Supplemental Information for

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

Kohei Sakata[1*], Minako Kurisu[2], and Yoshio Takahashi[3]

[1]Materials Science and Engineering, Graduate School of Engineering, Tokyo Denki University, 5 Senjyu-Asahi-Cho, Adachi-ku, Tokyo 120-8551, Japan.

[2]Atmosphere and Ocean Research Institute, The University of Tokyo, 5-1-5, Kashiwanoha, Kashiwa, Chiba 277-8564, Japan.

[3]Graduate School of Science, The University of Tokyo, 7-3-1, Hongo Bunkyo-ku, Tokyo 113-0033, Japan.

Email: kohei.sakata.33@mail.dendai.ac.jp

ORCID: 0000-0002-0103-9631

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Supporting Text

S1. Sampling method for size-fractionated aerosol particles.

This study provides unpublished total and dissolved Fe and Al concentrations in size-fractionated marine aerosol particles collected in the Pacific Ocean. Size-fractionated aerosol particles were collected during the research cruise of R/V Hakuho-Maru (KH-14-6). A high-volume air sampler (Model-123SL, Kimoto, Japan) was installed on the compass deck at 13 m above sea level. Aerosol samples were separated into seven-size fractions using a Sierra-type cascade impactor (TE-236, Tisch Environmental Inc., USA, Table S1). Aerosol particles in stages 1 to 6 were collected on the custom-made polytetrafluoroethylene filter (Sakata et al., 2018, NAFLON™ PTFE tape, Nichias Co., Ltd., Japan). The filter was rinsed following sequential procedures: ultrapure water, 3 mol/L of HNO3, 3 mol/L of HCl, and ultrapure water. The HNO_3 and HCl were diluted from concentrated acids with electric grade (Kanto Chemical Co. Inc., Japan). The filter was heated at 150°C for one day in all procedures. The finest fraction was collected on a cellulose filter (Whatman 41, 516 cm^2, GE Healthcare, USA), which was not pre-washed before sampling. The proportions of filter blank concentration relative to measurement concentration (=blank + sample) of T-Fe and T-Al were 9.1±4.9% and 2.6±2.7%, respectively.

The total digestion and water extraction of Fe and Al were performed based on Sakata et al. (2022). Briefly, total digestion and water extraction were performed using digested by a mixed acid (2 mL of 15.2 mol L^{-1} HNO_3, 2 mL of 9.3 mol L^{-1} HCl, and 1 mL of 20 mol L^{-1} HF, TAMAPURE-AA100). The mixed acid was evaporated to dryness, then the residue was redissolved in 5 mL of 0.15 mol L^{-1} HNO_3) and heated at 150°C for a few hours. The solution was filtrated by a hydrophilic PTFE membrane filter (ϕ: 0.20 μm, Dismic®, 25HP020AN, Advantec, Japan). Dissolvable Fe and Al were extracted by 5 mL of ultrapure water with ultrasonication for 30 min. The water-extracted solution was filtrated through the hydrophilic PTFE membrane filter, then the solution was acidified to 0.15 mol L^{-1} of HNO_3 by 15.2 mol L^{-1} of HNO_3. All sample treatments were performed in a clean booth with HEPA filtrated air condition (SS-MAC 15) installed in a clean room (Class-10000). Total and dissolvable concentrations of Fe and Al were determined by inductively coupled plasma mass spectrometry (ICP-MS, Agilent7700, Agilent, Japan). Iron and Al concentrations were determined by helium gas and non-gas collision modes, respectively.

Supplemental Figure

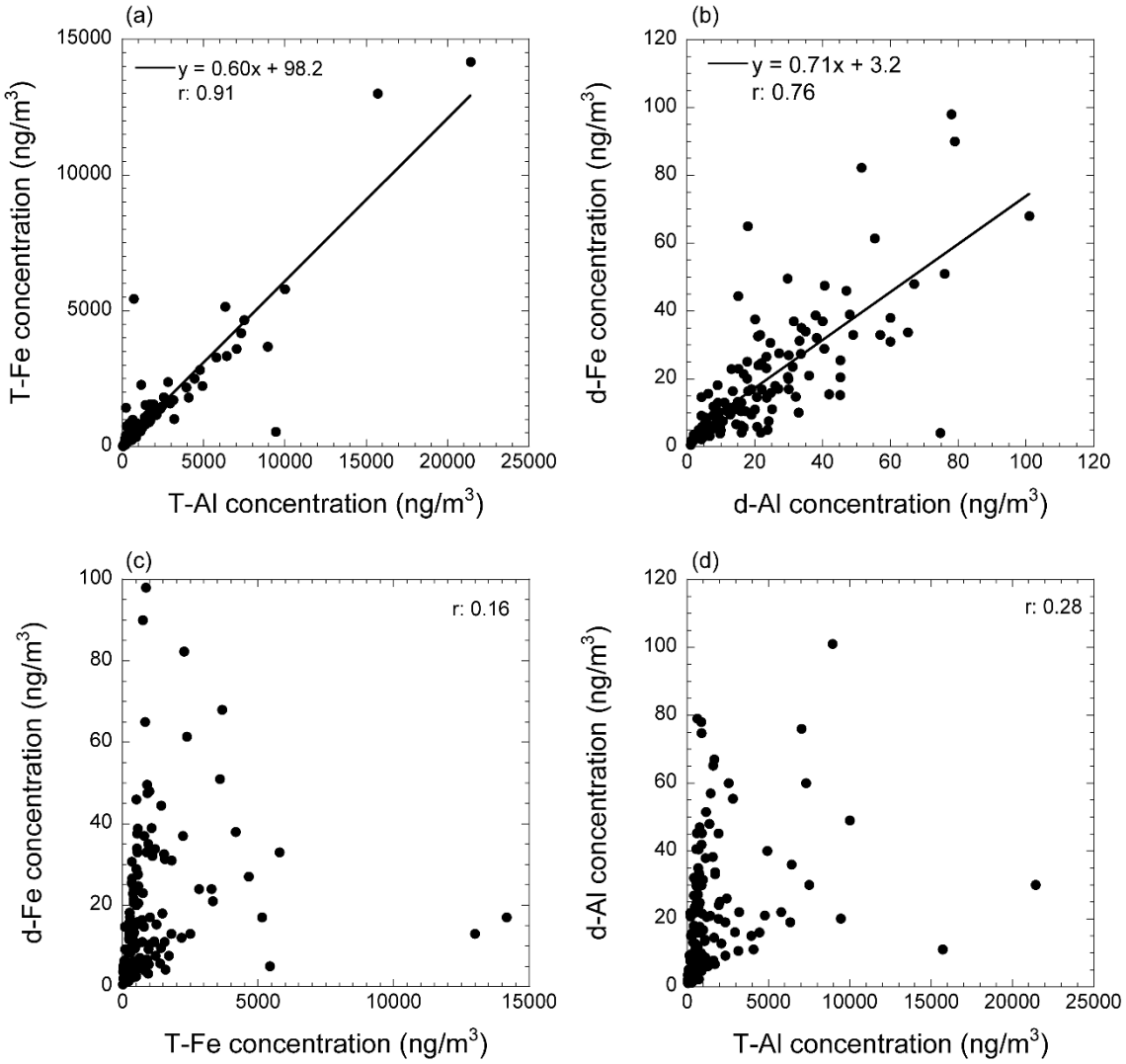


Figure S1. Scatter plots (a) between T-Al and T-Fe concentrations, (b) between d-Al and d-Fe concentratir4eons, (c) between T-Fe and d-Fe concentrations, and (d) between T-Al and d-Al concentrations in aerosol particles collected in East Asia.

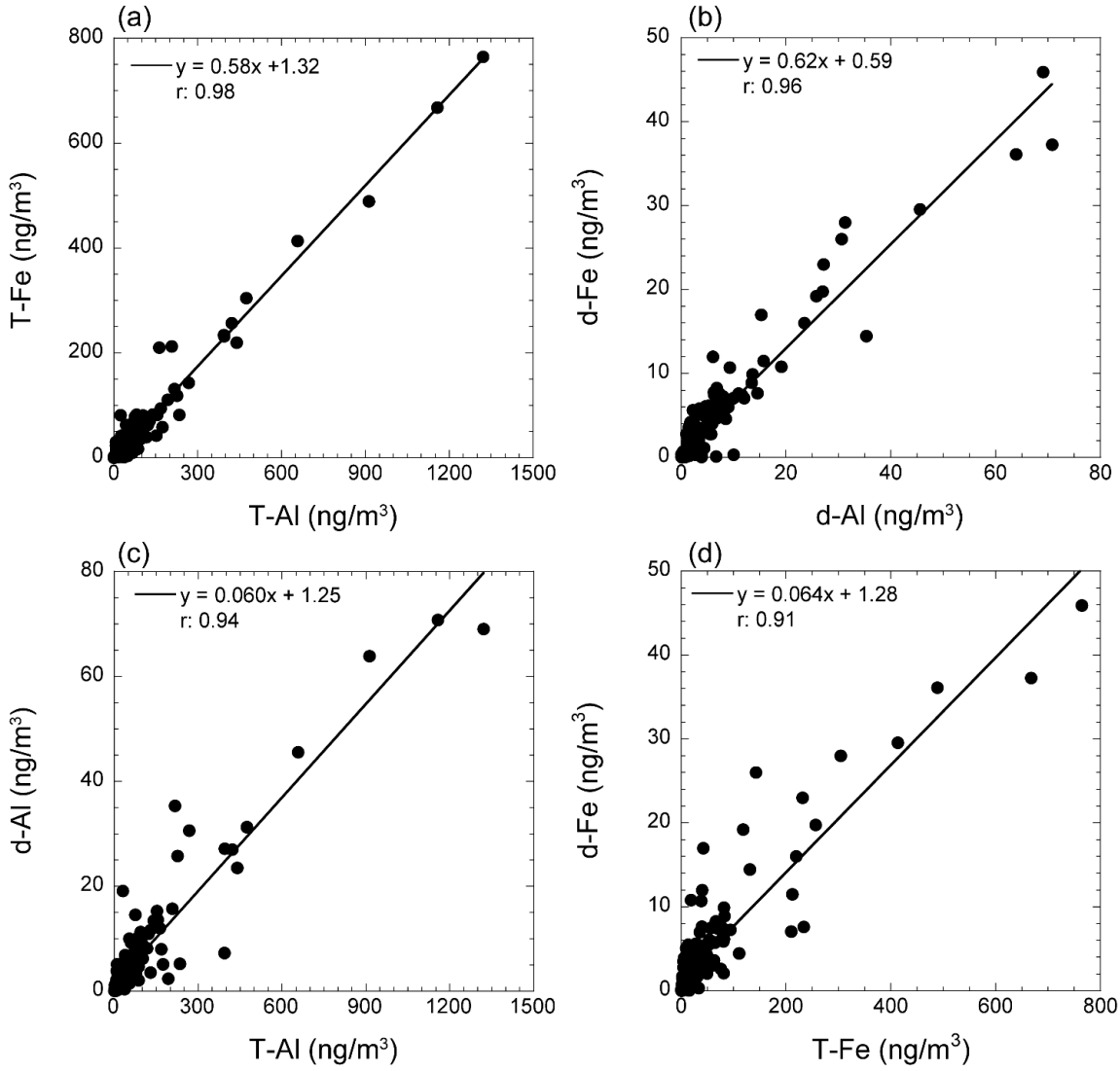
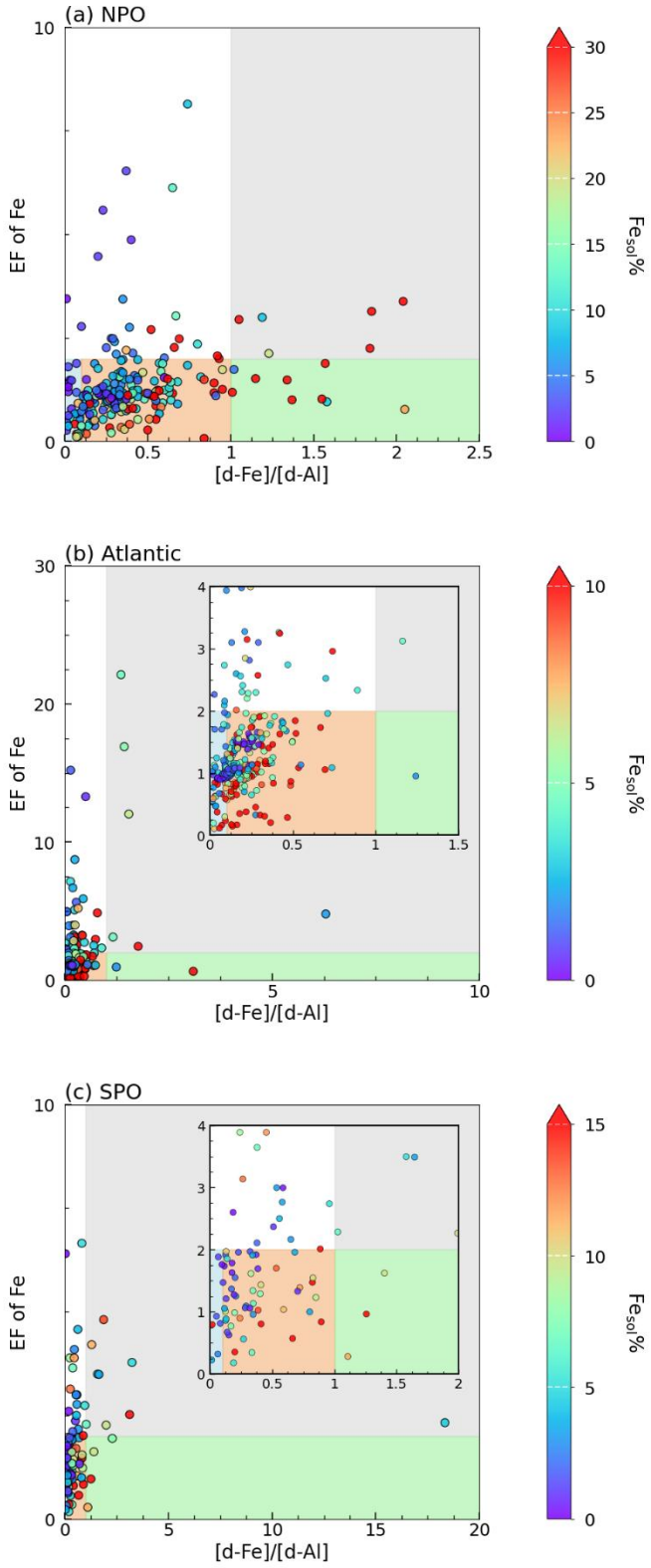


Figure S2. Scatter plots (a) between T-Al and T-Fe concentrations, (b) between d-Al and d-Fe concentrations, (c) between T-Fe and d-Fe concentrations, and (d) between T-Al and d-Al concentrations in aerosol particles collected in the North Pacific.



- **Figure S3.** 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.

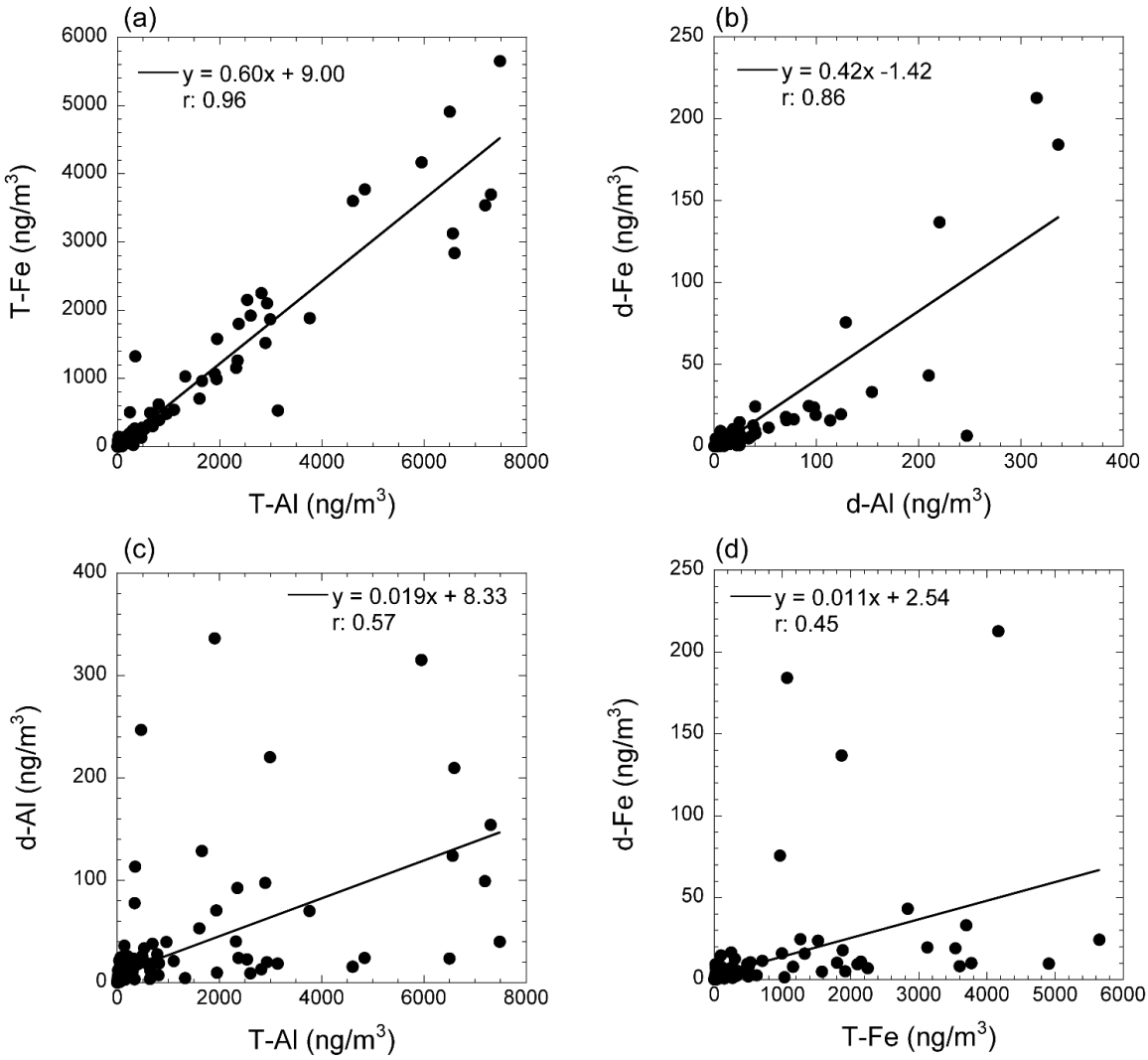


Figure S4. Scatter plots (a) between T-Al and T-Fe concentrations, (b) between d-Al and d-Fe concentrations, (c) between T-Fe and d-Fe concentrations, and (d) between T-Al and d-Al concentrations in aerosol particles collected in the Atlantic Ocean.

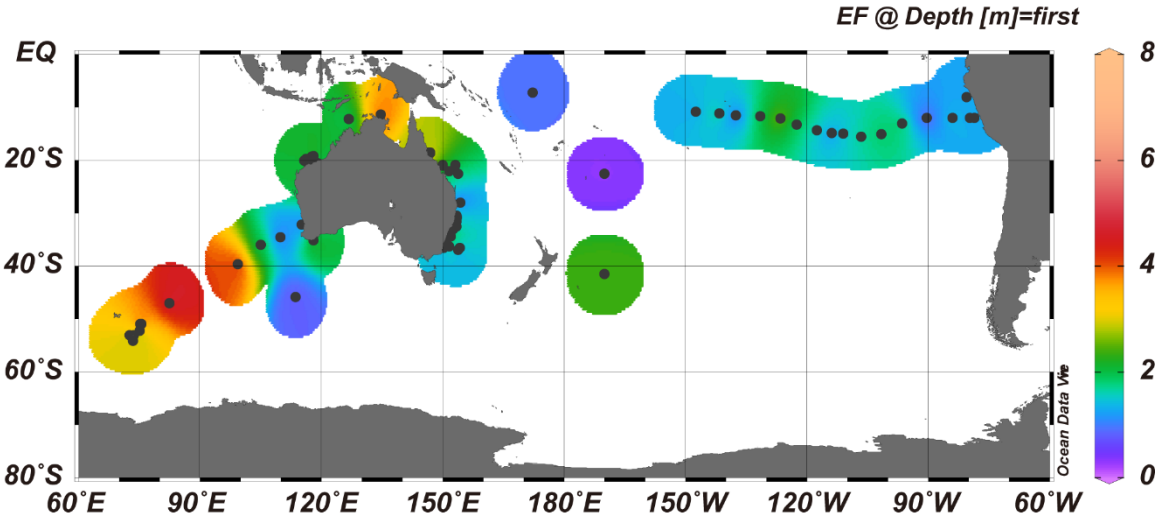


Figure S5. Spatial distribution of EFs in South Pacific TSP.

Reference

Sakata, K., Kurisu, M., Takeichi, Y., Sakaguchi, A., Tanimoto, H., Tamenori, Y. et al. (2022). Iron (Fe) speciation in size-fractionated aerosol particles in the Pacific Ocean: The role of organic complexation of Fe with humic-like substances in controlling Fe solubility, Atmospheric Chemistry Physics, 22, 9461–9482, https://doi.org/10.5194/acp-22-9461-2022.

Sakata, K., Kurisu, M., Tanimoto, H., Sakaguchi, A., Uematsu, M., Miyamoto, C., Takahashi, Y. (2018) Custom-made PTFE filters for ultra-clean size-fractionated aerosol sampling for trace metals. Marine Chemistry, 206, 100– 108. https://doi.org/10.1016/j.marchem.2018.09.009.