

Assessment of aerosol iron (Fe) solubility using global dataset, Part I: Mechanisms underlying the inverse relationship between Fe solubility and Fe concentration

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The authors have assembled an impressive dataset of aerosol concentration data from previous studies as well as unpublished data of their own.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.

- 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 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.
- 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.
- 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.
- P4, L82: typo in Buck and Paytan, 2012
- 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.
- P4, L93: Correct to “...we only compiled concentrations obtained...”
- 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)

- P5, L115-116: Is $T\text{-Fe}/T\text{-Al} > 5.18$ the calculated ratio of Fe/Al? As apposed to the enrichment factor? This is confusing.
- P6, L137: I believe a work is missing between encompasses and exhibiting.
- 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.
- P10, Fig. 2: Are the samples from the Indian/Southern Ocean not included?
- P11, L215: Are the fine aerosol T-Fe and T-Al plotted somewhere? “In contrast...” implies the reader can compare the results.
- P12, L235: Are the mean bulk Fe% significantly different? It appears they might not be in some cases.
- 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.
- P13, L261: The figure numbers and their corresponding references in the text break down from this point forward.
- 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?
- P18, L367: What is NEV?
- 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?
- 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.
- P20, Table 2: Add horizontal gridlines for clarity. In the caption, what are the italicized values indicative of?
- 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?
- 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?
- P26, L493: Provide a reference for the seasonal variability in Fe% being caused by the temperature dependence of aerosol pH.
- P28, L557: is Feanthro-dFe overestimated in the models? Please clarify.