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The authors have assembled an impressive dataset of aerosol concentration data from previous studies as well as unpublished data of their own.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 also apologize for the extended time required to complete the revision and sincerely appreciate the reviewer's patience and understanding. 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	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

appears to be a fundamental outcome of the study; however, it does not receive any mention 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.	point has been clarified by adding the following text to the first paragraph of the Implications section. The following text is included in Lines 621–638 of the revised manuscript. 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 Fe_{sol}% 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-Fe_{sol}% (<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 a 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. These revisions span multiple sections of the manuscript and therefore cannot all be listed here. They are described primarily in Sections 3.3.1, 3.4.1, and 3.4.3, among others. 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 as 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. I do not think “It is known...” is sufficient here.	We have added several papers that mentioned HF promote digestion of Si-O bond. The following text is included in Lines 89–92 of the revised manuscript. A critical consideration was whether hydrofluoric acid (HF) was incorporated in the digestion process. It is known that the incomplete digestion of Si-O bonds 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 the minerals (Chao and 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. As reported by previous studies (Clough et al., 2019; Perron et al., 2020b, Tang et al., 2025), Fe_{sol}% and Al_{sol}% extracted with acetate buffer may be higher than those obtained with MQ water. However, the increase in Fe_{sol}% caused by acetate buffer extraction varies widely among studies, ranging from 1.4- to 7.0-fold (Perron et al., 2020b; Tang et al., 2025), making it difficult to apply a consistent correction factor. Therefore, no correction was applied to the solubilities obtained by acetate buffer extraction in this study. By contrast, 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. The revised text below is provided in Lines 156–164 of the revised manuscript. Previous studies have reported that Fe_{sol}% and Al_{sol}% obtained by acetate buffer extraction are higher than those obtained by ultrapure water extraction (Clough et al.,

	2019; Perron et al., 2020b; Tang et al., 2025). However, the increase in Fe_{sol}% caused by acetate buffer extraction varies widely among studies, ranging from 1.4- to 7.0-fold (Perron et al., 2020b; Tang et al., 2025), making it difficult to apply a consistent correction factor. Therefore, no correction was applied to the solubilities obtained by acetate buffer extraction in this study. In addition, because the ratio of the increases in Fesol% and Alsol% does not exceed a factor of two (Tang et al., 2025), the effect of extraction method on the [d-Fe]/[d-Al] ratio is expected to be relatively small, and no correction was applied to the [d-Fe]/[d-Al] ratios obtained by acetate buffer extraction. Therefore, the influence of differences in extraction methods on the source apportionment of T-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. Yes, the value of 5.18 refers to the T-Fe/T-Al ratio in aerosol particles, not to the enrichment factor ($EF_{T\text{-}Fe}$). We have revised the text to clearly distinguish the measured T-Fe/T-Al ratio from $EF_{T\text{-}Fe}$. The revised text is provided in Lines 109–114 of the revised manuscript. In this study, a mean T-Fe/T-Al ratio of 0.52 ± 0.12, derived from five reported values for the upper continental crust, was used as the reference value to account for variability in crustal T-Fe/T-Al ratios (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 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 revised text is provided in Lines 140–142 of the revised manuscript. 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 proton-promoted and ligand-promoted dissolutions.

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 sources, T-Fe and d-Fe concentrations, Fe solubility (Fe_{sol}%), and anthropogenic contribution to d-Fe ($f_{anthro\text{-}dFe}$) in TSPs. Values are presented as mean ± standard deviation, with ranges in parentheses. Figure numbers below each region refer to figures showing the spatial distributions of these values.

Area	T-Fe source(s)	d-Fe source(s)	Mean T-Fe conc. (ng m^{-3})	Mean d-Fe conc. (ng m^{-3})	Mean Fe_{sol}% (%)	Mean $f_{anthro\text{-}dFe}$ (%)
East Asia	**TSP and coarse aerosol**: Mineral dust **Fine aerosol**: Mineral dust + anthro-Fe	TSP and coarse: Fe dissolution from mineral dust Fine aerosols: Fe dissolution from mineral dust and anthro-Fe emitted as insoluble 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
N. Pacific (Figure 6)	Asian outflow of mineral dust	Fe dissolution from mineral dust: Fe_{sol}% in coarse aerosols were enhanced by chemical alteration in the marine atmosphere 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 (Figure 9)	Saharan dust	Fe dissolution from mineral dust: cloud processing in high altitude Anthro-Fe mostly insoluble	387.2 ± 922.9 (0.2–5650.0)	6.9 ± 23.4 (<0.1–212.7)	6.2 ± 6.8 (0.1–50.8)	5.1 ± 9.7
S. Pacific (Figure 12)	Mineral dust and volcanic influence near Heard Island	Coastal Australia aerosols affected by anthro emissions acidified aerosols. Volcanic influence near Heard Island	17.6 ± 25.3 (0.1–129.8)	1.0 ± 23.4 (<0.1–8.3)	7.3 ± 6.4 (<0.1–41.9)	Up to 20% near Australia. Apparent high values near Heard Island are driven by volcanic ash (not anthro-Fe).

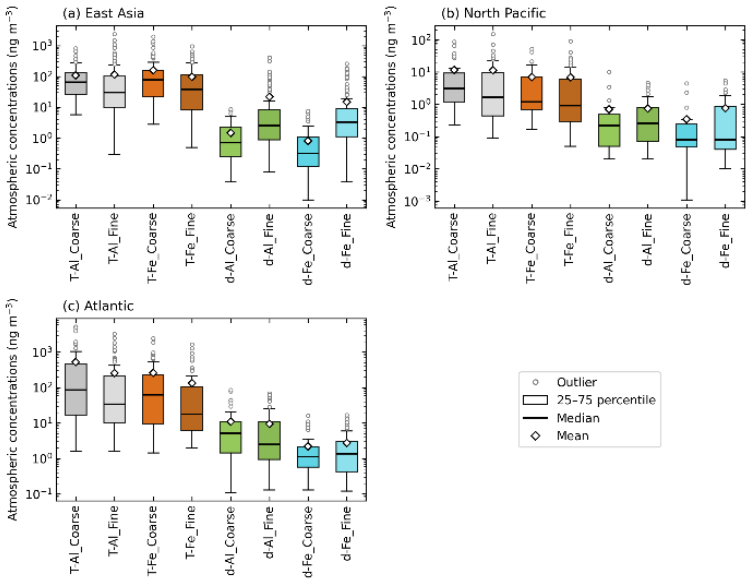
References. East Asia: Duvall et al., 2008; Kurisu et al., 2019; Hsieh et al., 2023; Sakata et al., 2023, 2025; Seo and Kim, 2023. The North Pacific: Buck et al., 2006, 2013; Marsay et al., 2022; Sakata et al., 2022; Kurisu et al., 2024. The Atlantic Ocean: Baker et al., 2006a, 2006b, 2013, 2020; Buck 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 S2, 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 TSP collected in Japan 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. The revised text is provided in Lines 263–272 of the revised manuscript. 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 during the 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 TSP collected in Japan during the Asian outflow season exhibited significantly higher Fe solubility than those collected in China and

	Korea: bulk Fe_{sol}% for the TSP samples collected in Japan (mean ± SD = 4.9 ± 3.6%) was higher than those in China and Korea (= 2.5 ± 2.5%; Holm-adjusted p = 0.0017). Significant differences were also observed for mineral-Fe_{sol}% (Japan: 6.2 ± 3.5%, China and Korea: 3.2 ± 4.1%; Holm-adjusted p = 0.0012) and anthro-Fe_{sol}% (Japan: 2.5 ± 4.2%, China and Korea: 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.	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 define the boundary are described in Lines 273–294 of the revised manuscript. Anthro-Fe_{sol}% 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 boundary line were operationally classified as non-combusted anthro-Fe, whereas

	those above the line 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-combusted 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-Fe_{sol}%, as well as between anthro-Fe loading and anthro-Fe_{sol}%, at the ocean-basin scale. Within this framework, inverse relationships were observed between mineral-Fe loading and mineral-Fe_{sol}% 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 (Figure 4 for North Pacific, Figure 8 for Atlantic, and Figure 11 for South Pacific). 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. The revised text is provided in Lines 406–412 of the revised manuscript. Most samples below the boundary were collected from February to May, when East Asian outflow is strong, suggesting a substantial influence of continental anthropogenic emissions. Their mean anthro-Fe_{sol}% was 1.0 ± 0.8%, and although anthro-Fe_{sol}% increased slightly with decreasing concentration, the maximum value was only 2.7%. These particles occurred in both coarse and fine aerosol particles, and their persistently low solubility resembled that of non-exhaust vehicle particles such as brake-pad debris (Figures 8b and 8c). Accordingly, non-combusted anthro-Fe, including non-exhaust vehicle particles, likely remains largely insoluble even after long-range transport over the marine atmosphere and probably contributes little to d-Fe supply in the North Pacific.
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 agree that factors other than specific surface area, including size-dependent differences in source mineralogy, may influence Fe solubility. We apologize that the original wording did not make this point clear. 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.

We have therefore revised the manuscript to consider possible source-related effects. Although size-dependent differences in the mineralogical composition of source dust may affect Fe solubility, differences in Fesol% appear to be minimal for freshly emitted mineral dust (Shi et al., 2011a). Additional anthropogenic inputs, such as ship-derived heavy-oil combustion, may also provide highly soluble Fe to the marine atmosphere (Sedwick et al., 2007). However, such emissions are expected to affect mainly fine aerosol particles and therefore cannot fully explain the preferential increase in solubility observed in coarse aerosol particles. We have clarified that the concurrent increases in mineral-Fesol% and Alsol% in coarse aerosol particles are consistent with further chemical alteration of mineral dust during marine transport. The revised text is provided in Lines 435–451 of the revised manuscript.

> The mean Fe_{sol}%, mineral-Fe_{sol}%, and anthro-Fe_{sol}% in North Pacific TSP were higher than those in East Asian aerosols collected during winter and spring, when Asian outflow is strongest (Table 2). This suggests that aerosols transported from East Asia undergo further chemical alteration during transport over the North Pacific. Size-resolved comparisons showed that both mineral-Fe_{sol}% and anthro-Fe_{sol}% increase in coarse aerosol particles during transport from Japan to the North Pacific, whereas no pronounced changes were observed in fine aerosol particles. A similar pattern was observed for Al_{sol}%, an indicator of mineral-dust alteration. Al_{sol}% in TSP and coarse aerosol particles was higher over the North Pacific than in Japan, whereas no pronounced difference was observed for fine aerosol particles between Japan and the North Pacific (Table 2). Although size-dependent differences in the mineralogical composition of source dust may potentially affect Fe solubility, differences in Fe_{sol}% appear to be minimal for freshly emitted mineral dust (Shi et al., 2011a). Additional input of anthro-Fe from ship-derived heavy-oil combustion may provide highly soluble Fe to the marine atmosphere (Sedwick et al., 2007). However, such emissions are expected to affect mainly fine aerosol particles and therefore cannot fully explain the preferential increase in solubility observed in coarse aerosol particles. Thus, the concurrent increases in mineral-Fe_{sol}% and Al_{sol}% in coarse aerosol particles suggest

	that mineral dust undergoes further chemical alteration during transport through the marine atmosphere. In contrast, the absence of substantial changes in solubility in fine aerosol particles between East Asia and the North Pacific suggests that their alteration pathway differs from that of coarse aerosol particles. Given that fine aerosol particles generally have larger specific surface areas and are therefore expected to be more reactive than coarse aerosol particles, the pronounced increases in Fe_{sol}% and Al_{sol}% specifically in coarse aerosol particles are particularly notable and require further explanation.
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 this important comment. We have added further discussion and references to clarify that Fe-binding organic ligands can be present in atmospheric aqueous phases relevant to cloud processing. Previous studies have suggested the presence of organic ligands capable of stabilizing Fe(II) and/or Fe(III) in rainwater collected over the ocean and in coastal regions. In addition, microorganisms in cloud water have been suggested to produce siderophores, which are strong Fe-binding ligands (Kieber et al., 2003; Willey et al., 2008; Cheize et al., 2012; Vinatier et al., 2016). In particular, Vinatier et al. (2016) investigated cloud water collected at the high-altitude Puy de Dôme station, providing evidence that such Fe-binding ligands can occur under conditions directly relevant to cloud processing. We have revised the manuscript accordingly. The revised text is provided in Lines 464–473 of the revised manuscript. 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 prevent the decrease in Fe solubility and maintain dissolved Fe over extended periods. 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, organic ligands present in cloud water may help preserve d-Fe during cloud processing and suppress the decrease in Fe solubility that would otherwise occur in the absence of strong organic ligands.
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.
P21, L424: In the water column, ligands keep iron in the dissolved fraction. Is that what happens in the atmosphere, or do the ligands contribute to the dissolution process?	Thank you for this useful comment. In the case of coarse aerosol particles, organic ligands 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

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)

	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, the North Pacific aerosol with high Fe_{sol}% exhibited high [d-Fe]/[d-Al] ratio, resulting in area (ii) of Figure S4a. Although further studies are needed, coarse aerosol samples in marine aerosols showing both high Fe_{sol}% and high [d-Fe]/[d-Al] ratios are considered to reflect enhanced Fe dissolution by strong ligands. The revised text is provided in Lines 483–492 of the revised manuscript. 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% in the presence of a strong Fe-binding ligand under seawater pH conditions. These results indicate that coarse aerosol particles retain a pool of Fe that can undergo further ligand-promoted dissolution. This interpretation is also consistent with the inter-laboratory comparison of Tang et al. (2025), which showed that, under the stronger leaching conditions of 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. Although the number of such samples was limited, the coexistence of high Fe_{sol}% and high [d-Fe]/[d-Al] ratios suggest that organic ligands in the marine atmosphere can enhance Fe dissolution from coarse aerosol particles during transport. Nevertheless, more direct observational evidence is 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 important comment. We agree that spatial variability in the sampling locations needs to be considered when interpreting the monthly variations in Atlantic aerosol concentrations. We therefore revised the manuscript to specify the relevant months, such as December–February and June–August, rather than using seasonal terms. We also clarified that the relatively high T-Fe and T-Al concentrations observed in December–February likely mainly reflect the influence of samples collected offshore of the Sahara Desert. Importantly,

	similar monthly variations in T-Fe concentrations have been reported from ground-based observations at Cape Verde, located in the same region, supporting the interpretation that the high T-Fe concentrations observed offshore of the Sahara Desert in December–February are reasonable. In contrast, satellite-derived DAOD over the same latitudinal region shows a maximum in June–August. We interpret this difference in terms of the seasonal variation in Saharan dust transport altitude, with dust transported mainly in the lower troposphere during December–May but at higher altitudes within the Saharan Air Layer during June–August. The manuscript has been revised accordingly. The revised text is provided in Lines 523–530 of the revised manuscript. Unfortunately, Atlantic TSP samples collected from February to April are not available in this dataset. However, shipboard observations showed that T-Fe and T-Al concentrations were clearly higher in December–February than in June–August (Figure 10a). Considering the spatial variability in the sampling locations, the high concentrations observed in December–February likely mainly reflect the influence of the region offshore of the Sahara Desert. Indeed, similar seasonal variations in T-Fe concentrations have also been reported from ground-based observations at Cape Verde, located offshore of the Sahara Desert (Carpenter et al., 2010; Fomba et al., 2013; Patey et al., 2015). In contrast, satellite observations of DAOD over the same latitudinal region indicate a June–August peak, which is also consistent with the seasonal variability of dust deposition fluxes recorded by sediment traps in seawater (Yu et al., 2019; van der Does et al., 2021).
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 Fe_{sol}% 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 revised text is provided in Lines 646–654 of the revised manuscript. 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, underestimation of mineral-Fesol% in fine particles may lead to an underestimation of mineral-derived d-Fe and, consequently, an overestimation of the relative contribution of anthro-Fe to total d-Fe. 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.