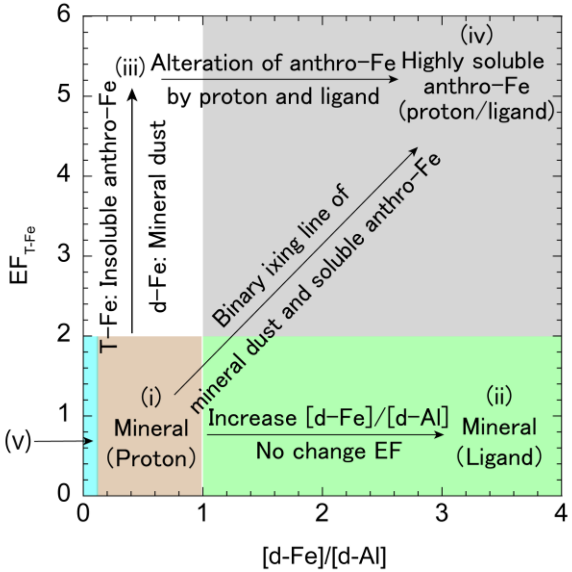
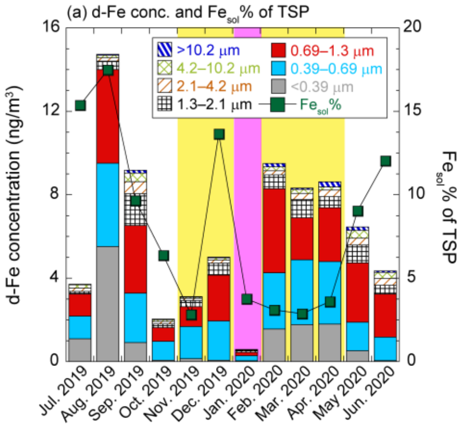
Reviewer 1

Reviewer's comments	Author's Reply
Overall comments: The manuscript presents a new concept discussing iron solubility of aerosol particles in the Pacific Ocean determined by atmospheric chemistry in East Asia. The work has been discussed in context to previous literature with appropriate references. However, the manuscript needs major revisions to improve clarity and structure for ease of interpretation. Key terminology has not been defined when introduced, with too many terms being used, causing confusion. Several figures have been incorrectly referenced and labeled throughout the publication. While the COVID-19 lockdown has been mentioned in the abstract and introduction, it lacks substantive discussion in the results and implications section, especially with respect to anthropogenic iron sources.	We sincerely thank the reviewer for the time and effort you have put into this review. We have carefully revised the manuscript with full consideration of the reviewer's comments and suggestions. Responses to the reviewer's comments are in black and not indented; corresponding revisions in the manuscript are in red and indented.
Line 104-108: Please add a clarification on when (pre or post sampling) and why filters were hydrophilized and treated with ethanol	During pre-sampling filter cleaning, the hydrophobic PTFE fiber filters did not sink in hydrochloric acid, making their proper cleaning difficult. Therefore, we hydrophilized the filters with ethanol so that they can be submerged in the acid. The ethanol evaporated during the air drying of the filters, returning them to a hydrophobic state at the time of sampling. PTFE filters are not properly wetted by cleaning solutions because they are hydrophobic. This situation has the potential to reduce cleaning efficiency. Therefore, the filters were hydrophilized with ethanol (99.5%, Wako First Class, Wako, Japan). The hydrophilized PTFE filters were soaked in 1 mol L−1 hydrochloric acid (EL grade, Kanto Chemical Co. Inc., Japan) and heated at 180 °C for one day. Subsequently, the filters were placed in ultrapure water and heated at 180 °C for one day. The rinsed filters were then air-dried in a clean booth. Air drying restored the hydrophobicity of the PTFE filters as a result of the complete removal of ethanol from the filters.
Line 136: Explain what $(\text{T-Fe/T-Al})_{\text{aerosol}}$ stands for in the equation? Clarify this term	We have added explanation of $(\text{T-Fe})/(\text{T-Al})_{\text{aerosol}}$. The enrichment factor of Fe (EFT-Fe) normalized by the mass ratio of Fe relative to that of Al in the upper continental crust (UCC) was calculated to evaluate the

	emission sources of Fe. The following equation was used for the calculation: $EF_{\mathrm{T-Fe}} = \frac{(\mathrm{T-Fe/T-Al})_{aerosol}}{(\mathrm{T-Fe/T-Al})_{UCC}},$ (Eq. 1) where (T-Fe/T-Al)aerosol represents the mass concentration of total Fe (= insoluble Fe + d-Fe in aerosol particles relative to the total Al).
Line 173: The y-axis should be correctly labeled as EF $_{\mathrm{T-Fe}}$ to be consistent with the text	Thank you for your suggestion. We have revised the y-axis label. The revised figure was attached below. 
Line 194: What are JPN+EAout, JPN, and EAout periods? Please specify the dates they comprise, as they have not been mentioned previously.	The sampling period in this study was separated into two categories: JPN, representing seasons dominated by air masses from within Japan, and EAout, representing seasons that are markedly influenced by air masses from East Asia, such as China. In the initial version of our manuscript, the detailed discussion on the origin of air masses was provided in Section 3.1. However, as pointed out in the comments, information on the origin of air masses was used in the PMF explanation. Therefore, Section 3.1 has been removed, and its contents have been moved to Section 2.1.
Line 261 Suggest using %Fe$_{\max}$ instead of [%FeT] for clarity. Avoid using too many terms if possible, as it is confusing.	As per your suggestion, %Femax has been rephrased with [%FeT].

Line 270: What is S/L ratio?	I apologize for not spelling out the S/L ratio. The S/L ratio refers to the solid-to-liquid ratio. The relevant sentence has been improved following. Under the assumption that the solid-to-liquid ratio of anthro-Fe is 0.06 g L^{-1}, which is comparable to that of mineral dust, the aerosol liquid water (ALW) content associated with hematite nanoparticles was quantified by using the following equation.
Line 327: There is no need to indicate bar graph and line graph with axis; this is self-explanatory in the legend.	Thank you for your suggestions. We revised the figure as following your comment. The revised figure is attached below. 
Line 350: Elaborate on what nss-SO42-/t-Fe represents before using the term. What is its importance?	I apologize for the insufficient explanation regarding the [nss-SO_4^{2-}]/[T-Fe] ratio. This ratio is used as an indicator of the acidity of iron-containing particles. An explanation of [nss-SO_4^{2-}]/[T-Fe] has been added to the relevant sentence. Indeed, the Fe_{sol}% of coarse aerosol particles was correlated with the [nss-SO_4^{2-}]/[T-Fe] ratio as an indicator of the acidity of Fe-bearing particles (Fig. S5a; Zhu et al., 2020, 2022; Liu et al., 2022).
Line 351: Smallest particle diameter (< 0.39 μm) does not consistently seem to have higher solubility except maybe in Feb 2020, as opposed to what is stated. Please clarify this discrepancy	The relevant sentence describes that in coarse aerosol particles (>1.3 μm), Fe_{sol}% increased with the decrease in particle shape (increase in surface area). The following sentence has been revised to clarify that Fe_{sol}% is being compared within the coarse particle fraction.

	The Fe$_{\text{sol}}$% of the finest particles did not increase likely because this size fraction contained a large amount of fresh Fe-bearing particles that had not experienced acidic conditions. This result is supported by Fe speciation analysis using macroscopic XANES spectroscopy, which showed that Fe(III)-sulfate was not present in all samples.
Line 392: Clarify what figure is being discussed here (Presumably 6b) Line 402: The figure caption for figure 6 is same as figure 5. Correctly describe Figure 6 and adjust the text accordingly. Line 405: The yellow regions labeled as the JPN period in fig 5 and 6 are incorrect, which complicates interpreting the results. Fix the labels and discussion accordingly. Line 630 and 640: Figures 10b and 10c have been incorrectly discussed as 11b and 11c throughout the text. There is no figure 11c. Please correctly state which figure is being referred to, and review the supporting text for consistency.	We sincerely apologize for the inconvenience caused by the errors in the figure numbers and captions. We have thoroughly reviewed the entire manuscript and corrected these issues.
Line 410: What is the chemical alteration being referred to? Is it only ocean acidification or other factors as well? Elaborate	We consider that aerosol acidification is the most dominant process to solubilize mineral dust in fine aerosol particles. Furthermore, an explanation for the limited impact of ligand-promoted Fe dissolution has been added at the end of the paragraph. Notably, ligand-promoted dissolution is considered a process that increases the Fe$_{\text{sol}}$% of mineral dust. However, the contribution of ligand-promoted Fe dissolution was likely small because there were almost no plots of aerosol particles in region (ii) of Fig. 5b, a region where this process is a major contributor to mineral dust.
Line 427: Please add more discussion on Fesol% from anthropogenic sources during the COVID-19 lockdown period. The datapoint in Figure 6c for the lockdown period is missing. Does anthro-Fe% drop significantly during COVID-19 lockdowns, and what does this imply about the primary sources of soluble anthro-Fe (e.g., industrial vs. vehicular emissions)?	The Fe$_{\text{sol}}$% of anthropogenic Fe could not be calculated because the EF$_{\text{T-Fe}}$ and [d-Fe]/[d-Al] ratio of fine aerosol particles were lower than the representative values for mineral dust, indicating that anthro-Fe had no contribution to fine aerosol particles (computationally, the abundance of anthro-Fe became negative). Therefore, we have added the reasons for the missing plots of anthro-Fe$_{\text{sol}}$% during the COVID-19 lockdown for caption of Fig. 6c. The plots of anthro-Fe$_{\text{sol}}$% in panel (c) are missing because either or both anthro-Fe or anthro-dFe concentrations were 0 due to the remarkable but small

	contributions of anthro-Fe during the COVID-19 lockdown period. Regarding to the primary source of anthro-Fe in fine aerosol particles, an important source of anthro-Fe, and its contribution factor during the COVID-19 period significantly decreased compared to other samples (Please see Figure S7). In the case of vehicular emissions, road dust encompassed in aged mineral dust factors (Figures 7b and 7e). Therefore, the factor contribution did not exhibit a reduction as pronounced as that of steelworks during the COVID-19 period.
Line 491: The term "Atmospheric Chemical alterations" is too vague. Specify the mechanisms driving Fe dissolution or discuss in more detail	Thank you for your comments. We have revised the relevant sentence in following. Given that the Fesol% values of brake ring and tire wear debris were less than 0.01% in the absence of chemical alterations, including proton- and ligand-promoted dissolutions (Shupert et al., 2013; Halle et al., 2021), the increase in the [d-Fe]/[d-Al] ratio of the aged dust factor may have been caused by the dissolution of Fe from these materials during chemical alterations in the atmosphere.
Methods Section: The methods are too lengthy and introduce many terms. Consider moving detailed protocols to the SI.	Some parts of the method section have been moved to Supplemental Information.
Implications Section: Please reword the implications section to place results in a broader context without introducing new terms like RDF that have not been mentioned previously.	In accordance with your feedback, the part concerning RDF has been deleted. The "Implication" section has been renamed to "Conclusions and future implications," and its content structure has undergone a substantial revision. Ther revised sentences were attached below. In this study, we investigated the factors controlling Fe_{sol}% in size-fractionated aerosol samples collected in the coastal region of the Sea of Japan. Our results showed that the T-Fe and d-Fe concentrations in TSP samples peaked in spring due to the substantial loading of Asian dust into the atmosphere. Steel industry and NEV particles, which were primarily composed of insoluble Fe, were important sources of T-Fe in fine aerosol particles. During the COVID-19 lockdown, the contribution of anthro-Fe (especially from the steel industry) to T-Fe decreased sharply, highlighting that anthro-Fe emitted from combustion and non-combustion sources was a major source of T-Fe in fine aerosol particles over East Asia. Fe_{sol}% was

higher in summer than spring, with high values mainly observed in fine aerosol particles, and correlated with the [nss-SO_4^{2-}]/[T-Fe] ratio, indicating that Fe in these fine particles was primarily dissolved through proton-promoted dissolution. Macroscopic and microscopic XANES spectroscopy revealed that the water-soluble Fe species in fine aerosol particles were Fe(II)-sulfate, Fe(III)-sulfate, and Fe(III)-oxalate and were also present in mineral dust and anthropogenic aerosols. Given the water insolubility of Fe species in freshly emitted mineral dust (aluminosilicates) and anthro-Fe (mainly Fe oxides), these water-soluble Fe species likely formed through aerosol acidification by H_2SO_4, a process supported by the strongly acidic conditions suggested by dissolution pH estimations. Therefore, chemical reactions, including aerosol acidification, play a critical role in the control of the Fe_{sol}% of aerosol particles in East Asia.

During the period of increased aerosol outflow from East Asia (November to April), the average Fe_{sol}% of TSPs collected at NOTOGRO (4.9%) was slightly lower than that of TSPs collected in the North Pacific. However, the Fe_{sol}% of fine aerosol particles increased substantially during transportation from East Asia to NOTOGRO, with their average Fe_{sol}% (14.3%) being comparable to that of fine aerosol particles collected in the western Pacific during a similar season (14.2%; Table S3). This finding suggests that the chemical alterations of Fe in mineral dust and anthro-Fe in fine aerosol particles mainly occurred over East Asia rather than during transport in the North Pacific. Therefore, long-term observations on the Fe_{sol}% of the fine aerosol particles collected at the rim of East Asia (i.e., entrance of the North Pacific) play an important role in understanding the controls on Fe_{sol}% supplied to the North Pacific. By contrast, the Fe_{sol}% of coarse aerosol particles were slightly higher in the western Pacific (average: 3.5%) than in NOTOGRO (average: 0.5%). This difference likely contributed to the difference in the Fe_{sol}% of TSPs between the two regions. Therefore, future research should also focus on the Fe dissolution processes in coarse aerosol particles during transport over the marine atmosphere to develop our understanding of aerosol Fe supply to the ocean

	surface because these differences may be a reason for the higher Fe_{sol}% of TSPs in the western Pacific than in East Asia.
Minor comments: Line 34: Use another word instead of ‘outside’ (e.g., except). Line 73, 89: Subscript ‘sol’ in Fe_{sol}% Line 596: Correct the spelling of ‘Dissolution’ and ‘Mineral’ in Figures 10a and 10b.	Thank you for your checking. These issues have been improved.