Response to the comments of Reviewer#2:

We appreciate the reviewer's helpful and constructive comments on the manuscript entitled "Trace elements in $PM_{2.5}$ aerosols in East Asian outflow in the spring of 2018: Emission, transport, and source apportionment". As the reviewers suggested, we have modified the manuscript. Major points for the revisions are listed as follows.

1) The improvements on the model predictions of Cu concentrations were added. Considering the Cu smelting as an additional source of Cu in the improved simulation allowed us to correct the underestimation of Cu concentrations in the base simulation.
2) In relation to 1), new figures were added to the supplement information (Figure S6 and S7 in the revised SI) and some figures shown in the original manuscript were modified.

*Note that the authors' replies were in red.

Abstract:

Ln 10: 1st sentence can be changed to 'Trace metals in aerosol particles impact Earth's radiative balance, ocean biogeochemistry, and human health.' (as mentioned in the introduction). Also removing 'therefore' from the second sentence is suggested since it is not a conclusion for the previous one.

We revised as suggested.

"Trace metals in aerosol particles impact Earth's radiative balance, human health, and ocean biogeochemistry. Semi-continuous measurements of the elemental composition of fine mode ($PM_{2.5}$) aerosols were conducted using an automated X-ray fluorescence analyzer on a remote island of Japan during the spring of 2018."

Ln 26: Using the word 'Minor' for anthropogenic contribution of Fe is not recommended. Maybe using the term 'not dominant' like used in conclusions is better.

We revised as suggested.

"However, despite the non-dominant anthropogenic contributions of Fe, they could adversely affect human health and ocean biogeochemistry owing to their higher water solubility."

Introduction:

Ln 42: Change 'has been concerned' to 'has been a concern'.

We revised as suggested.

"The oxidative potential of aerosol particles, which have the potential to generate ROS in cells and cause oxidative stress to cells, has been a concern."

Materials and methods:

Ln 118: Was the 4h analysis time period used so that sufficient mass could be collected on the tape, or for some other reason?

We operated the PX-375 for 4 hours of the sampling and 4000 sec of the following XRF analysis in this study. As suggested, the measurement needs the enough amounts of aerosols on a spot on the filter tape. At the same time the shorter time resolution was preferable to compare the temporal variations of trace metals with those of tracer compounds (BC and CO).

Ln 128: How were these uncertainties calculated?

The uncertainties were evaluated by considering the intercomparison between the XRF analyses by the PX-375 and the reference analyses (IC and ICP-MS). The linear regression slopes were regarded a measure of the accuracy and the ratio of 95% confidence intervals to the average of the slopes were regarded a measure of the precision. The uncertainties estimated in the original manuscript were evaluated by combining both (i.e., root-sum-square). Therefore, the uncertainties were evaluated relative to the reference analyses and cloud be rephrased by the relative errors. The sentence was revised as follows.

"Based on the intercomparison results (linear regression slopes and their variances), the relative errors in the on-site measurements of the concentrations of Cl, S, Fe, Pb, Mn, Cu, K, and Ca were evaluated to be 66%, 6%, 26%, 15%, 23%, 30%, 28%, and 33%, respectively"

Ln 146: Is there a particular reason why three-day APT was considered?

We followed our previous studies (Kanaya et al., 2016; 2020; Miyakawa et al., 2017). Typical transport time from the East Asian continent to the observation site in Fukue island was ~2 days. To cover the period of the air mass transport from the source regions, we set 3 days for calculating the APT.

Results:

Ln 250: States that 'Cu has different emission sources from BC, Pb and CO'. The correlations of Pb and Cu v/s BC/CO are not drastically different, so can that really imply a different emission source altogether?

Figures 3c and 3d indicated only the differences in r^2 of the correlation with BC or CO between Pb and Cu, indicating the possibility of the differences in the emission sources. We modified the sentence as follows.

"This indicates that Cu possibly has different emission sources from BC, Pb, and CO in East Asia, even though the similarity of their geospatial patterns was indicated by the CWTs for BC, Pb, and Cu."

Ln 690: Fig 4 and 5 give units of APT as mm, whereas Fig 3 gives it as mm h. Which is the correct one? Please check other figs in SI as well.

"mm" is correct. We modified as suggested.

Ln 295: Looking at Fig 5, it seems that Cu is simulated well by the IMPACT model, due to its consistent M/O across different APT ranges, but the text states otherwise. Can you elaborate more on this?

As suggested, only looking at Fig. 5 let us find that M/O of Cu did not largely vary across different APT ranges. However, we found the relationship between M/O and transport efficiency of Cu, as shown in Figure S6 (S8) in the original (revised) SI. (This point has been described at the sentences Ln. 295–297 of the original manuscript.) We thus concluded that the APT was not a strong forcing factor to account for the model underestimations of Cu concentrations but the removal processes of aerosols (as seen in TE_{BC}, TE_{Pb}, and TE_{Cu}) were not properly simulated in the base and improved versions of the IMPACT model. We modified the sentence Ln. 295–296 as follows.

"Although the APT was not a strong forcing factor of TE_{Cu}, the removal processes of Cu were not properly simulated by the IMPACT model, as indicated by the relationship between the M/O ratios and TE for BC, Pb, and Cu, as shown in **Figure S8**. We found that the wet removal of aerosols in the IMPACT model need to be revised."

Conclusions:

Ln 448: States that 'emission sources of these metals share the region where the large CO (and BC) emission sources are located.' I understand that this conclusion is based on CWT results. However, it is confusing to understand that Cu has a different emission source based on correlations but a similar region of emission. Please explain this part more clearly, and make edits to similar sentences made in the abstract and results too.

The sentence at Ln 17–19 was modified as follows.

"Positive correlations of Pb and Cu with BC and CO and the similarity of their concentration-weight trajectories indicated that the emission sources of these metals share the region where the large CO (and BC) emission sources are located and that CO can be regarded as a tracer of continental anthropogenic emissions."

The sentence at Ln 447–449 was modified as follows.

"Positive correlations of Pb and Cu with BC and CO and the similarity of their CWTs indicated that the emission sources of these metals share the region where the large CO (and BC) emission sources are located."

Ln 460: BC is used as a tracer for anthropogenic emissions. What is novel about this part?

BC is a well-known tracer for combustion sources (not only anthropogenic but also biomass burning (BB)). From the tagged tracer analysis in the IMPACT simulations, it was found in this study that BC mainly originated from anthropogenic combustion source (BB was not significant) in the East Asian outflow in the spring of 2018. To the best of our knowledge, for the first time, "the combination of BC and Si" in the multiple linear regression was used for the observation-based source apportionment of Fe and Mn.

Reference papers newly included in the revised manuscript

Barcan, V.: Nature and origin of multicomponent aerial emissions of the copper-nickel smelter complex. Environ. int., 28(6), 451–456, 2002.

Fang, T., Guo, H., Zeng, L., Verma, V., Nenes, A., and Weber, R. J.: Highly Acidic Ambient Particles, Soluble Metals, and Oxidative Potential: A Link between Sulfate and Aerosol Toxicity. Environ. Sci. Technol., 51(5), 2611–2620, https://doi.org/10.1021/acs.est.6b06151, 2017.

Hagino, H., Oyama, M., and Sasaki, S.: Laboratory testing of airborne brake wear particle emissions using a dynamometer system under urban city driving cycles. Atmos. environ., 131, 269-278, https://doi.org/10.1016/j.atmosenv.2016.02.014, 2016

Ito, A. and Feng, Y.: Role of dust alkalinity in acid mobilization of iron. Atmos. Chem. Phys., 10, 9237–9250, https://doi.org/10.5194/acp-10-9237-2010, 2010.

Ito, A. and Xu, L.: Response of acid mobilization of iron-containing mineral dust to improvement of air quality projected in the future. Atmos. Chem. Phys., 14, 3441–3459, https://doi.org/10.5194/acp-14-3441-2014, 2014.

Ito, A., and Kok, J. F.: Do dust emissions from sparsely vegetated regions dominate atmospheric iron supply to the Southern Ocean?. J. Geophys. Res. Atmos., 122, 3987–4002, https://doi.org/10.1002/2016JD025939, 2017.

Skeaff, J. M., Thibault, Y. and Hardy, D. J.: A new method for the characterisation and quantitative speciation of base metal smelter stack particulates. Environ. Monit. Assess., 177, 165–192, https://doi.org/10.1007/s10661-010-1627-9, 2011.

Sorooshian, A., Csavina, J., Shingler, T., Dey, S., Brechtel, F. J., Saez, A. E., and Betterton, E. A.: Hygroscopic and Chemical Properties of Aerosols Collected near a Copper Smelter: Implications for Public and Environmental Health. Environ. Sci. Technol., 46(17), 9473–9480, https://doi.org/10.1021/es302275k, 2012.

Yang, J., Ma, L., He, X., Au, W. C., Miao, Y., Wang, W.-X., and Nah, T.: Measurement report: Abundance and fractional solubilities of aerosol metals in urban Hong Kong - insights into factors that control aerosol metal dissolution in an urban site in South China. Atmos. Chem. Phys., 23, 1403–1419, https://doi.org/10.5194/acp-23-1403-2023, 2023.