



1 Persistent increases in bioaccessible iron deposition
2 from anthropogenic processing activities to the North
3 Pacific and Indian Oceans despite rapid decreases
4 from fuel combustion sources

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11 **Abstract**

12 The trend of atmospheric deposition of bioaccessible iron (BFe) from anthropogenic sources is
13 driven by changes in aerosol column loading of total Fe (TFe) and Fe bioaccessibility, primarily
14 due to the evolution of human activities. Black carbon (BC) is often used as a temporal and spatial
15 proxy for BFe emissions from anthropogenic aerosols. However, derived changes in BFe
16 concentrations from anthropogenic processing activities (e.g., smelting and abrasion) and fuel
17 combustion have not been explored separately because of a lack of evaluation of BFe aerosols for



18 separate anthropogenic sources. The Fe stable isotope ratio ($\delta^{56}\text{Fe}$) of size-segregated aerosols is
19 a useful tracer to disentangle anthropogenic Fe sources. Here, we evaluate the source contributions
20 predicted by an atmospheric chemical transport model using size-segregated aerosol observations
21 over the North Pacific. Using the improved model, we investigate the relative changes in BFe
22 aerosols from anthropogenic processing activities and fuel combustion sources in the major
23 continental outflow regions during 2002–2022. The model reveals a persistent increase in BFe
24 supply to the North Pacific and Indian Oceans from anthropogenic processing activity sources
25 from 2002 (18 Gg Fe yr⁻¹) to 2022 (20 Gg Fe yr⁻¹) despite a decrease from fuel combustion from
26 2012 (20 Gg Fe yr⁻¹) to 2022 (15 Gg Fe yr⁻¹). These findings may have important implications for
27 projections of future micronutrient supply to the oceans, which regulates the magnitude and
28 dynamics of marine primary productivity.

29 **Keywords**

30 iron, anthropogenic aerosol, mineral dust, air pollution, biomass burning

31 **Short summary**

32 Air pollution controls and industrial restructuring have reduced total iron emissions in aerosols
33 from anthropogenic sources. However, we reveal a persistent increase in bioaccessible iron
34 deposition from anthropogenic processing activities. These findings may have important
35 implications for assessing the impact of air quality on climate, marine ecosystems, and human
36 health and for exploring effective pollution control strategies.

37 **1. Introduction**

38 Atmospheric emissions of aerosol iron (Fe) from anthropogenic sources have been modulated
39 by changes in industrial activities and environmental policies. Thus, anthropogenic aerosol Fe



40 concentrations and deposition have changed substantially on a regional basis. Such aerosol Fe
41 changes could have important implications for a variety of environmental issues such as ocean
42 fertilization (Ito et al., 2021; Boyd et al., 2024), climate change (Ito et al., 2026; Moteki, 2023),
43 and human health risk (Campbell et al., 2025). However, significant uncertainties remain in the
44 source apportionment due to a lack of source-specific evaluation of Fe-laden aerosols. Thus,
45 constraining the anthropogenic source apportionment is critical in assessing the evolution of
46 atmospheric Fe deposition to the ocean due to climate change and human activity.

47 Previous studies have analyzed historical model simulations to investigate aerosol Fe trends on
48 regional and global scales, mostly based on the evaluation of total Fe (TFe) and bioaccessible Fe
49 (BFe) concentrations without distinguishing anthropogenic aerosol sources between
50 “anthropogenic processing activity” and “fuel combustion” (Luo et al., 2008; Ito and Shi, 2016;
51 Hamilton et al., 2020; Rathod et al., 2022; Bergas-Massó et al., 2023). Thus, attribution of the
52 estimated trends and spatial variability to specific aerosol Fe sources remains elusive. Here, we
53 regard “anthropogenic processing activity” as the metal smelting, ground transportation, and
54 shipping transportation sectors and “fuel combustion” as the energy production, industry,
55 Residential, Commercial, and Other (RCO) (excluding biofuel and waste), and biofuel/waste
56 combustion sectors, respectively.

57 A distinction in Fe stable isotope ratios ($\delta^{56}\text{Fe}$) for size-segregated aerosols between natural and
58 anthropogenic aerosols provides constraints on model predictions of aerosol source types (Wei et
59 al., 2024; Zhang et al., 2025b; Kurisu et al., 2026b). Given the strong dependence of $\delta^{56}\text{Fe}$ on
60 particle size, it is crucial that these aerosol source characterizations are size-segregated for models
61 to reproduce the lighter $\delta^{56}\text{Fe}$ in the smaller particles compared to the heavier $\delta^{56}\text{Fe}$ in the larger
62 particles (Hsieh and Ho, 2024; Kurisu et al., 2026b). Additionally, high-time-resolution field



63 observations of trace elements have been used to verify the contribution of lithogenic (mineral
64 dust) and anthropogenic sources to aerosol TFe (Ito and Miyakawa, 2023; Zhang et al., 2024).

65 East Asia and Southeast Asia have intensive anthropogenic emissions of aerosol Fe from fuel
66 combustion, metal production, and transportation, which deliver BFe to the North Pacific. Among
67 the high nitrate, low chlorophyll (HNLC) regions, the subarctic Pacific displays high sensitivity to
68 Fe fertilization experiments, depending on biogeochemical processes such as the selection of the
69 dominant phytoplankton group or species, the effects of grazing, interactions within the plankton
70 community, and the effects of nutrient concentrations on phytoplankton growth (Yoon et al., 2018;
71 Ito et al., 2023). Anthropogenic aerosol Fe has been shown to alter phytoplankton bloom dynamics
72 and nutrient supply as far as the North Pacific transition zone (Pinedo-González et al., 2020;
73 Hawco et al., 2025). Along this outflow pathway, size-segregated aerosol measurements over the
74 East China Sea indicate that anthropogenic and lithogenic aerosol Fe exhibit distinct $\delta^{56}\text{Fe}$ values
75 and solubilities among different particle sizes and leaching methods (Hsieh et al., 2023; Hsieh and
76 Ho, 2024). The size-fractionated aerosol $\delta^{56}\text{Fe}$ studies indicate that anthropogenic, lithogenic, and
77 pyrogenic (open biomass burning) sources constitute a substantial and variable fraction of the Fe
78 delivered to the North Pacific (Kurusu et al., 2024; Bunnell et al., 2025). In China, air pollution
79 controls and industrial restructuring have altered the relative contribution of anthropogenic and
80 lithogenic sources to atmospheric Fe over the past decade (Zhang et al., 2025a; Cui et al., 2025),
81 necessitating a thorough, source-resolved evaluation of the contribution of East and Southeast
82 Asian anthropogenic emissions to BFe deposition in the North Pacific. Consequently, it is crucial
83 to understand the trend and the driving factors to assess the impact accurately and develop
84 informed environmental management strategies. Thus, an interdisciplinary approach combining
85 modeling and observations is one of the best methods for this evaluation.



86 South Asia and the Middle East also have intensive anthropogenic, lithogenic, and pyrogenic
87 emissions of aerosols, which supply BFe to the northern Indian Ocean. Influenced by the winter
88 and summer monsoon circulation, aerosols from the Indo-Gangetic Plain are transported over the
89 Bay of Bengal, where anthropogenic sources contribute to the high fractional solubility of aerosol
90 Fe (Ito et al., 2019; Kumar et al., 2024). The tropical Indian Ocean and Arabian Sea are among the
91 ocean regions where atmospheric nutrient supply constrains marine nitrogen fixation and primary
92 productivity (Seok et al., 2026). Model studies further indicate that anthropogenic Fe deposition
93 has substantially altered the ecosystem and carbon balance of the Indian Ocean over the industrial
94 era (Pham and Ito, 2021). Under a future scenario with high emissions of near-term climate forcings
95 (SSP3-7.0), a global model suggests increasing BFe deposition to the Indian Ocean (Bergas-Massó
96 et al., 2023). However, aerosol deposition fluxes to this basin remain among the least constrained
97 by both observations and models, with deposition to the Indian Ocean underestimated by $34 \pm 22\%$
98 (He et al., 2025). Consequently, a source-resolved investigation that distinguishes anthropogenic
99 processing activity and fuel combustion contributions is indispensable for future research to
100 accurately assess the impact of these emissions on Indian Ocean biogeochemistry.

101 Here, our main objective is to characterize the trends and spatial variability of BFe deposition
102 from anthropogenic processing activities and fuel combustion sources, separately, in the major
103 continental outflow regions to the North Pacific and Indian Oceans over the last two decades. To
104 verify the source apportionment of aerosol BFe, we combine the model results with ship-based
105 observations of size-segregated aerosol chemical compounds. We focus on the model validation
106 of aerosol BFe from different anthropogenic and natural sources over the North Pacific in the
107 downstream region of East Asian outflow. Our findings portray an increase in BFe deposition from



108 anthropogenic processing activities to the North Pacific and Indian Oceans despite a decrease from
109 fuel combustion from 2012 to 2022.

110 **2. Methods**

111 We estimate trace metal and BFe concentrations and deposition for Fe-containing aerosols from
112 anthropogenic processing activities and fuel combustion sources, separately. We describe the
113 atmospheric chemical transport model and the emission database of Fe aerosols for anthropogenic
114 sources in Section 2.1, the method to estimate the contribution of each source to aerosol Fe in
115 Section 2.2, and the observational dataset in Section 2.3.

116 **2.1 Atmospheric chemical transport model**

117 This study used the Integrated Massively Parallel Atmospheric Chemical Transport (IMPACT)
118 model (Ito et al., 2026, and references therein) with updated anthropogenic Fe emissions. The
119 model simulations were performed using a horizontal resolution of $2.0^{\circ} \times 2.5^{\circ}$ for latitude by
120 longitude and 47 vertical layers. The simulations were conducted for comparison with the field
121 measurements in 2017 and 2021, as well as for sensitivity simulations in 2002, 2012, and 2022.
122 The Modern-Era Retrospective analysis for Research and Applications 2 reanalysis meteorological
123 data from the National Aeronautics and Space Administration Global Modeling and Assimilation
124 Office (Gelaro et al., 2017) were used as the meteorological conditions to drive the atmospheric
125 chemical transport model. The model simulated the emissions, chemistry, transport, and deposition
126 of aerosols for four distinct aerosol size bins (<1.26 , $1.26\text{--}2.5$, $2.5\text{--}5$, and $5\text{--}20$ μm in diameter)
127 and their precursor gases for anthropogenic, pyrogenic, and lithogenic sources. The model
128 estimates BFe as the potential for Fe to be released from the particulate phase in aerosol water and

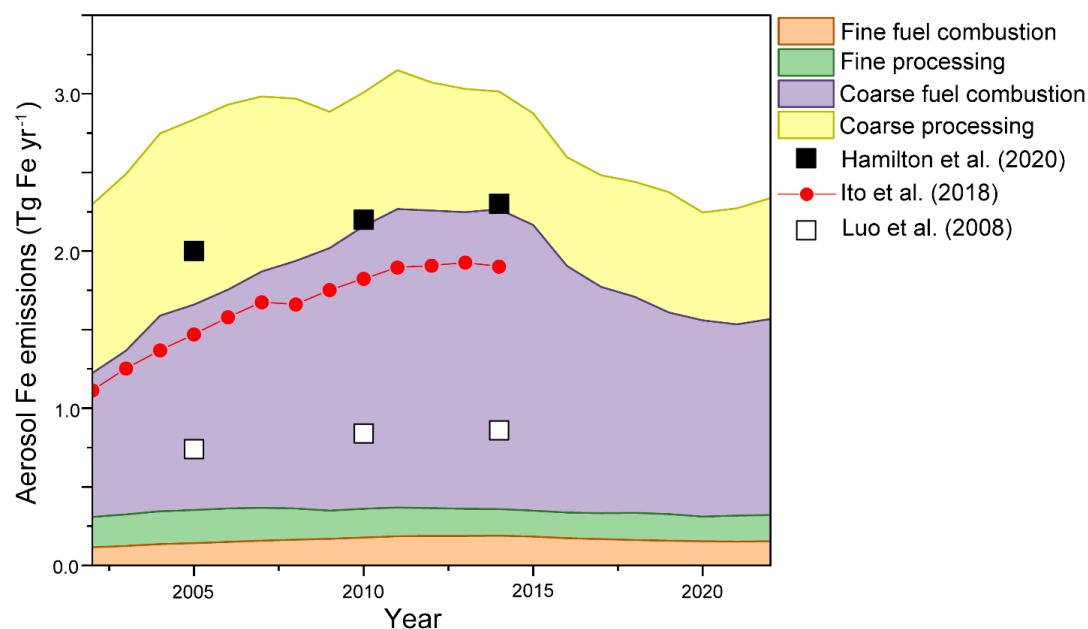


129 become available for biological uptake under environmentally relevant conditions, whereas water-
130 soluble Fe refers to an operationally defined measurement of Fe released into ultrapure water.

131 Anthropogenic and pyrogenic aerosol emissions were prescribed, whereas lithogenic emissions
132 were dynamically simulated to consider the weather conditions at each model time step.
133 Anthropogenic aerosol emissions in the fine mode were estimated for each country, sector, fuel
134 (i.e., process, biomass, coal, heavy oil, diesel oil, light oil, and natural gas), and time using BC
135 emissions and the fraction of BC in PM_{2.5} (Hoesly et al., 2025). The fraction of BC in PM₁₀ for
136 each sector (Dubois et al., 2025) was used to estimate anthropogenic aerosol emissions in the
137 coarse mode. The metal content of aerosols from anthropogenic sources was obtained from the
138 compilation of source-specific aerosol measurements (Ito and Miyakawa, 2023; Dubois et al.,
139 2025; Li et al., 2026) (Table S1). The metal smelting sector represents manufacturing processes
140 such as the production of iron and steel, aluminum, and other nonferrous metals. The emissions
141 from metal smelting were extrapolated in time using the Fe emissions in 2018 (Rathod et al., 2020;
142 Ito and Miyakawa, 2023) and BC emissions from processes (2C1_Iron-steel-alloy-prod,
143 2C3_Aluminum-production, 2C4_Non-Ferrous-other-metals in Hoesly et al., 2025) during the
144 historical period. The transportation sector includes exhaust and non-exhaust emissions (Dubois
145 et al., 2025). The former includes non-energetic combustion of engine lubricants, whereas the latter
146 mainly consists of abrasion from brakes, tires, and the road surface itself. The energy production,
147 industry, and RCO sectors were separated into biofuel and other fuel (i.e., coal, oil, and gas)
148 combustion sources. The aerosols emitted from anthropogenic processing activities (i.e., metal
149 smelting, road, and rail sectors) have higher Fe fractions than those from fuel combustion (i.e.,
150 biofuel, oil and coal) (Table S1).



151 The aerosol Fe emissions in this study, based on the CMIP7 emission database, are higher than
 152 those based on CMIP6 (Ito et al., 2018; Hamilton et al., 2020), largely because of additional
 153 anthropogenic processing activity emissions (Fig. 1). Coarse-mode emissions dominate the total
 154 for most sectors, particularly for the coal/oil (energy/industry/RCO) and transportation categories
 155 (Table S2). The rising phase of fuel combustion emissions from 2002 ($1.0 \text{ Tg Fe yr}^{-1}$) to 2014 (2.1
 156 Tg Fe yr^{-1}) based on CMIP7 agrees closely with the trend based on CMIP6 and shifts the dominant
 157 source from anthropogenic processing activities in 2002 ($1.3 \text{ Tg Fe yr}^{-1}$) to fuel combustion
 158 sources after 2004. After the peak, fuel combustion sources decline rapidly to 2022 (1.4 Tg Fe
 159 yr^{-1}), while anthropogenic processing activity emissions remain nearly constant ($0.9 \text{ Tg Fe yr}^{-1}$).
 160 The contrasting trends result in larger fine-mode anthropogenic processing activity emissions in
 161 2022 ($0.17 \text{ Tg Fe yr}^{-1}$) than fuel combustion emissions ($0.15 \text{ Tg Fe yr}^{-1}$).



162
 163 **Figure 1.** Annual aerosol Fe emissions from anthropogenic processing activity and fuel
 164 combustion in fine and coarse particles (this work) and previous studies from 2002 to 2022.



165 **2.2 Model estimate of each source contribution to anthropogenic Fe**

166 To estimate changes in anthropogenic Fe concentrations and deposition, we performed five
167 sensitivity experiments (from Experiment 1 to Experiment 5 in Table 1). The five experiments are
168 grouped into two source categories used in the source-attribution trend analysis: Experiments 1–3
169 represent anthropogenic processing activities, corresponding to the metal smelting, ground
170 transportation, and shipping transportation sectors, while Experiments 4–5 represent fuel
171 combustion, corresponding to the energy/industry/RCO (excluding biofuel and waste) and
172 biofuel/waste combustion sectors.

173 The model used different tagged tracers for different source sectors and mineralogy from
174 anthropogenic sources (Ito and Miyakawa, 2023; Ito et al., 2026). The Fe-bearing phases included
175 highly reactive amorphous Fe hydroxide, crystalline Fe oxide, Fe associated with magnetite, and
176 the remaining aluminosilicate minerals (Baldo et al., 2022). The previous model assumed that
177 anthropogenic Fe aerosols were emitted by high-temperature evaporation processes and prescribed
178 a $\delta^{56}\text{Fe}$ of -4.7‰ for all anthropogenic emissions (Kurusu et al., 2021). Consequently, the previous
179 version of the IMPACT model reproduced the light $\delta^{56}\text{Fe}$ in fine particles but estimated lighter
180 $\delta^{56}\text{Fe}$ in coarse particles compared to observations, probably because of the different
181 characteristics of anthropogenic Fe aerosols for different sources emitted by different processes
182 (Kurusu et al., 2026b). Subsequently, the revised version of the IMPACT model considered the
183 mixing of evaporation (i.e., light $\delta^{56}\text{Fe}$) and abrasion (i.e., heavy $\delta^{56}\text{Fe}$) and prescribed a $\delta^{56}\text{Fe}$ of
184 $-0.4 \pm 0.1\text{‰}$ for traffic emissions based on observations of fine particles collected in a tunnel
185 (Kurusu et al., 2016). As a result, the revised version of the IMPACT model reproduced the heavier
186 endmember value of anthropogenic Fe ($-3.3 \pm 1.2\text{‰}$) estimated from observations of fine particles
187 (Kurusu et al., 2026b).



188 Here, we assume that aerosol concentrations are log-normally distributed within each
189 observation size stage, and use that distribution to estimate what fraction of the observation size
190 stage's mass is allocated to each model bin whenever a model-bin boundary falls inside that
191 observation size stage. Accordingly, the values of $\delta^{56}\text{Fe}$ from traffic emissions were estimated to
192 be -0.65‰ for bin1, 0.10‰ for bin2, 0.22‰ for bin3, and 0.25‰ for bin4 from the size-segregated
193 measurements (Kurusu et al., 2016). Further, the IMPACT model results indicate the dissolution
194 of surface amorphous materials and the exposure of mineral surfaces that could enhance the ice
195 nucleation activity of crystalline Fe-containing particles (Ito et al., 2026). The surface exposure
196 was inferred from simulated Fe-bearing phase changes (Baldo et al., 2022), which were associated
197 with morphology changes (Chen et al., 2012). The last stable phases in Fe-bearing particles are
198 not produced by evaporation but by melting of source materials and subsequent cooling. Therefore,
199 we prescribed the same $\delta^{56}\text{Fe}$ for the aluminosilicate clay minerals in each anthropogenic source
200 as in lithogenic and pyrogenic Fe (0.1‰). Overall, $\delta^{56}\text{Fe}$ values were prescribed for (i) metal
201 smelting and fossil fuel combustion sectors (-4.7‰) excluding crystalline Fe-containing particles,
202 (ii) ground and shipping transport sectors (-0.65‰ for bin1, 0.10‰ for bin2, 0.22‰ for bin3, and
203 0.25‰ for bin4), (iii) biofuel and waste combustion sectors (0.1‰), and (iv) crystalline Fe-
204 containing particles in anthropogenic Fe (0.1‰).

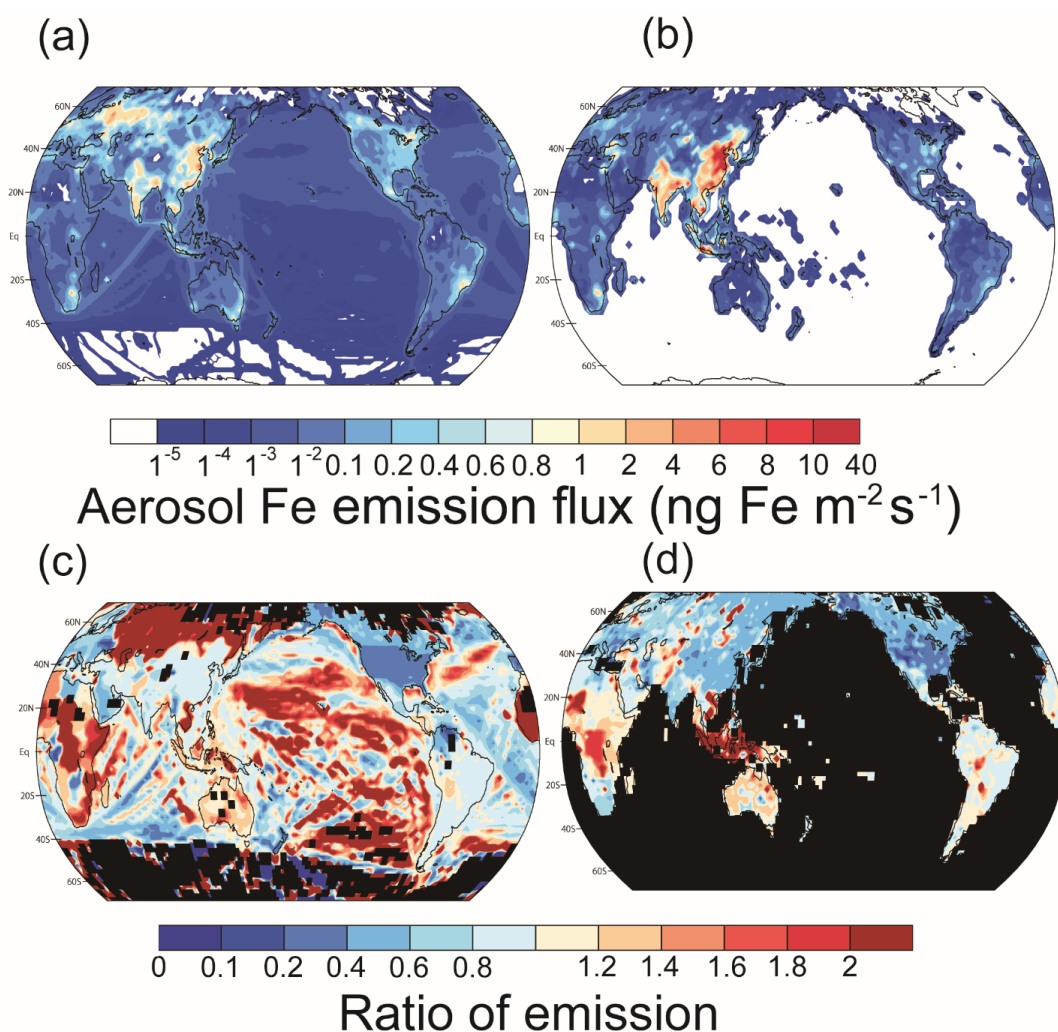
205 **Table 1.** Summary of model experiments performed in this study.

Simulation	Category	Sector
Experiment 1	Anthropogenic processing	Metal smelting
Experiment 2	Anthropogenic processing	Ground transportation
Experiment 3	Anthropogenic processing	Shipping transportation
Experiment 4	Fuel combustion	Energy, industry, and RCO, except biofuel and waste
Experiment 5	Fuel combustion	Biofuel and waste combustion

206 RCO represents Residential, Commercial, and Other sectors.



207 This sector-based grouping underlies two categories of anthropogenic Fe emissions from
208 anthropogenic processing activity and fuel combustion (Fig. 2). Anthropogenic processing activity
209 emissions are more spatially widespread than fuel combustion emissions, showing shipping
210 emissions over most ocean basins together with distinct hotspots over South and East Asia. On the
211 other hand, fuel combustion emissions are more sharply concentrated over South Asia and parts
212 of East and Southeast Asia. The 2012–2022 ratio of anthropogenic processing activity emissions
213 exceeds 1 across most low- to mid-latitude ocean regions, including the tropical Pacific and Indian
214 Oceans and parts of Africa and the Middle East, indicating a broad increase over the decade, with
215 only limited regions (e.g., off the eastern coast of North America) showing a decrease. The ratio
216 of fuel combustion emissions shows a more spatially patchy pattern, with increases concentrated
217 over parts of Africa, the Maritime Continent, and Australia and decreases over Europe and North
218 America.



219

220 **Figure 2.** Comparison of anthropogenic Fe emission fluxes ($\text{ng m}^{-2} \text{s}^{-1}$) from (a) anthropogenic
 221 processing activity sources in 2022, (b) fuel combustion sources in 2022, (c) ratio of anthropogenic
 222 processing activity sources from 2012 to 2022, and (d) ratio of fuel combustion sources from 2012
 223 to 2022. The black-shaded areas denote grid cells with negligible fuel combustion emissions in
 224 2012.



2.3 Observational dataset of size-segregated aerosols over the North Pacific Ocean

Measurements of size-segregated aerosol concentrations used in this study were obtained from two ship-board atmospheric sampling efforts undertaken in 2017 (R/V Hakuho-maru KH-17-3 cruise, KH2017, circles) (Kurusu et al., 2024) and 2021 (R/V Mirai MR21-01 cruise, MR2021, stars) (Kurusu et al., 2026a) over the North Pacific Ocean (Fig. 3). The KH-17-3 track extends into the eastern North Pacific along a single zonal transect, whereas the MR21-01 track forms a looped pattern closer to the Japanese coast. Thus, the two cruises provide seasonally distinct (summer versus late winter) and largely non-overlapping spatial coverage of the North Pacific outflow region.

Size-segregated aerosol samples were collected using a Sierra-type cascade impactor (TE-235 or TE-236, Tisch Environmental Inc., USA) installed on a high-volume air sampler (KIMOTO, Model-120SL, Japan) at a flow rate of 1132 L min^{-1} . The particles were separated into six stages (i.e., Stage 1, $>7.2 \mu\text{m}$; Stage 2, $3.0\text{--}7.2 \mu\text{m}$; Stage 3, $1.5\text{--}3.0 \mu\text{m}$; Stage 4, $0.95\text{--}1.5 \mu\text{m}$; Stage 5, $0.49\text{--}0.95 \mu\text{m}$; and Stage 6, $<0.49 \mu\text{m}$) for KH-17-3 and seven stages (i.e., Stage 1, $>15 \mu\text{m}$; Stage 2, $7.2\text{--}15 \mu\text{m}$; Stage 3, $3.2\text{--}7.2 \mu\text{m}$; Stage 4, $1.7\text{--}3.2 \mu\text{m}$; Stage 5, $1\text{--}1.7 \mu\text{m}$; Stage 6, $0.54\text{--}1 \mu\text{m}$; and Stage 7, $<0.24 \mu\text{m}$) for MR21-01, respectively.

Acid digestion of the aerosol particles for total elemental concentration analysis, leaching of aerosol particles for the analysis of the water-soluble fraction of the trace elements, and iron isotope analysis are described in previous studies (Kurusu et al., 2024). Briefly, aerosol TFe concentrations were analyzed using inductively coupled plasma quadrupole mass spectrometry (ICPMS) after mixed-acid digestion. The aerosol water-soluble Fe concentrations were analyzed using a batch extraction method in ultrapure water with ultrasonic treatment for 30 min. After 24

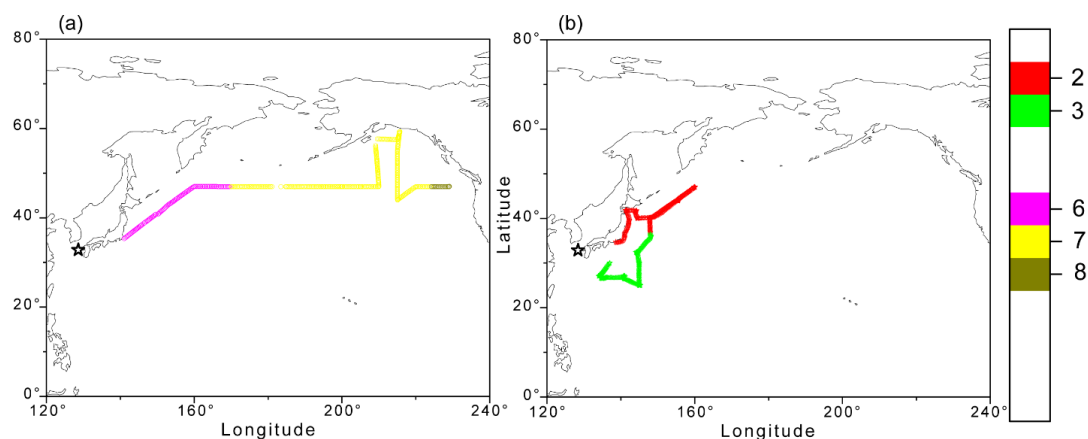


h, the samples were filtered with a 0.2 µm hydrophilic PTFE filter. Based on the analysis of three replicate samples, relative standard deviations (accounting for the uncertainty of the sampling method and the leaching protocol) were estimated for TFe (5–20%) and soluble Fe (5–10%) concentrations. $\delta^{56}\text{Fe}$ is expressed as:

$$\delta^{56}\text{Fe} = \left(\frac{(^{56}\text{Fe}/^{54}\text{Fe})_{\text{sample}}}{(^{56}\text{Fe}/^{54}\text{Fe})_{\text{IRMM-014}}} - 1 \right) \times 1000 \quad (\text{‰}),$$

where IRMM-014 (obtained from the Institute for Reference Materials and Measurements) was used as the standard material. After Fe separation using an anion exchange resin, the Fe isotope ratio was analyzed using a multi-collector ICPMS (Neptune Plus, Thermo Fischer Scientific, Germany) at JAMSTEC.

The measurement datasets of elemental concentrations of $\text{PM}_{1.0}$ aerosols were obtained from the same MR21-01 cruise (Miyakawa et al., 2025; Miyakawa and Kinase, 2026), whereas those of atmospheric BC particles are available at Fukue Island (32.75° N, 128.68° E) (Kanaya et al., 2020). The metal concentrations (Fe, Ti, Cu, and Pb) of fine-mode aerosols were measured using a continuous particulate monitor with X-ray fluorescence (XRF) analysis (PX-375, Horiba Ltd., Kyoto, Japan).



262

263 Figure 3. Sampling periods (month) along the cruise tracks and location of the Fukue observational
 264 station (black star: 32.75° N, 128.68° E). (a) June to August in 2017 (cruise number: KH-17-3,
 265 circles). (b) February to March in 2021 (cruise number: MR21-01, stars). White colors represent
 266 no observational data from the cruises.

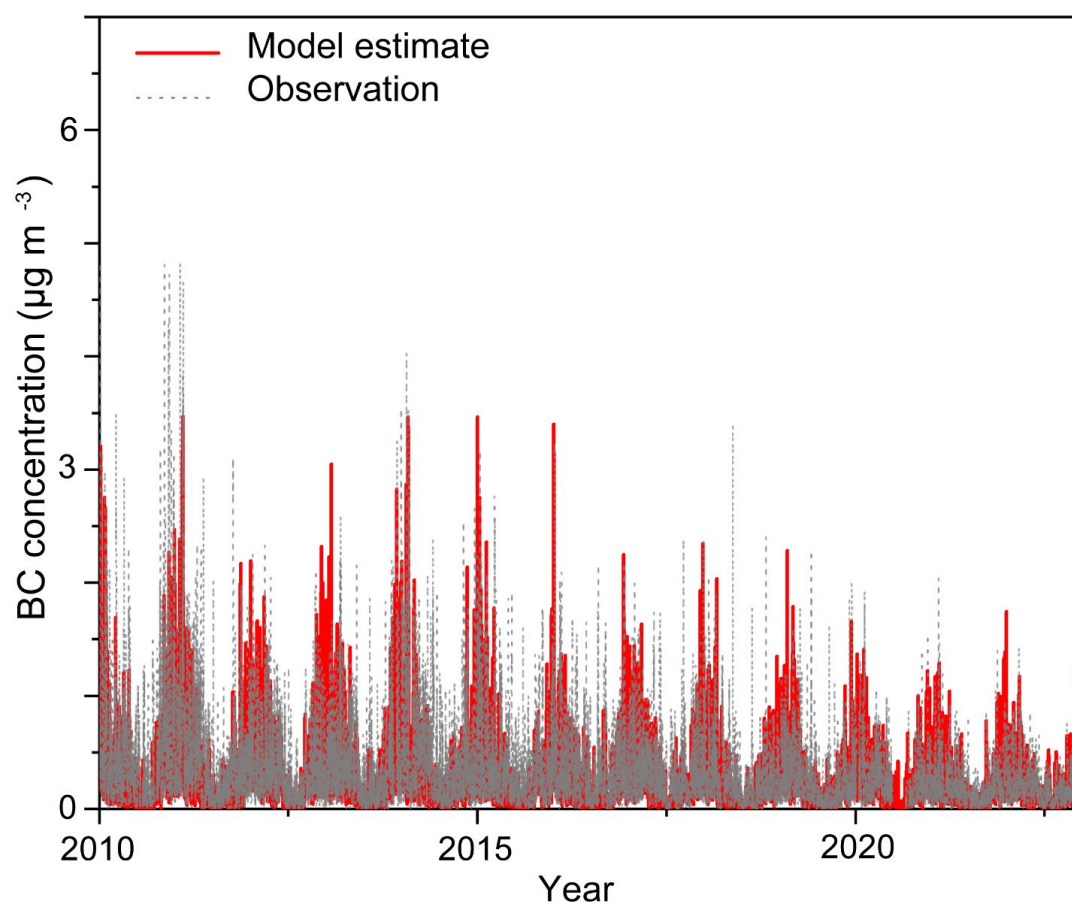
267 3. Results and discussion

268 3.1 Evaluation of black carbon concentrations at Fukue

269 The long-term model estimates of black carbon (BC) concentrations were compared with field
 270 observations for aerosol samples collected at Fukue station, whereas the short-term simulation
 271 results were thoroughly evaluated using the same observational dataset (Ito and Miyakawa, 2023;
 272 Miyakawa et al., 2023, 2025). The model generally reproduced BC concentrations within a factor
 273 of 10 (Fig. 4 and Table S3), primarily owing to the improvement in the emission database (see
 274 Section 2.1). Both time series show strong day-to-day variability superimposed on a recurring
 275 winter–spring maximum and summer minimum. The model reproduces the timing of most
 276 observed peaks, though it sometimes underestimates the polluted episodes. Peak concentrations in
 277 both model and observations decrease gradually over the record, consistent with a recent reduction
 278 in regional combustion emissions (Kanaya et al., 2020).



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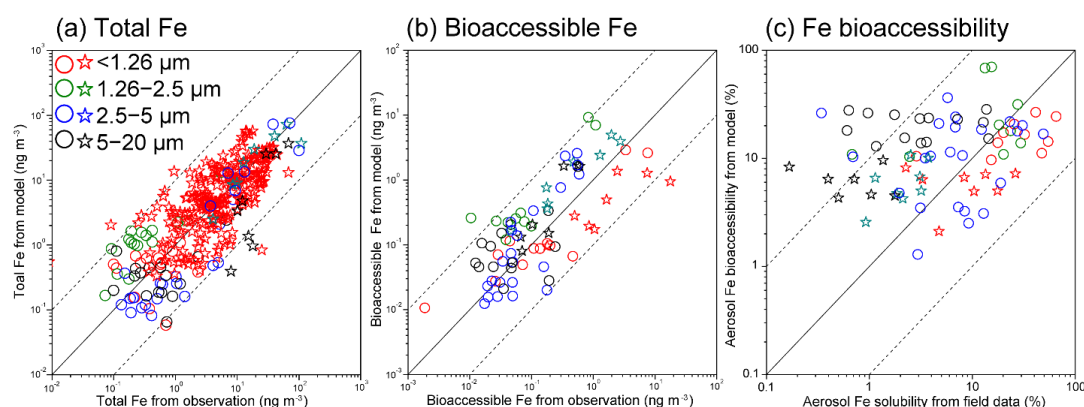
281 **Figure 4.** BC concentrations at Fukue station from model estimates (median: 0.19) and
 282 observations (median: 0.20) from 2010 to 2022. Summary of model statistics is available in Table
 283 S3.

284 3.2 Evaluation of trace metals and $\delta^{56}\text{Fe}$ over the North Pacific Ocean

285 We compared TFe, BFe, and Fe bioaccessibility ($= \text{BFe} / \text{TFe}$) for the 2017 (circles) and 2021
 286 (stars) cruises (Fig. 5 and Table S4). The model reproduced aerosol TFe and BFe concentrations
 287 over the North Pacific within a factor of 10. On the other hand, Fe bioaccessibility was more widely



288 scattered, with the closest agreement for fine particles, and tended to be overestimated by the
 289 model for the coarser size bins. The results are consistent with previous measurements, which
 290 indicate that solubilities obtained with ultrapure water are often lower than those obtained with
 291 buffer and Berger leaches in coarser particles, but the deviations are relatively small for fine
 292 particles (Hsieh et al., 2023; Hsieh and Ho, 2024).

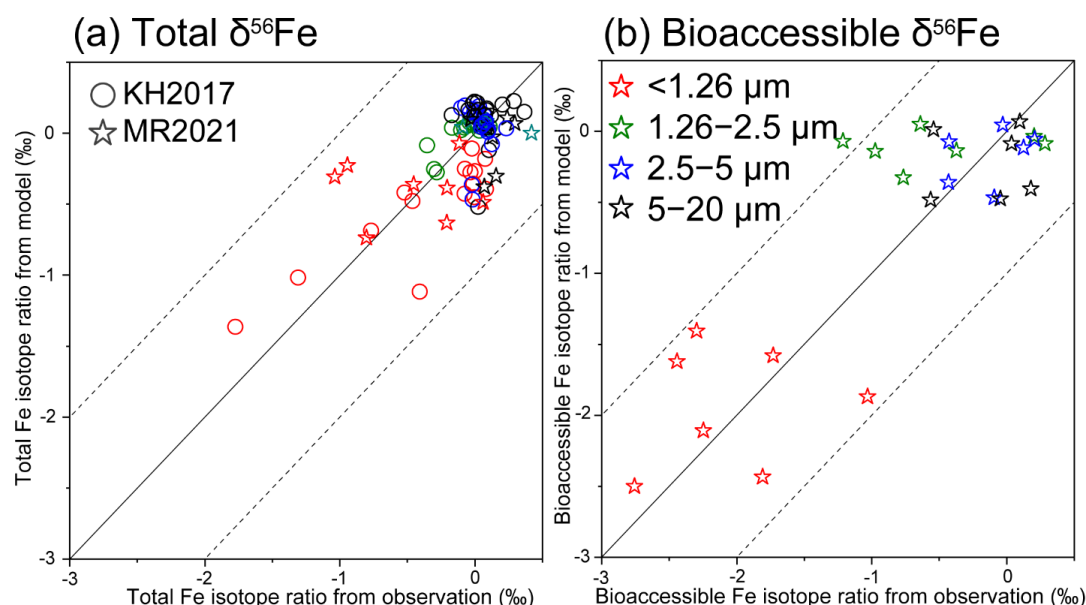


293 **Figure 5.** Comparison of (a) total Fe concentrations, (b) bioaccessible Fe concentrations, and (c)
 294 Fe bioaccessibility for 2017 (circle) and 2021 (star) particles. Data points are colored by aerosol
 295 size bin (red: <1.26 μm; green: 1.26–2.5 μm; blue: 2.5–5 μm; black: 5–20 μm). Total Fe
 296 concentrations of fine-mode aerosols are measured using PX-375, whereas aerosol concentrations
 297 for each bin are estimated by assuming a log-normal distribution within each size stage for samples
 298 analyzed using ICPMS. Summary of model statistics is available in Table S4.

300 We compared total and bioaccessible $\delta^{56}\text{Fe}$ for size-segregated aerosols between model
 301 estimates and observations for KH2017 (circle) and MR2021 (star) (Fig. 6 and Table S5). Fine
 302 particles (<1.26 μm, red) are significantly isotopically lighter than the coarser particles for both
 303 TFe and BFe, which is consistent with an anthropogenic source signature. On the other hand,
 304 coarse particles larger than 1.26 μm are close to crustal values ($\delta^{56}\text{Fe} \approx 0$) in both the model and

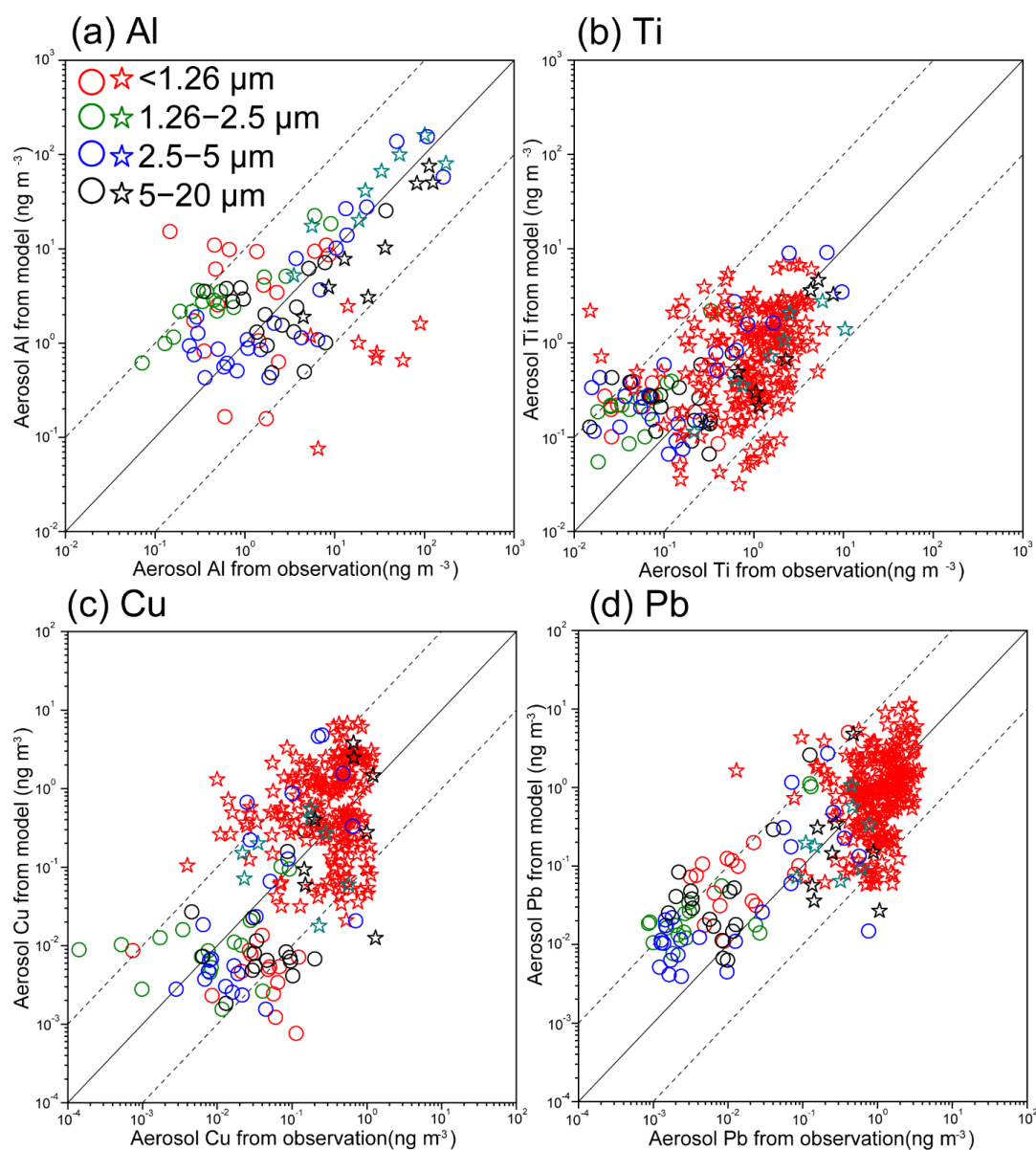


305 observations partly owing to the size-segregated $\delta^{56}\text{Fe}$ for the transport sector and crystalline Fe-
 306 containing particles in the model.



307
 308 **Figure 6.** Comparison of total and bioaccessible $\delta^{56}\text{Fe}$ (‰) for size-segregated aerosols between
 309 model estimates and observations. Data points are colored by aerosol size bin (red: <1.26 μm ;
 310 green: 1.26–2.5 μm ; blue: 2.5–5 μm ; black: 5–20 μm). Summary of model statistics is available
 311 in Table S5.

312 We compared aluminum (Al), titanium (Ti), copper (Cu) and lead (Pb) concentrations for size-
 313 segregated aerosols between model estimates and observations for KH2017 (circle) and MR2021
 314 (star) (Fig. 7 and Table S6). The model generally reproduced crustal tracers (Al and Ti) and
 315 anthropogenic tracers (Cu and Pb) over the North Pacific.



316

317 **Figure 7.** Comparison of Al, Ti, Cu and Pb concentrations between model estimates and
 318 observations. Data points are colored by aerosol size bin (red: $<1.26 \mu\text{m}$; green: $1.26\text{--}2.5 \mu\text{m}$;
 319 blue: $2.5\text{--}5 \mu\text{m}$; black: $5\text{--}20 \mu\text{m}$). Total Ti, Cu and Pb concentrations of fine-mode aerosols are
 320 measured using PX-375, whereas aerosol concentrations for each bin are estimated by assuming a



log-normal distribution within each size stage for samples analyzed using ICPMS. Summary of model statistics is available in Table S6.

3.4 Contribution of anthropogenic processing activity and fuel combustion sources

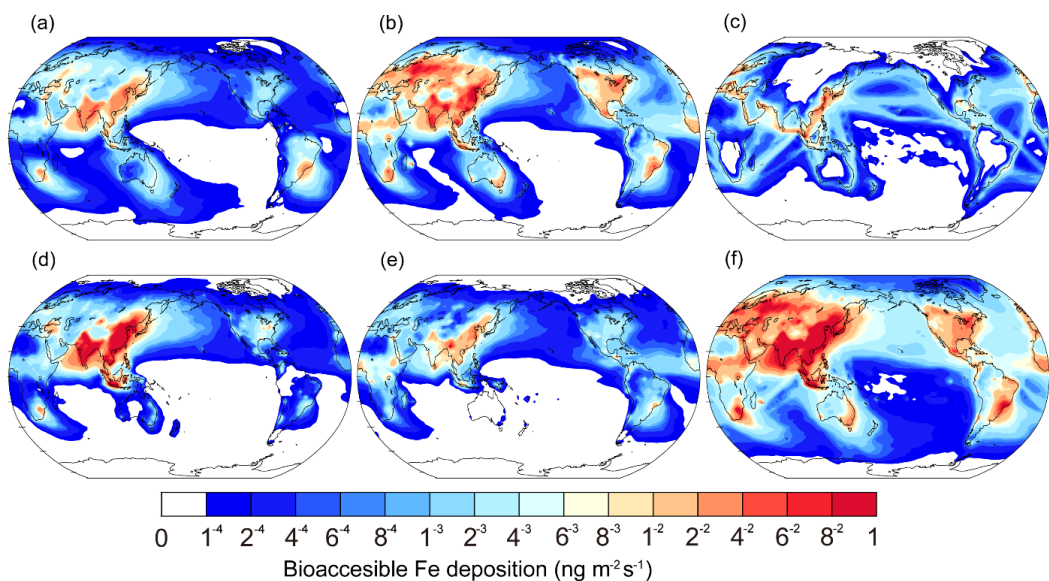


Figure 8. Deposition fluxes of bioaccessible Fe ($\text{ng Fe m}^{-2} \text{s}^{-1}$) from (a) metal smelting (10 Gg Fe yr^{-1}), (b) ground transportation (11 Gg Fe yr^{-1}), (c) shipping transportation (11 Gg Fe yr^{-1}), (d) energy/industry/RCO (excluding biofuel and waste) (13 Gg Fe yr^{-1}), (e) biofuel/waste combustion (5 Gg Fe yr^{-1}), and (f) the sum of the anthropogenic sources (50 Gg Fe yr^{-1}). The parentheses represent bioaccessible Fe deposition to the global ocean in 2022. Summary of model statistics is available in Table S7.

Of the five individual sectors, energy/industry/RCO combustion (13 Gg Fe yr^{-1}) delivers the largest BFe deposition to the global ocean, closely followed by ground transportation (11 Gg Fe yr^{-1}), shipping transportation (11 Gg Fe yr^{-1}) and metal smelting (10 Gg Fe yr^{-1}), while



334 biofuel/waste combustion (5 Gg Fe yr^{-1}) contributes the least (Fig. 8, Table S7). BFe from shipping
335 transportation is directly emitted over the surface ocean. Moreover, BFe even in the coarse mode
336 (percentage of the coarse mode in total Fe emissions: 98% in Table S2 vs. 14% in Ito, 2013) is
337 mostly supplied along the major oceanic shipping lanes. Thus, this sector extends anthropogenic
338 Fe deposition farther into the open ocean than the emission sources on land. On the other hand,
339 BFe in coarse particles from the other four sectors is mostly deposited on land and is partly
340 delivered to the ocean in the downstream region of the major combustion and industrial sources.

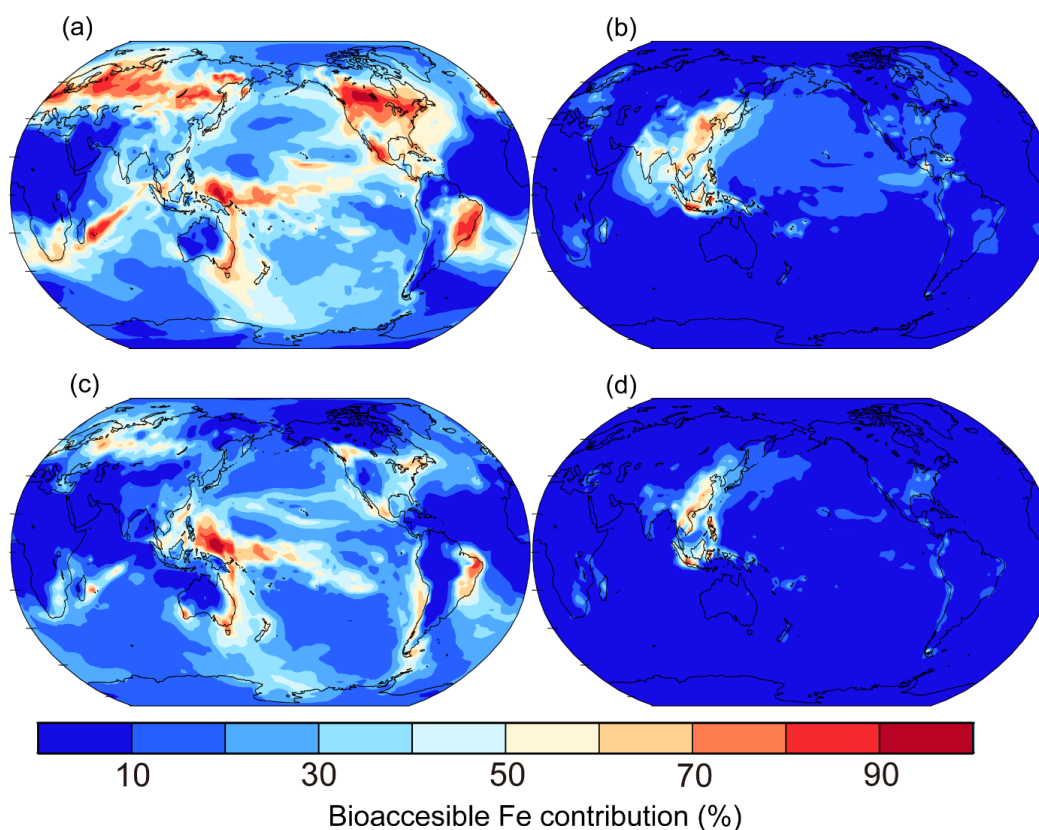
341 Metal smelting produces markedly higher Fe bioaccessibility (41%: the average Fe
342 bioaccessibility of deposition to the oceans) than the other sectors after atmospheric aging,
343 primarily because of the high content of magnetite (90%) and its faster dissolution compared with
344 crystalline Fe oxide and the stable aluminosilicate minerals (Ito and Miyakawa, 2023) (Fig. S1).
345 On the other hand, the other four sectors show much lower Fe bioaccessibility over the open ocean.
346 Among these lower-bioaccessibility sectors, shipping transportation (25%) has a higher value than
347 ground transportation (16%), biofuel/waste combustion (14%), and energy/industry/RCO
348 combustion (11%), primarily because of the high content of Fe sulfate (72% for heavy oil vs. 16%
349 for diesel oil) and its faster dissolution. The spatial pattern of the summed anthropogenic sources
350 (17%) closely resembles that of the transportation and combustion sectors rather than that of metal
351 smelting, reflecting the comparable deposition of metal smelting relative to the other sectors
352 despite its higher Fe bioaccessibility (Fig. 8).

353 Consequently, BFe deposition from anthropogenic processing activity (32 Gg Fe yr^{-1}) is nearly
354 double that of fuel combustion (18 Gg Fe yr^{-1}) in 2022, although the global TFe emissions from
355 anthropogenic processing activity ($936 \text{ Gg Fe yr}^{-1}$) is smaller than that from fuel combustion (1401



356 Gg Fe yr⁻¹). The North Pacific and Indian Oceans predominantly receive atmospheric BFe from
357 anthropogenic processing activity (20 Gg Fe yr⁻¹) and fuel combustion (15 Gg Fe yr⁻¹) compared
358 with the other oceans combined (12 and 3 Gg Fe yr⁻¹, respectively).

359 Despite TFe deposition comparable to or lower than the previous estimate (Ito et al., 2018) in
360 most ocean basins, our BFe deposition is generally higher, most notably in the North Pacific (0.048
361 vs. 0.021 Tg Fe yr⁻¹) and the Indian Ocean (0.106 vs. 0.09 Tg Fe yr⁻¹) (Table S7). The largest
362 basin-level discrepancy in TFe occurs in the North Atlantic, where our estimate (11.9 Tg Fe yr⁻¹)
363 is more than double the ensemble estimate (5.07 Tg Fe yr⁻¹), partly because of larger particles in
364 the IMPACT model than in the other models (Myriokefalitakis et al., 2018). At the global scale,
365 these estimates of TFe and BFe deposition fall within a relatively narrow range (15–27 Tg Fe yr⁻¹
366 and 0.24–0.27 Tg Fe yr⁻¹, respectively) (Table S7).



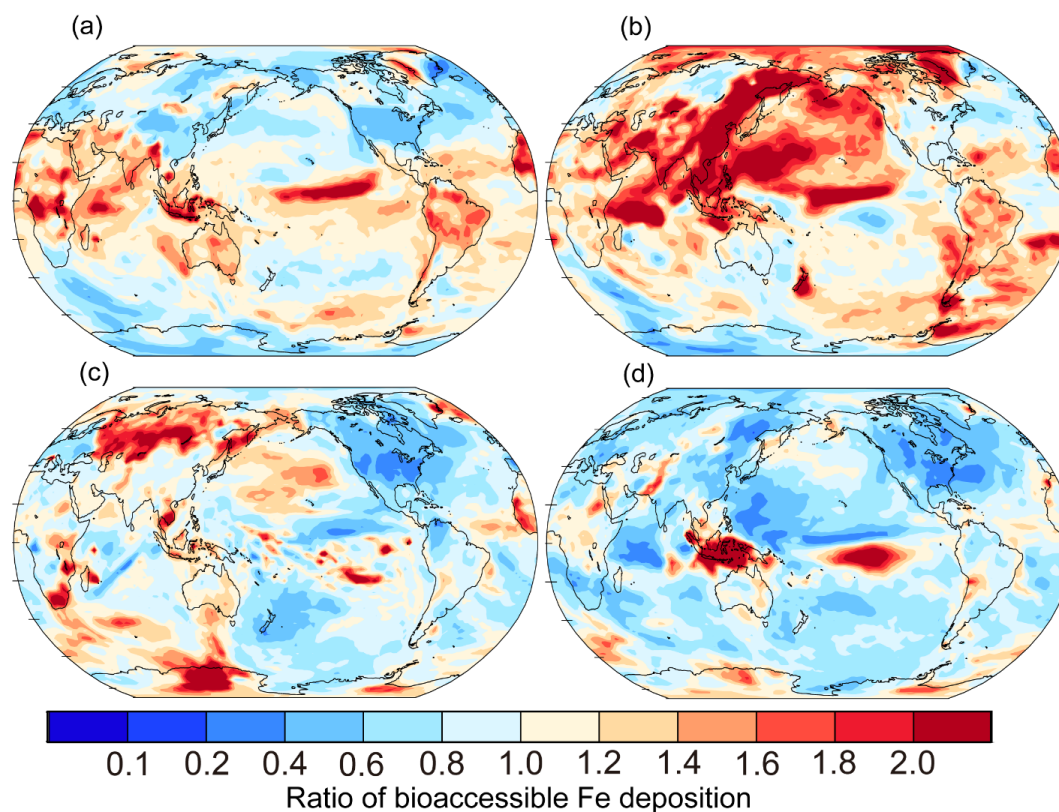
367

368 **Figure 9.** Seasonal percentage contribution of bioaccessible Fe deposition fluxes from (a)
 369 anthropogenic processing activity sources (Experiment 1, Experiment 2, Experiment 3) during
 370 December–February (DJF) (33%), (b) fuel combustion sources (Experiment 4 and Experiment 5)
 371 during DJF (27%), (c) anthropogenic processing activity sources during June–August (JJA) (8%),
 372 and (d) fuel combustion sources during JJA (5%) to total sources (%) in 2022. The parentheses
 373 represent the contribution of bioaccessible Fe deposition fluxes to the sum of the North Pacific and
 374 Indian Oceans.

375 Anthropogenic processing activity sources contribute roughly 30–50% of the sum of BFe
 376 deposition from anthropogenic, lithogenic, and pyrogenic sources extending across much of the



377 North Pacific from the East Asian coast toward the central ocean, together with an elevated band
 378 across the tropical Indian Ocean and the Maritime Continent during DJF (Fig. 9). This broad
 379 pattern weakens substantially in JJA, whereas isolated patches of high contribution remain near
 380 the Maritime Continent where the lithogenic and pyrogenic contributions are low. Fuel combustion
 381 sources, in contrast, are tightly confined to the Maritime Continent (Indonesia and Malaysia),
 382 contributing comparatively little to the broader North Pacific and northern Indian Oceans in the
 383 downstream region of Asian outflow.



384
 385 **Figure 10.** Ratio of bioaccessible Fe deposition fluxes from (a) anthropogenic processing activity
 386 sources from 2002 to 2012 (1.1), (b) fuel combustion sources from 2002 to 2012 (1.6), (c)
 387 anthropogenic processing activity sources from 2012 to 2022 (1.0), and (d) fuel combustion



388 sources from 2012 to 2022 (0.7). The parentheses represent ratios of bioaccessible Fe deposition
 389 fluxes to the sum of the North Pacific and Indian Oceans.

390 BFe deposition from anthropogenic processing activities shows a small change from 2002 to 2012
 391 (ratio 1.1 vs. ratio 1.6 for processing activities vs. fuel combustion), with increases over the
 392 northern Indian Ocean (Fig. 10). Over the same period, fuel combustion shows an intense increase
 393 across nearly the entire North Pacific from the East Asian coast to the open ocean, the northern
 394 Indian Ocean, and the Maritime Continent. From 2012 to 2022, this pattern largely reverses for
 395 combustion sources (ratio around 0.6–0.8), while a strong localized increase persists over the
 396 Maritime Continent. Over the same period, anthropogenic processing activity sources show little
 397 net change on average for the region (ratio 1.0), reflecting a partial offset between an increase in
 398 the central North Pacific, the Bering Sea, and the southern Indian Ocean, and a decrease in the
 399 eastern North Pacific near North America and shipping lanes in the Indian Ocean. Overall, fuel
 400 combustion drives the sharp rise in BFe deposition to the North Pacific and Indian Oceans up to
 401 2012 and its subsequent decline, while anthropogenic processing activities contribute a
 402 comparatively small and spatially variable signal.

403 **4. Implications of bioaccessible Fe deposition from human activities**

404 To estimate the contribution of anthropogenic processing activity and fuel combustion sources
 405 to BFe deposition, we used the IMPACT model that could reproduce the ship-based measurements
 406 of trace metal concentrations, Fe bioaccessibility, and $\delta^{56}\text{Fe}$. The model reproduced the lighter
 407 $\delta^{56}\text{Fe}$ (-0.48 ± 0.32 ‰ vs. -0.39 ± 0.49 ‰ for TFe and -0.52 ± 0.36 ‰ vs. -0.35 ± 0.53 ‰ for
 408 BFe) in the finer particles (<1.26 μm in diameter) compared with the coarser particles ($0.05 \pm$
 409 0.15 ‰ vs. 0.04 ± 0.13 ‰ for TFe and 0.00 ± 0.21 ‰ vs. -0.01 ± 0.23 for BFe) over the North
 410 Pacific. The model estimates of anthropogenic BFe deposition in the North Pacific and Indian



Oceans indicate an increase from 2002 (30 Gg Fe yr⁻¹) to 2022 (35 Gg Fe yr⁻¹). On the other hand, the model displays a decrease in anthropogenic BFe deposition in the sum of the other oceans from 2002 (19 Gg Fe yr⁻¹) to 2022 (17 Gg Fe yr⁻¹). Specifically, the model estimates of anthropogenic BFe deposition from fuel combustion in the North Pacific and Indian Oceans showed a rapid increase from 2002 (12 Gg Fe yr⁻¹) to 2012 (20 Gg Fe yr⁻¹) and a decrease to 2022 (15 Gg Fe yr⁻¹). In contrast, the model for anthropogenic processing activity sources revealed a persistent increase in anthropogenic BFe deposition in the North Pacific and Indian Oceans in 2002 (18 Gg Fe yr⁻¹), 2012 (19 Gg Fe yr⁻¹), and 2022 (20 Gg Fe yr⁻¹). Further, the model results suggested that the contribution of the anthropogenic processing activity sources to BFe deposition in the North Pacific and Indian Oceans was pronounced in winter (austral summer) (33% in 2022) when the lithogenic and pyrogenic contributions were low, compared to summer (8% in 2022). These findings may have important implications for projections of future micronutrient supply to the oceans in the context of a warming climate, which is projected to decrease the internal supply of nutrients to the surface ocean. Therefore, this study calls for further studies to elucidate the role of BFe from anthropogenic processing activity, develop Fe dissolution parameterizations that account for atmospheric aging, and revisit the measurements of Fe isotopes for size-segregated aerosols. This is especially crucial for assessing the impact of air quality on climate, marine ecosystems, and human health and for exploring effective pollution control strategies. A future Model Intercomparison Project of Atmospheric Nutrient Deposition (MIPAND) is planned, in which we will share the anthropogenic emission database developed in this study and provide marine biogeochemical models with deposition data from atmospheric models in conjunction with field observations.



433 **Data availability**

434 Observational data are available online (<https://doi.org/10.17632/p566r8vp7c.1>,
435 <https://www.jamstec.go.jp/egcr/j/atmos/observation/topics1.html>,
436 <https://doi.org/10.5281/zenodo.19471817>, <https://doi.org/10.17632/p566r8vp7c.1>). Additional
437 data related to this paper may be requested from the authors.

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440 **Author contributions**

441 AI conceived the modeling study and analyzed the observational data. KM contributed measured
442 data. The manuscript was drafted by AI and commented on by KM.

443 **Competing interests**

444 Authors declare that they have no competing interests.

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456 **References**

457 Baldo, C., Ito, A., Krom, M. D., Li, W., Jones, T., Drake, N., Ignatyev, K., Davidson, N., and Shi,
458 Z.: Iron from coal combustion particles dissolves much faster than mineral dust under simulated
459 atmospheric acidic conditions, *Atmos. Chem. Phys.*, 22, 6045–6066, [https://doi.org/10.5194/acp-](https://doi.org/10.5194/acp-22-6045-2022)
460 22-6045-2022, 2022.

461 Bergas-Massó, E., Gonçalves-Ageitos, M., Myriokefalitakis, S., Miller, R. L., van Noije, T., Le
462 Sager, P., Montané Pinto, G., and Pérez García-Pando, C.: Pre-industrial, present and future



- 463 atmospheric soluble iron deposition and the role of aerosol acidity and oxalate under CMIP6
464 emissions, *Earth's Future*, 11, e2022EF003353, <https://doi.org/10.1029/2022EF003353>, 2023.
- 465 Boyd, P. W., Arrigo, K. R., Ardyna, M., Halfter, S., Huckstadt, L., Kuhn, A. M., Lannuzel, D.,
466 Neukermans, G., Novaglio, C., Shadwick, E. H., Swart, S., and Thomalla, S. J.: The role of biota
467 in the Southern Ocean carbon cycle, *Nat. Rev. Earth Environ.*, 5, 390–408,
468 <https://doi.org/10.1038/s43017-024-00531-3>, 2024.
- 469 Bunnell, Z. B., Sieber, M., Hamilton, D. S., Marsay, C. M., Buck, C. S., Landing, W. M., John, S.
470 G., and Conway, T. M.: The influence of natural, anthropogenic, and wildfire sources on iron and
471 zinc aerosols delivered to the North Pacific Ocean, *Geophys. Res. Lett.*, 52, e2024GL113877,
472 <https://doi.org/10.1029/2024GL113877>, 2025.
- 473 Campbell, S. J., Uttinger, B., Barth, A., Leni, Z., Zhang, Z.-H., Resch, J., Li, K., Steimer, S. S.,
474 Banach, C., Gfeller, B., Wragg, F. P. H., Westwood, J., Wolfer, K., Bukowiecki, N., Ihalainen, M.,
475 Yli-Pirilä, P., Somero, M., Kortelainen, M., Louhisalmi, J., Sklorz, M., Czech, H., di Bucchianico,
476 S., Streibel, T., Delaval, M. N., Rüger, C., Bäumlin, N., Salathé, M., Fang, Z., Pardo, M., D'Aronco,
477 S., Giorio, C., Shi, Z., Harrison, R. M., Green, D. C., Kelly, F. J., Rudich, Y., Paulson, S. E.,
478 Sippula, O., Zimmermann, R., Geiser, M., and Kalberer, M.: Short-lived reactive components
479 substantially contribute to particulate matter oxidative potential, *Sci. Adv.*, 11, eadp8100,
480 <https://doi.org/10.1126/sciadv.adp8100>, 2025.
- 481 Chen, H., Laskin, A., Baltrusaitis, J., Gorski, C. A., Scherer, M. M., and Grassian, V. H.: Coal fly
482 ash as a source of iron in atmospheric dust, *Environ. Sci. Technol.*, 46, 2112–
483 2120, <https://doi.org/10.1021/es204102f>, 2012.
- 484 Cui, W., Song, X., Su, Y., Chen, X., Wu, D., and Li, Q.: Soluble iron in source-based
485 anthropogenic PM_{2.5} predominantly from steel industry and residential combustion in China,
486 *Geophys. Res. Lett.*, 52, e2025GL118603, <https://doi.org/10.1029/2025GL118603>, 2025.
- 487 Dubois, N., Foret, G., Siour, G., Uzu, G., Couvidat, F., Vida, M., André, J.-M., Moukhtar, S.,
488 Sirina-Leboine, N., Aujay Plouzeau, R., Alleman, L., Conil, S., Pallares, C., Salque Moreton, G.,
489 Stratigou, E., Alastuey, A., Querol, X., Ito, A., Jaffrezo, J.-L., Favez, O., and Beekmann, M.:
490 Construction of copper, iron and manganese anthropogenic emission inventories for Europe,
491 *Atmos. Environ.*, 363, 121582, <https://doi.org/10.1016/j.atmosenv.2025.121582>, 2025.
- 492 Gelaro, R., McCarty, W., Suárez, M. J., Todling, R., Molod, A., Takacs, L., Randles, C. A.,
493 Darmenov, A., Bosilovich, M. G., Reichle, R., Wargan, K., Coy, L., Cullather, R., Draper, C.,
494 Akella, S., Buchard, V., Conaty, A., da Silva, A. M., Gu, W., Kim, G.-K., Koster, R., Lucchesi,
495 R., Merkova, D., Nielsen, J. E., Partyka, G., Pawson, S., Putman, W., Rienecker, M., Schubert, S.
496 D., Sienkiewicz, M., and Zhao, B.: The Modern-Era Retrospective Analysis for Research and
497 Applications, Version 2 (MERRA-2), *J. Climate*, 30, 5419–5454, <https://doi.org/10.1175/JCLI-D-16-0758.1>, 2017.
- 499 Hamilton, D. S., Scanza, R. A., Rathod, S. D., Bond, T. C., Kok, J. F., Li, L., Matsui, H., and
500 Mahowald, N. M.: Recent (1980 to 2015) trends and variability in daily-to-interannual soluble iron
501 deposition from dust, fire, and anthropogenic sources, *Geophys. Res. Lett.*, 47, e2020GL089688,
502 <https://doi.org/10.1029/2020GL089688>, 2020.



- 503 Hawco, N. J., Conway, T. M., Coesel, S. N., Barone, B., Seelen, E. A., Yang, S.-C., Bundy, R. M.,
504 Pinedo-Gonzalez, P., Bian, X., Sieber, M., Lanning, N. T., Fitzsimmons, J. N., Foreman, R. K.,
505 König, D., Groussman, M. J., Allen, J. G., Juranek, L. W., White, A. E., Karl, D. M., Armbrust, E.
506 V., and John, S. G.: Anthropogenic iron alters the spring phytoplankton bloom in the North Pacific
507 transition zone, *Proc. Natl. Acad. Sci. USA*, 122, e2418201122,
508 <https://doi.org/10.1073/pnas.2418201122>, 2025.
- 509 He, Y., Kadko, D., Stephens, M. P., Sheridan, M. T., Buck, C. S., Marsay, C. M., Landing, W. M.,
510 Zheng, M., and Liu, P.: Constraining aerosol deposition over the global ocean, *Nat. Geosci.*, 18,
511 966–974, <https://doi.org/10.1038/s41561-025-01785-2>, 2025.
- 512 Hoesly, R. M., Smith, S. J., Ahsan, H., Prime, N., O'Rourke, P., Crippa, M., Klimont, Z., Guizzardi,
513 D., Feng, L., Harkins, C., McDonald, B. C., and Wang, S.: CEDS v_2025_04_18 Gridded Data
514 0.1 degree [data set], Zenodo, <https://doi.org/10.5281/zenodo.15001545>, 2025.
- 515 Hsieh, C.-C. and Ho, T.-Y.: Contribution of anthropogenic and lithogenic aerosol Fe in the East
516 China Sea, *J. Geophys. Res.-Oceans*, 129, e2024JC021113,
517 <https://doi.org/10.1029/2024JC021113>, 2024.
- 518 Hsieh, C.-C., You, C.-F., and Ho, T.-Y.: The solubility and deposition flux of East Asian aerosol
519 metals in the East China Sea: the effects of aeolian transport processes, *Mar. Chem.*, 253, 104268,
520 <https://doi.org/10.1016/j.marchem.2023.104268>, 2023.
- 521 Ito, A.: Global modeling study of potentially bioavailable iron input from shipboard aerosol
522 sources to the ocean, *Global Biogeochem. Cy.*, 27, 1–10, <https://doi.org/10.1029/2012GB004378>,
523 2013.
- 524 Ito, A., Kawana, K., Miyakawa, T., Matsumoto, K., and Kanaya, Y.: Contribution of crystalline
525 iron-containing particles and bioaerosols to ice-nucleating particles over the North Pacific, *Prog.*
526 *Earth Planet. Sci.*, 13, 47, <https://doi.org/10.1186/s40645-026-00838-3>, 2026.
- 527 Ito, A., Lin, G., and Penner, J. E.: Radiative forcing by light-absorbing aerosols of pyrogenetic
528 iron oxides, *Sci. Rep.-UK*, 8, 7347, <https://doi.org/10.1038/s41598-018-25756-3>, 2018.
- 529 Ito, A. and Miyakawa, T.: Aerosol iron from metal production as a secondary source of
530 bioaccessible iron, *Environ. Sci. Technol.*, 57, 4091–4100,
531 <https://doi.org/10.1021/acs.est.2c06472>, 2023.
- 532 Ito, A., Miyazaki, Y., Taketani, F., Iwamoto, Y., and Kanaya, Y.: Marine aerosol feedback on
533 biogeochemical cycles and the climate in the Anthropocene: lessons learned from the Pacific
534 Ocean, *Environ. Sci.: Atmos.*, 3, 782–798, <https://doi.org/10.1039/D2EA00156J>, 2023.
- 535 Ito, A., Myriokefalitakis, S., Kanakidou, M., Mahowald, N. M., Scanza, R. A., Hamilton, D. S.,
536 Baker, A. R., Jickells, T., Sarin, M., Bikkina, S., Gao, Y., Shelley, R. U., Buck, C. S., Landing, W.
537 M., Bowie, A. R., Perron, M. M. G., Guieu, C., Meskhidze, N., Johnson, M. S., Feng, Y., Kok, J.
538 F., Nenes, A., and Duce, R. A.: Pyrogenic iron: The missing link to high iron solubility in aerosols,
539 *Sci. Adv.*, 5, eaau7671, <https://doi.org/10.1126/sciadv.aau7671>, 2019.



- 540 Ito, A. and Shi, Z.: Delivery of anthropogenic bioavailable iron from mineral dust and combustion
541 aerosols to the ocean, *Atmos. Chem. Phys.*, 16, 85–99, <https://doi.org/10.5194/acp-16-85-2016>,
542 2016.
- 543 Ito, A., Ye, Y., Baldo, C., and Shi, Z.: Ocean fertilization by pyrogenic aerosol iron, *npj Clim.*
544 *Atmos. Sci.*, 4, 30, <https://doi.org/10.1038/s41612-021-00185-8>, 2021.
- 545 Kanaya, Y., Yamaji, K., Miyakawa, T., Taketani, F., Zhu, C., Choi, Y., Komazaki, Y., Ikeda, K.,
546 Kondo, Y., and Klimont, Z.: Rapid reduction in black carbon emissions from China: evidence from
547 2009–2019 observations on Fukue Island, Japan, *Atmos. Chem. Phys.*, 20, 6339–6356,
548 <https://doi.org/10.5194/acp-20-6339-2020>, 2020.
- 549 Kumar, A., Susann, T., Fernandes, O. S., Biswas, H., Girach, I., Roxy, M. K., Kurian, S.,
550 Marandino, C. A., Sarma, V. V. S. S., and Shenoy, D. M.: Surface ocean-lower atmospheric
551 processes in the Indian Ocean: Current understanding, knowledge gaps, and future directions,
552 *Elem. Sci. Anth.*, 12, 00041, <https://doi.org/10.1525/elementa.2023.00041>, 2024.
- 553 Kurisu, M., Nishioka, J., Sugie, K., Murayama, A., and Obata, H.: Seasonal changes in iron sources
554 to the surface ocean in the subarctic western North Pacific revealed by iron isotopes, Abstract
555 OS26-A011, AOGS2026, 2026a.
- 556 Kurisu, M., Sakata, K., Miyamoto, C., Takaku, Y., Iizuka, T., and Takahashi, Y.: Variation of iron
557 isotope ratios in anthropogenic materials emitted through combustion processes, *Chem. Lett.*, 45,
558 970–972, <https://doi.org/10.1246/cl.160451>, 2016.
- 559 Kurisu, M., Sakata, K., Uematsu, M., Ito, A., and Takahashi, Y.: Contribution of combustion Fe
560 in marine aerosols over the northwestern Pacific estimated by Fe stable isotope ratios, *Atmos.*
561 *Chem. Phys.*, 21, 16027–16050, <https://doi.org/10.5194/acp-21-16027-2021>, 2021.
- 562 Kurisu, M., Sakata, K., Nishioka, J., Obata, H., Conway, T. M., Hunt, H. R., Sieber, M., Suzuki,
563 K., Kashiwabara, T., Kubo, S., Takada, M., and Takahashi, Y.: Source and fate of atmospheric
564 iron supplied to the subarctic North Pacific traced by stable iron isotope ratios, *Geochim.*
565 *Cosmochim. Ac.*, 378, 168–185, <https://doi.org/10.1016/j.gca.2024.06.009>, 2024.
- 566 Kurisu, M. and Takahashi, Y.: Testing iron stable isotope ratios as a signature of biomass burning,
567 *Atmosphere*, 10, 76, <https://doi.org/10.3390/atmos10020076>, 2019.
- 568 Kurisu, M., Zhu, C., Miyakawa, T., Ito, A., Suzuki, K., Kashiwabara, T., Kubo, S., Takahashi, Y.,
569 Harada, N., Foret, G., Dubois, N., and Kanaya, Y.: Identification of anthropogenic Fe originated
570 from East Asia using Fe stable isotope ratios of aerosols collected on Fukue Island, *Atmos.*
571 *Environ.*, 373, 121893, <https://doi.org/10.1016/j.atmosenv.2026.121893>, 2026b.
- 572 Li, R., Plaas, H. E., Zhang, Y., Chen, Y., Zhang, T., Yang, Y., Rathod, S., Zhang, G., Wang, X.,
573 Hamilton, D. S., and Tang, M.: Residential burning is a potentially significant source of soluble
574 iron to the ocean, *Atmos. Chem. Phys.*, 26, 9037–9060, <https://doi.org/10.5194/acp-26-9037-2026>,
575 2026.



- 576 Luo, C., Mahowald, N., Bond, T., Chuang, P. Y., Artaxo, P., Siefert, R., Chen, Y., and Schauer,
577 J.: Combustion iron distribution and deposition, *Global Biogeochem. Cy.*, 22, GB1012,
578 <https://doi.org/10.1029/2007GB002964>, 2008.
- 579 Miyakawa, T., Ito, A., Zhu, C., Shimizu, A., Matsumoto, E., Mizuno, Y., and Kanaya, Y.: Trace
580 elements in PM_{2.5} aerosols in East Asian outflow in the spring of 2018: emission, transport, and
581 source apportionment, *Atmos. Chem. Phys.*, 23, 14609–14626, [https://doi.org/10.5194/acp-23-](https://doi.org/10.5194/acp-23-14609-2023)
582 14609-2023, 2023.
- 583 Miyakawa, T., Kinase, T., Kurisu, M., Taniguchi, K., Katsumata, M., Takashima, H., Taketani, F.,
584 and Kanaya, Y.: Natural and continental influence on aerosol distributions over the Northwestern
585 Pacific Ocean in the late winter and early spring of 2021, *Prog. Earth Planet. Sci.*, 12, 77,
586 <https://doi.org/10.1186/s40645-025-00752-0>, 2025.
- 587 Miyakawa, T. and Kinase, T.: Chemical/elemental compositions and particle type classifications
588 of fine mode aerosols over the Northwestern Pacific Ocean [data set], Zenodo,
589 <https://doi.org/10.5281/zenodo.19471817>, 2026.
- 590 Miyakawa, T., Zhu, C., Yokokawa, M., Mukawa, M., Ito, A., Shimizu, A., Nishizawa, T., and
591 Kanaya, Y.: Size-resolved characterizations of Fe in aerosols in East Asian outflow in winter and
592 spring: Source apportionment and bioaccessibility, *EGUsphere* [preprint],
593 <https://doi.org/10.5194/egusphere-2026-3778>, 2026.
- 594 Moteki, N.: Climate-relevant properties of black carbon aerosols revealed by in situ measurements:
595 a review, *Prog. Earth Planet. Sci.*, 10, 12, <https://doi.org/10.1186/s40645-023-00544-4>, 2023.
- 596 Myriokefalitakis, S., Ito, A., Kanakidou, M., Nenes, A., Krol, M. C., Mahowald, N. M., Scanza,
597 R. A., Hamilton, D. S., Johnson, M. S., Meskhidze, N., Kok, J. F., Guieu, C., Baker, A. R., Jickells,
598 T. D., Sarin, M. M., Bikkina, S., Shelley, R., Bowie, A., Perron, M. M. G., and Duce, R. A.:
599 Reviews and syntheses: the GESAMP atmospheric iron deposition model intercomparison study,
600 *Biogeosciences*, 15, 6659–6684, <https://doi.org/10.5194/bg-15-6659-2018>, 2018.
- 601 Pham, A. L. D. and Ito, T.: Anthropogenic iron deposition alters the ecosystem and carbon balance
602 of the Indian Ocean over a centennial timescale, *J. Geophys. Res.-Oceans*, 126, e2020JC016475,
603 <https://doi.org/10.1029/2020JC016475>, 2021.
- 604 Pinedo-González, P., Hawco, N. J., Bundy, R. M., Armbrust, E. V., Follows, M. J., Cael, B. B.,
605 White, A. E., Ferrón, S., Karl, D. M., and John, S. G.: Anthropogenic Asian aerosols provide Fe
606 to the North Pacific Ocean, *Proc. Natl. Acad. Sci. USA*, 117, 27862–27868,
607 <https://doi.org/10.1073/pnas.2010315117>, 2020.
- 608 Rathod, S. D., Hamilton, D. S., Mahowald, N. M., Klimont, Z., Corbett, J. J., and Bond, T. C.: A
609 mineralogy-based anthropogenic combustion-iron emission inventory, *J. Geophys. Res.-Atmos.*,
610 125, e2019JD032114, <https://doi.org/10.1029/2019JD032114>, 2020.
- 611 Rathod, S. D., Hamilton, D. S., Li, L., Mahowald, N. M., Matsui, H., Pierce, J. R., and Bond, T.
612 C.: Atmospheric radiative and oceanic biological productivity responses to increasing
613 anthropogenic combustion-iron emission in the 1850–2010 period, *Geophys. Res. Lett.*, 49,
614 e2022GL099323, <https://doi.org/10.1029/2022GL099323>, 2022.



- 615 Seok, M.-W., Li, L., Doney, S. C., Kim, T.-W., and Mahowald, N. M.: Atmospheric soluble iron
616 sustains nitrogen fixation in oligotrophic oceans, *Commun. Earth Environ.*, 7, 656,
617 <https://doi.org/10.1038/s43247-026-03657-y>, 2026.
- 618 Wei, T., Dong, Z., Zong, C., Liu, X., Kang, S., Yan, Y., and Ren, J.: Global-scale constraints on
619 the origins of aerosol iron using stable iron isotopes: A review, *Earth-Sci. Rev.*, 258, 104943,
620 <https://doi.org/10.1016/j.earscirev.2024.104943>, 2024.
- 621 Yoon, J.-E., Yoo, K.-C., Macdonald, A. M., Yoon, H.-I., Park, K.-T., Yang, E. J., Kim, H.-C., Lee,
622 J. I., Lee, M. K., Jung, J., Park, J., Lee, J., Kim, S., Kim, S.-S., Kim, K., and Kim, I.-N.: Reviews
623 and syntheses: Ocean iron fertilization experiments – past, present, and future looking to a future
624 Korean Iron Fertilization Experiment in the Southern Ocean (KIFES) project, *Biogeosciences*, 15,
625 5847–5889, <https://doi.org/10.5194/bg-15-5847-2018>, 2018.
- 626 Zhang, T., Liu, J., Xiang, Y., Liu, X., Zhang, J., Zhang, L., Ying, Q., Wang, Y., Wang, Y., Chen,
627 S., Chai, F., and Zheng, M.: Quantifying anthropogenic emission of iron in marine aerosol in the
628 Northwest Pacific with shipborne online measurements, *Sci. Total Environ.*, 912, 169158,
629 <https://doi.org/10.1016/j.scitotenv.2023.169158>, 2024.
- 630 Zhang, T., Xiang, Y., Liu, Y., Yan, C., Zhu, B., Liu, X., Fan, X., Wang, Y., Zhang, H., Wang, Y.,
631 Chen, S., Chai, F., Zhu, T., and Zheng, M.: Source apportionment of atmospheric iron in northern
632 China based on 3-year continuous measurements, *Geophys. Res. Lett.*, 52, e2024GL114462,
633 <https://doi.org/10.1029/2024GL114462>, 2025a.
- 634 Zhang, Y., Li, R., Bunnell, Z. B., Chen, Y., Zhu, G., Ma, J., Zhang, G., Conway, T. M., and Tang,
635 M.: A critical review of the use of iron isotopes in atmospheric aerosol research, *Atmos. Chem.*
636 *Phys.*, 25, 11067–11086, <https://doi.org/10.5194/acp-25-11067-2025>, 2025b.