

Reviewer 1

	Comments	Reply
1.1.	<p>Aerosol Fe solubility is a key parameter for impacts of aerosol Fe on marine biogeochemistry, is still not well constrained. Several sources and processes may contribute to dissolved aerosol Fe, but quantitative explanations are still difficult. Sakata et al. explored the possibility to use the ratio of dissolved Al to dissolved Fe to understand factors which control aerosol Fe solubility in fine and coarse particles, and then discussed sources of dissolved aerosol Fe. This idea is novel, and the results are very interesting. This manuscript can still be substantially improved to increase its readability, clarity and impacts. I also posted a community comment 28 March, and the authors may also need to take it into account.</p> <p>Overall, I urge the authors to check this manuscript carefully and thoroughly, as there are a lot of language issues. Professional editing is also recommended. In the community comment I posted on 28 March, I provided a few examples.</p>	<p>We sincerely thank the reviewer for the time and effort put into this review. We have carefully revised the manuscript with full consideration of the comments and suggestions provided. We also reply to your comments posted as community comments in this sheet. Please find the point-to-point replies listed below. We apologize for the inconvenience for your peer-preview processes due to our poor English. English editing was conducted by a native English speaker prior to submission.</p> <p>“Revised text as it appears in the text (in quotes, blue font)”</p>
1.2.	<p>Line 15-17: The second sentence in the abstract largely repeats the first sentence. In addition, there are some typos elsewhere in this manuscript (for example, line 305-306; line 29: should there be "for fine particles" after "from 0.386 to 4.67"?). The authors may want to check the language thoroughly.</p>	<p>Thank you for pointing out. We have removed the first sentence in abstract.</p>
1.3.	<p>Line 155-159: It would be nice to provide values used for $[X]/[Na^+]$ for seawater and references.</p>	<p>The molar ratios of target ions relative to Na^+ have been added in the manuscript.</p> <p>The molar ratios of $[K^+]/[Na^+]$, $[Mg^{2+}]/[Na^+]$, $[Ca^{2+}]/[Na^+]$, and $[SO_4^{2-}]/[Na^+]$ were 0.0213, 0.113, 0.0213, and 0.0596, respectively (Nozaki, 2001).</p>
1.4.	<p>Line 267-268: Although these numbers can be found in the SI, could the author provide average EF values of Fe in TSP for different seasons?</p>	<p>Thank you for the suggestions. We provided EF values in TSP derived from the Japanese atmosphere (in summer and fall) and East Asia.</p> <p>The average EF of Fe in the TSP samples collected from summer to fall (June to October) was 2.84 ± 0.83 when air masses mainly originated from Japan (Fig. S6a). By contrast, the average EF of Fe in the TSP derived from East Asia in winter and spring was 1.57 ± 0.35 (Fig. S6a). Thus, the influences of</p>

		anthropogenic emission on the total Fe in TSP are more significant in the air mass derived from Japan than that from East Asia (Figs. S1, S2, and S6a).
1.5.	Line 305-306: This sentence “The d-Fe and d-Al of fine aerosol particles were $72.0 \pm 8.87\%$ and $53.1 \pm 9.90\%$, respectively” may need revision.	<p>Thank you for pointing out. The sentence has been improved as below.</p> <p>The average d-Fe and d-Al concentrations of fine aerosol particles were 27.3 ± 17.4 and 14.3 ± 10.9 ng m⁻³, which accounted for $72.0 \pm 8.9\%$ and $53.1 \pm 9.9\%$ of d-Fe and d-Al in the TSP, respectively.</p>
1.6.	<p>Line 311-318: Our previous work (Zhang et al., 2022; Zhang et al., 2023) also found and try to explain enrichment of dissolved Fe in fine particles, when compared to total Fe.</p> <p>Recently we published two papers (Zhang et al., 2022; Zhang et al., 2023), which discussed sources of dissolved Fe and Fe solubility for fine and coarse particles and are highly relevant to the work presented by Sakata et al. Our two papers reported many similar results as Sakata et al., including what Sakata et al. presented in Sections 3.3-3.4. There are also some differences between our work and the work by Sakata et al. (2023). Therefore, I would like to bring the authors' attention to our recent work (Zhang et al., 2022; Zhang et al., 2023).</p> <p>Section 3.4.2: Our previous studies (Zhang et al., 2022; Zhang et al., 2023) also suggested that chemical aging played a more important role in regulating Fe solubility in coarse particles than fine particles.</p>	<p>These three comments are similar and have been combined into one. Thank you for bringing our attention to the two papers. We briefly compared the result between our study and your previous studies, and the following sentence was added to the manuscript.</p> <p>These results are reasonable and supported by previous studies reporting that mineral dust and anthropogenic Fe in fine aerosol particles were covered by sulfate, and internal mixing of these particles with sulfate promoted the dissolution of Fe (Sullivan et al., 2007; Fitzgerald et al., 2015; Li et al., 2017, Zhu, Y. et al., 2020, 2022). However, compared with the correlation factor of Fesol% with [nss-SO4²⁻]/[total Fe], coarse aerosol particles yielded a higher correlation factor than fine aerosol particles (Figs. 4a and 4c). Similar results have been reported by previous studies on Fesol% in China, which indicated the presence of anthropogenic Fe with high Fesol% as one of the reasons for the low value of the correlation factor (Zhang et al., 2022, 2023). Therefore, the low correlation between Fesol% and [nss-SO4²⁻]/[total Fe] of fine aerosol particles implied that acidification of mineral dust was not the sole factor controlling Fesol%.</p> <p>There are several different points in the results between this and your studies, but we did not discuss the reasons. This is because (i) it is not easy to compare results between this and your studies due to many differences in the sampling methods (e.g., stage numbers of impactors, sampling periods, and date), and (ii) detailed comparisons between these studies would make this paper longer and consequently conflict with your other comment (No. 1.7).</p>

		<p>The difference between our and your studies would be whether $\text{Fe}_{\text{sol}}\%$ is correlated with $[\text{NO}_3^-]/[\text{total-Fe}]$ or not. In the case of coarse aerosol particles, we believed that HNO_3 mainly reacted with CaCO_3 in mineral dust as described in Supplemental Note. In the case of fine aerosol particles, no relationship between the $\text{Fe}_{\text{sol}}\%$ and $[\text{NO}_3^-]/[\text{total-Fe}]$ seems to be found because NO_3^- in aerosols prefers the vapor phase due to its low solubility in water under high temperature and acidic conditions. Indeed, Zhang et al. (2023) have reported that the correlation between $\text{Fe}_{\text{sol}}\%$ and $[\text{NO}_3^-]/[\text{total-Fe}]$ in summer was worse. Therefore, the nitrate evaporation associated with pH cycling during aerosol transport is likely the reason for no correlation between $[\text{NO}_3^-]/[\text{total-Fe}]$, but further research is needed.</p>
1.7.	<p>It may increase the readability to group Sections 3.5-3.7 into a new section. The current Section 3 is very long, and contents in Sections 3.5-3.7 are different from Section 3.1-3.4?</p> <p>In addition, I feel the current manuscript is very long, and the quality of some figures is not good (they contain many panels and are very busy). Sections 3.5-3.7 contain key information in this manuscript, but are not easy to follow. Perhaps the authors could improve them during the revision, but I cannot provide specific suggestions.</p>	<p>Sections 3.5-3.7 of the first version were rearranged into a new section, Section 4 (title: Establishing the source estimation method for d-Fe using $[\text{d-Fe}]/[\text{d-Al}]$) in the revised version. This section has been improved to increase the readability.</p> <p>Regarding the paper length, the section on the ion concentration and several figures were moved to Supplemental Information. We hope you could understand our decision that we did not reduce the text length for the other sections to avoid ambiguity in the significance of the newly established indicators for source apportionment of d-Fe.</p>