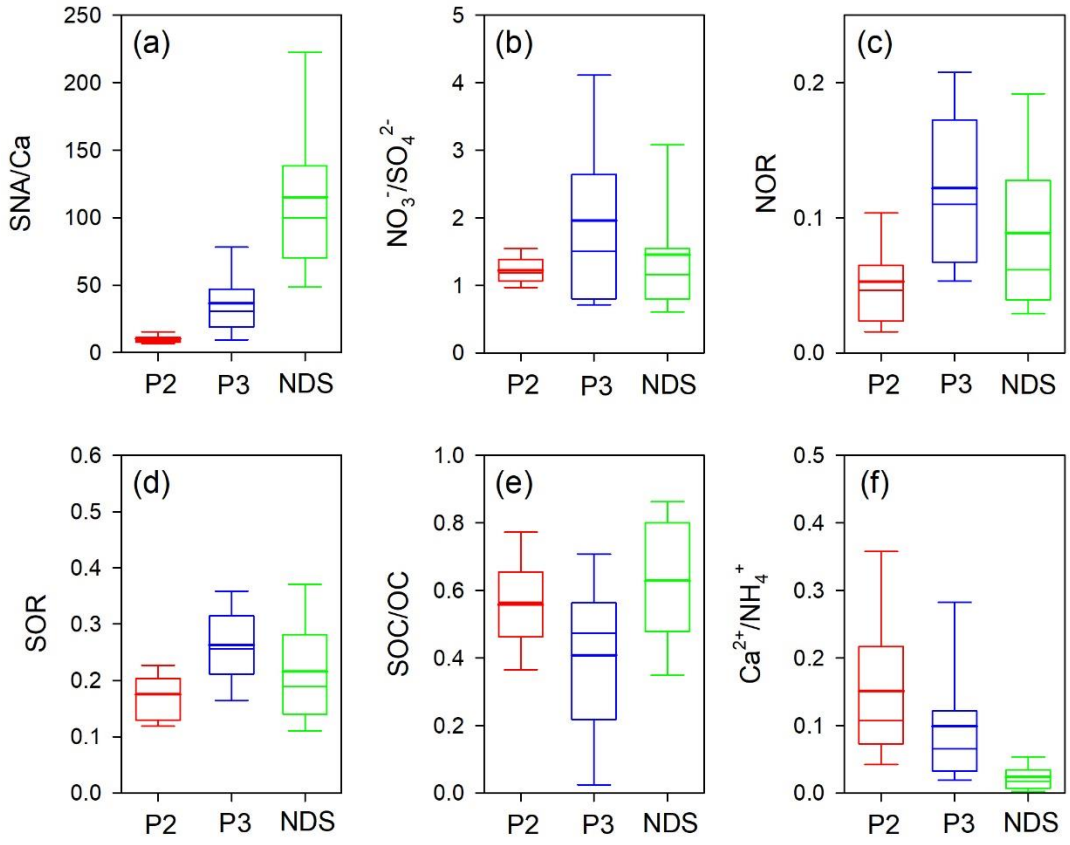
3.3.2. Dust backflows during P3

In this section, Ternary diagrams of typical metals are added to support the analysis of dust backflows. Please check the detailed writing in **Lines 541 – 581.**



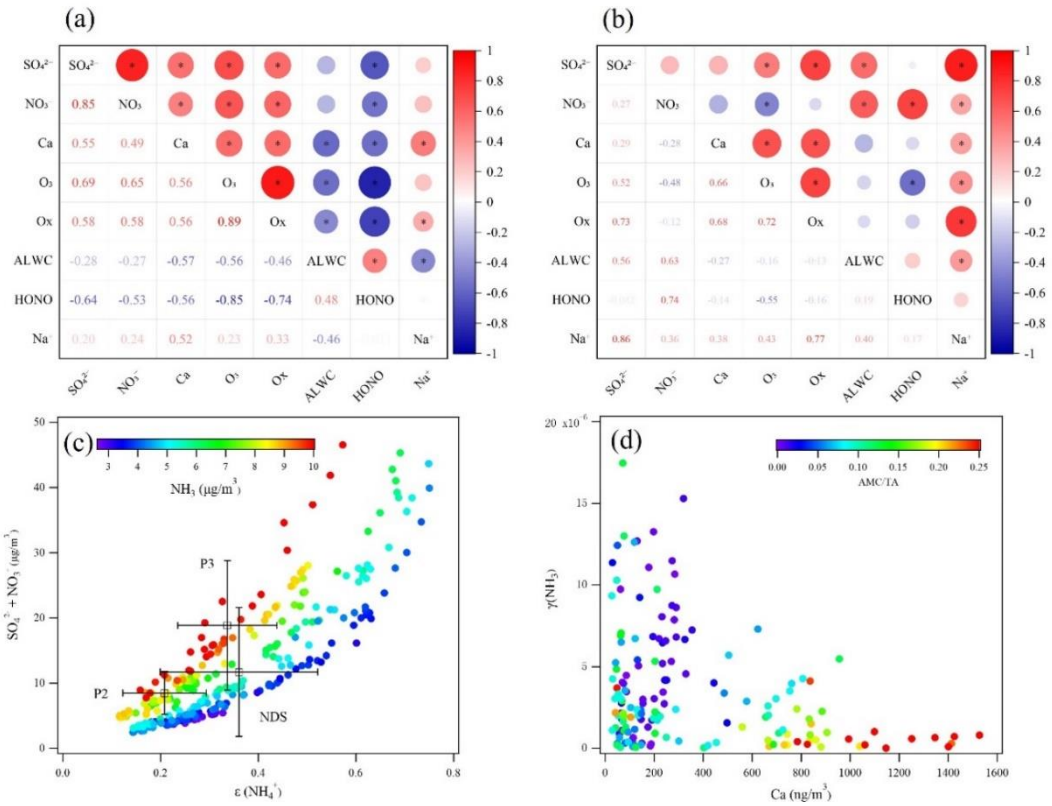
3.4.1. Comparison of typical chemical tracers

In this section, we analyzed a set of chemical tracers to gain insights into the formation characteristics of secondary aerosols in different stages, please check the full writings in **Lines 583 – 621.**



3.4.2. Distinct formation processes of secondary aerosols between P2 and P3

In this section, we further analyze the formation mechanism and key influencing factors of secondary components during P2 and P3. Major changes are made as below. (1) The correlation heatmaps of SO_4^{2-} and NO_3^- with various parameters are compared between P2 and P3. (2) The role of the gas-particle partitioning of NH_3 in the formation of secondary aerosols is analyzed. (3) The uptake coefficient of NH_3 on particles is applied to explain the aerosol formation potentials in different stages. Please check the full writings in **Lines 623 – 705.**



The whole paper needs to be rewritten in scientific language and vocabularies, and it is necessary that a native English speaker need to help the authors to revised it word by word.

This manuscript is now fully rewritten after polishing the language and vocabularies. It is revised by a native English speaker. Please check the track changed version to see all the changes.

Response to Reviewer #2's Comments

Desert dust aerosol has great impacts on atmospheric chemistry. Although there are numerous field measurements in this field, the work by Lu et al. is very interesting as the dust event examined was very unique, characterized by low wind speed, high RH, higher concentration of reactive trace gases, and dust backflow. This work can be published after the following comments are addressed.

We thank for the reviewer's positive comments and helpful suggestions on this manuscript. Based on the specific comments, we have responded to all the comments point-by-point and made corresponding changes in the manuscript as highlighted in the track change mode. Please check the detailed responses to all the comments as below.

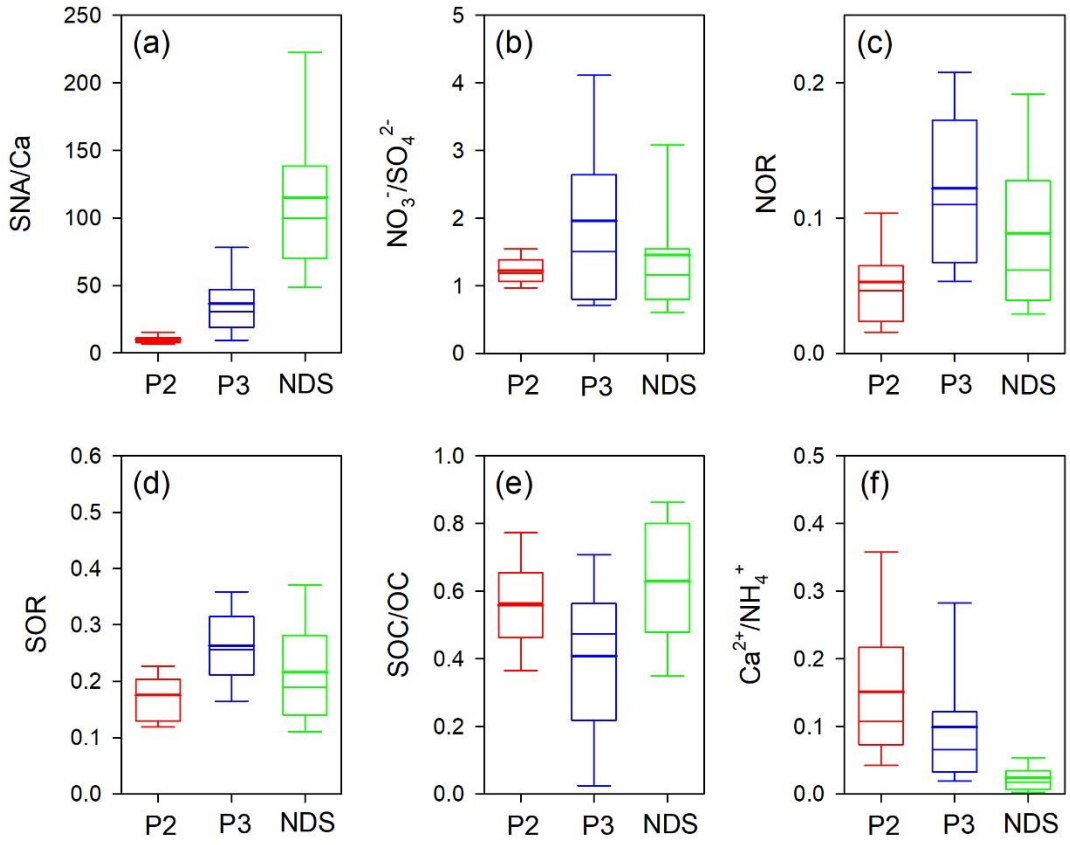
Major comments: I have to admit that I am not familiar with field observation work, and therefore the editor may invite colleagues with field observation expertise to assess this manuscript. In my opinion, the phenomena Lu et al. observed and tried to interpret is very interesting, but the discussion can still be improved. For example, the two most important sections (3.3 and 3.4) are quite descriptive, and conclusions they drew are not very well supported in the current version. Therefore, further data analysis is encouraged, and the authors may think about if they could deeper insights into dust chemistry from the unique event they observed.

Thanks for the comments. We quite agree with the reviewer that the analysis in the original version was quite descriptive and more deeper data analysis is needed. In the revision, we have removed all the descriptive analysis and related discussions in Section 3.3 and 3.4. Instead, we added two new sections in the revised manuscript.

3.4.1. Comparison of typical chemical tracers

In this section, we analyzed a set of chemical tracers to gain insights into the formation characteristics of secondary aerosols in different stages, please check the full writings

in **Lines 583 – 621.**



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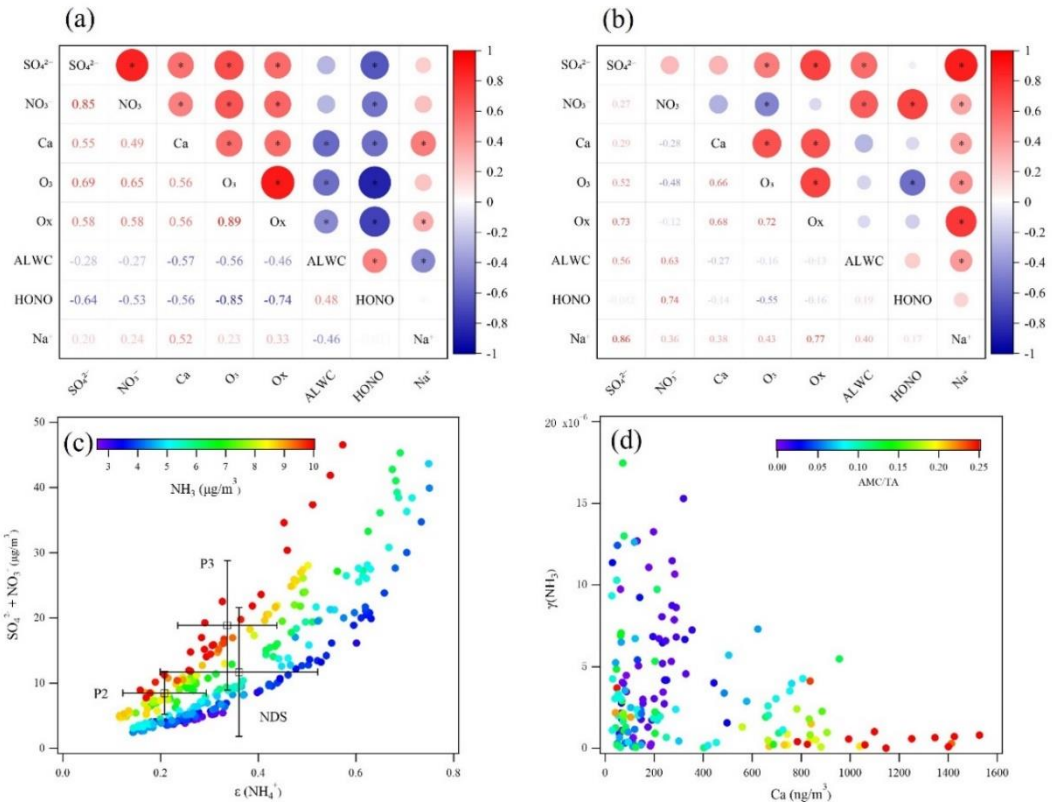

Minor comments:

Line 53: The authors may consider citing a recent review paper (Tang et al., 2017) which discussed impacts of dust on tropospheric chemistry.

Thanks for proving the information. The reference is added as below.

Tang et al. (2017) conducted a comprehensive review on the effect of dust heterogeneous reactions on the tropospheric oxidation capacity. They proposed that high RH (e.g., > 80%) and a wider range of temperature should be considered in the laboratory studies of heterogeneous reactions of mineral dust. Also, more comprehensive kinetic models should be developed to understand the complex multiphase reactions.

Line 71: Guo et al. (2019) investigated hygroscopicity of a number of Ca and Mg salts, and suggested that atmospheric aging could greatly enhance hygroscopicity of mineral dust.

Thanks for proving the information. The reference is added as below.

Guo et al. (2019) investigated the dependence of deliquescence relative humidities on temperature and hygroscopic properties of eight Ca- and Mg-containing salts. It was found that the hygroscopic growths of some salts were significant at 90 % RH, implying that favorable environmental conditions could greatly enhance the hygroscopicity of mineral dust.

References:

Tang, M. J., Huang, X., Lu, K. D., Ge, M. F., Li, Y. J., Cheng, P., Zhu, T., Ding, A. J., Zhang, Y. H., Gligorovski, S., Song, W., Ding, X., Bi, X. H., and Wang, X. M.: Heterogeneous reactions of mineral dust aerosol: implications for tropospheric oxidation capacity, Atmos. Chem. Phys., 17, 11727-11777, 2017.

Guo, L., Gu, W., Peng, C., Wang, W., Li, Y. J., Zong, T., Tang, Y., Wu, Z., Lin, Q., Ge, M., Zhang, G., Hu, M., Bi, X., Wang, X., and Tang, M.: A comprehensive study of hygroscopic properties of calcium- and magnesium-containing salts: implication for hygroscopicity of mineral dust and sea salt aerosols, Atmos. Chem. Phys., 19, 2115–2133, https://doi.org/10.5194/acp-19-2115-2019, 2019.

Line 379-382, line 395-398: Sulfate and nitrate formation is quite complicated in the troposphere. These conclusions currently drawn by the authors are merely based on correlations, and further discussion is needed.

Thanks for the comments and we quite agree. As responded above, we have removed almost all the original writings and added deeper analysis in Section 3.4. Please check the detailed writings in **Lines 583 – 705.**

Line 436-451: Sea spay aerosols also contain soluble Ca2+. The authors may want to subtract the contribution of sea spray aerosol on soluble Ca2+ using the Ca2+/Na+ ratio in sea water.

Thanks for the comment. It is a good idea that the contribution of sea spray aerosol on soluble Ca^{2+} can be quantified using the Ca^{2+}/Na^{+} ratio in sea water. However, considering that during the dust period, Na^{+} could be derived from both dust and sea salts. Thereby, the contribution of sea spray aerosol on soluble Ca^{2+} could be overestimated by using $(Ca^{2+}/Na^{+})_{seawater} * Na^{+}_{aerosol}$. In the revision, we have added the sentence "Additionally, the backflow transport pathway facilitated the entrainment of sea salts and contributed to the increase of soluble calcium." in **Lines 558 - 559.**

Line 493-494: This sentence sound strange. I am not sure if sulfate can be aged in the atmosphere, as sulfate cannot be further oxidized. In addition, the co-variation of sulfate with Na+ may indicate important contribution of sea spray aerosol (which is a major source of Na+) to sulfate.

Thanks for pointing out this incorrect statement. We didn't mean sulfate can be aged but meant to say part of sulfate was formed in the upstream regions and transported with the sea spray aerosols. In the revision, we have re-written this paragraph in Lines 655 - 667.

In addition, unlike P2, both SO_4^{2-} and NO_3^{-} showed moderate to significant correlations with Na^{+}. Since neither SO_4^{2-} nor NO_3^{-} correlated with Ca, it can be inferred that sea salts played a more important role in the transport of air pollutant during the dust backflow over the ocean. To assess whether dust or sea salts participated in the heterogeneous reactions of secondary aerosol during P3, the ISORROPIA II model was run with different scenarios. Figure S5 shows the model performance for SO_4^{2-}, NO_3^{-}, NH_4^{+}, and NH_3 based on the $SO_4^{2-}-NO_3^{-}-NH_4^{+}-Cl^{-}-NH_3-HCl-HNO_3$ system. After adding Ca^{2+} into this thermodynamic equilibrium system, the correlations between the simulations vs observations for all four species were lowered with different extents (Figure S6). If Na^{+} was added into the thermodynamic equilibrium system. the model performance was slightly improved (Figure S7). This corroborated that the heterogeneous reactions on dust were very limited while sea salts were intensively involved in the formation of secondary inorganic aerosols during the dust backflow.

References: Tang, M. J., Huang, X., Lu, K. D., Ge, M. F., Li, Y. J., Cheng, P., Zhu, T., Ding, A. J., Zhang, Y. H., Gligorovski, S., Song, W., Ding, X., Bi, X. H., and Wang, X. M.: Heterogeneous reactions of mineral dust aerosol: implications for tropospheric oxidation capacity, Atmos. Chem. Phys., 17, 11727-11777, 2017.