Response to Reviewer 2 Comments

Dear Reviewer,

We would like to express our sincere gratitude for your time to review our manuscript and for the helpful suggestions to improve the article entitled "***Source-resolved atmospheric metal emissions, concentrations, and their deposition fluxes into the East Asian Seas***". We have carefully reviewed all the comments and revised the article accordingly. Please find the detailed responses below in blue and the corresponding revisions in track changes in the revised manuscript.

The reviewer's comments are in black.

The author's responses are in blue.

Revisions in the manuscript are in ***italics and bold*** (line numbers before and inside the bracket refer to those in revised manuscript with and without track of changes, respectively).

Specific comments:

1. First, the validation of the modeling results should be further discussed, which I think is very important for modeling work. Tables S9-10 were not discussed sufficiently. If there are measurements on surface concentrations of metals over lands and oceans, some detailed comparisons are needed (site/region/total average). You can refer to some similar works on N emissions, in which the validation works were made between modeling and measurements on land and oceans (see the supporting materials of papers: https://doi.org/10.1073/pnas.2121998119; https://doi.org/10.1073/pnas.2221459120).

   Response:

   Thank you for pointing this out. As you suggested, we have added a comparative analysis of the long-term monitoring data of $PM_{2.5}$-bound metals from the Shanghai Pudong site with the atmospheric metal concentrations obtained from the modelling to Page 10, Lines 254-258 of the revised manuscript, with a comparative graph Figure S4 (Supplementary Information Page 6, Lines 50-52). In addition to concentration comparisons over land, we used navigational measurements of atmospheric iron concentrations from the sea area for validation, as there are few long-term measurements in the sea area. On Page 10, Lines 252-254 of the revised manuscript *"The concentration of Fe was 201.1 ng·m^{-3} in the YS and 92.17 ng·m^{-3} in the ECS, and the contribution of land anthropogenic sources to the Fe concentration was 71.6% in the ECS, similar with the values reported by previous study (Zhang et al., 2024)."*

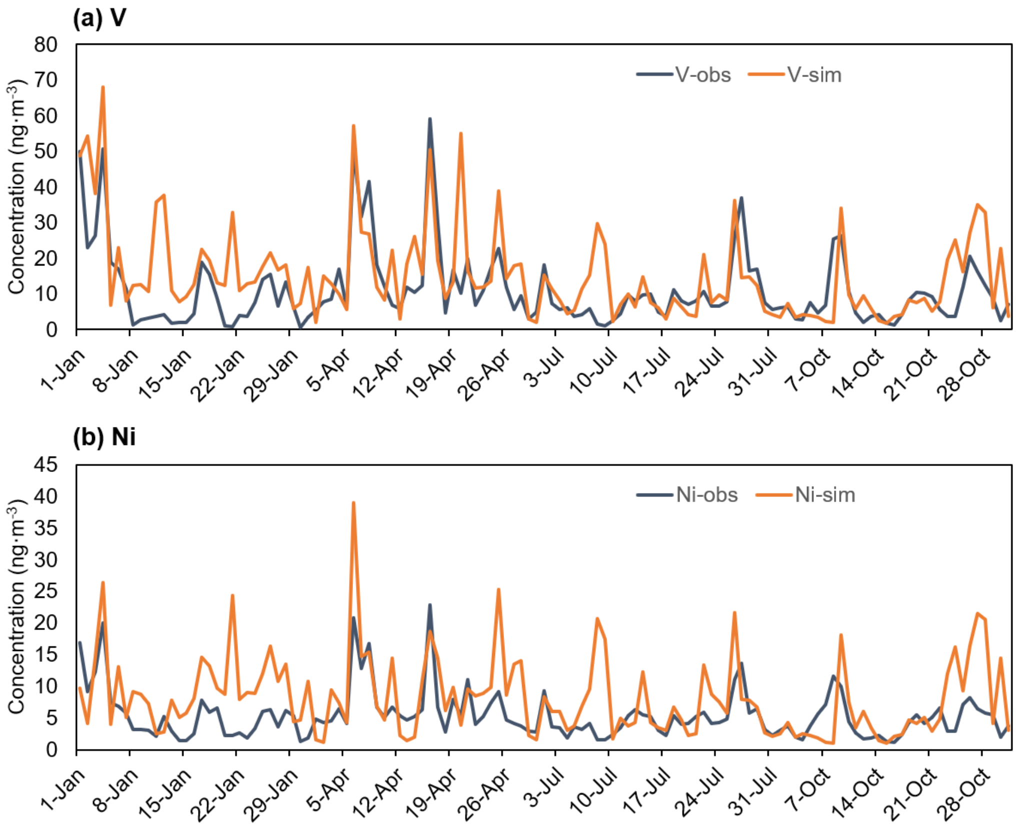
Revisions in the manuscript:

1. Page 11 (10), Lines 273-276 (254-258):

The available long-term and near real-time concentration monitoring data of V and Ni in the fine mode at the Pudong site (in Shanghai, China) obtained by Zou et al. (2020) were used to further validate the simulation of the model. As presented in Fig.S4, the simulated concentrations of V and Ni were in good agreement with the monitoring data, with respective normalized mean fractional bias (NMFB) and normalized mean fractional error (NMFE) of -0.31 and 0.37 for V and -0.38 and 0.40 for Ni.

2. Supplementary Information Page 6, Lines 50-52 (50-52):

Figure S4. Comparison of simulated daily concentrations of V (a) and Ni (b) with observations at the Pudong site (31.2331°N, 121.5447° E, Shanghai, China).



2. Second, are there any social-economic drivers for seasonal changes in metal emissions? For instance, when we talked about NH_3, it's usually controlled by increasing population and food production (N fertilizer, livestock). NH_3 seasonal changes are mainly affected by temperature and fertilizer applications. I hope to see some additional discussions on metal seasonal changes. How the urbanization affects metal emissions and pollution? (see refs on social-economic drivers on agricultural N emissions: L. Liu. 2023 Nature, https://doi.org/10.1038/d41586-023-02753-9; Deng et al. 2024 Nature communications, https://doi.org/10.1038/s41467-023-44685-y).

Response:

Thanks for your suggestions. After reading the literature you recommended we can understand the effect of socio-economic drivers on the seasonal characteristics of emissions. Especially for NH_3, which is highly correlated with agriculture, factors such as urbanization, population, fertilizer application, etc. can significantly affect emissions. As the base year of the emission inventory established in this study is 2017, there is no long-term data to support the analysis of the impact of industrialization progress or urbanization on emissions. Therefore, we limit our discussion to the seasonal changes of emissions, and a detailed discussion has been added on Page 6, Lines 166-183 of the revised manuscript.

Revisions in the manuscript:

1. Pages 6-7 (6), Lines 179-197 (166-183):

The monthly emission statistics of the three sources are detailed in Table S5-S9. *According to Table S5-S7, the overall quantity of metals emitted by ships was predominantly higher in summertime (July and August),* ***followed by*** *wintertime (November and December), while it was relatively lower in September.* ***This is related to the activities of ships, which are more active in the summer months and have higher emissions, a trend that has been reported in previous studies (Chen et al., 2018; Zhai et al., 2023). In terms of land anthropogenic sources, the emissions of all metallic elements in the fine mode were greater in winter (December and January) due to elevated heating demand, a seasonal feature consistent with previous studies (Luo et al., 2022; Zhao et al., 2021). The emissions of Fe, Al, and Ni in the coarse mode showed the same seasonal characteristics, while the highest emissions of V, Cu, and Zn occurred in April and October. Overall, the monthly variation of metal emissions from land anthropogenic sources was not as significant as that from ship sources, suggesting that***

metals could be emitted from stable sources such as industrial combustion (Zhang et al., 2018). Dust emissions were mainly concentrated in April, accounting for about 45% of the total annual emissions. In consideration of the significant seasonal variation, we counted the contribution of metals from the three emission sources in spring, as shown in Fig.S2. Dust sources were identified as the primary contributor to the coarse mode emissions of Fe and Al, accounting for a higher proportion of spring emissions than of annual emissions, 90.0% and 94.2% respectively. For the fine mode springtime emissions of these two metals, dust sources accounted for 51.9% and 61.8%, respectively, and were also the most significant source of emissions. There were also relatively high emissions in July and May, with the remaining months being insignificant. This is related to the fact that the dust events in East Asia occur mainly in spring (Gui et al., 2022; Hsu et al., 2010; Kang and Wang, 2005; Kang et al., 2016) and studies have also reported dust events in summer (Chen et al., 2014) and autumn (Zhang et al., 2015) in certain years.

References

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