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This manuscript presents field observations of total and water-soluble Fe in PM_{2.5} and PM₁₀ aerosols and attempts to distinguish their sources using a multilinear regression approach. The topic is relevant to atmospheric iron cycling and aerosol source apportionment. However, the manuscript currently overstates several conclusions, and its novelty is not clearly demonstrated. In addition, several interpretations require stronger evidence, and comparisons with previous studies are insufficient. Therefore, I recommend revision before the manuscript can be considered for publication.

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.

1. In the Abstract, the authors repeatedly refer to "trace metals", whereas the study actually focuses exclusively on Fe concentrations and Fe solubility. This terminology is misleading because no other trace metals are investigated. I recommend replacing "trace metals" with iron (Fe) throughout the Abstract and other relevant sections.

As suggested, we rephrased trace metals with "Fe" at all instances.

2. The title, "Size-resolved characterizations of Fe in aerosols," overstates the scope of the study. The manuscript only compares water-soluble Fe concentrations and fractions between PM_{2.5} (fine-mode) and PM₁₀ (total aerosols), rather than measuring a true size distribution using size-segregated aerosol samples.

As suggested, the resolution of the size classification was not so high in this study. “Size-resolved” was replaced by “Size-segregated”.

3. The source apportionment results shown in Figure 6 require further clarification. For several PM₁₀ samples, the contribution of dust exceeds 100%, while these samples simultaneously exhibit relatively high Fe solubility. These results appear physically questionable. The authors should explain whether these points are statistical outliers or result from the regression method itself, and provide additional validation of the source contribution estimates. At present, I am not fully convinced by these results.

As R_{dustFe} was estimated by the data analyses using the MLR, the determinations of R_{dustFe} have uncertainties. So, exceedance from 100% of R_{dustFe} can occur due to the MLR-based estimation. Only one data point exceeds 100% of dust contribution in Fig. 6 of the original manuscript and $f_{\text{Fe,sol}}$ was around 0.04 (not so high in our data set). The data points strongly affected by Asian dust (e.g., R_{dustFe} values > 0.8) showed the variations of $f_{\text{Fe,sol}}$ from 0.01 to 0.06. This is caused by variabilities in $f_{\text{Fe,sol}}$ which was accounted for by atmospheric processing as well as the source attributions of Fe in dust particles. To clarify this, we have added the following sentence.

“Note that $f_{\text{Fe,sol}}$ varied from 0.01 to 0.06 for some data points with $R_{\text{dustFe}} > 0.8$ (i.e., strongly affected by Asian dust) in **Fig. 6**. These relatively large variations in $f_{\text{Fe,sol}}$ were likely derived from the atmospheric processing of Fe in Asian dust particles, which is further discussed in the following.”

Exceedances from 100% of R_{dustFe} were also found in Fig. 7b, which was related to the estimation of R_{dustFe} for coarse mode (2.5–10 μm) aerosols by the subtraction (“The R_{dustFe} for coarse mode particles was estimated based on the differences in R_{dustFe} between PM_{2.5} and PM₁₀ aerosol particles.” at Line 406–407 of the original manuscript). Because of indirect derivation (i.e., subtraction), the precision to determine the values of R_{dustFe} became worse and we acknowledged that the derived values cannot well resolve the changes in R_{dustFe} for coarse mode aerosols. We added the following sentences at the end of the second paragraph (of the revised manuscript) of section 3.4.

“The relationship between $f_{\text{Fe,sol}}$ and R_{dustFe} for coarse mode aerosols was unclear (**Fig. 7b**), which is partly because the precision to indirectly derive R_{dustFe} for coarse mode aerosols was not enough

to resolve small changes in the values of R_{dustFe} .”

And we modified the second sentence in the third paragraph (of the revised manuscript) of Section 3.4 as follows.

“These relationships suggest that air mass aging rather than source attribution can affect the variability in $f_{\text{Fe,sol}}$, especially for coarse-mode particles.”

4. It is necessary to include the heterogeneous reactions on dust particles which already have been aged in continental air of East China before it can reach up the North Pacific Ocean (Nat. Sci. Rev 2025;12:nwaf221; Environ. Sci. Technol. 2026; 60:8007-8017.).

We appreciate an important suggestion.

As suggested, we integrated this point in Section 3.4.

“This can be observational evidence that aqueous-phase processing affects the variability in $f_{\text{Fe,sol}}$ for mineral dust particles (e.g., Sakata et al., 2025; Shi et al., 2015). 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. Substantial aging involving nitric acid uptake by Asian dust can modify the physicochemical properties of Asian dust particles and enhance $f_{\text{Fe,sol}}$ (e.g., Li et al., 2025; Zhi et al., 2026), which can occur over the continent even without further aging during transport to the outflow regions. It is indicated that transport pathways from the desert to the receptor points also have great importance accounting for the variations in $f_{\text{Fe,sol}}$ for coarse particles.”

5. The sentence "The R_{dustFe} values were compared with $f_{\text{Fe,sol}}$ for total, fine-, and coarse-mode particles" is difficult to understand. The manuscript does not clearly explain what R_{dustFe} represents, why it should be compared with Fe solubility, or what scientific implication can be drawn from this comparison.

R_{dustFe} was defined as “the contribution of dust to Fe (L405–406, P15 of the original manuscript)”. However, this parameter was introduced at L366, P15 in Section 3.3.2, “The dust contribution to total Fe~ respectively.”.

The definition was not clearly stated in the earlier version. Hence, we have modified this sentence

as follows.

“The dust contribution to total Fe (R_{dustFe}) in $\text{PM}_{2.5}$ and PM_{10} aerosols ($g_{\text{Si,FePM}_{2.5}}[\text{Si}_{\text{PM}_{2.5}}]_t/[\text{Fe}_{\text{PM}_{2.5}}]_t$ and $g_{\text{Si,FePM}_{10}}[\text{Si}_{\text{PM}_{10}}]_t/[\text{Fe}_{\text{PM}_{10}}]_t$, respectively) were, on average, 60% ($\pm 25\%$) and 72% ($\pm 25\%$), respectively”

The reason why this parameter was compared with the Fe fractional solubility is that Asian dust aerosol typically has low Fe water solubility, but anthropogenic combustion aerosols exhibited the higher Fe water solubility than Asian dust, as described in Introduction section (Line 64–66 of the original manuscript). We modified the sentence in section 3.4 as follows.

“The R_{dustFe} values were compared with $f_{\text{Fe,sol}}$ for total and size-segregated (fine and coarse) particles (**Figs. 6 and 7a–b**) to investigate the impacts of Asian dust, which has typically very low $f_{\text{Fe,sol}}$ (e.g., Kurisu et al., 2021), on size-segregated $f_{\text{Fe,sol}}$.”

6. The novelty of this study is not sufficiently highlighted in either the Abstract or the Conclusions. From the current presentation, the main findings appear to be that continental combustion sources enhance soluble Fe concentrations in outflow regions, while elevated total Fe concentrations in spring are mainly associated with dust particles. However, these conclusions have already been well established in previous atmospheric Fe studies. If the primary innovation is the newly developed multilinear regression method for separating the sources of total and soluble Fe, the authors should explicitly compare this method with existing source apportionment techniques, such as PMF, and demonstrate its advantages and limitations. Otherwise, it is difficult to understand what new scientific insight this study provides.

We appreciate important suggestions to improve the manuscript.

Related to the comment 7 in the following, we consider that the novelties of this study beyond our previous study are (1) high-temporal resolution measurements of $\text{PM}_{2.5}$ and PM_{10} aerosol elemental compositions using a newly developed system, and (2) the combinations of such data sets and the offline characterizations of fractional solubility of Fe and total Fe concentrations in size-segregated aerosols. By combining (1) and (2), we investigated the evolution of fractional solubility of Fe in terms of sources and atmospheric processing of Fe, and the relationship between water-soluble Fe and BC concentrations in the East Asian outflow. Furthermore, using the obtained data sets, the capabilities of the IMPACT model to simulate the source apportionment and atmospheric processing of Fe in aerosols were validated for better prediction of water-soluble

Fe concentrations. We thus modified our previous works in Introduction (see the response to Comment 7 in the following).

Note that source apportionment analyses were developed in our previous paper (Miyakawa et al., 2023) but this study thoroughly validated the results with independent lidar measurements (dust PM mass concentrations vs extinction coefficients) and by comparing the results (Fe content in dust particles and anthropogenic Fe/BC enhancement ratios) with those in previous studies.

The descriptions in the first paragraph of Section 3.3.1 were also modified.

“The simple source apportionment 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 for PM_{2.5} aerosols 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:”

Because of the word limit of the Abstract, we modified mainly Conclusions.

Here is the revised version of Conclusions

“For the first time, high-temporal resolution measurements of the elemental composition of both PM_{2.5} and PM₁₀ aerosols were conducted using a newly developed system based on the automated aerosol elemental analyzer PX-375 on a remote island of Japan in the winter–spring of 2023–2024. These measurements were combined with high-volume air sampling (5-d) and offline chemical analyses to characterize the estimation of Asian dust impacts and water-soluble concentrations and fractions of Fe in the East Asian outflow region. Using the observational data sets, the IMPACT model was validated in terms of the predictions of size-segregated

concentrations and the water-soluble fraction of Fe. Mass concentrations of total aerosol and Fe for PM_{2.5} (PM₁₀) aerosols reached 85 (205) and 2.7 (7.4) $\mu\text{g m}^{-3}$, respectively (April 18–20, 2024). Backward trajectories and satellite observations during this period indicated a strong influence of Asian dust transport on the outflow region. The values of $f_{\text{Fe,sol}}$ varied greatly from 3 % to 40 % for PM_{2.5} aerosols and were below 1.5 % for coarse mode aerosols ($>2.5 \mu\text{m}$), indicating that mineral dust dominated the coarse mode and that R_{dustFe} temporally varied for PM_{2.5} aerosols. Temporal variations in Fe concentrations for PM_{2.5} and PM₁₀ aerosols were analyzed based on MLR using the concentrations of tracer species (Si and BC) and were classified into dust and non-dust Fe contributions for PM_{2.5} and coarse-mode aerosols. The evolution of size-segregated $f_{\text{Fe,sol}}$ in the East Asian outflow was investigated using R_{dustFe} and indicators for aging processes (transport time, RH, and increases in sulfate mass). The source apportionment (dust vs. non-dust contributions) was essential to account for the $f_{\text{Fe,sol}}$ variations for PM_{2.5} aerosols, whereas coarse mode aerosols, which were mainly apportioned to dust, were largely affected by aging processes, especially aqueous phase processes associated with RH higher than 75–80 %. Temporal variations in Fe_{sol} concentrations were well correlated with those in BC concentrations, indicating the significance of combustion processes (mainly from continental sources) in enhancing Fe_{sol} concentrations in the outflow regions. $\Delta\text{Fe}_{\text{sol}}/\Delta\text{BC}$ was 53.8 (± 11) $\text{ng } \mu\text{g}^{-1}$ on average during the observation period and increased from $\sim 40 \text{ ng } \mu\text{g}^{-1}$ in winter to $\sim 90 \text{ ng } \mu\text{g}^{-1}$ in spring, which could be reproduced by the IMPACT model. The modeled data analyses suggest that the observed seasonal changes in $\Delta\text{Fe}_{\text{sol}}/\Delta\text{BC}$ are primarily associated with the increased dust contribution to Fe_{sol} in spring, and secondarily with the larger contribution of the residential sector to BC than to Fe_{sol} . Our observations provide combined datasets of size-segregated Fe and Fe_{sol} concentrations and the evolution of $f_{\text{Fe,sol}}$ through aging processes. These datasets are useful constraints for further developing, validating, and refining numerical models that simulate the detailed behavior of Fe in aerosols over regions under the East Asian winter monsoon influences.”

7. how this work advances beyond the authors' previous publication (e.g., <https://doi.org/10.5194/acp-23-14609-2023>)? Rather than simply presenting another set of observations, the authors should discuss the methodological improvements, new scientific findings, and additional atmospheric implications of the present study compared with their earlier work.

In Introduction, we described our previous works including others as follows.

“Recently, the IMPACT model was refined to simulate the multi-elemental compositions of atmospheric aerosols (Ito and Miyakawa, 2023; Miyakawa et al., 2023), and it has been evaluated and updated in terms of source apportionment, bioaccessibility, and removal processes of Fe in East Asian outflow using high-temporal resolution measurements of Fe in fine aerosols and tracer compounds (Miyakawa et al., 2023) and size-resolved (fine and coarse) characterizations of stable isotope ratios of aerosol Fe and water-soluble Fe (Kurusu et al., 2026).”

Our previous work (Miyakawa et al., 2023) measured the high-temporal resolution variations of concentrations of Fe and other trace metals (Pb, Cu, and Mn) only in PM_{2.5} aerosols and analyzed the data with other measurements of tracer species like carbon monoxide and BC and the comparisons with the model simulations, which demonstrated the usefulness of the high-temporal resolution measurements of trace metals in terms of the characterizations of emission and removal processes of trace metals and the validations and refinements of the model simulations. But the paper did not include the investigation of the chemical characteristics (i.e., fractional solubility) of Fe content in aerosols, suggesting that our previous study is a different work from this study.

This study improved the high-temporal resolution measurements of Fe by adding the function of alternate characterizations of both PM_{2.5} and PM₁₀ aerosols and integrated the offline measurements of the size-segregated fractional solubility of Fe using high-volume air sampling and the following chemical analyses. We analyzed the evolution of the size-segregated fractional solubility of Fe with the auxiliary meteorological data (in relation to backward trajectories) and the source attributions of dust and non-dust contributions to Fe. This indicates that this study extended our previous work to a wider range of sizes from only fine to fine and coarse particles and to the investigation of the relationship with the fractional solubility of Fe. We modified the last sentence and added the following sentences at the end of third paragraph of Introduction section.

“Recently, the IMPACT model was refined to simulate the multi-elemental compositions of atmospheric aerosols (Ito and Miyakawa et al., 2023; Miyakawa et al., 2023), and it has been evaluated and updated in terms of source apportionment, bioaccessibility, and removal processes of Fe in East Asian outflow using high-temporal resolution measurements of Fe only in fine particles and tracer compounds (Miyakawa et al., 2023) and size-segregated (fine and coarse) characterizations of stable isotope ratios of aerosol Fe and water-soluble Fe (Kurusu et al., 2026). The high-temporal resolution variations of concentrations of Fe in both fine and coarse particles and their combination with the offline chemical analyses have not been investigated in the East

Asian outflow in our previous works.”

8. The discussion lacks sufficient comparison with previous observations of total Fe concentrations and Fe solubility reported in East Asia and other outflow regions. A comprehensive comparison with published datasets would allow readers to better evaluate whether the reported values are reasonable and would place the present observations into a broader atmospheric context.

We appreciate the suggestion.

Comparisons with some previous studies were discussed in section 3.4. The major reasons why only the selected study (Zhu et al., 2020) was compared in Fig. 7 are the differences in the size segregation definition to classify fine and coarse particles among studies and the purpose of the comparison that is to show the reference data near emission sources. Furthermore, the size cut accuracy of TE-231 cascade impactor (discussed in section 3.1.1) is another reason. Some outflow data sets over the North Pacific Ocean, at the other outflow sites/regions (NOTOGRO site, Japan, 37.451° N, 137.359° E, Gosan site in Jeju island, South Korea, 33.292° N, 126.162° E, and over East China Sea, 22–50° N, 140° E–150° E), and a continental site in China (Qingdao) can be compared when we focus on the analyses of TSP (or sum of fine and coarse) samples (Fig. S12 of the revised manuscript). The data information was summarized in Table S5 of the revised SI.

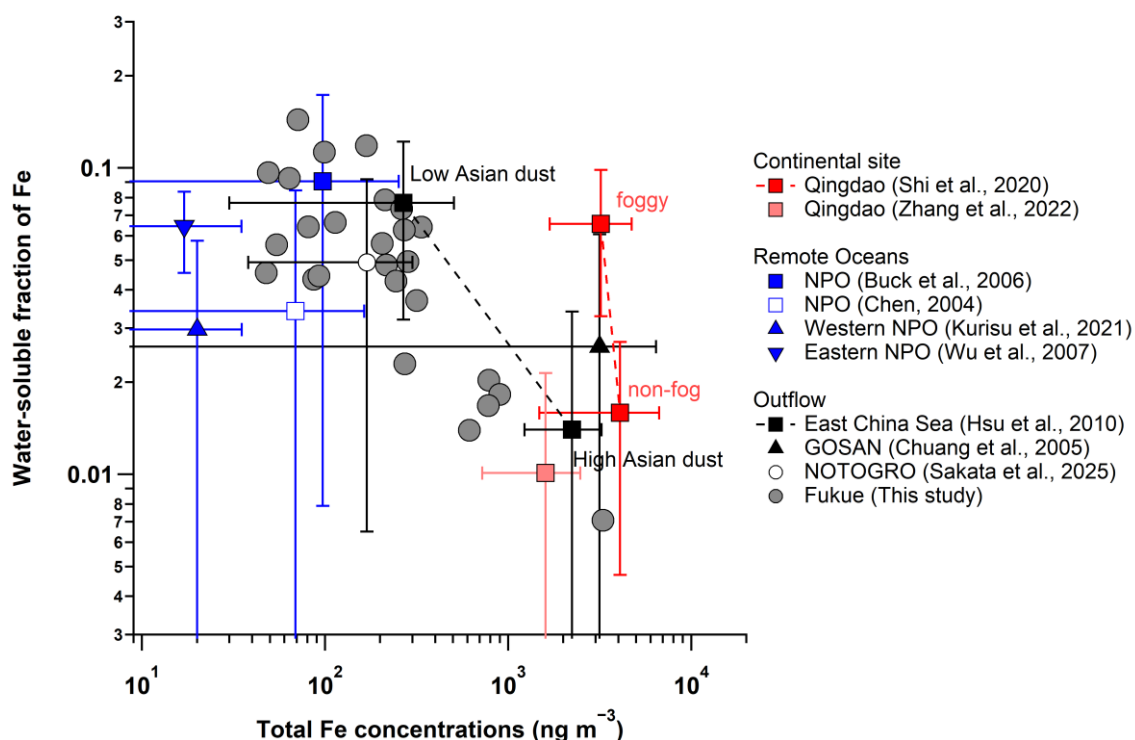


Fig. S12. The relationship between the fractional solubility and concentrations of Fe for TSP samples from this study (shaded circles) and previous studies in a continental site, Qiangdao (red (Shi et al., 2020) and pink (Zhang et al., 2022) squares), remote North Pacific Ocean (blue filled squares (Buck et al., 2006), open squares (Chen 2004), triangles (Kurusu et al., 2021), and inverse triangles (Wu et al., 2007)), and outflow regions (black squares (Hsu et al., 2010), triangles (Chuang et al., 2005), and open circles (Sakata et al., 2025)). All the data in previous studies were represented as the average with bars showing the standard deviations.

We included this comparison in the first paragraph of section 3.4 and added the following in the revised manuscript.

“The fractional solubility of Fe ($f_{\text{Fe,sol}}$) was analyzed as a proxy of bioaccessibility of Fe. The observed relationship between $f_{\text{Fe,sol}}$ and total Fe concentrations for TSP (i.e., the sum of fine and coarse), which has been typically discussed in many previous studies (e.g., Sholkovitz et al., 2012), were shown in Fig. S11. The levels of $f_{\text{Fe,sol}}$ and total Fe concentrations for TSP and its tendency were basically consistent with those in previous studies, showing the inverse relationship that Fe concentrations decreased but $f_{\text{Fe,sol}}$ increased from near sources (e.g., Qingdao) to remote oceans (wide area of North Pacific Ocean). The averaged $f_{\text{Fe,sol}}$ and total Fe concentrations for TSP in other outflow studies (NOTOGRO in Japan and GOSAN in South Korea) and over East China Sea (Hsu et al., 2010) overlapped those evaluated in this study.

In the following, possible factors controlling the variations in $f_{\text{Fe,sol}}$ were further investigated. R_{dustFe} for $\text{PM}_{2.5}$ and PM_{10} aerosol particles were evaluated using the MLR analyses of Fe (see Section 3.3.2). The R_{dustFe} for coarse mode particles was estimated based on the differences in R_{dustFe} between $\text{PM}_{2.5}$ and PM_{10} aerosol particles. The R_{dustFe} values were compared with $f_{\text{Fe,sol}}$ for total, fine-, and coarse-mode particles (**Figs. 6 and 7a–b**) to investigate the impacts of Asian dust, which has typically very low $f_{\text{Fe,sol}}$ (e.g., Kurisu et al., 2021), on size-segregated $f_{\text{Fe,sol}}$.”

Table S5. The summary of previous studies compared in Fig. S11.

Place/Region	Latitude and longitude	Seasons and years	Size Classification	Leaching	Air mass category	Reference
Qingdao, China	36.10° N, 120.55° E	Winter-spring of 2012-2013	TSP	Ultrapure water	Continent	Shi et al. (2020)
Qingdao, China	36.34° N, 120.67° E	Winter of 2019	Fine < 1 µm, Coarse > 1 µm	DI water	Continent	Zhang et al. (2020)
GOSAN, South Korea	33.29° N, 126.16° E	Spring of 2001	TSP	Ultrapure water	Continental outflow	Chuang et al. (2005)
NOTOGRO, Japan	37.45° N, 137.36° E	Winter-spring of 2019–2020	Fine < 1.3 µm, Coarse > 1.3 µm	Ultrapure water	Continental outflow	Sakata et al. (2025)
Fukue, Japan	32.75° N, 128.68° E	Winter–spring of 2023–2024	Fine < 2.5 µm, Coarse > 2.5 µm	Ultrapure water	Continental outflow	This study
East China Sea	22–50° N, 140° E–150° E	Spring of 2005, 2006, and 2007 and November 2006	TSP	DI water	Continental outflow	Hsu et al. (2010)
North Pacific	22–50° N, 140° E–150° W	Late spring–early summer of 2002	TSP	DI water	Remote ocean	Buck et al. (2006)
North Pacific	15–30° N, 170° E–160° W	April 2001, July, September, October 2002, August 2003	Fine < 2.5 µm, Coarse > 2.5 µm	pH 4.5 formate buffer	Remote ocean	Chen (2004)
Northwestern Pacific	20–47° N, 140° E–167° E	Winter of 2013-2014 and summer of 2014	Fine < 2.5 µm, Coarse > 2.5 µm	Ultrapure water	Remote ocean	Kurisu et al. (2021)
Northeastern Pacific	22–32° N, 123° W–158° W	Spring of 2006	TSP	Seawater	Remote ocean	Wu et al. (2007)

9. The interpretation of Fe solubility remains largely descriptive and does not adequately discuss the atmospheric mechanisms controlling Fe dissolution.

We appreciate the suggestion.

In Introduction, we have described the following.

“Although only a small fraction of total aerosol Fe is water-soluble (e.g., Sholkovitz et al., 2012), the fractional solubility of Fe varies substantially depending on the aerosol source, atmospheric processing such as acidification, and cloud processing during long-range transport (e.g., Ito et al., 2020; Mahowald et al., 2018).”

To include more about the background of aerosol Fe dissolutions affecting the fractional solubilities of Fe, we modified this as follows.

“Although only a small fraction of total aerosol Fe is generally water-soluble (e.g., Sholkovitz et al., 2012), its fractional solubility varies substantially with aerosol source, particle size, and atmospheric processing. Fe solubilization can be promoted by acidification with sulfuric and nitric acids, while being suppressed by neutralization using alkaline components such as calcium carbonate (e.g., Meskhidze et al., 2005; Ito and Feng, 2010). Complexation with organic ligands can further promote Fe dissolution and stabilize dissolved Fe (e.g., Sakata et al., 2025), while photochemical reactions can alter Fe dissolution and redox speciation (e.g., Chen and Grassian, 2013). These processes may be further facilitated by aqueous-phase processing in clouds and fog during long-range atmospheric transport (e.g., Shi et al., 2015, 2020).”

We modified the last sentence of third paragraph of Section 3.4 as follows.

“Even though some processes affecting $f_{\text{Fe,sol}}$, especially the impacts of organic ligands (like oxalic acid) and photochemical reactions, were not elucidated in this study, the observed relationships between $f_{\text{Fe,sol}}$ and air mass histories can be useful constraints for evaluating and refining model simulations of the processes controlling $f_{\text{Fe,sol}}$ (e.g., aqueous phase reactions)”

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