Reviewer 1

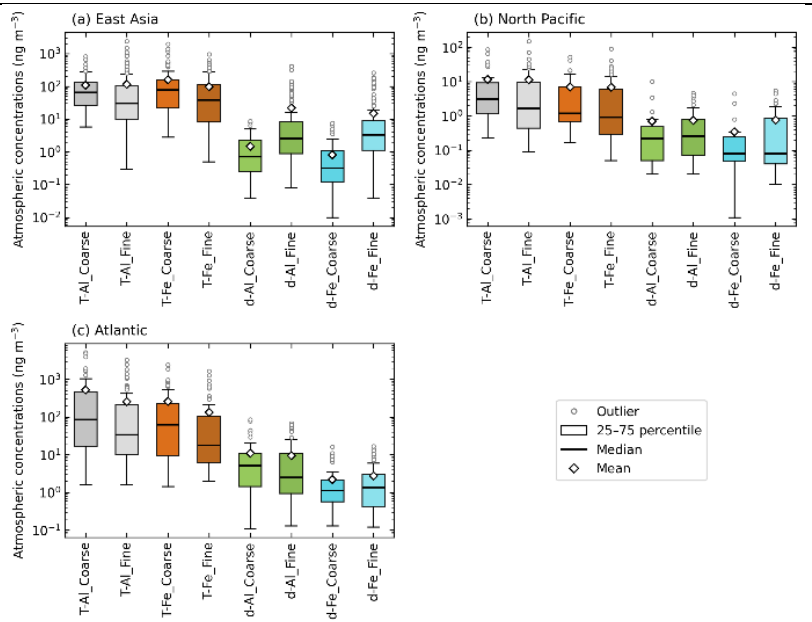
	Reviewer's comments	Author's reply
	This is a synthesis study on Aerosol Iron fractional solubility, compiling all available soluble Al and Fe data over the Atlantic and Pacific oceans. This study has high scientific significance, improving our current understanding of aerosol iron solubility. This paper 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 EFT-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. We regret the extended time required for the revision and are grateful for the reviewer's patience throughout this process. 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	The authors considered size-segregated samples from their unpublished data for this study, described in line 75.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 in the literatures. In particular, no other size-segregated samples are available for which all of the T-Fe, T-Al, d-Fe, and d-Al concentrations are available in the Southern Hemisphere. The following texts were added in S1 of Supplemental Information. 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 classified as coarse aerosol particles, whereas those 2.1 μm and smaller were classified as fine aerosol particles.
2.	The author considers that "the ultrapure water and ammonium acetate buffer were	We address the two questions together below. Although these comments concern Section 2.1,

3.	uniformly classified as d-Fe, with the same approach applied for Al." (line 100). A 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	we added the relevant explanation to Section 2.3, where this issue is discussed more directly. 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.
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 statements are included in Lines 124–128 of the revised manuscript. These areas differ based on the dissolution mechanisms of mineral dust. Area (i): [d-Fe]/[d-Al] ranging 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): [d-Fe]/[d-Al] over 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 S2, 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.	“Similar Fe” refers to the comparison of Fesol% between mineral-Fe and anthro-Fe at comparable concentrations. Based on this comparison, the samples were divided into two groups: one in which anthro-Fesol% was lower than mineral-Fesol%, and the other in which anthro-Fesol% was similar to or higher than mineral-Fesol%. The detailed classification method is addressed in our responses to Comments 10 and 11.
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 two 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-combustion-derived anthro-Fe after accounting for atmospheric alteration of both Fe components. In contrast, 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 variation than mineral-Fe_{sol}% (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, the use of a fixed threshold such as 0.1% was avoided. Instead, a boundary line parallel to and below the log–log regression line between mineral-Fe concentration and mineral-Fe_{sol}% was defined so 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).

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, a larger number of samples plotted in area (v) compared with the other oceanic regions likely reflects the influence of Saharan dust. The following text is included in Lines 512–518 of the revised manuscript. 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. The following text is included in Lines 145–146 of the revised manuscript. Additionally, some samples from the Sharan Desert showed values below 0.1, suggesting that these sources may also occasionally contribute (Shi et al., 2011a; 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 4, 8, and 11 in revised version.
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	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

	follow the season. Do this correction for all the possible places like line-497,501...	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_{sol}%".	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 (iii)"of what? It should be mentioned.	Thank you for pointing this out. We have clarified that areas (i) and (iii) refer to those in the $EF_{T\text{-}Fe}$–[d-Fe]/[d-Al] diagram of Figure 5c. Area (i) represents mineral-dust-derived T-Fe and d-Fe produced by its proton-promoted dissolution, whereas area (iii) represents poorly soluble anthro-Fe-dominated T-Fe and d-Fe mainly produced by the proton-promoted

		dissolution of mineral dust. Further details are provided in Section 2.3. The following text is included in Lines 301–306 of the revised manuscript. The mean Fe_{sol}% in coarse aerosol particles was 1.0 ± 1.1%, with higher values observed from June to August (Figure 4c). This value was lower than the mean mineral-Fe_{sol}% of 2.3 ± 2.9% but higher than the mean anthro-Fe_{sol}% of 0.1 ± 0.2% (Figure 4c). As observed for TSP, most coarse aerosol samples were distributed within regions (i) and (iii) of the [d-Fe]/[d-Al]–$EF_{T\text{-}Fe}$ diagram (Figure 5c). These results indicate that T-Fe in East Asian coarse aerosol particles was derived primarily from mineral dust and poorly soluble anthro-Fe, whereas d-Fe was supplied mainly through proton-promoted dissolution 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 $EF_{T\text{-}Fe}$ 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.