

RC2

This study conducted observations at Fukue Island, located in the East Asian outflow region, from December 2023 to May 2024, covering both winter and spring seasons. The authors collected aerosol samples with different size fractions and analyzed a wide range of chemical components. Overall, this work provides a valuable observational dataset for improving our understanding of aerosol chemistry in the East Asian outflow region and its potential implications for ocean biogeochemistry. However, the current manuscript could be further improved in terms of data analysis and the interpretation of the results. Below, I provide several comments and suggestions that may help strengthen the manuscript.

We appreciate the reviewer's careful review of our manuscript and useful comments which helped improve its quality. The major revisions are summarized as follows:

1. The title was changed to "Size-segregated characterizations of Fe in aerosols in East Asian outflow in winter and spring: Source apportionment and bioaccessibility"
2. Revisions of the supporting information to support the data analyses such as multilinear regression used in this study and data interpretations.
3. Comparing Fe concentrations and fractional solubilities with those in several previous studies were included in Section 3.4.

*The comments from the reviewers are presented in black font in this document and the authors' responses to the comments are presented in red font.

Major Comments

(1) Line 262: The authors reported an uncertainty of 60% for Si. It would be helpful if the authors could provide further details on how this uncertainty was estimated. In addition, since Si was subsequently used as an important tracer for estimating dust contributions, the authors may consider discussing whether such a relatively large uncertainty could introduce substantial impacts on the derived dust contributions.

This value was derived from the comparison of Si concentrations with the independent measurements using an LVS at a different sampling site (Tamanoura) in Fukue island (Table S3 in SI of the original manuscript). As this comparison was not performed at the same site, this evaluation was not formal, and indeed we consider that this was a pessimistic evaluation. We modified this and relevant sentences as follows.

“The high-temporal resolution measurements of Si very well captured the variations in Si concentrations compared to those at Tamanoura site ($r^2 = 0.91$) but showed the systematic biases (about 60%). This can be caused by that measurements for Si quantification using PX-375 were not rigorously calibrated using ambient samples (see Section 2.1). We discussed the impacts of the uncertainties related to the systematic biases in Si concentrations at our site on the data analyses in Sections 3.3.1 and 3.3.2.”

As to the impacts on the multilinear regression analysis, this uncertainty does not affect the results so much. This is because the relative variations in concentrations are much more critical to constrain the regressed results than the absolute values of concentrations (as demonstrated in our previous study (Miyakawa et al., 2023)). Table S3 and Fig. S7 indicate that the high-temporal resolution measurements very well captured the variations in Si concentrations ($r^2 = 0.91$). This indicated that the data analysis using the temporal variations in Si concentrations was acceptable. In this study, we tested the case reducing the Si concentrations for $PM_{2.5}$ and PM_{10} aerosols and summarized the important fitted coefficients in Eqs. 1–4 in the following table. This table was included in the revised SI.

Table S4. The results of the sensitivity tests using different Si concentrations (base case and divided by 1.6) in the MLR analysis

Fitted coefficients	Base case	[Si]/1.6 case
$g_{BCPM_{2.5}}$	13.12	13.12
$g_{sulfatePM_{2.5}}$	1.78	1.78
$C_{PM_{2.5}}$	2.34	2.34
$g_{BCPM_{10}}$	12.12	12.07
$g_{sulfatePM_{10}}$	2.02	2.03
$C_{PM_{10}}$	3.75	3.76
$g_{BC,FePM_{2.5}}$	0.11	0.11
$g_{BC,FePM_{10}}$	0.15	0.15

It was found from this sensitivity test that the systematic difference in Si concentrations did not affect the source apportionments results to estimate the dust contributions to total and Fe mass for $PM_{2.5}$ and PM_{10} particles. In Sections 3.3.1 and 3.3.2, we added the following sentences,

respectively.

“The impacts of the uncertainties (i.e., possible systematic biases around 60% discussed in Section 3.1.2) related to Si concentrations ($[Si_{PM2.5}]_t$ and $[Si_{PM10}]_t$) to the MLR results of total mass (Eqs. 1 and 2) were evaluated to be very small (**Table S4**) by conducting the MLR analyses using the concentrations of Si divided by 1.6.”

“The impacts of the uncertainties related to Si concentrations to the MLR results of Fe content (Eqs. 3 and 4) were evaluated to be very small (**Table S4**) by the same manner as used for total mass (Section 3.3.1).”

Although Al was not measured in this study, the authors may consider evaluating the applicability of alternative dust tracers, such as Ti or non-sea-salt Ca (nss-Ca), for source identification. Furthermore, given that the observations were conducted at an island site, if the relevant data are available, the use of nss-SO₄²⁻ instead of total SO₄²⁻ as a tracer for secondary aerosol sources may also be considered.

We appreciate the suggestion. Although the suggestion is reasonable, in this study, sodium was not measured by the PX-375 instrument. So, the estimation of non-sea-salt components could not be performed. As suggested, Ti can be a candidate for the tracer of mineral dust particles. However, a previous study suggested the non-dust component substantially contributed to Ti-content in aerosols in the East Asian outflow region (Sakata et al., 2021).

(2) Section 3.3.1: For the source apportionment of PM_{2.5} and PM₁₀, the authors only considered BC, sulfate, and Si as source tracers, which may not fully represent the diverse sources contributing to atmospheric particulate matter. The authors have acknowledged this limitation and indicated that the intercept term in the multiple linear regression model represents contributions from other sources, such as sea salt. However, the use of the term “background levels (line 328)” in the current statement may require further clarification, as it could imply that contributions from these additional sources remain relatively constant. In fact, even sea salt emissions can exhibit temporal variability, for example, due to variations in wind speed and meteorological conditions across different seasons.

As suggested, the sources of PM_{2.5} and PM₁₀ aerosols are diverse and not always well represented

using the simple sets of tracers, especially near source regions. As the purpose of this analysis was to evaluate the contributions of dust to total aerosols for the transboundary transport events in the outflow regions, detailed source characterizations of total mass and Fe content of PM_{2.5} and PM₁₀ aerosols are out of the scope of this study. So, we modified the title of subsections 3.3.1 (Estimating the dust contribution to total aerosol mass) and 3.3.2 (Estimating the dust contribution to Fe).

Polluted air masses which are affected by multiple sources over the continent are advected to our observation site in a remote island in western Japan. Therefore, concentrations of various atmospheric components can temporally co-vary (for example carbon monoxide and sulfate, Miyakawa et al., 2017). In fact, we found the positive correlations of BC and sulfate (Miyakawa et al., 2023 and also this study (Fig. S9 of the revised manuscript)). This indicates that the temporal variations of BC concentrations can account for those of both anthropogenic aerosol and a part of secondary aerosol concentrations. We modified the relevant descriptions to explain this point as follows.

“The simple source apportionment of total mass of PM_{2.5} and PM₁₀ aerosols was conducted using the observed BC, Si, and sulfate concentrations ($[BC]_t$, $[Si]_t$, and $[SO_4^{2-}]_t$, respectively) to evaluate the impacts of Asian dust to total mass for PM_{2.5} and PM₁₀ aerosols. This is basically the same approach developed in Miyakawa et al. (2023). Because concentrations of BC positively correlated with those of sulfate (**Fig. S9**), we assumed that the temporal variations in BC concentrations could account for those of both anthropogenic aerosols and a part of secondary aerosol concentrations. We further assumed that sulfate concentrations could be used to constrain the secondary formation component through aqueous phase reactions inside aerosol particles and cloud droplets, which cannot be represented by temporal variations in BC concentrations (Miyakawa et al., 2023). Si concentrations work as a good indicator for the impacts of Asian dust in the East Asian outflow (Miyakawa et al., 2023). The validity of selecting Si, BC, and SO_4^{2-} as input variables for the multiple linear regression (MLR) models was described by Miyakawa et al. (2023). The observed temporal variations in PM_{2.5} and PM₁₀ aerosol concentrations ($[PM_{2.5}]_t$ and $[PM_{10}]_t$) were fitted to the following MLR model:”

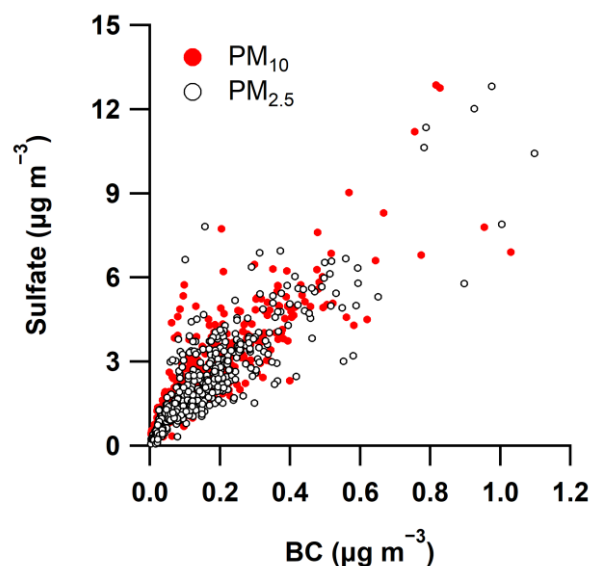


Fig. S9. Correlations between sulfate and BC concentrations for $\text{PM}_{2.5}$ (red markers) and PM_{10} (open markers) particles.

The reconstructed particle mass and Fe of $\text{PM}_{2.5}$ and PM_{10} aerosols well reproduced those observed (Fig. S10 of the revised manuscript). The dust contribution to PM mass was very well validated by the comparison with lidar measurements (Fig. 4). The dust contribution to Fe was indirectly evaluated by calculating the percent mass contribution of Fe-content in dust aerosols (Eqs. 5 and 6) with its typical range revealed by previous studies. The anthropogenic Fe to BC ratios were also evaluated with those derived from previous studies using independent methods. All of these evaluations indicated that the estimation of dust contribution to total particle mass and Fe for $\text{PM}_{2.5}$ and PM_{10} aerosols were reasonable.

The variations in relative differences in the concentrations between reconstructed and observed PM concentrations ($\equiv (\text{reconstructed} - \text{observations})/\text{observations}$) were investigated to analyze the impacts of unaccounted part of PM mass by using the MLR. (Fig. S11 of the revised manuscript) It was found that there was no clear seasonal transition in the relative differences (median levels were within ± 0.1) and that the relative differences increased only when $\text{PM}_{2.5}$ and PM_{10} mass concentrations were lower than a few $\mu\text{g m}^{-3}$. This suggests that the contributions of the unaccounted part of the temporal variations were very small especially for the transboundary transport cases with increased aerosol concentrations and cannot affect the major conclusions in this study.

We added the following sentence in Section 3.3.1.

“The variations in relative differences in the concentrations between reconstructed and observed PM concentrations were investigated to analyze the impacts of unaccounted part of PM mass by using the MLR (**Fig. S11**). It was found that the relative differences increased only when $\text{PM}_{2.5}$ and PM_{10} mass concentrations were lower than a few $\mu\text{g m}^{-3}$ and that there was no clear seasonal transition in the relative differences (median levels were within ± 0.1). This suggests that the contributions of the unaccounted part of the temporal variations were very small, especially for the transboundary transport cases with increased aerosol concentrations, which was a major target of this study.”

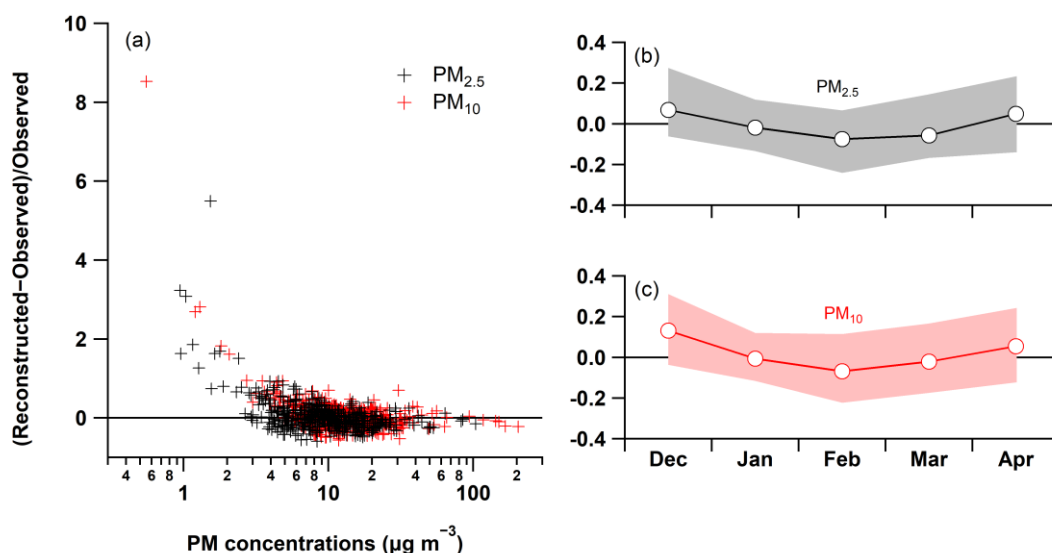


Fig. S11. (a) The relationship between relative differences between reconstructed and observed PM concentrations ($\equiv (\text{reconstructed} - \text{observed}) / \text{observed}$) and PM concentrations for $\text{PM}_{2.5}$ and PM_{10} particles (black and red markers, respectively). (b) and (c) the seasonal progression of monthly median and interquartile ranges of relative differences between reconstructed and observed PM concentrations for $\text{PM}_{2.5}$ and PM_{10} particles, respectively.

In addition, the statement that BC represents both anthropogenic sources and a part of secondary sources (line 313) may require further justification. The authors may consider providing a clearer explanation of the rationale for using BC as a tracer for secondary sources. Although BC can undergo atmospheric aging and some source apportionment studies have identified aged BC as a

source factor, the carbonaceous core of aged BC is still associated with primary emissions rather than secondary formation.

The statement was not sufficient. As described in the above, at remote sites, it is difficult to separate the source signatures only from the temporal variations of the compounds. So, the point is not secondary processes of BC but the temporal co-variation between BC and secondary aerosols like sulfate.

Therefore, the authors may consider incorporating additional source tracers to achieve a more comprehensive source apportionment of atmospheric particulate matter, if such measurements are available. Alternatively, the authors may consider performing separate multiple linear regression analyses for winter and spring to account for potential seasonal variations in the contributions represented by the intercept term.

As described above, unaccounted parts of PM concentrations did not show substantial seasonal variations and showed a significant fraction only when the absolute PM mass concentrations are very low. The detailed comprehensive source apportionments are out of the scope of this study.

(3) Section 3.3.2: Similar to the above comment, the authors may consider conducting separate multiple linear regression analyses for winter and spring to evaluate the source contributions of Fe. This consideration is particularly relevant because previous studies have demonstrated that the regression coefficients between BC and anthropogenic Fe can differ substantially between winter and spring (Zhang et al., Source Apportionment of Atmospheric Iron in Northern China Based on 3-Year Continuous Measurements). Therefore, accounting for seasonal variations in the regression relationships may help improve the accuracy and reliability of the estimated Fe source contributions.

We appreciate the suggestion of the very important studies.

We have discussed this point in Section 3.5 of the original manuscript as follows.

“It should be noted that the predicted reductions in the BC emissions from residential sectors from winter to spring imply the potential uncertainty in assuming the fixed fitting coefficients (e.g., $g_{BC,FePM2.5}$) for the BC component in the MLR model throughout the observation period (section 3.3.2). These coefficients can be higher (lower) than the calculated values in spring (winter), but the difference cannot be large ($< \sim 20\%$).”

We removed these sentences here to clarify this explanation for interpreting the correlation slope of anthropogenic Fe and BC concentrations.

And we compared the observation-based results with the model simulations. The following figure (Fig. S17 of the revised manuscript) is added in the SI of the revised manuscript. This shows the correlation between modeled-BC and -anthropogenic Fe concentrations. The observation-based estimations ($g_{BC,FePM_{2.5}}$ and $g_{BC,FePM_{10}}$) were also overlayed.

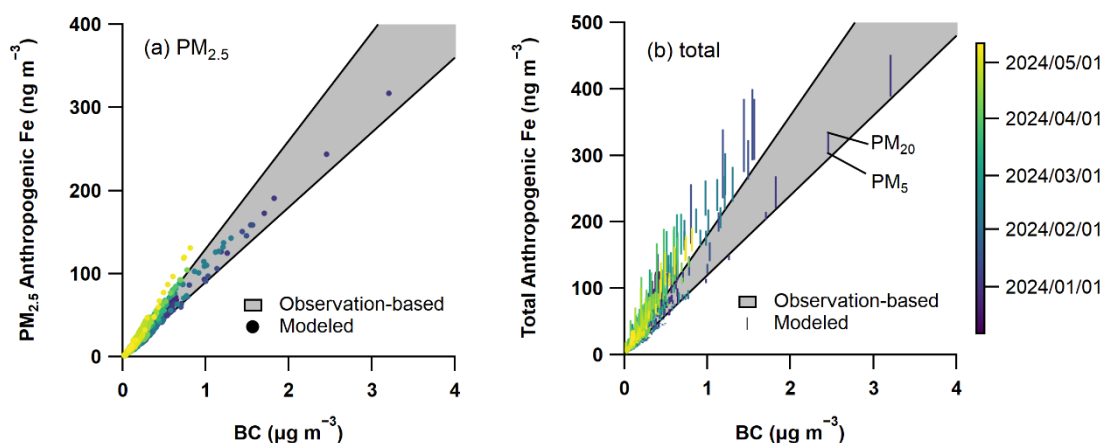


Fig. S17. (a) The correlation of $PM_{2.5}$ anthropogenic Fe and BC concentrations from the model simulations (markers) and the observation-based estimation (shaded region showing the 95% confidence interval range of the fitted coefficient $g_{BC,FePM_{2.5}}$). (b) The correlation of total anthropogenic Fe and BC concentrations from the model simulations (bars between PM_5 and PM_{20}) and the observation-based (PM_{10}) estimation (shaded region showing the 95% confidence interval range of the fitted coefficient $g_{BC,FePM_{10}}$). The data points or bars for the model simulations was colored by the date (based on Japanese Standard Time).

The modeled variabilities of the slope of the correlations showed clear seasonal pattern for $PM_{2.5}$ aerosols and were comparable to 95% confidence interval ranges of the observation-based estimations. When we use the smaller set of the observed data by dividing the seasons in the same analysis, the confidence interval ranges will be larger than the current analysis and it cannot resolve such small changes in the correlation slopes. We acknowledged the limitation of the method used in this study and should further describe the importance of more data collections to obtain statistically significant results of changes in the slope with the suggested reference (Zhang et al., 2025). We added the following sentence in section 3.5.

“This is also consistent with the comparison results of correlations of anthropogenic Fe and BC concentrations between the observation-based estimations of the slopes ($g_{BC,FePM_{2.5}}$ and $g_{BC,FePM_{10}}$) and model simulations (**Fig. S17**). Possible seasonal changes in BC emissions from residential sector in China slightly affected the slope of correlations between anthropogenic Fe and BC concentrations. It should be noted that the predicted reductions in the BC emissions from residential sectors from winter to spring imply the potential uncertainty in assuming the fixed fitting coefficients (e.g., $g_{BC,FePM_{2.5}}$) for the BC component in the MLR model throughout the observation period (section 3.3.2). As the seasonal variations of the slope for the model simulations are comparable to 95% confidence interval ranges of $g_{BC,FePM_{2.5}}$ and $g_{BC,FePM_{10}}$ determined by the MLR, our observations and data analyses using the MLR have limitations to elucidate such small differences derived from seasonal changes in BC emissions from residential sectors, indicating the requirement of more observations for the longer periods (e.g., Zhang et al., 2025).”

(4) Figure 7: The use of “transport time from the continent” as an indicator of aerosol aging degree should be interpreted with caution. For different air masses, variations in meteorological conditions (e.g., temperature and relative humidity) and chemical compositions can substantially influence atmospheric processing, making transport time alone difficult to serve as a metric for aerosol aging.

We agreed with the reviewer’s comment and thus have considered some relevant parameters such as meteorological conditions (relative humidity and precipitation) and relative amount of sulfate to dust and BC as shown in Figs. 8 and S14 of the revised manuscript.

Since the authors have quantified secondary source contributions in PM_{2.5} and PM₁₀, they may consider using these contributions as an alternative indicator of aerosol aging. Besides, the molar ratio of secondary inorganic anions [$NO_3^- + 2 \times nss-SO_4^{2-}$] to total Fe may also serve as a potential indicator of atmospheric aging.

As stated in the above, we could not estimate the contribution of non-sea-salt components because of the lack of online measurements of sodium. Although nitrate is an important component to affect the aging processes of Fe-containing particles (e.g., Zhi et al., 2026; Zhu et al., 1997), we did not measure nitrate concentrations using online instruments, which are required for accurate

quantification of fine mode fraction of nitrate aerosols (e.g., ammonium nitrate). We investigated the mass ratios of sulfate to dust and to BC (Fig. S11 in SI of the original manuscript). Taking into account the possible role of nitrate is a future target. We thus added the following sentences to acknowledge the importance of nitrate in the aging processes.

“Although nitrate as well as sulfate can effectively promote Fe dissolution (e.g., Zhi et al., 2026; Zhu et al., 1997), we could not include the information of nitrate in the data analysis because of the lack of online measurements.” in Section 3.4

“Despite the usefulness of our data sets, additional observations of auxiliary parameters are required to better characterize Fe_{sol} . For example, online measurements of inorganic aerosol components including sodium and nitrate ions are essential for a more thorough evaluation of the impacts of aerosol acidity on the atmospheric processing of Fe.” in Section 4.4

(5) Lines 658-660, The authors suggested that the increase in $\Delta Fe_{sol}/\Delta BC$ from $\sim 40 \text{ ng } \mu\text{g}^{-1}$ in winter to $\sim 90 \text{ ng } \mu\text{g}^{-1}$ in spring was primarily associated with an increased contribution of dust to Fe_{sol} , and secondarily with the larger contribution of the residential sector to BC compared with Fe_{sol} . It would be helpful if the authors could provide further evidence or quantitative analysis to support the conclusion that the former factor plays a more important role.

Assuming that the further verifications beyond the model data analyses are suggested by the reviewer, currently we have no further direct evidence to accurately indicate the impacts of dust Fe_{sol} on the seasonal progression of $\Delta Fe_{sol}/\Delta BC$. We consider that observational constraints are essential to validate the model simulations. When we assume the coarse mode Fe_{sol} work well as an indicator of dust contribution to Fe_{sol} , coarse mode fraction of Fe_{sol} was analyzed. It increased from 3% in January–February, 2024 to 6% in March–April, 2024. This feature can further support the interpretation of the increases in the $\Delta Fe_{sol}/\Delta BC$. We modified the sentence in L475–476 of the original manuscript as follows in Section 3.4.

“However, the dust Fe concentrations increased from winter to spring during the observation period (Fig. 5) and were much higher than the anthropogenic Fe in spring, indicating the significant contributions of dust to Fe_{sol} in spring, regardless of their low water solubility. Indeed, coarse mode fraction of Fe_{sol} increased from 3% in winter (January–February) to 6% in spring (March–April), suggesting that the increased dust contribution to Fe_{sol} in spring is a cause of the seasonal changes in $\Delta Fe_{sol}/\Delta BC$.”

In addition, the authors may consider clarifying whether this interpretation represents a general seasonal pattern or is mainly influenced by the strong dust event observed in April 2024. Since dust events may substantially alter soluble Fe concentrations but have a relatively limited impact on BC concentrations, the contribution of this episodic event to the observed increase in $\Delta\text{Fe}_{\text{sol}}/\Delta\text{BC}$ should be carefully evaluated.

As shown in Fig. 9 of the original manuscript, the observed enhancements of $\Delta\text{Fe}_{\text{sol}}/\Delta\text{BC}$ during spring season were not limited to the specific event which was suggested by the reviewer (April 18–20, 2024, the information during this period is included in single data point of Fig. 9).

Minor Comments

(1) The manuscript contains several statements that could benefit from more precise and rigorous wording. For example, in line 65, the authors state that “anthropogenic aerosols often contain less Fe but show substantially higher solubility due to atmospheric processing”. The phrase “due to atmospheric processing” may require further clarification, as it appears to attribute the higher Fe solubility of anthropogenic aerosols primarily to atmospheric processing, while potentially overlooking the fact that Fe from some anthropogenic sources may already exhibit relatively high solubility upon emission.

As suggested, this statement is not accurate. For example, Fe-containing particles from residential sources potentially have high water solubility (e.g., Cui et al., 2025). We thus modified the sentence as follows.

“Asian dust aerosols are typically enriched in total Fe but exhibit low solubility, whereas anthropogenic aerosols often contain less Fe but show substantially higher solubility due to source types and atmospheric processing during transport (e.g., Cui et al., 2025; Kurisu et al., 2021).”.

(2) Line 487: The term “significantly correlated” is used. However, the statistical significance tests are not provided in Table S4. The authors may consider adding the relevant statistical results or revising the wording accordingly.

We simply rephrased “significantly” by “positively”.

(3) Lines 495-498: “This can be partly derived from the uncertainties in the dry deposition fluxes of coarse particles in the model (Kok et al., 2021). As the high temporal resolution variations in the source-resolved Fe concentrations were reasonably well simulated, we concluded that the removal processes as well as emissions were also reasonably simulated”. The authors first attribute the discrepancy partly to uncertainties in dry deposition fluxes, but subsequently conclude that the removal processes were reasonably simulated. The authors may consider clarifying how these two statements can be reconciled.

We referred to Kok et al. for describing the possible uncertainties in the simulations of coarse mode aerosols and interpreting the poorer correlations in coarse mode aerosols than those in fine mode aerosols. We think that there was no contradiction. However, these sentences were not clear and needed to be softened. We thus modified as follows.

“The poorer correlations in the comparison for total Fe can be partly derived from the uncertainties in the dry deposition fluxes of coarse particles in the model (Kok et al., 2021). Although the high temporal resolution variations in the source-resolved Fe concentrations were reasonably well simulated, further verifications of the removal processes in the model are required in future.”

(4) Lines 508-509 “Therefore, the possible reason for the discrepancies between the observations and model simulations (NMB of 21 %) in this study was the type of leaching method used to derive the Fe_{sol} concentrations.” The statement may be too definitive. The authors may consider revising it to indicate that this is one of the possible reasons contributing to the discrepancy.

This sentence has unclear explanation about the discrepancy between the observations and model simulations. This comparison was made for total aerosols which were not segregated by the size range. For the size-segregated comparisons, the size cut performances as well as analytical errors (the leaching method and the following analyses themselves) can affect the comparison results. We thus modified this sentence as follows.

“Therefore, one of the possible reasons for the discrepancies of total Fe_{sol} concentrations between the observations and model simulations (NMB of 21 %) in this study was the type of leaching method used to derive the Fe_{sol} concentrations.”

(5) Lines 555-558 “The model simulations implied that the long-range transport of dust Fe from the continent in East Asia and its deposition in the outflow oceanic regions over the NWPO can control the atmospheric supply of bioaccessible Fe to the ocean surface in the HNLC region and contribute to the significant amount of bioavailable Fe required for sustaining the springtime ocean biological activities in the NWPO.” This statement represents a broad conclusion. However, the current analysis may not provide sufficient evidence to fully support this statement.

In this study, by comparing relevant processes supplying Fe to the surface sea water (e.g., intermediate water intrusion) at K2 site in the NWPO (from our previous study, Nagashima et al., 2023), we suggested the possible significant influence of atmospheric supply of Fe to the ocean biological activities in this region, however, this sentence is too strong as suggested. We hence modified the sentence to soften the message, as follows.

“The model simulations implied that the long-range transport of dust Fe from the continent in East Asia and its deposition in the outflow oceanic regions over the NWPO can control the atmospheric supply of bioaccessible Fe to the ocean surface in the HNLC region and can contribute to the significant amount of bioavailable Fe affecting the springtime ocean biological activities in the NWPO.”

(6) Line 655: The authors state that “indicating the significance of continental combustion sources in enhancing Fe_{sol} concentrations in the outflow regions.” The authors may consider whether the term “continental” is sufficiently precise in this context, as BC in the outflow regions may also include contributions from ship emissions.

Major sources of BC transported to Fukue island from the continent in East Asia were anthropogenic combustion sources over the continent (e.g., Miyakawa et al., 2019). Ship emissions were less dominant for the impact on BC concentrations at Fukue site, which was estimated by a tagged tracer experiment of the IMPACT model simulations (Miyakawa et al., 2023).

We modified this as follows.

“indicating the significance of combustion processes (mainly from continental sources) in enhancing Fe_{sol} concentrations in the outflow regions”

(7) The authors may consider revising several expressions throughout the manuscript to ensure more accurate terminology. For example, the term “total (PM₁₀) aerosols” may need clarification, as PM₁₀ is generally not considered equivalent to the total aerosol fraction. In addition, terms such as “PM_{2.5} aerosols” and “Si aerosols” may need to be revised for greater terminological accuracy. The authors may consider using “PM_{2.5} particles” or “the PM_{2.5} fraction” and “Si-containing particles”.

We modified the relevant phrases. See the revised manuscript for details.

(8) When describing the agreement between two datasets, the authors frequently use expressions such as “within ~30%”, “< ±30%”, and “within ±30%”. The authors may consider using a consistent expression throughout the manuscript and providing a clear definition of these terms. For example, it is unclear whether “within ±30%” refers to a relative bias (e.g., NMB/NME less than 30%), or whether it indicates that the slope of the linear regression between the two datasets falls within the range of 0.7–1.3.

We modified the relevant sentences as follows.

“The results were in good agreement (the linear regression slope of 0.96 ± 0.05 and $r^2 = 0.79$) (Fig. S3a).”

“These were in good agreement (the linear regression slope of 1.01 ± 0.14) and had good correlations ($r^2 = 0.89$).”

“In general, PX-375-derived PM₁₀ concentrations for almost all elements (except Cr and Pb) agreed well (the linear regression slopes were 0.67–1.39 and $r^2 = 0.86$ –0.98) with HVS-derived total concentrations, even though the size cut definition was different between the two methods (PM₁₀ vs. the sum of fine and coarse modes).”

“Although measurements/sampling time resolutions and site locations differed, the levels and temporal variations of most components, except Si and BC, reasonably agreed well between the two sites, showing the ratios of the period averages within ±30 % (Table S3 and Fig. S7).”

(9) If lidar depolarization ratio measurements are available, the authors may consider comparing

them with the dust contributions derived from PM source apportionment. Such a comparison could provide additional independent evidence for evaluating the reliability of the estimated dust contributions. However, this analysis is not essential and could be considered as a potential improvement if the relevant data are available.

The data analysis of the lidar data in this study uses the depolarization ratio information to separate the impacts of dust and spherical (non-dust) aerosols from total extinction coefficients (See L141–148, P5 in section 2.1 of the original manuscript in detail). And the separated lidar results were compared with the ground-based source apportionment results in section 3.3.1 (and Figure 4 of the original manuscript). As discussed in section 3.3.1, the lidar-derived dust extinction coefficients (converted mass concentrations) very well correlated with the PM₁₀ dust concentrations ($r^2 = 0.95$). We considered that this data analysis supported the validity of the source apportionments.

(10) I am interested in the use of particle-laden spots on the filter roll tape from the PX-375 for subsequent IC and ICP-MS analyses. Since the filter is collected on a roll tape, the authors may consider clarifying how potential cross-contamination between particle-loaded spots from different positions on the roll (e.g., inner and outer layers) was avoided or evaluated during sample handling and analysis.

The filter roll used for PX-375 was developed by Horiba and designed for minimizing the chemical artifacts and the cross contamination (Asano et al., 2017; and references therein). So, we have carefully handled the filter roll minimizing the contact with metallic stuff. For example, the collected filter roll was preserved in a plastic bag, and the individual filter spot was prepared using ceramic scissors.

References for the responses to the reviewers' comments

Asano, H., Aoyama, T., Mizuno, Y., Shiraishi, Y.: Highly Time-Resolved Atmospheric Observations Using a Continuous Fine Particulate Matter and Element Monitor. ACS Earth Space Chem. 1 (9): 580–590. <https://doi.org/10.1021/acsearthspacechem.7b00090>, 2017

Buck, C. S., Landing, W. M., Resing, J. A., Lebon, G. T.: Aerosol iron and aluminum solubility in the northwest Pacific Ocean: Results from the 2002 IOC cruise, Geochem. Geophys.

Geosyst., 7, Q04M07, <https://doi.org/10.1029/2005GC000977>, 2006.

Chen, Y.: Sources and Fate of Atmospheric Nutrients over the Remote Oceans and Their Role on Controlling Marine Diazotrophic Microorganisms, (PhD dissertation). University of Maryland, College Park., 2004

Chen H, Grassian V. H.: Iron dissolution of dust source materials during simulated acidic processing: the effect of sulfuric, acetic, and oxalic acids. *Environ Sci Technol.* 47(18): 10312-21. <https://doi.org/10.1021/es401285s>, 2013

Chuang, P. Y., Duvall, R. M., Shafer, M. M., and Schauer, J. J.: The origin of water soluble particulate iron in the Asian atmospheric outflow, *Geophys. Res. Lett.*, 32, L07813, <https://doi.org/10.1029/2004GL021946>, 2005.

Cui, W., Song, X., Su, Y., Chen, X., Wu, D., Li, Q.: Soluble iron in source-based anthropogenic PM_{2.5} predominantly from steel industry and residential combustion in China. *Geophys. Res. Lett.*, 52, e2025GL118603. <https://doi.org/10.1029/2025GL118603>, 2025

Hsu S.-C., Wong G.T.F., Gong G.-C., Shiah F.-K., Huang Y.-T., Kao S.-J., Tsai F., Candice Lung S.-C., Lin F.-J., Lin I.I., Hung C.-C., and Tseng C.-M.: Sources, solubility, and dry deposition of aerosol trace elements over the East China Sea, *Mar. Chem.*, 120, 116–127, <https://doi.org/10.1016/j.marchem.2008.10.003>, 2010.

Hsu, S.-C., et al: Effects of acidic processing, transport history, and dust and sea salt loadings on the dissolution of iron from Asian dust, *J. Geophys. Res.*, 115, D19313, <https://doi.org/10.1029/2009JD013442>, 2010

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

Kok, J. F., Adebisi, A. A., Albani, S., Balkanski, Y., Checa-Garcia, R., Chin, M., Colarco, P. R., Hamilton, D. S., Huang, Y., Ito, A., Klose, M., Li, L., Mahowald, N. M., Miller, R. L., Obiso, V., Perez Garcia-Pando, C., Rocha-Lima, A., and Wan, J. S.: Contribution of the world's main dust source regions to the global cycle of desert dust, *Atmos. Chem. Phys.*, 21, 8169–8193, <https://doi.org/10.5194/acp-21-8169-2021>, 2021.

Kurisu, M., Sakata, K., Uematsu, M., Ito, A., and Takahashi, Y.: Contribution of combustion Fe in marine aerosols over the northwestern Pacific estimated by Fe stable isotope ratios, *Atmos. Chem. Phys.*, 21, 16027–16050, <https://doi.org/10.5194/acp21-16027-2021>, 2021.

Kurisu, M., Zhu, C., Miyakawa, T., Ito, A., Suzuki, K., Kashiwabara, T., Kubo, S., Takahashi, Y., Harada, N., Foret, G., Dubois, N., and Kanaya, Y.: Identification of anthropogenic Fe originated from East Asia using Fe stable isotope ratios of aerosols collected on Fukue Island, *Atmos. Environ.*, 373, 15, <https://doi.org/10.1016/j.atmosenv.2026.121893>, 2026.

Li, W., Ito, A., Wang, G., Zhi, M., Xu, Li., Yuan, A., Zhang, J., Liu, L., Wu, F., Laskin, A., Zhang, D., Zhang, X., Zhu, T., Chen, J., Mihalopoulos, N., Bougiatioti, A., Kanakidou, M., Wang, G., Hu, H., Zhao, Y., Shi, Z.: Aqueous-phase secondary organic aerosol formation on mineral dust, *Nat. Sci. Rev.*, 12, 7, nwaf221, <https://doi.org/10.1093/nsr/nwaf221>, 2025

Meskhidze, N., Chameides, W. L., and Nenes, A.: Dust and pollution: A recipe for enhanced ocean fertilization?, *J. Geophys. Res.*, 110, D03301, <https://doi.org/10.1029/2004JD005082>, 2005.

Miyakawa, T., Oshima, N., Taketani, F., Komazaki, Y., Yoshino, A., Takami, A., Kondo, Y., and Kanaya, Y.: Alteration of the size distributions and mixing states of black carbon through transport in the boundary layer in East Asia, *Atmos. Chem. Phys.*, 17, 5851–5864, <https://doi.org/10.5194/acp-17-5851-2017>, 2017.

Miyakawa, T., Komazaki, Y., Zhu, C., Taketani, F., Pan, X., Wang, Z., and Kanaya, Y.: Characterization of carbonaceous aerosols in Asian outflow in the spring of 2015: importance of non-fossil fuel sources, *Atmos. Environ.*, 214, 116858, <https://doi.org/10.1016/j.atmosenv.2019.116858>, 2019.

Miyakawa, T., Ito, A., Zhu, C., Shimizu, A., Matsumoto, E., Mizuno, Y., and Kanaya, Y.: Trace elements in PM_{2.5} aerosols in East Asian outflow in the spring of 2018: Emission, transport, and source apportionment, *Atmos. Chem. Phys.*, 23, 14609–14626, <https://doi.org/10.5194/acp-23-14609-2023>, 2023.

Nagashima, K., Kawakami, H., Sugie, K., Fujiki, T., Nishioka, J., Iwamoto, Y., Takemura, T., Miyakawa, T., Taketani, F., and Aita, M. N.: Asian dust-deposition flux to the subarctic Pacific estimated using single quartz 870 particles, *Sci. Rep.* 13, 15424, <https://doi.org/10.1038/s41598-023-41201-6>, 2023.

Sakata, K., Takahashi, Y., Takano, S., Matsuki, A., Sakaguchi, A., Tanimoto, H.: First X-ray Spectroscopic Observations of Atmospheric Titanium Species: Size Dependence and the Emission Source. *Environ. Sci. Technol.* 55 (16): 10975–10986. <https://doi.org/10.1021/acs.est.1c02000>, 2021

Sakata, K., Takano, S., Matsuki, A., Takeichi, Y., Tanimoto, H., Sakaguchi, A., Kurisu, M., and Takahashi, Y.: Atmospheric chemistry in East Asia determines the iron solubility of aerosol particles supplied to the North Pacific Ocean, *Atmos. Chem. Phys.*, 25, 11087–11107, <https://doi.org/10.5194/acp-25-11087-2025>, 2025.

Shi, Z., Krom, M. D., Bonneville, S., L. G.: Atmospheric Processing Outside Clouds Increases Soluble Iron in Mineral Dust. *Environ. Sci. Technol.* 49 (3): 1472–1477. <https://doi.org/10.1021/es504623x>, 2015.

Shi, J., Guan, Y., Ito, A., Gao, H., Yao, X., Baker, A. R., and Zhang, D.: High production of soluble iron promoted by aerosol acidification in fog, *Geophys. Res. Lett.*, 47, e2019GL086124, <https://doi.org/10.1029/2019GL086124>, 2020.

Sholkovitz, E. R., Sedwick, P. N., Church, T. M., Baker, A. R., and Powell, C. F.: Fractional solubility of aerosol iron: Synthesis of a global-scale dataset, *Geochim. Cosmochim. Ac*, 89, 173–189, <https://doi.org/10.1016/j.gca.2012.04.022>, 2012.

Wu, J., Rember, R., Cahill C.: Dissolution of aerosol iron in the surface waters of the North Pacific and North Atlantic oceans as determined by a semicontinuous flow-through reactor method, *Global Biogeochem. Cycles*, 21, GB4010, <https://doi.org/10.1029/2006GB002851>, 2007.

Zhang, H., Li, R., Dong, S., Wang, F., Zhu, Y., Meng, H., et al.: Abundance and fractional solubility of aerosol iron during winter at a coastal city in northern China: Similarities and contrasts between fine and coarse particles. *J. Geophys. Res.: Atmos.*, 127, e2021JD036070. <https://doi.org/10.1029/2021JD036070>, 2020.

Zhang, T., Xiang, Y., Liu, Y., Yan, C., Zhu, B., Liu, X., et al.: Source apportionment of atmospheric iron in northern China based on 3-year continuous measurements. *Geophys. Res. Lett.*, 52, e2024GL114462. <https://doi.org/10.1029/2024GL114462>, 2025

Zhi, M., Baldo, C., Shi, Z., Wang, G., Xu, L., Li, K., Shao, L., Niu, H., Yuan, Q., Zhang, J., Fu, P., Ito, A., Fu, H., Li, W.: Enrichment in More Soluble Ferric Iron (Fe(III)_s) during Dust Transport by Nitric Acid Uptake. *Environ. Sci. Technol.* 60 (10): 8007–8017. <https://doi.org/10.1021/acs.est.5c11063>, 2026

Zhu, X. R., Prospero, J. M., Millero, F. J.: Diel variability of soluble Fe(II) and soluble total Fe in North African dust in the trade winds at Barbados, *J. Geophys. Res.*, 102(D17), 21297–21305, <https://doi.org/10.1029/97JD01313>, 1997