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Reply on RC1

Liang Xu et al.

Author comment on "Long-range transport of anthropogenic air pollutants into the marine air: insight into fine particle transport and chloride depletion on sea salts" by Liang Xu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-623-AC1, 2021

Anonymous Referee #1

General Response: We thank the Referee for your helpful comments. We have addressed all comments and provided point by point response below. The revised manuscript is presented in below Response.

This manuscript presented the characteristics of aerosol particles collected from eastern China sea (ECS) and northwestern Pacific Ocean (NWPO) based on particle analysis by TEM. The results showed during the cruise the samples collected in ESC contained more anthropogenic particles than those in NWPO. The chloride depletion in the sea salt particles was also discussed. The manuscript find that the particles may have been impacted by the long range transported anthropogenic pollutants. This study provides a case study in these areas for the better understanding in the impact of anthropogenic emission on the marine environments. The scope of this manuscript is suitable for this journal. A set of issues and comments need to be considered before publication.

Answer: We appreciated the Referee#1's comments which significantly improve the quality of the manuscript. We carefully answer them one by one as below. The modifications were highlighted in red in the revised manuscript.

Comments:

1, The manuscript presented a new set of data for this area, but the general implications in the current version are somewhat limited or not clearly discussed. It is suggested to re-categorize the manuscript as a measurement report. The manuscript stated that the acidic gas precursors transported for a longer distance and thus posed a significant impact on particles in NWPO which can't directly concluded from the evident or data of particle characteristics presented. The conclusion in Line 31-35 is overstated. In addition, in those

11 samples from NWPO, there are only two samples that contain S-rich particles over 15% in number. Also, most of these from 11 samples from NWPO are originated from Sea of Japan and were transported to NWPO within two days, that means those sea salt particles may have been aged when passing Japan.

Answer: (1) Our primary thought about this paper was to discuss the impacts of long-range transported anthropogenic air pollutants to the marine air. We carefully revised the paper and significantly improved the quality. We believe that the revised manuscript is more suitable as a research article.

(2) The conclusion in Line 31-35 was revised to avoid overstatement.

Context: Our results highlight that anthropogenic acidic gases in the troposphere (e.g., SO_2, NO_x, and volatile organic compounds) could be transported to remote marine air and exert a significant impact on aging of SSA particles in the NWPO. The study shows that anthropogenic particles and gases from East Asia significantly perturb different aerosol chemistry from coastal to remote marine air.

(3) Samples 11 and 12 contain S-rich particles over 15% in number. The high S-rich fraction in these two samples was attributed to new particle formation event caused by long-range transported continental gases (Zhu et al., 2019, New particle formation in the marine atmosphere during seven cruise campaigns, Atmospheric Chemistry and Physics).

(4) If sea salt particles were from Sea of Japan, passing through Japan to our sampling site, we would expect more anthropogenic aerosols (e.g., S-rich, soot, metal, etc.) found in the NWPO samples. However, anthropogenic aerosols accounted for small fraction. We do think that sea salt particles aging when passing Japan was not the case in our study.

2, Line 27-28, I didn't find the definition for the primary and secondary anthropogenic aerosols in the text based on these TEM analysis or particle classification.

Answer: In this study, primary anthropogenic aerosols include OM, soot, metal, and fly ash particles. Secondary anthropogenic aerosols are S-rich and OM coating particles. In the manuscript, we revised the "primary and secondary anthropogenic aerosols" to "anthropogenic aerosols" and provided a brief definition of anthropogenic aerosols in Abstract and Section 3.1.

Context: Anthropogenic aerosols (e.g., sulfur (S)-rich, S-soot, S-metal/fly ash, organic matter (OM)-S, and OM coating particles) were identified and dramatically declined from 87% to 8% by number from the ECS to remote NWPO.

3, Line 35-38, The statement doesn't provide useful information or conclusion for this manuscript.

Answer: We revised the sentence.

Context: More attention should be given to the modification of SSA particles in remote marine areas due to the influence of anthropogenic gaseous pollutants.

4, Line 49-51, Chloride depletion doesn't release I-containing compounds.

Answer: Sorry for the misleading, we removed the sentence from the manuscript.

5, Line 55-58, The references are not related to ice nuclei and so the statement need more related references for aged SSA serving as IN.

Answer: We added more references about SSA serving as IN.

Kanji, Z. A., L. A. Ladino, H. Wex, Y. Boose, M. Burkert-Kohn, D. J. Cziczo, and M. Krämer (2017), Overview of Ice Nucleating Particles, Meteorological Monographs, 58, 1.1-1.33.

Kong, X., M. J. Wolf, M. Roesch, E. S. Thomson, T. Bartels-Rausch, P. A. Alpert, M. Ammann, N. L. Prisle, and D. J. Cziczo (2018), A continuous flow diffusion chamber study of sea salt particles acting as cloud nuclei: deliquescence and ice nucleation, Tellus B: Chemical and Physical Meteorology, 70(1), 1-11.

6, Line 72, Please rewrite this sentence, it is not very clear what that means.

Answer: We revised the sentence for better understanding.

Context: Therefore, it is important to understand the physicochemical properties of long-range transported anthropogenic aerosol particles in marine air.

7, Line 152-155, Line 176-180, the classification scheme is not clear and somehow mixed. Please use the same definition for the particle types. Line 157-159, how does two types of OM particles were distinguished based on the TEM analysis?

Answer: Sorry for the confusion.

S-rich, OM, soot, metal, fly ash, mineral , and sea salt (Line 152-155) are seven basic aerosol components in this study. However, most of the aerosol particles observed in this study contained two or more different aerosol components. We further defined six internally mixed particles to elucidate the mixing states of aerosol particles (Line 176-180). S-metal refers to metal particles mixed with sulfate (Figure 2c). S-fly ash refers to fly ash particles mixed with sulfate (Figure 2d). S-soot refers to soot particles mixed with sulfate (Figure 2e). OM coating refers to secondary organic matter coated on sulfate (Figure 2f). OM-S refers to primary OM particle mixed with sulfate. S-rich refers to secondary inorganic particles (e.g., $(NH_4)_2SO_4$ and NH_4NO_3), which are formed from their gaseous precursors, such as SO_2, NO_x, and NH_3 (Figure 2h).

In the TEM images, the POM particles normally have a spherical or irregular shape. The SOM particles display a core-shell structure, which usually represents an inorganic core (e.g., sulfate) coated by secondary organics.

We also revised these sentences in the manuscript for clear understanding.

Context: To elucidate the mixing structure of the non-SSA particles, we further defined six types of internally mixed particles: S-metal, metal particles mixed with sulfate (Figure 2c); S-fly ash, fly ash particles mixed with sulfate (Figure 2d); S-soot, soot particles

mixed with sulfate (Figure 2e); OM coating, secondary organic matter coated on sulfate (Figure 2f); OM-S, primary OM particle mixed with sulfate (Figure 2g); and S-rich, secondary inorganic sulfate and nitrate particles (e.g., $(NH_4)_2SO_4$ and NH_4NO_3) (Figure 2h).

In the TEM images, the POM particles normally have a spherical or irregular shape. The SOM particles display a core-shell structure, which usually represents an inorganic core (e.g., sulfate) coated by secondary organics (Li et al., 2016b).

8, Line 198-199, Is the 5% significantly higher than 2% based on the number of particles analysis using TEM? Have you considered the uncertainties in the classification since the total number of particles analyzed are low?

Answer: Although the total number of mineral particles analyzed in this study is small, the uncertainty in mineral particles based on manual identification is low. We can clearly distinguish mineral particles under high-resolution TEM due to their unique morphology and chemical composition. In order to avoid potential misunderstanding, we revised the "twice as high as" to "higher than".

Context: Altogether, the number fraction of mineral particles was 5% in the ECS, higher than that in the NWPO.

9, Line 231, there is no evidence showing that S-rich particles are formed from the precursors.

Answer: Sorry for the confusion. We acknowledge that we do not have evidence to prove it. Thus, we removed the sentence from the manuscript.

10, Line 310, This statement doesn't need a figure here. There is no further description for this Figure 9.

Answer: We added one more paragraph to describe the schematic diagram in Figure 9.

Context: Based on the results and discussion above, a conceptual model was proposed to summarize the impact of long-range transported anthropogenic air pollutants on marine aerosols. Both anthropogenic gases and aerosol particles could be transported to downwind marine areas. Anthropogenic aerosol particles from the continent significantly influence the ECS air. During the transport, aerosol particles could be scavenged due to dry or wet deposition while some reactive gases can be transported further to the NWPO air and influence the aging of SSA particles.

11, Line 323-326, It is not clear what does the authors try to explain.

Answer: Sorry for the confusion.

Particles above the dash line represent that Cl in the SSA particles was not only replaced

by S deposition. For the fully aged SSA particles, 70% of them in the ECS were above the dash line, while the proportion increased to 87% in the NWPO. The result indicates that S deposition could not compensate Cl-depletion in most of the fully aged SSA particles in the ECS and NWPO. There must be other acids leading to Cl-depletion in the fully aged SSA particles besides reaction with H_2SO_4.

Context: Thus, particles above the dash line represent that Cl in these SSA particles was not only replaced by S deposition, other chemical processes (e.g., react with HNO_3 and organic acids) also contributed to the Cl-depletion. The number of fully aged SSA particles above the dash line was further counted. For the fully aged SSA particles, 70% of them in the ECS were above the dash line, while the proportion increased to 87% in the NWPO (Figure 6). The result indicates that S deposition could not compensate for Cl-depletion in most of the fully aged SSA particles in both ECS and NWPO. There must be other acids leading to Cl-depletion in the fully aged SSA particles besides the reaction with H_2SO_4.

Here we also revised the Figure 6.

Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2021-623/acp-2021-623-AC1-supplement.pdf