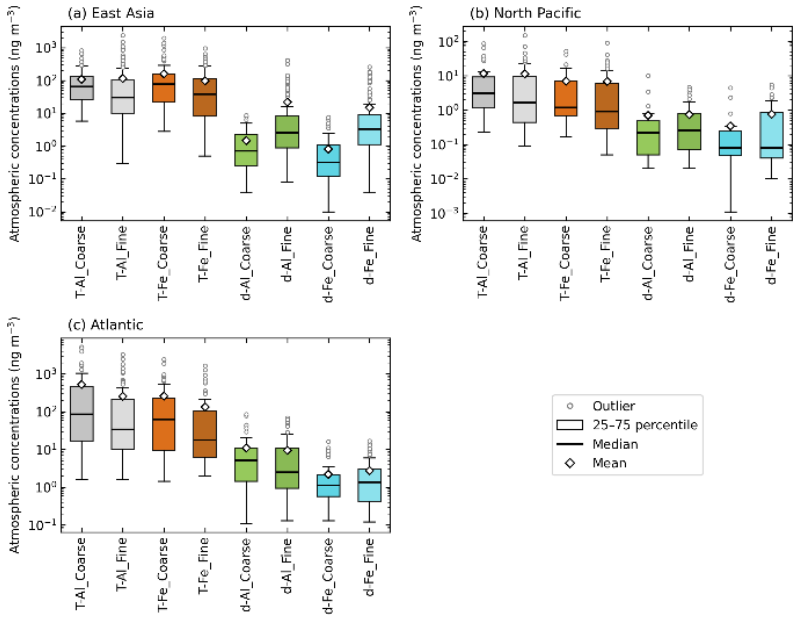
Reviewer 1

	Reviewer's comments	Author's reply
	This is a synthesis study on Aerosol Iron fractional solubility, compiling all available soluble Al and Fe data over the Atlantic and Pacific oceans. This study has high scientific significance, improving our current understanding of aerosol iron solubility. This paper improves the understanding of the traditional hyperbolic relation between soluble iron and dust/Aluminum. The scientific quality and presentation quality of the paper are excellent and need minimal improvement. The synthesis of a relationship diagram between the EF T-Fe and [d-Fe]/[d-Al] ratio over a large geographical area has global significance. This study will also motivate atmospheric geochemists to make concordant observations of dissolved iron and aluminium. Before the publication of this manuscript, the author should address the following major corrections, which will improve the manuscript. In general, all the axis labels for similar plots should be the same, which should agree with the text description.	We sincerely thank the reviewer for the constructive and encouraging comments on our manuscript. We appreciate the reviewer's positive assessment of the scientific significance of this synthesis study, particularly the compilation of soluble Fe and Al data over the Atlantic and Pacific Oceans and the development of the $EF_{T\text{-}Fe}$–[d-Fe]/[d-Al] diagram. In response to the reviewer's comments, we have carefully revised the manuscript. We have addressed all the major and technical comments as described below. A comprehensive list of changes, including grammatical corrections, is provided in the attached response sheet. Red text shows the text after revision.

Specific comments

	Reviewer's comments	Author's reply
1 .	The authors considered size-segregated samples from their unpublished data for this study, described in line 75. In the supplementary material, they described that the samples were collected in 7 different sizes. Which size fraction do they consider in this study, and why do they consider it? This information is missing.	Thank you for this comment. We have added information on the size fractions of the aerosol samples from the unpublished dataset to the Supplementary Information. The reason for including these samples is that size-segregated aerosol datasets are extremely limited, and, in the Southern Hemisphere, no other samples are available for which all of the T-Fe, T-Al, d-Fe, and d-Al concentrations are available. The aerodynamic diameter ranges for stages 1–7 were >10.2 μm, 4.2–10.2 μm, 2.1–4.2 μm, 1.3–2.1 μm, 0.69–1.3 μm, 0.39–0.69 μm, and <0.39 μm, respectively. In this study, particles larger than 2.1 μm were grouped as coarse aerosol particles, whereas particles of 2.1 μm and smaller were grouped as fine aerosol particles.
2.	The author considers that "the ultrapure water and ammonium acetate buffer were uniformly classified as d-Fe, with the same approach applied for Al." (line 100). A	We address the two questions together below. Although these comments concern Section 2.1, we added the relevant explanation to Section 2.3, where this issue is discussed more directly.

3.	recent assessment of leaching methodology highlighted the impact of different leaching media on aerosol samples (Tang, Mingjin, et al., 2025). The source of Al is traditionally considered to be of crustal origin, whereas Fe can be derived from multiple sources; this can impact the leachability of these metals, which is also discussed in this manuscript. How, then, do the authors justify this difference in terms of d-Fe and d-Al concentrations? The authors should discuss this hypothesis. In addition, they should evaluate the potential changes caused by the different leaching media in their current study. If they use the correlation factor 1.4 proposed by Perron et al. (2020b) for Ammonium acetate leach, do they observe any difference in their current observation (line 104)? It should be explained, and this explanation will be helpful for future observations	Acetate buffer was mainly used for samples collected over the Atlantic Ocean, where Saharan dust influence is substantial. Therefore, as reported by Tang et al. (2025), Fe_{sol}% and Al_{sol}% extracted with acetate buffer may be higher than those obtained with MQ water. However, we consider that this effect has little influence on the source apportionment in this study, particularly on the $EF_{T\text{-}Fe}$–[d-Fe]/[d-Al] diagram. This is because the increases in Fe and Al extracted by acetate buffer relative to MQ water are generally similar. Therefore, even if acetate buffer increases both Fe and Al solubilities, the [d-Fe]/[d-Al] ratio itself is not expected to change substantially. In addition, Clough et al. (2019) reported that extraction with gluconic acid, which is a relatively weak ligand similar to acetate, increased Fe and Al solubilities by approximately 1.5 and 1.6 times, respectively, relative to MQ water. Thus, relatively weak ligands such as acetate and gluconic acid likely increase Fe and Al solubilities to a similar extent, and therefore have only a limited effect on the [d-Fe]/[d-Al] ratio. In contrast, an increase in the [d-Fe]/[d-Al] ratio due to ligand-promoted dissolution is observed in area (ii) of Figure 2. This is likely because strong ligands such as oxalate enhance Fe solubility more than Al solubility. Similar trends have also been reported in aerosol metal extraction experiments using strong ligands, including the Berger method (Clough et al., 2019; Perron et al., 2020b; Tang et al., 2025). As described above, this study treats Fe_{sol}% and Al_{sol}% obtained by ultrapure water and acetate buffer extractions in the same manner. Recent studies have reported that acetate buffer extraction yields higher Fe_{sol}% and Al_{sol}% than ultrapure water extraction (Clough et al., 2019; Perron et al., 2020b; Tang et al., 2025). However, the increases in Fe_{sol}% and Al_{sol}% caused by acetate buffer extraction are generally similar, and thus the [d-Fe]/[d-Al] ratio is not expected to differ substantially between MQ water and acetate buffer extractions. Therefore, the influence of differences in extraction methods on the source apportionment of Total-Fe and d-Fe based on this diagram is considered to be limited.
4. 5.	Line-125, include references for each discussed process. Line-138, put references for these observations, where aluminosilicate glasses emitted from the combustion process, coal burning .. having EF of Fe <2 but [d-Fe]/[d-Al] <0.1.	I have added appropriate references for each dissolution process. These areas differ based on the dissolution mechanisms of mineral dust. Area (i), where [d-Fe]/[d-Al] ranges from 0.1 to 1.0, is associated with proton-promoted dissolution

		(Kodama and Schnitzer, 1973; Desboeufs et al., 2001; Lowson et al., 2005; Duvall et al., 2008; Shi et al., 2011a, Bibi et al., 2015; Bray et al., 2015). Area (ii), where [d-Fe]/[d-Al] exceeds 1.0, is associated with ligand-promoted dissolution, for example by oxalate (Kodama and Schnitzer, 1973; Bray et al., 2015).
6.	In Eq.3 define *F*mineral and Fanthro	We apologize for the ambiguity. Here, $f_{mineral}$ and f_{anthro} refer to $f_{mineral\text{-}dFe}$ and $f_{anthro\text{-}dFe}$, respectively. In addition, as suggested in the technical comment, we have revised the notation by changing uppercase *F* to lowercase *f* throughout the relevant parts of the manuscript.
7.	Table 1, *Fanthro-dFe* (%) Write an equation for this calculation.	We apologize for the confusion caused by the inconsistent notation of $F_{anthro\text{-}dFe}$. In the revised manuscript, we have standardized the notation as $f_{anthro\text{-}dFe}$ throughout the text and tables. $f_{anthro\text{-}dFe}$ is defined by Eq. (2), and we have clarified this in the relevant part of the manuscript.
8.	Line-215, Author described the role of different particle size towards dissolve metal contribution, It should be supplemented by a figure.	We have added Figure S3, which presents box-and-whisker plots of T-Fe, T-Al, d-Al, and d-Fe concentrations in coarse and fine aerosol particles for each region/ocean. 
9.	Line-240, “similar Fe” which type of Fe authors are reefing, total Fe or dissolve Fe.	We have clarified that “similar Fe” refers to the component-specific Fe concentration plotted on the x-axis, not dissolved Fe concentration. Specifically, anthro-Fe was divided into two groups relative to mineral-Fe at comparable Fe concentrations: one with lower Fe_{sol}% than

		mineral-Fe and the other overlapping with mineral-Fe in both Fe concentration and Fe_{sol}%. Anthro-Fe in TSP could be further divided into two groups in samples where anthro-Fe and mineral-Fe concentrations were comparable, based on the Fe_{sol}% of anthro-Fe relative to that of mineral-Fe. One group showed lower Fe_{sol}% than mineral-Fe, whereas the other showed Fe_{sol}% similar to mineral-Fe.
10. 11.	Line-240-243, The boundary between…and mineral-Fesol%". Rewrite this line, it is hard to follow. Line-276-278, How is the boundary line between combusted and non-combusted anthro-Fe defined?	Given that these three comments concern related issues, we respond to them collectively below. In this study, anthro-Fe with lower solubility than mineral-Fe was operationally classified as non-combusted anthro-Fe after accounting for the atmospheric alteration of both Fe components. Anthro-Fe with solubility comparable to or higher than that of mineral-Fe was considered to be primarily derived from high-temperature combustion. The classification criteria and the method used to determine the boundary have been clarified in the revised manuscript, as described below. Anthro-Fe in East Asian TSP exhibited a wider range of anthro-Fe_{sol}% than mineral-Fe (Figure 4b). Although combustion-derived anthro-Fe is generally considered to be more soluble than mineral-Fe, some samples showed anthro-Fe_{sol}% lower than mineral-Fe_{sol}% and below 0.1%, the approximate lower limit reported for combustion-derived Fe. Such low-solubility anthro-Fe was likely derived from sources other than high-temperature combustion. To account for changes in the solubility of mineral-Fe and anthro-Fe during atmospheric transport and to avoid imposing a fixed threshold such as 0.1%, a boundary line parallel to and below the log–log regression line between mineral-Fe concentration and mineral-Fe_{sol}% was defined such that 5% of the mineral-Fe samples fell below it. For each mineral-Fe sample, the relative deviation from the regression relationship was quantified as the logarithm of the observed-to-predicted mineral-Fe_{sol}% ratio [log(observed mineral-Fe_{sol}%/predicted mineral-Fe_{sol}%)]. The 5th percentile of these relative deviations was then added to the intercept of the regression equation to define the boundary line. Based on this boundary, anthro-Fe samples below the line were operationally classified as non-combusted anthro-Fe, whereas those above it were classified as high-temperature combustion-derived anthro-Fe, with solubility

		comparable to or higher than that of mineral-Fe (Figure 4b). In the non-combusted anthro-Fe group below the boundary, all anthro-Fe_{sol}% values were below 1.0%, with an average of 0.3 ± 0.2%. Several samples with high anthro-Fe concentrations showed particularly low values of less than 0.1%. Such low solubility is consistent with non-combustion anthro-Fe, including Fe associated with brake-pad debris and tire-wear particles (Halle et al., 2013; Shupert et al., 2021; Cui et al., 2025). By contrast, the high-temperature combustion-derived anthro-Fe group above the boundary had an average anthro-Fe_{sol}% of 4.0 ± 5.7%, higher than the average mineral-Fe_{sol}% of 2.4 ± 4.1% in East Asian TSP. This characteristic is consistent with the generally higher solubility of anthro-Fe derived from high-temperature combustion. However, anthro-Fe_{sol}% was generally only approximately 1% even in high-concentration samples dominated by relatively fresh aerosol particles. This observation implies that these particles were emitted with relatively low initial solubility rather than in a highly soluble form and that they subsequently underwent solubilization through chemical processing during atmospheric transport (Sholkovitz et al., 2009; Ito et al., 2021).
12.	Line-444-447, The supplementary Figure S3b clearly shows a significant number of samples were being plotted in area (v), Add a description on these samples.	The [d-Fe]/[d-Al] ratios of mineral dust collected in the Sahara Desert and mineral dust-rich aerosols are reported to be 0.10 ± 0.04, and values below 0.1 are frequently observed (Shi et al., 2011; Desboeufs et al., 2024). Therefore, the larger number of samples plotted in area (v) compared with the other oceanic regions likely reflects the influence of Saharan dust. Atlantic aerosols showed a relatively large number of samples in area (v), where the influence of fly ash derived from high-temperature combustion is generally expected to be significant, compared with the other oceanic regions (Figure S4b). Considering that Saharan dust often exhibits [d-Fe]/[d-Al] ratios below 0.10, the samples plotted in area (v) likely reflect the contribution of Saharan dust with low [d-Fe]/[d-Al] ratios, rather than fly ash derived from high-temperature combustion. This interpretation is consistent with the near absence of samples in the area characterized by high-temperature combustion-derived $EF_{T\text{-}Fe}$ and anthro-Fe with high [d-Fe]/[d-Al] ratios. By contrast, aerosols with

		high $EF_{T\text{-}Fe}$ were mainly plotted in area (iii), indicating that anthro-Fe over the Atlantic was likely present mainly as insoluble Fe. We have also revised the Methods section to mention that the [d-Fe]/[d-Al] ratio of Saharan dust can be lower than 0.1. Saharan dust has a [d-Fe]/[d-Al] ratio of 0.10 ± 0.04, close to the lower limit of the [d-Fe]/[d-Al] ratio observed for aluminosilicate minerals, and values below 0.1 are frequently reported (Shi et al., 2011; Desboeufs et al., 2024).
13.	In Figure 5, 8 and 11correlation plot should contain the boundary line equation.	As recommended, we have added the equations of the boundary lines to the correlation plots in Figures 5, 8, and 11.
14.	Line-472, As per my understanding, the figure number is wrong, correct the figure numbers in this line and as well as all subsequent lines.	We apologize for the inconvenience. We have rechecked the figure numbers and corrected them.
15.	Line-472 and 491, Instead of using the word summer season, either use months or define the summer months. The study area includes both the hemisphere; it is hard to follow the season. Do this correction for all the possible places like line-497,501...	Thank you for this suggestion. We agree with your comment and have revised the text to specify the relevant months, such as July–August, instead of using seasonal terms. For consistency, we have also applied the same revision to regions other than the Atlantic Ocean.
16.	Line-516-517, Heard Island located at southwest of Australia change the statement according to that.	Thank you for pointing this out. I have revised it.

Technical comments

	Reviewer's comments	Author's reply
1.	In line 66, the authors mentioned they used "the molar ratio of d-Fe to dissolved Al…" Is it the molar ratio or the mass ratio?	In this study, we used the molar concentration ratio of dissolved Fe to dissolved Al. The molar concentration ratio was adopted for practical reasons, as it provides a simple and intuitive range of 0.1–1.0 for acid-promoted dissolution of mineral dust.
2.	Line-124, label EFT-Fe does not match the Figure1 vertical axis. Change the vertical axis label in Figure 1.	Thank you for your comment. We have revised all diagrams so that the y-axis label is consistently shown as $EF_{T\text{-}Fe}$.
3.	Line-162, correct the notation *Fmineral* and *Fanthro*. In Eq. 2 and line 142 use the notation "*Fmineral-dFe*"for the same parameter. Instead of using "F", it is suggested to use "*f*" or some other symbol for smooth reading.	Thank you for this suggestion. We have changed *F* to lowercase *f* throughout the relevant parts of the revised manuscript.

4.	Line-140 change the 0.10 to 0.1.	We appreciate the reviewer's comment. We have corrected 0.10–1.0 to 0.10–1.00 for consistency with the notation used elsewhere.
5.	Line-192 expands the "EF" either to Enrichment factor or EFT-Fe.	Thank you for pointing this out. We have rephrased EF as $EF_{T\text{-}Fe}$.
6. 7. 8.	Figure 2(b) correct the Y axis to EFT-Fe. Figure 3 Put the a and b caption on the figure. Figure 5 (d,e,f) solid black line should be included in the legend	Thank you for pointing this out.
9. 10. 12.	Line-230, Typo correction "mineral-F_{esol}%" Line-239, Typo error "Fesol%". Line-275, Typo error" Fe_{so}l%".	Thank you for pointing out. I have revised them. We have revised these figures to address the issues pointed out by the reviewer.
11.	Line-258,"plotted in the area(i) and (ii)"of what? It should be mentioned.	Thank you for pointing this out. We have clarified the meaning of "areas (i) and (iii)" in this sentence and made similar revisions in the other relevant parts of the manuscript. This result indicated that T-Fe in East Asina TSPs was mainly originated from mineral dust and insoluble anthro-Fe, and d-Fe was derived from proton-promoted dissolutions of mineral dust.
13. 14. 15. 16.	Line-311, There is no Figure 4f. Line-436, Refer the figure number for the correlation slope. Line-443, Correct the figure number. Line-435, The Figure 8a is about North Pacific Fe? Correct the Figure number.	We apologize for any inconvenience caused. We have checked these issues and revised the manuscript accordingly.
17.	Line-445-446, Describe the area(iii) and area (iv)of which type of figure.	We have labeled areas (i)–(v) in the figure and revised the figure caption as follows. Figure S4. Diagrams between [d-Fe]/[d-Al] ratio and EFT-Fe in TSP collected above (a) the North Pacific, (b) the Atlantic, and (c) the South Pacific Oceans. Areas (i)–(v) indicate different Fe sources and dissolution processes: (i) proton-promoted dissolution of mineral dust, (ii) ligand-promoted dissolution of mineral dust, (iii) insoluble anthro-Fe, (iv) highly soluble anthro-Fe, and (v) aluminosilicate glass derived from sources such as coal fly ash.
18.	Line-477-478, Insert the figure number.	We have added the corresponding figure numbers at the end of the relevant sentences.
19.	Line 546 and line 548, instead of pelagic, use the word open ocean. The pelagic region can be misleading.	We have revised them.

20.	Typographic error in Supplementary Figure S1. “(b)between d-Al and d-Fe concentratir4eons”	We have revised it.

Reviewer 2

Reviewer's comments	Author's reply
The authors have assembled an impressive dataset of aerosol concentration data from previous studies as well as unpublished data of their own. Focusing on total and soluble aerosol iron, along with supporting data that includes total and soluble aluminum, they have attempted to better understand the long recognized inverse relationship between the amount of aerosol iron and its solubility. They explain the data using a previously published framework that associates the calculated enrichment factor of aerosol iron relative to the upper continental crust and the ratio of soluble iron to soluble aluminum. Based on this analysis, they conclude that observed increases in aerosol iron solubility are indicative of atmospheric processing during transport rather than simple mixing of high abundance, low solubility mineral aerosol with low abundance, high solubility anthropogenic aerosol. The question of what processes and factors control aerosol iron solubility has bedeviled the research community and hampered progress in our ability to model this important input to the surface ocean. The methodology applied in this study shows promise, though its broad applicability is not clear. The soluble aerosol observations utilized in this study are biased toward 'mild' leaching techniques which may not be indicative of the ultimate aerosol solubility, and more so, the bioavailability of these aerosol derived trace elements. This comment is not an indictment of the study, and certainly does not place it out of step with the current state of the art in the field. The work is an important contribution that could be improved with revision as detailed in the comments to follow.	We sincerely thank the reviewer for the careful evaluation of our manuscript and for the constructive and positive comments regarding the significance of our dataset and the proposed framework for interpreting aerosol Fe solubility. We appreciate the reviewer's recognition that our study provides an important contribution toward understanding the processes controlling aerosol Fe solubility. We agree that measured aerosol Fe solubility depends on the extraction protocol and that operationally defined soluble Fe cannot be regarded as equivalent to the ultimately bioavailable fraction. Nevertheless, relatively mild extraction methods, such as ultrapure water and ammonium acetate extractions, have an important advantage for compiling a broad aerosol dataset. Their limited acidity and complexing capacity minimize additional dissolution of Fe and Al during the extraction procedure, allowing the fraction already to present in a soluble form to be evaluated with relatively straightforward interpretation. More aggressive extraction methods may dissolve additional Fe from initially insoluble particles, making it difficult to distinguish Fe that was already soluble in the aerosol from Fe mobilized by the extraction itself. Because dissolution is also a necessary step for the biological utilization of aerosol-derived Fe, we consider ultrapure water and ammonium acetate extractions to provide a reasonable and internally interpretable basis for the comprehensive comparison conducted in this study, while recognizing that they do not directly quantify ultimate bioavailability. We have addressed all the major and technical comments as described below. A comprehensive list of changes, including grammatical corrections, is provided in the attached response sheet. Red text shows the text after revision.
The abstract states that both mineral-Fe and anthro-Fe showed inverse relationships between aerosol concentration and solubility and that this indicates atmospheric processing, rather than two-component mixing, is driving trends in increasing aerosol iron dissolution. This finding appears to be a fundamental outcome of the study; however, it does not receive any mention	Thank you for this valuable comment. The inverse relationships between concentration and solubility for both mineral-Fe and anthro-Fe are important findings of this study, because they may provide insights into the factors controlling the solubility of each Fe component. This point has been clarified by adding the following text to the first paragraph of the Implications

in the “Implications” section which concludes the manuscript. I suggest the authors incorporate additional concluding remarks to drive home the important findings of the study.	section. The factors controlling Fe_{sol}% in aerosol particles were investigated by compiling data for total and dissolved Fe and Al in aerosol particles. An inverse relationship between total Fe and Fesol% was observed, as has been reported in previous studies (Sholkovitz et al., 2009; Mahowald et al., 2018). When similar plots were constructed separately for mineral-Fe and anthro-Fe, inverse relationships between concentration and solubility were found for both components. The inverse relationship for mineral-Fe is likely explained by chemical alteration during atmospheric transport, because freshly emitted or unaged mineral particles generally have low Fe_{sol}%. In contrast, the anthro-Fe results indicated more diverse behavior. In samples with comparable anthro-Fe and mineral-Fe concentrations, two major groups were identified: one in which anthro-Fe_{sol}% was lower than mineral-Fe_{sol}%, and another in which anthro-Fe_{sol}% was comparable to mineral-Fe_{sol}%. The former group was mainly found in coarse aerosol particles collected in East Asia and showed extremely low anthro-Fesol% (<0.1%), suggesting contributions from brake-pad wear from vehicles and related sources. The latter group was mainly found in fine aerosol particles and showed a high average anthro-Fe_{sol}% of 22.2 ± 24.0%. Although anthro-Fe in fine particles is generally associated with high-temperature combustion processes, only a limited number of samples had anthro-Fe_{sol}% higher than that reported for highly soluble emissions such as heavy oil combustion (>37.0%). This suggests that part of the anthro-Fe emitted with initially low solubility from solid fuel combustion, including coal combustion and the steel industry, was solubilized during atmospheric transport. Similar inverse relationships between concentrations and solubilities for mineral-Fe and anthro-Fe were also observed in marine aerosol particles. Therefore, inverse-correlation plots for mineral-Fe and anthro-Fe may provide a useful approach for evaluating source-dependent variations in Fe_{sol}% and for distinguishing anthro-Fe derived from high-temperature combustion from that derived from non-high-temperature combustion sources.

P4, L67: Are monthly mean concentrations appropriate? How was the spatial variability in the observations accounted for? The manuscript should include greater detail in how observations were binned both temporally and spatially.	Thank you for this important comment. We acknowledge that the Fe and Al concentration data in aerosols used in this study were not obtained from repeated observations at fixed stations, but were compiled from observations over broad spatial regions. We also recognize that aerosol transport from continental source regions to the marine atmosphere can vary seasonally and interannually. Therefore, the monthly mean values were not interpreted as seasonal cycles specific to individual sampling locations, but as averages of broadly distributed observations grouped by calendar month. In the revised manuscript, we clarified that the discussion focuses on the North Pacific region, which is strongly influenced by East Asian outflow, and on the Atlantic region, particularly 0–30°N, where Saharan dust strongly affects aerosol composition. We also interpreted the shipboard observations in relation to monthly variations in Fe and Al concentrations and solubilities observed at or near the major source regions. The similarity in seasonal variations between ground-based and shipboard observations supports the interpretation that the observed monthly patterns reflect broad seasonal changes in aerosol sources, transport, and atmospheric processing, rather than only spatial sampling bias. Although this compiled dataset has limitations, long-term, fixed-location, high-time-resolution observations of marine aerosols from research vessels are extremely difficult to obtain. We therefore believe that compiling shipboard observations over broad oceanic regions is important for constraining the magnitude and seasonal variability of Fe and Al concentrations and solubilities over ocean basins, which are otherwise often evaluated mainly using chemical transport models.
P4, L81-83: Can you elaborate on what is meant by filter choice influencing the solubility results? Presumably, all the data was blank corrected for the filter background as appropriate.	We apologize that our original statement was unclear. We intended to refer to the differences in Fe solubility among filter materials reported by Buck and Paytan (2012). However, after further consideration, we concluded that it was not possible to quantitatively evaluate the effects of filter material in the present study. Accordingly, we have removed the relevant text from the revised manuscript.
P4, L82: typo in Buck and Paytan, 2012	I have corrected it. “Pytan” has been changed to “Paytan”.
P4, L91: Cite a reference supporting the point that HF is required to break down Si-O bonds.	We have added several papers that mentioned HF promote digestion of Si-O bond.

I do not think “It is known...” is sufficient here.	A critical consideration was whether hydrofluoric acid (HF) was incorporated in the digestion process. It is known that the incomplete digestion of Si-O bond present in aluminosilicates, due to the omission of HF, leads to an underestimation of the T-Fe and T-Al concentrations that are substituted or trapped within it (Chaoand Sanzolne, 1992; Mitra and Rimstidt, 2009).
P4, L93: Correct to “...we only compiled concentrations obtained...”	Thank you for pointing out. I have revised it.
P5, L100: The authors might discuss any reported discrepancies between ultrapure water soluble and ammonium acetate soluble trace elements. Tang et al. (doi.org/10.5194/amt-18-6125-2025, 2025)	Acetate buffer was mainly used for samples collected over the Atlantic Ocean, where Saharan dust influence is substantial. Therefore, as reported by Tang et al. (2025), Fesol% and Alsol% extracted with acetate buffer may be higher than those obtained with MQ water. However, we consider that this effect has little influence on the source apportionment in this study, particularly on the EFT-Fe–[d-Fe]/[d-Al] diagram. This is because the increases in Fe and Al extracted by acetate buffer relative to MQ water are generally similar. Therefore, even if acetate buffer increases both Fe and Al solubilities, the [d-Fe]/[d-Al] ratio itself is not expected to change substantially. In addition, Clough et al. (2019) reported that extraction with gluconic acid, which is a relatively weak ligand similar to acetate, increased Fe and Al solubilities by approximately 1.5 and 1.6 times, respectively, relative to MQ water. Thus, relatively weak ligands such as acetate and gluconic acid likely increase Fe and Al solubilities to a similar extent, and therefore have only a limited effect on the [d-Fe]/[d-Al] ratio. In contrast, an increase in the [d-Fe]/[d-Al] ratio due to ligand-promoted dissolution is observed in area (ii) of Figure 2. This is likely because strong ligands such as oxalate enhance Fe solubility more than Al solubility. Similar trends have also been reported in aerosol metal extraction experiments using strong ligands, including the Berger method (Clough et al., 2019; Perron et al., 2020b; Tang et al., 2025). As described above, this study treats Fe_{sol}% and Al_{sol}% obtained by ultrapure water and acetate buffer extractions in the same manner. Recent studies have reported that acetate buffer extraction yields higher Fe_{sol}% and Al_{sol}% than ultrapure water extraction (Clough et al., 2019; Perron et al., 2020b; Tang et al., 2025). However, the increases in Fe_{sol}% and Al_{sol}% caused by acetate buffer extraction are generally similar, and thus the

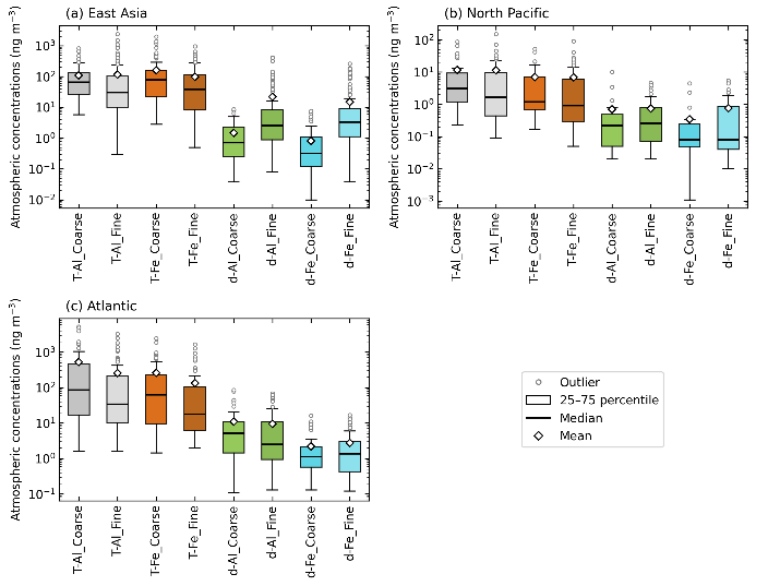
	[d-Fe]/[d-Al] ratio is not expected to differ substantially between MQ water and acetate buffer extractions. Therefore, the influence of differences in extraction methods on the source apportionment of Total-Fe and d-Fe based on this diagram is considered to be limited.
P5, L115-116: Is T-Fe/T-Al > 5.18 the calculated ratio of Fe/Al? As apposed to the enrichment factor? This is confusing.	Thank you for this comment. The text has been revised to distinguish $EF_{T\text{-}Fe}$ from the measured T-Fe/T-Al ratio, as follows: In this study, the mean T-Fe/T-Al ratio of 0.52 ± 0.12 in five studies was employed with the consideration of variability of the T-Fe/T-Al ratio in the upper continental crust (Turekian, 1961; Taylor, 1964; Taylor and McLennan, 1995; Wedepohl, 1995; Rudnick and Gao, 2003). $EF_{T\text{-}Fe}$ values exceeding 10 are commonly used to indicate a substantial contribution from anthro-Fe. This threshold corresponds to a measured T-Fe/T-Al ratio in aerosol particles exceeding 5.2. In this study, a lower $EF_{T\text{-}Fe}$ threshold of 2.0 was adopted to include samples with possible anthro-Fe contributions; this threshold corresponds to a measured T-Fe/T-Al ratio in aerosol particles exceeding 1.04.
P6, L137: I believe a work is missing between encompasses and exhibiting.	We apologize for the error in the original text. We have revised it as follows. The readily soluble anthro-Fe discussed here includes both initially soluble anthro-Fe with high Fe_{sol}% at emission and initially insoluble anthro-Fe plotted in area (iii), which subsequently becomes more soluble through atmospheric chemical processing.
P9, Table 1: The differentiation between areas is not clear in the table. What samples are “East Asia” and which are “N. Pacific”? A map showing the locations of the observations would help. Add horizontal gridlines separating the information for each area from one another. As is, the d-Fe source cells are hard to distinguish.	Thank you for pointing this out. “East Asia” refers exclusively to aerosol samples collected at terrestrial sites in China, Korea, and Japan, whereas “N. Pacific” refers to marine aerosol samples collected east of Japan during research cruises. We apologize for the poor readability of the original table. Table 1 has been reformatted to make the regional classifications and the information for each area easier to distinguish.

Table 1 Basin-scale summary of aerosol Fe (T-Fe and d-Fe ranges), Fe_{sol}%, and $f_{anthro\text{-}dFe}$ in East Asian and marine TSPs.

T-Fe sources	d-Fe sources	Mean T-Fe conc. ($ng\ m^{-3}$)	Mean d-Fe conc. ($ng\ m^{-3}$)	Fe_{sol}% (%)	$f_{anthro\text{-}dFe}$ (%)
East Asia: Only terrestrial aerosol samples collected in East Asia (e.g., China, Korea, and Japan)					
TSP and Coarse: Mineral-Fe Fine: Dust + anthro-Fe	TSP and coarse: Fe dissolution of dust Fine aerosols: Fe dissolution of dust and combusted anthro-Fe	1101.3 ± 1861.7 (15.6-14166.7)	12.6 ± 27.0 (0.6-98.0)	3.5 ± 3.2 (0.1-17.5)	7.9 ± 10.3
North Pacific: Open ocean aerosol collected during research cruise (Fig. 6)					
Mineral-Fe in Asian dust	Coarse: Enhanced Fe_{sol}% through chemical alteration in the marine atmosphere Fine: Mineral-Fe and anthro-Fe solubilized over East Asia and episodic anthro-Fe supply from shipping	50.0 ± 104.4 (0.2–764.5)	4.5 ± 7.3 (<0.1–45.9)	13.9 ± 14.4 (0.4–74.2)	6.1 ± 10.4
Atlantic: Open ocean aerosol collected during research cruise (Fig X)					
Mineral-Fe in Saharan dust	Fe dissolution from mineral-Fe through cloud processes	387.2 ± 922.9 (0.2–5650.0)	6.9 ± 23.4 (<0.1–212.7)	0.1–50.8 (6.2 ± 6.8)	5.1 ± 9.7
South Pacific: Open ocean aerosol collected during research cruise (Fig X)					
Mineral-Fe and volcanic-Fe from Heard Island		17.6 ± 25.3 (0.1–129.8)	1.0 ± 1.6 (<0.1–8.3)	7.3 ± 6.4 (<0.1–41.9)	5.1 ± 9.7

References. East Asia: Duvall et al., 2008; Kurisu et al., 2019; Hsieh et al., 2023; Sakata et al., 2023, 2025; Seo and Kim, 2023. The North Pacific: Buck et al., 2006, 2013; Marsay et al., 2022; Sakata et al., 2022; Kurisu et al., 2024. The Atlantic Ocean: Baker et al., 2006a, 2006b, 2013, 2020; Buck et al., 2010a, 2010b; Chance et al., 2015. The South Pacific: Buck et al., 2013, 2019; Sakata et al., 2022; Perron et al., 2020a, 2021.

P10, Fig. 2: Are the samples from the Indian/Southern Ocean not included?	We recognize that several observational studies on aerosol Fe have been conducted in the Indian Ocean and the Southern Ocean. However, because T-Al and d-Al concentrations were often not measured in these studies, they could not be incorporated into the present dataset.
P11, L215: Are the fine aerosol T-Fe and T-Al plotted somewhere? “In contrast...” implies the reader can compare the results.	Thank you for pointing this out. To address this comment, we have added Figure SX, showing box-and-whisker plots of the size distributions of T-Fe, T-Al, d-Fe, and d-Al in each region and oceanic area.

	
P12, L235: Are the mean bulk Fe% significantly different? It appears they might not be in some cases.	Thank you for this comment. Because the data of Fe_{sol}%, mineral-Fe_{sol}% and anthro-Fe_{sol}% were not normally distributed, differences of them between Chinese/Korean and Japanese aerosols were evaluated using Mann–Whitney U tests with Holm correction. Bulk Fe_{sol}%, mineral-Fe_{sol}%, and anthro-Fe_{sol}% were all significantly higher in Japanese TSP than in Chinese and Korean TSP (adjusted p = 0.0017, 0.0012, and 0.0057, respectively). The text has been revised to clarify that the reported mean ± SD values are descriptive statistics and that the statistical tests evaluated differences in the distributions of the two groups. These results indicate that the inverse relationship between Fe_{sol}% and T-Fe in East Asian TSP cannot be explained by simple source mixing alone, but instead reflects enhanced Fe dissolution of both mineral dust and anthro-Fe during chemical processing in atmospheric transport. To evaluate regional differences, two-sided Mann–Whitney U tests with Holm correction were applied because the data were not normally distributed. The analysis showed that Japanese TSP collected during the Asian outflow season exhibited significantly higher Fe solubility than Chinese and Korean TSP. Bulk Fesol% was higher in Japanese TSP than in Chinese and Korean TSP, with mean ± SD values

	of 4.9 ± 3.6% and 2.5 ± 2.5%, respectively (Holm-adjusted p = 0.0017). Significant differences were also observed for mineral-Fe_{sol}% (6.2 ± 3.5% vs. 3.2 ± 4.1%; Holm-adjusted p = 0.0012) and anthro-Fe_{sol}% (2.5 ± 4.2% vs. 0.9 ± 1.2%; Holm-adjusted p = 0.0057). Together, these results support the progressive solubilization of both mineral-Fe and anthro-Fe during transport from the East Asian continent.
P12, L241: Why was this particular boundary chosen to divide anthro-Fe into two groups? Is there a clearer way to articulate the choice? "...lower 5th percentile limit of the power-law relationship..." is not straightforward.	Given that these three comments concern related issues, we respond to them collectively below. In this study, anthro-Fe with lower solubility than mineral-Fe was operationally classified as non-combusted anthro-Fe after accounting for the atmospheric alteration of both Fe components. Anthro-Fe with solubility comparable to or higher than that of mineral-Fe was considered to be primarily derived from high-temperature combustion. The classification criteria and the method used to determine the boundary have been clarified in the revised manuscript, as described below. Anthro-Fe in East Asian TSP exhibited a wider range of anthro-Fe_{sol}% than mineral-Fe (Figure 4b). Although combustion-derived anthro-Fe is generally considered to be more soluble than mineral-Fe, some samples showed anthro-Fe_{sol}% lower than mineral-Fe_{sol}% and below 0.1%, the approximate lower limit reported for combustion-derived Fe. Such low-solubility anthro-Fe was likely derived from sources other than high-temperature combustion. To account for changes in the solubility of mineral-Fe and anthro-Fe during atmospheric transport and to avoid imposing a fixed threshold such as 0.1%, a boundary line parallel to and below the log–log regression line between mineral-Fe concentration and mineral-Fe_{sol}% was defined such that 5% of the mineral-Fe samples fell below it. For each mineral-Fe sample, the relative deviation from the regression relationship was quantified as the logarithm of the observed-to-predicted mineral-Fe_{sol}% ratio [log(observed mineral-Fe_{sol}%/predicted mineral-Fe_{sol}%)]. The 5th percentile of these relative deviations was then added to the intercept of the regression equation to define the boundary line. Based on this boundary, anthro-Fe samples below the line were operationally classified as non-combusted anthro-Fe, whereas those above it were classified as high-temperature combustion-derived anthro-Fe, with solubility

	comparable to or higher than that of mineral-Fe (Figure 4b). In the non-combusted anthro-Fe group below the boundary, all anthro-Fe_{sol}% values were below 1.0%, with an average of 0.3 ± 0.2%. Several samples with high anthro-Fe concentrations showed particularly low values of less than 0.1%. Such low solubility is consistent with non-combustion anthro-Fe, including Fe associated with brake-pad debris and tire-wear particles (Halle et al., 2013; Shupert et al., 2021; Cui et al., 2025). By contrast, the high-temperature combustion-derived anthro-Fe group above the boundary had an average anthro-Fe_{sol}% of 4.0 ± 5.7%, higher than the average mineral-Fe_{sol}% of 2.4 ± 4.1% in East Asian TSP. This characteristic is consistent with the generally higher solubility of anthro-Fe derived from high-temperature combustion. However, anthro-Fe_{sol}% was generally only approximately 1% even in high-concentration samples dominated by relatively fresh aerosol particles. This observation implies that these particles were emitted with relatively low initial solubility rather than in a highly soluble form and that they subsequently underwent solubilization through chemical processing during atmospheric transport (Sholkovitz et al., 2009; Ito et al., 2021).
P13, L261: The figure numbers and their corresponding references in the text break down from this point forward.	We apologize for the inconvenience. We have rechecked the figure numbers and corrected them.
P13, L264: Because the observations were not made at a single location, how was spatial and temporal variability accounted for? For example, how could a hypothetical sample from 45 ◦N collected in August be compared to a sample from 10 ◦N in December? Did any of the observations track a dust plume to observe changes in solubility?	We thank the reviewer for raising this important point. In this study, we did not perform backward-trajectory analysis or satellite-based AOD analysis to track specific dust plumes or emission sources for individual samples. We have revised the manuscript to clarify that our analysis does not track individual plumes, but evaluates the relationships between Fe loading and Fe solubility across the compiled aerosol dataset. Therefore, these results should be interpreted as dataset-level relationships, rather than as evidence for the temporal evolution of Fe solubility within individual dust plumes. Sholkovitz et al. (2012) compiled aerosol Fe loading and Fe solubility data from different sampling locations, collection periods, and analytical methods, and showed that a robust inverse relationship can emerge at regional to global scales despite these differences. In this

	section, we extend this framework to coarse aerosol particles and fine aerosol particles, and separately examine the relationships between mineral-Fe loading and mineral-Fesol%, as well as between anthro-Fe loading and anthro-Fesol%, at the ocean-basin scale. Within this framework, inverse relationships were observed between mineral-Fe loading and mineral-Fesol% in coarse aerosol particles. Similar relationships were also found for anthro-Fe in coarse aerosol particles and for both Fe types in fine aerosol particles. These consistent relationships suggest that common processes during atmospheric transport operate at the ocean-basin scale and influence the solubility of both mineral and anthropogenic Fe.
P18, L367: What is NEV?	We apologize for not spelling out the abbreviation. NEV denotes non-exhaust vehicle particles, and we have now added the full term along with representative examples. By contrast, the group below the boundary frequently showed anthro-Fe_{sol}% values of <0.1%, with a mean of 0.9 ± 0.8%, indicating the influence of non-combustion anthro-Fe, including brake pad and tire wear debris in non-exhaust vehicle particles, as also inferred for coarse aerosol particles (Kajino et al., 2020; Sakata et al., 2025).
P19, L400: Are there other factors to consider beyond surface area? Are there differences in source mineralogy which would explain higher solubility in coarse over fine particles?	Thank you for this helpful comment. We apologize that the original wording was unclear. Our intention was not to argue that Fe and Al solubility are simply higher in fine aerosol particles than in coarse aerosol particles, nor to attribute the observed size dependence solely to surface area. Rather, we intended to emphasize that, although fine aerosol particles generally have larger specific surface areas and may therefore be expected to undergo chemical alteration more readily, the increases in Fesol% and Alsol% from East Asia to the North Pacific were observed mainly in coarse aerosol particles. As suggested by the reviewer, source-related differences in mineralogy may also influence Fe solubility. We have therefore revised the text to consider possible source effects, including additional anthropogenic inputs such as ship-derived heavy-oil fly ash. However, because such inputs are expected to affect fine aerosol particles more strongly, they cannot fully explain the preferential increase observed in coarse aerosol particles. We also clarified that the increases in mineral-Fesol% and Alsol% suggest further chemical alteration of mineral dust in coarse

	aerosol particles during marine transport. The mean Fe_{sol}%, mineral-Fe_{sol}%, and anthro-Fe_{sol}% of North Pacific TSP were higher than those of East Asian aerosols during December–May, when Asian outflow is strongest (Table 2). In coarse aerosol particles, both mineral-Fe_{sol}% and anthro-Fe_{sol}% increased during marine atmospheric transport, and a similar pattern was observed for Al_{sol}%, an indicator of mineral dust alteration. In contrast, the corresponding values in fine aerosol particles showed little change (Table 2). Ship-derived heavy-oil fly ash may affect anthro-Fesol%, but its influence is expected to be greater in fine aerosol particles. Thus, the increase in anthro-Fe_{sol}% in coarse aerosol particles is unlikely to be explained by additional ship-derived inputs alone and instead suggests chemical alteration of anthro-Fe during marine transport. The increase in mineral-Fe_{sol}% and Al_{sol}% further indicates that mineral dust in coarse aerosol particles also underwent chemical alteration during transport through the marine atmosphere. Fine aerosol particles generally have larger specific surface areas than coarse aerosol particles and may therefore be expected to undergo chemical alteration more readily. Thus, this size-dependent behavior cannot be explained solely by surface area and suggests that additional dissolution processes in coarse aerosol particles must also be considered.
P20, L413: How high in the atmosphere does marine organic matter typically reach? A cite showing that organic matter is present at heights typical of cloud processing would help here.	Thank you for your comment. We have added this point to the manuscript, noting that previous studies have suggested the presence of organic ligands that stabilize Fe(II) and/or Fe(III) in rainwater collected over the ocean and in coastal regions, and that microorganisms in cloud water may produce siderophores (Kieber et al., 2003; Willey et al., 2008; Cheize et al., 2012; Vinatier et al., 2016). Supporting this idea, Wu et al. (2023) showed that the Fe_{sol}% of fine aerosol particles collected on Matsu Island remained high for 10 days in seawater amended with deferoxamine, whereas it decreased to below 1% without deferoxamine. This finding suggests that strong organic ligands can enhance the persistence of soluble Fe after aerosol deposition into seawater. Consistent with this interpretation, previous studies have reported or suggested organic ligands capable of stabilizing Fe(II) and/or Fe(III)

	in marine and coastal rainwater. Microorganisms in cloud water have also been suggested to produce siderophores, a class of strong Fe-binding ligands (Kieber et al., 2003; Willey et al., 2008; Cheize et al., 2012; Vinatier et al., 2016). Fe(III)-HULIS complexes have also been detected in fine aerosol particles over the North Pacific (Sakata et al., 2022; Kurisu et al., 2024). Because these Fe-organic complexes can remain soluble over a wide pH range, complexation with marine organic matter and cloud-water-derived ligands may help preserve d-Fe during cloud processing and facilitate the subsequent transfer of soluble Fe to the ocean.
P20, Table 2: Add horizontal gridlines for clarity. In the caption, what are the italicized values indicative of?	As suggested, we revised the layout of Table 2 and added horizontal gridlines for clarity. The caption has also been revised to clarify that the italicized values are means based on small sample sizes ($N \leq 10$) and should be interpreted with caution due to their greater uncertainty. **Table 2.** Annual and December–May means of Fe_{sol}, mineral-Fe_{sol}, anthro-Fe_{sol}, and $f_{anthro\text{-}dFe}$ in total suspended particles (TSP), coarse aerosol particles, and fine aerosol particles collected in East Asia and the North Pacific. Values are given as mean ± standard deviation, with the number of samples (N) shown in parentheses. Italicized values denote means calculated from 10 or fewer samples ($N \leq 10$) and should be interpreted with caution because of their greater uncertainty.

Variable	Period	TSP		Coarse aerosol particles		Fine aerosol particles	
		East Asia	North Pacific	East Asia	North Pacific	East Asia	North Pacific
Fe_{sol} (%)	Annual	3.5 ± 3.2 (N = 139)	13.9 ± 14.4 (N = 155)	1.0 ± 1.0 (N = 110)	9.2 ± 12.8 (N = 31)	21.0 ± 23.6 (N = 181)	24.3 ± 24.9 (N = 47)
	Dec.–May	2.8 ± 2.7 (N = 91)	10.5 ± 6.5 (N = 73)	0.8 ± 0.7 (N = 57)	*6.0 ± 4.6 (N = 10)*	16.3 ± 19.3 (N = 86)	13.1 ± 16.4 (N = 20)
mineral-Fe_{sol} (%)	Annual	4.7 ± 4.5 (N = 137)	16.3 ± 16.6 (N = 155)	2.3 ± 2.9 (N = 110)	8.9 ± 11.0 (N = 31)	27.9 ± 28.0 (N = 181)	28.5 ± 28.7 (N = 47)
	Dec.–May	3.8 ± 4.2 (N = 91)	13.4 ± 10.7 (N = 73)	2.8 ± 2.7 (N = 57)	*4.8 ± 10.0 (N = 10)*	22.5 ± 23.4 (N = 86)	15.2 ± 18.1 (N = 20)
anthro-Fe_{sol} (%)	Annual	1.7 ± 4.1 (N = 91)	12.1 ± 21.8 (N = 69)	0.1 ± 0.2 (N = 45)	*11.3 ± 30.2 (N = 10)*	16.0 ± 22.4 (N = 56)	26.2 ± 32.0 (N = 23)
	Dec.–May	1.0 ± 2.0 (N = 81)	5.4 ± 8.9 (N = 44)	0.1 ± 0.1 (N = 21)	*0.9 ± 3.0 (N = 3)*	12.3 ± 20.1 (N = 181)	*17.4 ± 27.5 (N = 8)*
$f_{anthro\text{-}dFe}$ (%)	Annual	2.9 ± 2.4 (N = 139)	10.5 ± 9.3 (N = 155)	2.2 ± 2.5 (N = 110)	11.8 ± 16.2 (N = 31)	16.3 ± 15.0 (N = 181)	20.2 ± 17.0 (N = 47)
	Dec.–May	2.5 ± 2.6 (N = 91)	8.5 ± 5.1 (N = 73)	1.5 ± 1.8 (N = 57)	*11.4 ± 15.3 (N = 10)*	13.2 ± 11.7 (N = 86)	13.5 ± 13.1 (N = 20)

P21, L424: In the water column, ligands keep iron in the dissolved fraction. Is that what	Thank you for this useful comment. In the case of coarse aerosol particles, organic ligands

happens in the atmosphere, or do the ligands contribute to the dissolution process?	may have contributed not only to the stabilization of dissolved Fe but also to Fe dissolution from aerosol particles because a larger fraction of low-solubility mineral Fe likely remained. This is consistent with the experimental results of Wu et al. (2023), which showed that Fe dissolution from low-solubility aerosols was promoted in the presence of strong Fe-binding ligands. In addition, Tang et al. (2025) also suggested that, under strong leaching by acetic acid (Berger method), the increase in Fe dissolution was larger than that in Al dissolution, and therefore the [d-Fe]/[d-Al] ratio could become higher than under ligand-free conditions (ultrapure water extraction). Indeed, coarse aerosol particles with high Fe_{sol}% exhibited high [d-Fe]/[d-Al] ratio, resulting in area (ii) of Figure SX. Although further studies are needed, coarse aerosol samples in marine aerosols showing both high Fesol% and high [d-Fe]/[d-Al] ratios are considered to reflect enhanced Fe dissolution by strong ligands. Experimental results from Wu et al. (2023) support this interpretation, showing that the Fe_{sol}% of coarse aerosol particles from Matsu Island increased from a few percent to about 10% when exposed to a simulated marine organic ligand under seawater pH conditions. These results indicate that Fe in coarse aerosol particles retains the capacity for further dissolution by organic ligands. The observed pattern is also consistent with the inter-laboratory comparison of Tang et al. (2025), which showed that, under strong-ligand conditions using the Berger method, the increase in Fe_{sol}% relative to ultrapure water extraction was larger than that in Al_{sol}%. Such preferential enhancement of Fe relative to Al dissolution can increase [d-Fe]/[d-Al] ratios under strong-ligand conditions. Although the number of such samples was limited, the coexistence of high Fe_{sol}% and high [d-Fe]/[d-Al] ratios suggests that organic ligands in the marine atmosphere can substantially enhance Fe dissolution in coarse aerosol particles during transport. Nevertheless, more direct observational evidence is still needed to constrain the importance of this process over the North Pacific.
P23, L453: Higher aerosol loading in the wintertime (presumably Northern Hemisphere, this should be specified) is surprising. Is this a bias because there a few high latitude, low loading observations during NH winter?	Thank you for this suggestion. We agree with your com ment and have revised the text to specify the relevant months, such as July–August, instead of using seasonal terms. For consistency, we have also applied the same revision to regions other than the Atlantic Ocean.

P26, L493: Provide a reference for the seasonal variability in Fe% being caused by the temperature dependence of aerosol pH.	We have added three reference papers showing that Fesol% can be controlled by the temperature dependence of aerosol pH (Tao and Murphy, 2019b; Zhang et al., 2023; Yang and Weber, 2023).
P28, L557: is Feanthro-dFe overestimated in the models? Please clarify.	The predicted mineral-Fe_{sol}% of fine mineral particles in Earth system models and related models, typically less than 10%, is lower than the mean mineral-Fe_{sol}% observed for fine aerosol particles in our dataset of the marine aerosols. This difference may lead to an underestimation of mineral-derived d-Fe concentrations and consequently bias the relative contribution of anthro-Fe ($f_{anthro\text{-}dFe}$) to total d-Fe high. We have revised the manuscript to make this interpretation clearer. The discrepancy in $f_{anthro\text{-}dFe}$ between models and observations may be partly explained by the lower mineral-Fe_{sol}% represented in the models compared with values derived from field observations. Representative model calculations generally predict mineral-Fe_{sol}% of less than 10% for fine aerosol particles over the North Pacific. In contrast, mineral-Fe_{sol}% in fine aerosol particles frequently exceeded 10% in this study, with mean values of 28.5 ± 28.7% in the North Pacific and 11.3 ± 16.7% in the Atlantic. Thus, underestimating the mineral-Fe_{sol}% in fine particles may lead to an underestimation of the mineral contribution to d-Fe and consequently bias the relative contribution of anthro-Fe to total d-Fe high. Incorporating a size-dependent scheme capable of representing higher mineral-Fe_{sol}% in fine particles may therefore improve the representation of atmospheric Fe supply to the surface ocean and its source apportionment in Earth system models.

Community comments

Reviewer's comments	Author's reply
We sincerely apologize for the time it took to revise the manuscript. We would also like to express our sincere gratitude for your careful and constructive review. Our point-by-point responses to your comments are provided in the attached PDF file. We would greatly appreciate it if you could kindly refer to the file for details.	Thank you for your comment. In the revised manuscript, we have added the following text and cited the paper you recommended. Although several recent studies have examined the seasonal variability of Fesol% (Takahashi et al., 2013; Sakata et al., 2023, 2025; Zhang et al., 2023; Chen et al., 2026), such observations remain scarce globally, contributing to uncertainty in identifying the factors controlling Fe solubility.