

1 **Residential burning is a **potentially** significant source of soluble iron to the**
2 **ocean**

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32

33 **Abstract**

34 Understanding the physicochemical processes that supply atmospheric aerosol iron (Fe) to the
35 ocean is crucial for understanding of global biogeochemical cycles. Anthropogenic activity
36 contributes significant fluxes of aerosol Fe to the atmosphere, the soluble fraction of which can
37 modulate marine primary productivity upon its deposition to the ocean surface. However,
38 anthropogenic aerosol Fe solubility remains poorly constrained, due in part to a lack of direct
39 measurements spanning a multitude of anthropogenic sources. We measured solubility of
40 aerosol Fe from several distinct anthropogenic combustion processes and fuel types. The
41 median Fe solubility varied widely by source, ranging from 0.03% for power plant coal fly ash
42 to 55.87% for biofuel burning; furthermore, residential coal burning aerosol possessed much
43 higher Fe solubility than power plant coal fly ash. Using the new Fe solubilities reported herein,
44 we updated parameters for anthropogenic aerosol Fe within the Community Earth System
45 Model. Anthropogenic combustion is estimated to contribute up to 20% of the global soluble
46 Fe flux to the ocean in the present day. Furthermore, we identified residential coal burning as
47 a previously neglected but potentially important source with regional flux contributions
48 ranging from <1% to 21%. Our work underscores the need to further refine understanding of
49 aerosol Fe properties from a wide variety of anthropogenic sources by increasing observations
50 in more novel aerosol regimes, with a focus on residential coal burning. This understanding
51 will in turn aid in characterizing the influences of anthropogenic activity on past, present, and
52 future atmospheric nutrient inputs to marine ecosystems.

53

54 **1 Introduction**

55 Anthropogenic activities have altered the atmospheric burden and deposition fluxes of
56 biogeochemically relevant trace metals, including iron (Fe) (Bergas-Massó et al., 2023;
57 Hamilton et al., 2020b). **The quantity of** Fe in ocean waters plays a particularly important role
58 in modulating the spatiotemporal distribution of primary productivity in ocean ecosystems,
59 which has downstream impacts on marine fisheries and carbon sequestration (Ito et al., 2021;
60 Tagliabue et al., 2014; Tagliabue et al., 2017). Energy-production, transportation, shipping,
61 and manufacturing (e.g., steel production) are all characterized sources of anthropogenic
62 aerosol Fe (Ito and Miyakawa, 2023; Ito and Shi, 2016; Rathod et al., 2024). These differing
63 combustion fuel types possess distinct physicochemical properties that influence their impact
64 on radiative forcing and nutrient supply (Al-Abadleh et al., 2023; Ito et al., 2018; Matsui et al.,
65 2018; Rathod et al., 2020).

66 To assess the potential nutritional impact of atmospheric Fe deposition on ocean
67 ecosystems, atmospheric aerosol research primarily focuses on tracing the soluble Fe content
68 in aerosol (Baker et al., 2020; Ito et al., 2019; Mahowald et al., 2018). Soluble Fe content is
69 often expressed as the fraction of soluble to total Fe in aerosol and then reported as a percentage
70 solubility (Baldo et al., 2022; Liu et al., 2022; Mahowald et al., 2009). Several key processes
71 control solubility of aerosol Fe over the course of its lifetime: 1) Fe mineralogy, 2) interactions
72 with acidic and organic species in aerosol and cloud water, and 3) particle size and surface area
73 to volume ratios (Bergas-Massó et al., 2023; Journet et al., 2008; Mcdaniel et al., 2019).
74 Anthropogenic combustion not only alters the magnitude and spatial distribution of Fe fluxes
75 from the atmosphere and **to the** surface ocean, but also influences the composition of the

76 atmosphere, that in turn, influences dissolution chemistry of aerosol Fe both directly and
77 indirectly. Mixing of aerosol Fe with acidic (e.g., sulfates or nitrates) and organic species (e.g.,
78 oxalate) co-emitted during anthropogenic combustion increases its solubility during transport
79 (Bergas-Massó et al., 2023; Chen et al., 2024; Itahashi et al., 2022; Li et al., 2017; Longo et al.,
80 2016). Furthermore, diverse technologies utilized during combustion processes (i.e., variable
81 combustion temperatures, boilers vs. furnaces, degree of emission control, and the fuel quality)
82 also influence the physicochemical properties of aerosol Fe beyond the composition of fuel
83 alone. As a result, **studies examining** socioeconomic, technology, and policy driven changes to
84 anthropogenic fuel-burning **are** needed to anticipate impacts on the global Fe cycle (Hamilton
85 et al., 2020b).

86 When compared to mineral dust, anthropogenic emissions of aerosol Fe are several orders
87 of magnitude lower at the global scale; however, anthropogenic Fe has a higher fractional
88 solubility (Ito et al., 2021), **and the relative contribution of soluble Fe from anthropogenic**
89 **combustion** is spatially distinct **from dust** (Hamilton et al., 2020b; Hamilton et al., 2019).
90 Therefore, anthropogenic activity can be a major contributor to Fe fluxes in many high nutrient
91 low chlorophyll (HNLC) ocean regions (Hawco et al., 2025; Liu et al., 2022).

92 Despite the importance of understanding anthropogenic Fe fluxes, the fractional solubility
93 of aerosol Fe emitted from various anthropogenic sources remains poorly understood
94 (Desboeufs et al., 2005; Li et al., 2022b; Oakes et al., 2012); consequently, Fe solubility
95 parameterizations in modeling studies for anthropogenic Fe vary widely (Ito et al., 2019;
96 Myriokefalitakis et al., 2018). **In this work**, we measured the Fe content and solubility **for**
97 aerosol emitted by several important anthropogenic sources (i.e., coal power plants, steelwork

98 industry, municipal water combustion, oil combustion, residential coal, and biofuel burning).
99 Then, using an Earth System Model, we applied the experimental results by updating Fe
100 solubility parameters for distinct anthropogenic combustion fuel-sources. Simulated Fe
101 concentrations and solubilities were validated against a global observational Fe aerosol dataset
102 at the regional scale. Then, the model was used to quantify and bound uncertainties in emission
103 and deposition fluxes of soluble Fe under three anthropogenic combustion emission scenarios
104 spanning past (pre-industrial) to future (Shared Socioeconomic Pathway 3-7.0, SSP370)
105 conditions.

106 **2 Methodology**

107 The experimental and modelling methods employed in this work are described in Sections
108 2.1 and 2.2, respectively.

109 **2.1 Experimental methods**

110 This work examined six types of anthropogenic combustion aerosol, which were classified
111 into two broad categories. The first category, fly ash, included power plant coal fly ash,
112 steelwork fly ash, municipal waste fly ash, and oil fly ash. The second category, residential fuel
113 sources, included residential coal and biofuel combustion. Biofuels examined in this work were
114 limited to straw, wood, grasses and leaves, and we did not examine other biofuels such as dung.

115 **2.1.1 Fly ash and bottom ash samples**

116 The volume-mean diameters, determined using diameter light scattering, were found to
117 be 16.9-67.6, 4.7-176.4, 21.2-115.9 and 15.4 μm for power plant coal fly ash (n=31), steelwork
118 fly ash (n=29), municipal waste fly ash (n=3), and oil fly ash (n=1) samples, respectively (Li,
119 2025).

120 Power plant coal fly ash samples were obtained from electrostatic precipitators or
121 baghouse rows in coal power plants in 29 provinces in China (Li et al., 2021; Liu et al., 2021);
122 one coal power plant was selected in each province except for Guangdong and Shandong where
123 two coal power plants were selected for each province. As a result, 31 power plant coal fly ash
124 samples were examined in total. In addition, we examined 29 steelwork fly ash samples
125 collected from different iron and steel plants, three municipal waste fly ash samples (Li et al.,
126 2022b; Li et al., 2021), and two oil fly ash samples which were PM_{2.5} samples emitted by heavy
127 oil and diesel fuel combustion in the engine of a cargo ship (Wu et al., 2018), and one oil
128 bottom ash sample (Fu et al., 2012). As the numbers of municipal waste and oil ash samples
129 were limited, we include their results data in the supplement, but do not discuss them further
130 in the main paper due to a lack of statistical significance.

131 Fly ash and bottom ash samples (~10 mg for each sample) were digested and then
132 analyzed using inductively coupled plasma mass spectrometry (ICP-MS) to determine their Fe
133 content. Experimental procedures for sample digestion and total Fe measurement can be found
134 elsewhere (Li et al., 2022c). Soluble Fe was leached and determined using the procedure
135 described in our previous work (Li et al., 2022b). In brief, fly ash and bottom ash samples (~20
136 mg for each sample) were individually leached in 20 mL sodium acetate buffer (5 mmol/L, pH
137 = 4.3) for 2 h, during which an orbital shaker (300 rpm) was used to stir the solution. The
138 aqueous mixture was centrifuged (3000 rpm) for 15 min, and a pH paper (range: 3.5-6.8;
139 precision: 0.3; Macherey-Nagel, Germany) was used to measure the pH of the solution and no
140 measurable pH change occurred after leaching. The aqueous solution was filtered through a
141 polyethersulfone filter (pore size: 0.22 μ m), acidified to contain 1% (v/v) nitric acid and then

142 analyzed by ICP-MS to measure soluble Fe. In this work, fractional solubility of Fe was
143 reported as the ratio (in %) of soluble Fe to total Fe.

144 A wide range of protocols, differing in leaching solution, filter pore size, and so on, were
145 used to extract soluble Fe, and the results obtained using different leaching protocols could be
146 substantial (Tang et al., 2025). Sodium acetate buffer, instead of ultrapure water, was used in
147 the present work as the leaching solution, because its pH did not change during leaching due
148 to its larger buffering capacity compared to ultrapure water.

149 **2.1.2 Residential coal and biofuel combustion aerosols**

150 Generation and collection of residential coal and biofuel combustion aerosols are detailed
151 in the **Supplement** (Text S1). In brief, we burned coal and biofuel in a commercial cook stove
152 widely used in rural areas in China and collected PM_{2.5} samples (aerosol particles with
153 aerodynamic particle diameters below 2.5 μm) onto pre-cleaned Whatman 41 (W41) cellulose
154 filters using a medium volume aerosol sampler (TH-150C, Tianhong Co.).

155 Our work examined three types of coal (anthracite, semibituminous coal, and bituminous
156 coal) and nine types of biofuel (wheat straw, rice straw, corn straw, rape straw, cogongrass,
157 China fir trunk, pine trunk, poplar trunk, and pine needle) commonly found in China. We
158 collected eight filter samples for each fuel type, except anthracite for which we only collected
159 two filter samples. We had to combine some filter samples in our experimental analysis to meet
160 the detection limit for soluble Fe; as a result, the number of effective filter samples (for which
161 Fe content and solubility were reported) were usually <8 for each fuel type (see **Tables S2 and**
162 **S4 for further information**).

163 After aerosol collection, the filters were individually placed in a pre-cleaned Petri dish
164 and then stored in a desiccator for 60 h to remove particle-associated water. The mass of filters
165 before and after aerosol collection were measured (accuracy of 0.1 mg), and the mass of
166 particles collected ranged from 2.5 to 432.7 mg. Each filter was then divided into two equal
167 parts. To determine the soluble Fe content, the first half of a filter was leached in 20 mL sodium
168 acetate buffer (5 mmol/L, pH = 4.3) for 2 h (Section 2.1.1) and analyzed using ICP-MS. Fe
169 concentrations in some leaching solutions were low; as a result, these leaching solutions (~15
170 mL for each solution) were combined for the same fuel type and then pre-concentrated to a
171 volume of 6 mL, in order to increase Fe concentration in the solution used for ICP-MS analysis.
172 The second half of a filter was digested and analyzed by ICP-MS to determine total Fe, and the
173 experimental procedure used can be found in our previous work (Zhang et al., 2022). If leaching
174 solutions were combined for the first parts of these filters, their second parts were also
175 combined and digested together to allow direct comparison.

176 **2.1.3 Quality assurance and quality control**

177 The detection limit of Fe in solution was determined to be 0.5 µg/L in this work. A
178 reference solution (NIST 1643f) was used to check the accuracy of ICP-MS analysis, and the
179 difference between actual and measured concentrations was found to be <1%. Furthermore,
180 three blanks (with no fly ash or filters not loaded with any particles) were used in each batch
181 when we measured total or soluble Fe. The background levels of soluble Fe were always below
182 the detection limit; the background levels of total Fe, ranging from 4.3-5.7 µg/L, were much
183 lower than total Fe concentrations for most of our samples and subtracted when we reported
184 our results.

185 2.2 Model simulations

186 2.2.1. Atmospheric Fe model description

187 Earth System Models can investigate the spatiotemporal distribution and fluxes of key
188 atmospheric nutrients under various climatological regimes (Hamilton et al., 2020b; Hamilton
189 et al., 2022; Wu et al., 2020). To test the impact of new soluble Fe parameters (Section 2.1) on
190 modeled fluxes of soluble aerosol Fe to the atmosphere and marine ecosystems, we used the
191 Mechanism of Intermediate complexity for Modeling Iron (MIMI). MIMI is an Fe aerosol-
192 chemistry module embedded within the atmospheric component (Community Atmosphere
193 Model version 6, CAM6) of the Community Earth System Model version 2 (CESM2)
194 (Danabasoglu et al., 2020; Hamilton et al., 2019). Mineral dust, anthropogenic combustion,
195 and wildfire emissions are currently represented as **distinct** sources of aerosol Fe in MIMI. The
196 current dust emission scheme within MIMI includes an updated soil moisture submodule
197 within the land component of the model that prognostically calculates dust aerosolization as a
198 function of soil moisture (Li et al., 2022a). **Following the implementation of a new soil-**
199 **moisture scheme, dust was rescaled to attain a global climatological mean dust aerosol optical**
200 **depth of ~0.03 (Ridley et al., 2016), consistent with all previous versions of the MIMI model.**
201 The inclusion of these improvements to dust and updated anthropogenic Fe sources represents
202 a new working version of MIMI v1.1, as described herein, **and detailed validation efforts are**
203 **reported in the Supplement (Figure S3 and Table S11).**

204 A comprehensive overview of model details and parameters is provided in Hamilton et al.
205 (2019); in brief, MIMI simulates the emission, atmospheric transport, and deposition of Fe-
206 containing aerosol within three distinct particle size modes (Aitken, accumulation, and coarse

207 modes). Within each source of aerosol Fe (dust, wildfire, and anthropogenic combustion), both
208 the insoluble and soluble fractions are carried as separate tracers, and the soluble fraction of Fe
209 for each aerosol source is assigned at the point of emission. Prior to deposition and during
210 atmospheric transport, Fe solubility is further modified via non-reversible multiphase reactions
211 with acidic and organic species. Acidic processing is a function of aerosol pH and temperature
212 (Meskhidze et al., 2005), while organic processing is an aqueous phase chemistry reaction that
213 depends on oxalate concentrations which are calculated based on the concentrations of
214 secondary organic aerosol present (Johnson and Meskhidze, 2013; Scanza et al., 2018).

215 The model is gridded in a 3-dimensional space at a resolution of 0.96×1.25 degrees
216 (latitude \times longitude) and includes 56 vertical pressure levels from the surface to 2 hPa at the
217 highest altitude. Meteorology is forced in all the simulations using Modern-Era Retrospective
218 analysis for Research and Applications Version 2 (MERRA-2), and a 1-year model spin up
219 was undertaken for all simulations.

220 **2.2.2 Global pyrogenic Fe emission inventories and input dataset development**

221 While dust Fe emissions are calculated prognostically within MIMI, anthropogenic and
222 wildfire (sum of these being pyrogenic) emissions are prescribed using emissions inventories.
223 Annual mean anthropogenic Fe emission fluxes were inputted to the model using a modified
224 version of an inventory first developed in Rathod et al. (2020) and further detailed in Rathod
225 et al. (2024). In this inventory, Fe content in combustion aerosol was empirically derived for
226 the present day (PD; climatological year 2010) using the Speciated Pollution Emissions Wizard
227 (SPEW) (Bond et al., 2007; Bond et al., 2004), which characterizes anthropogenic emissions
228 of particulate matter by fuel-source and combustion technology. Soluble and insoluble Fe

229 content **are** dependent on fuel-type and **also** segregated by key sectors: 1) industrial fossil fuel
 230 (coal), 2) industrial and vehicular fossil fuels (oil), 3) smelting operations (steel/iron), and 4)
 231 residential cooking/heating (biofuel/biomass/wood) (Rathod et al., 2020). Industrial oil
 232 emissions were separated by land- and sea-based emissions to distinguish terrestrial
 233 transportation from shipping. Wildfire-Fe emission parameters are detailed in Hamilton et al.
 234 (2019), and in this work we used the CMIP6 (Coupled Model Intercomparison Project phase
 235 six) fire emission **inventory** for PD simulations (Van Marle et al., 2017).

236 **Here, for the first time, we separated anthropogenic coal Fe into distinct industrial and**
 237 **residential sources and tested two approaches to constrain the magnitude of the residential Fe**
 238 **signal, i.e. a high emission scenario and a low emission scenario. To create the first inventory**
 239 **(‘high-residential’ emissions), we multiplied a series of spatially-resolved (1° x 1° grid box)**
 240 **ratios of residential-to-industrial black carbon (BC) emissions to the Rathod et al. (2024) Fe**
 241 **inventory emissions for coal. The BC ratios (Eqs. 1a and 1b) were derived from the CMIP6 BC**
 242 **emission dataset (Hoesly et al., 2018):**

$$243 \quad [Fe_{RESI}]_{i,j,HIGH} = \frac{[BC_{RESI}]_{i,j,H}}{[BC_{IND}]_{i,j,H}} \times [Fe_{IND+RESI}]_{i,j,R} \quad (1a)$$

$$244 \quad [Fe_{IND}]_{i,j,HIGH} = \frac{[BC_{IND}]_{i,j,H}}{[BC_{RESI}]_{i,j,H}} \times [Fe_{IND+RESI}]_{i,j,R} \quad (1b)$$

245 **Where i and j represent the longitudinal and latitudinal coordinates, $RESI$ and IND represent**
 246 **residential and industrial sources, $HIGH$ represents the high-residential inventory being**
 247 **constructed, H represents the Hoesly et al. (2018) dataset, R represents the Rathod et al. (2024)**
 248 **dataset, and $[Fe]$ and $[BC]$ represent their respective speciated fluxes in units of $\text{kg m}^{-2} \text{s}^{-1}$.**
 249 **This approach was chosen to capture spatial variations in coal burning technologies within the**

250 inventory but assumed that residential Fe-to-industrial Fe emissions track residential BC-to-
251 industrial BC.

252 To create the second inventory ('low-residential' emissions) we globally scaled the high-
253 residential inventory to reflect potential differences in fuel-type specific emissions. To do so,
254 we used fuel-type specific Fe emissions reported in Rathod et al. (2020) and fuel-type specific
255 BC emissions reported in Bond et al. (2004). The CMIP6 anthropogenic emissions dataset of
256 Hoesly et al. (2018) classifies emission sectors as: 0) agriculture, 1) energy, 2) industrial, 3)
257 terrestrial transportation, 4) residential/commercial/other, 5) solvents production/application,
258 6) waste, and 7) international shipping. Therefore, emissions labeled either commercial or
259 residential in Rathod et al. (2020) were grouped into 'residential' to pair them with CMIP6.
260 We also treated 'energy' and 'industrial' sources together as industrial coal BC and ocean-
261 masked 'international shipping' with land-masked 'terrestrial transportation' together as oil BC
262 sources. Hoesly et al. (2018) does not separate residential biofuel (wood) from residential coal,
263 and these two sources are under separate investigation within this study; as a result, we used
264 the ratio of residential coal to all residential sources reported in Bond et al. (2004) to separate
265 the Hoesly residential BC emissions (Table S8), assuming parity between BC datasets. Then,
266 we calculated the ratio of residential Fe-to-residential BC and industrial Fe-to-industrial BC
267 (Eq. 2a; Table S8), resulting in a scaling factor of 0.035. This scaling factor was then applied
268 to the high-residential emissions to create the low-residential Fe emissions inventory (Eqs. 2b
269 and 2c):

$$270 \quad \frac{[Fe_{RESI}]_{R^*} / [BC_{RESI}]_{H^*}}{[Fe_{IND}]_{R^*} / [BC_{IND}]_H} = 0.035 \quad (2a)$$

$$271 \quad [Fe_{RESI}]_{i,j,LOW} = [Fe_{RESI}]_{i,j,HIGH} * 0.035 \quad (2b)$$

$$[Fe_{IND}]_{i,j,LOW} = [Fe_{IND}]_{i,j,HIGH} * (1 - 0.035) \quad (2c)$$

where H^* denotes data from Hoesly et al. (2018) scaled by the Bond et al. (2004) BC inventory to isolate residential coal, R^* denotes data from Rathod et al. (2020), and LOW denotes the low-residential inventory. By redistributing residential and industrial coal Fe emissions using these sector-specific ratios of Fe-to-BC to create the low-residential emissions inventory, residential coal emissions were reduced by one order of magnitude at the global scale when compared to the high-residential emissions inventory (16 Gg a⁻¹ vs 460 Gg a⁻¹; Table S9). Both inventories were applied and tested with each PD model simulation to perform a sensitivity analysis that quantified the uncertainty in residential coal Fe emissions introduced by each new inventory.

Once sector-specific emissions inventories were read into the model, fuel-sources were summed to total one anthropogenic tracer. This tracer is then separated into soluble and insoluble fractions with three modes within the model code (Eqs. 3a and 3b). This results in six anthropogenic combustion-Fe tracers in total to be transported within the model, as follows:

$$[Fe_{insol}]_a = \Sigma\{[Fe_X]_{i,j,a,b} * (1 - sol_b)\} \quad (3a)$$

$$[Fe_{sol}]_a = \Sigma ([Fe_X]_{i,j,a,b} * sol_b) \quad (3b)$$

where X denotes the emissions scenario, a represents the aerosol mode (fine or coarse), b represents the fuel-source (industrial oil, industrial coal, residential coal, residential biofuel, or smelting), $insol$ represents the insoluble fraction, sol represents the soluble fraction, and sol_b represents the fractional solubility for each fuel-source (b). As a final step, the fine mode was split into accumulation and Aitken modes by applying a ratio of 9:1.

293 2.2.3 Model simulations performed

294 **Thirteen** model simulations were performed to evaluate the impact of anthropogenic
 295 combustion aerosol **solubility updates** (Section 2.1) on atmospheric soluble Fe fluxes to key
 296 marine ecosystems (Table 1). For all simulations, we set the model climatology to present-day
 297 (PD) conditions, spanning 2009-2011 **inclusive**. Simulations were distinguished as cases
 298 (variable Fe solubility parameterizations) within different emission scenarios (variable
 299 anthropogenic combustion emission fluxes).

300

301 **Table 1.** Description of model simulations performed using MIMI with emission scenarios and
 302 emission **inventories either directly inputted (Fe) to the model or utilized to generate the Fe**
 303 **inventory (BC)**. PD = present day (2010 CE), PI = pre-industrial (1750 CE), SSP370 = Shared
 304 Socioeconomic Pathway scenario 3-7.0, MID = midcentury (2040-2050 CE) and END = end
 305 century (2090-2100 CE). NA = assumed industrial activity is zero at 1750 CE.

Emissions Scenario	Simulation	Fe Emissions Inventory	BC Emissions database	BC Emission (Tg a ⁻¹)
PD	PD-BASE	Rathod et al., 2024	Hoesly et al., 2018	6.46
PD	PD-RESI	High-Residential	Hoesly et al., 2018	6.46
PD	PD-BIOF	High-Residential	Hoesly et al., 2018	6.46
PD	PD-IND	High-Residential	Hoesly et al., 2018	6.46
PD	PD-BASE	Rathod et al., 2024	Bond et al., 2004 & Hoesly et al., 2018	6.46
PD	PD-RESI	Low-Residential	Bond et al., 2004 & Hoesly et al., 2018	6.46
PD	PD-BIOF	Low-Residential	Bond et al., 2004 & Hoesly et al., 2018	6.46
PI	PI-BASE	Hamilton et al., 2020a	NA	NA
PI	PI-BIOF	Hamilton et al., 2020a	NA	NA
FU (2050)	MID-SSP370-BASE	High-Residential	SSP3.70	8.30
FU (2050)	MID-SSP370-BIOF	High-Residential	SSP3.70	8.30
FU (2100)	END-SSP370-BASE	High-Residential	SSP3.70	6.33
FU (2100)	END-SSP370-BIOF	High-Residential	SSP3.70	6.33

306

307 The first seven simulations (i.e., PD simulations) aimed to assess the impact of each new
308 solubility parameter on the ability of the model to capture ship-based observations of total Fe,
309 soluble Fe, and Fe solubility. These simulations applied the high-residential emissions
310 inventory (Section 2.2.2), and were repeated using the low-residential Fe inventory with the
311 exception of PD-IND (Table 1). The first PD case in each set (i.e., PD-BASE) served as a
312 baseline, i.e., no changes were made to solubility when compared to previous studies using
313 MIMI. Residential coal was distinguished from industrial coal emissions, but this had no
314 impact on soluble Fe fluxes since the same fractional solubility (0.2%) was applied to both
315 sources (Rathod et al., 2020; Rathod et al., 2024). In the next three PD cases (PD-RESI, PD-
316 BIOF, PD-IND), fractional solubility was updated incrementally for individual sectors to
317 assess fuel-type specific impacts to soluble Fe fluxes, which are later detailed in Section 3.3;
318 results from PD-IND are reported in the Supplement. Information on model validation and
319 constraint to ship-based observations of aerosol Fe is provided in Section 2.2.5.

320 Using both pre-industrial (PI; 1750 CE) and future (FU; 2050 and 2100 CE) anthropogenic
321 emissions scenarios, we performed six model simulations to examine the impacts of changes
322 in anthropogenic activity on Fe fluxes over time. In each pair of simulations, we applied
323 the -BASE solubility parameters to establish a baseline and the -BIOF solubility parameters to
324 examine an upper bound for residential soluble Fe fluxes. Accordingly, we also used the high-
325 residential Fe emissions inventory framework in development of the FU emissions inventory
326 to further establish a maximum estimate for anthropogenic soluble Fe through the end of the
327 21st century. To isolate how changes in soluble aerosol Fe fluxes responded to changes in

328 emission parameterizations and subsequent dissolution chemistry **in the model**, PI and FU
329 simulations were conducted with meteorological and climatological conditions identical to **the**
330 PD (2009-2011).

331 **The PI simulations (PI-BASE and PI-BIOF) served as a reference point** for comparison to
332 PD and FU simulations, **per** minimal influence on the Fe cycle by anthropogenic emissions
333 (Table 1). MID-SSP370-BASE, MID-SSP370-BIOF, END-SSP370-BASE, and END-
334 SSP370-BIOF were conducted to evaluate the projected impact of **population increases** and
335 socioeconomic changes to energy production and fuel-usage **over the course of the century**.
336 We **selected the highest air pollution** emissions scenario **of the Shared Socio-economic**
337 **Pathways (SSPs)** as detailed in **SSP 3-7.0 (SSP370)**, the “regional rivalry” scenario, which
338 represents anticipated sociopolitical and environmental changes resulting in an increase to
339 radiative forcing by $3-7.0 \text{ W m}^{-2}$ by the year of 2100 (Riahi et al., 2017). Given that BC
340 emissions are anticipated to peak in the midcentury (2040-2050) but return to PD-comparable
341 emissions by 2100 (Turnock et al., 2020), we assessed projected changes to Fe emissions at
342 both the mid-point (2050) and endpoint of the 21st century (2100). **Dust fluxes in future Fe**
343 **emission scenarios were adjusted to account for dust-climate feedback using a scaling factor**
344 **ranging between 1.0-1.1, as described in Hamilton et al. (2020a).**

345 **2.2.4 Preindustrial (PI) and future (FU) Fe emission estimates**

346 For PI simulations, we used a pre-developed Fe combustion emission inventory (Hamilton
347 et al., 2020b). Only residential **(wood)** biofuel burning served as an anthropogenic source of
348 Fe due to a presumable lack of industrialized anthropogenic emissions (i.e., fossil fuels and
349 smelting; **(Hamilton et al., 2020a)**; global emission **for anthropogenic combustion** was 0.7×10^7

350 ³ Gg Fe a⁻¹ and only occupied the fine aerosol mode (i.e., sum of Aitken and accumulation
351 modes). Details on the development of the PI Fe combustion emission inventory are provided
352 in Hamilton et al. (2020b).

353 For FU simulations, we developed two new Fe emissions datasets (for 2050 and 2100)
354 which were both derived from our high-residential emissions inventory developed for the PD
355 simulations. Fe emissions were linearly scaled for all combustion sources according to
356 projected changes in anthropogenic BC emissions via the decadal CMIP6 anthropogenic BC
357 emission dataset for 2040-2050 and 2090-2100 (Hoesly et al., 2018; Riahi et al., 2017). BC
358 emissions labeled ‘residential, commercial and other’ were separated into residential coal and
359 residential biofuel sources of BC based on the grid-cell specific ratios of residential coal Fe to
360 residential biofuel Fe in our high-residential emissions inventory.

361 Following a similar approach to the PD high-residential emissions inventory, using a grid-
362 cell resolved dynamic ratio of Fe-to-BC, that was grouped based on sector and aerosol size
363 fraction, we calculated FU Fe emissions tracking CMIP6 projected BC emissions (Eq. 4), as
364 follows:

$$365 \quad \frac{[Fe_X]_{i,j,a,b}}{[BC_X]_{i,j,a,b}} = \frac{[Fe_{PD}]_{i,j,a,b}}{[BC_{PD}]_{i,j,a,b}} \quad (4)$$

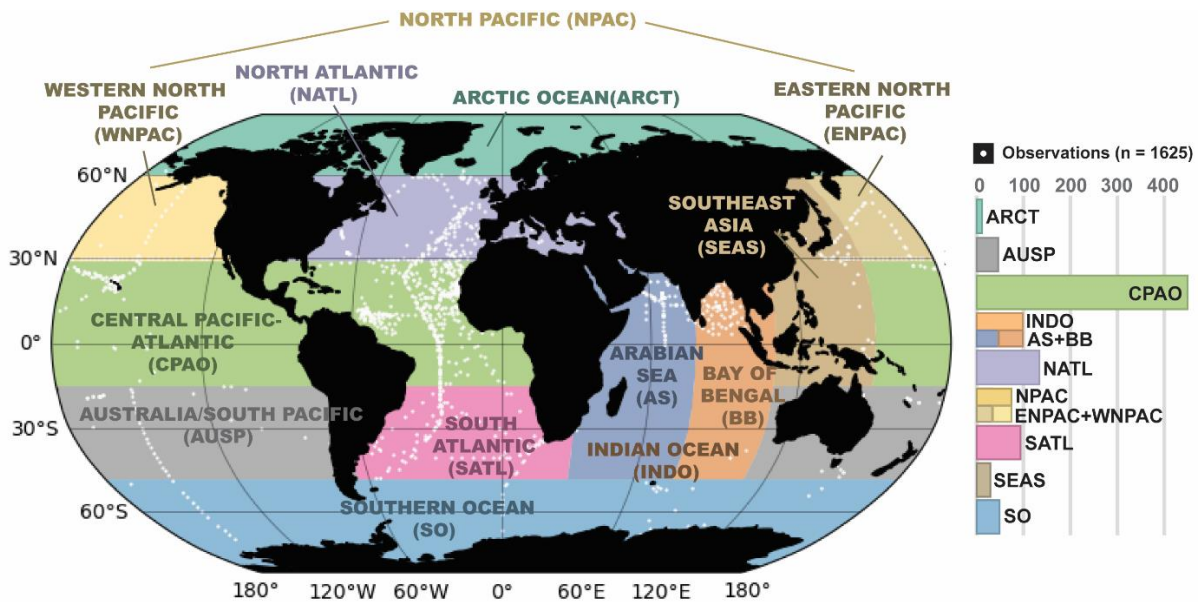
366 where X denotes the emissions scenario (MID-SSP370 or END-SSP370), *i* and *j* represent the
367 longitudinal and latitudinal coordinates, *a* represents the aerosol mode (fine or coarse), *b*
368 represents the fuel-source (industrial oil, industrial coal, residential coal, residential biofuel, or
369 smelting), and [Fe] and [BC] represent the speciated fluxes (kg m⁻² s⁻¹). BC emissions from
370 smelting operations were not directly available for PI or FU projections; therefore, they were
371 set to 0.0 in the PI and maintained at PD levels in the FU. By using SSP370, the -BIOF case

372 solubility parameters, and the high-residential emissions inventory, our FU simulations
373 established an upper bound estimate for future anthropogenic soluble Fe fluxes as based on
374 current observational uncertainties. The SimFire inventory, coupled to the LPJ-GUESS (Lund-
375 Potsdam-Jena General Ecosystem Simulator) vegetation model, was used to prescribe wildfire-
376 Fe emissions during the PI era (Hamilton et al., 2018; Hamilton et al., 2020a; Knorr et al., 2016)
377 For wildfire-Fe emissions in FU scenarios, we used the CMIP6 fire emission datasets for MID-
378 SSP370 and END-SSP370 (Bergas-Masso et al., 2025; Hamilton et al., 2024).

379 **2.2.5 Model validation**

380 To evaluate model performance, we compared global observations of total Fe
381 concentration, soluble Fe concentration, and Fe solubility to modeled values for each PD
382 simulation, grouping data by key aerosol deposition and ocean biogeochemistry regions. The
383 observational dataset of Fe content in aerosol was reported in Hamilton et al. (2019) and
384 updated herein to include measurements from Srinivas et al. (2012) and more recent studies
385 published between 2021 and 2024 (n = 1624) (Desboeufs et al., 2024; Elliott et al., 2024;
386 Kurisu et al., 2021; López-García et al., 2021; Marafante et al., 2024; Panda et al., 2022; Perron
387 et al., 2022; Rodríguez et al., 2021; Sakata et al., 2022; Seo and Kim, 2023; Winton et al., 2022;
388 Wu et al., 2023; Zhang et al., 2024). Observed Fe solubility in aerosol spans five orders of
389 magnitude (Perron et al., 2024), and one reason for this large range is due to differences in
390 experimental procedures during quantification (Tang et al., 2025). To facilitate a more direct
391 comparison between modeled and observed soluble Fe content, we removed observations from
392 the global dataset that did not measure soluble Fe directly. When multiple observations fell
393 within a model grid cell, values were aggregated to climatological averages, using medians to

394 be most representative of expected variations in Fe fluxes across time and space (final n = 990;
 395 **Figure 3**). For final evaluation of the model capability in simulating surface Fe concentrations,
 396 both model and observational data were grouped into key ocean regions (Figure 1), based on
 397 predominant sources of atmospheric aerosol and phytoplankton nutrient limitation dynamics
 398 (i.e., HNLC regions) as revealed in Hamilton et al. (2019) and Hamilton et al. (2023). **To**
 399 **quantitatively evaluate model skill, root mean square errors (RMSE) were calculated for the**
 400 **high-residential inventory cases and are provided in the Supplement.**



401
 402 **Figure 1.** Regional groupings for model-observation comparisons of surface Fe concentrations
 403 (ship-based, in aerosol). The coordinates for individual Fe observations are indicated with a
 404 white circle. Number of soluble Fe observations within each region are provided by the
 405 histogram (n=990).

406

407 **3 Results and Discussion**

408 Sections 3.1 and 3.2 present Fe content and solubility measured in our experimental work.

409 **The numbers of samples examined in our work are very small for municipal waste fly ash and**

410 oil fly/bottom ash, and the results may not be representative; therefore, these results are mainly
 411 presented in the Supplement. Modeling results are presented in Section 3.3.

412 3.1 Fe content by fuel type

413 This work quantified the Fe content in particles from six different combustion and
 414 anthropogenic sources, including power plant coal fly ash, residential coal combustion aerosol,
 415 steelwork fly ash, residential biofuel burning aerosol, municipal waste fly ash, and oil fly ash
 416 (Table 2; Fe content in individual samples is provided in Tables S1-S5).

417
 418 **Table 2.** Summary of Fe content and solubility for power plant coal fly ash, residential coal
 419 combustion aerosol, steelwork fly ash, and biofuel burning aerosol examined in our work (*n*:
 420 number of samples examined in our work). Fe content and solubility for other samples
 421 examined in this work (i.e. municipal waste fly ash, oil fly ash and oil bottom ash) can be found
 422 in the Supplement.

sample type	<i>n</i>	range	average	median
Fe content (mg/g)				
power plant coal fly ash	31	20.7-103.8	37.2±16.8	35.0
residential coal combustion aerosol	10	0.025-0.101	0.044±0.023	0.038
steelwork fly ash	29	5.8-918.9	312.6±246.1	346.5
biofuel burning aerosol	27	0.002-0.101	0.023±0.026	0.013
Fe solubility (%)				
power plant coal fly ash	31	0.002-0.17	0.05±0.05	0.03
residential coal combustion aerosol	10	7.03-100	33.30±27.71	28.45
steelwork fly ash	29	0.007-10.64	1.37±2.77	0.07

423

424

425 **3.1.1 Power plant coal fly ash**

426 Fe content ranged from 20.7 to 103.8 mg/g for the 31 power plant coal fly ash samples
427 examined in our work, with average and median values being 37.2 ± 16.8 and 35.0 mg/g,
428 respectively. As shown in Table S6, Fe content ranged from 16.0 to 52.0 mg/g ($n = 3$) in one
429 study (Baldo et al., 2022), with mean and median values being 33.0 ± 18.0 and 31.0 mg/g; in
430 another study (Goodarzi, 2006), the median value of Fe content was determined to be 34.4
431 mg/g ($n = 7$). Fe content measured by these two studies (Baldo et al., 2022; Goodarzi, 2006)
432 agreed well with our work. Some other studies (Dutta et al., 2009; Fu et al., 2012; Jankowski
433 et al., 2006; Meij, 1994) found higher mean or median Fe content for power plant coal fly ash
434 (Table S6), but the reported ranges overlapped with our work. For example, Fe content were
435 found to range from 38.3 to 98.6 mg/g ($n = 7$) in one study (Li et al., 2022b), with mean and
436 median values being 62.1 ± 26.7 and 43.2 mg/g; in another study (Moreno et al., 2005), Fe
437 content were found to range from 18.2 to 112.0 mg/g ($n = 23$), with mean and median values
438 being 57.8 ± 22.7 and 52.5 mg/g.

439 In summary, the mean or median Fe content reported in different studies are typically in
440 the range of 30-70 mg/g for power plant coal fly ash, and this variability is likely due to
441 difference in coal (Wang et al., 2015; Ward, 2016) and combustion conditions (Blissett and
442 Rowson, 2012; Kutchko and Kim, 2006). Fe content in power plant coal fly ash was set to ~70

443 mg/g in some modeling studies (Luo et al., 2008; Rathod et al., 2020), being consistent with
444 experimental results.

445 **3.1.2 Residential coal combustion aerosol**

446 For the 10 residential coal combustion aerosol samples (PM_{2.5}) we examined, Fe content
447 ranged from 0.025 to 0.101 mg/g (Table 2), with average and median values being 0.044 ±
448 0.023 and 0.038 mg/g, respectively. Only a few previous studies measured Fe content in
449 residential coal combustion aerosols (Table S6). The average Fe content was determined by
450 Patil et al. (2013) to be 0.048 ± 0.035 mg/g (n = 3) for PM_{2.5} and 0.061 ± 0.044 mg/g (n = 3)
451 for PM₁₀, being similar to or slightly higher than our result. In another two studies (Watson et
452 al., 2001; Zhang et al., 2012), the average Fe content was measured to be 0.671 ± 0.023 mg/g
453 (n = 4) and 0.7 ± 0.1 mg/g (n = 5), significantly higher than our result, and such differences
454 may be attributed to variations in coal types and combustion conditions. Overall, our and
455 previous studies suggest that the Fe content in residential coal combustion aerosols is very low,
456 typically below 1 mg/g. Fe content were set to 1 and 0.5 mg in previous modeling studies (Luo
457 et al., 2008; Rathod et al., 2020), being broadly consistent with experimental results.

458 Fe content in power plant coal fly ash is much higher than residential coal combustion
459 aerosols, primarily due to differences in combustion conditions (Rathod et al., 2020). Power
460 plant coal fly ash has very low carbon content and is mainly composed of metals and minerals
461 (Ahmaruzzaman, 2010; Li et al., 2022c; Patil et al., 2013); in contrast, residential coal
462 combustion aerosol particles contain a large fraction of carbonaceous materials due to
463 incomplete combustion, and thus the content of metals, including Fe, are much lower (Patil et
464 al., 2013; Zhang et al., 2012). Furthermore, combustion temperature typically ranges from 1200

465 to 1700 °C for coal-fired power plant, enabling Fe in coal to enter fly ash particles through
466 volatilization-condensation (Blissett and Rowson, 2012); residential coal combustion occurs at
467 much lower temperatures which are insufficient for Fe to enter aerosols through this process
468 (Rathod et al., 2020), also leading to lower Fe content.

469 **3.1.3 Steelwork fly ash**

470 For the 29 steelwork fly ash samples we examined, Fe content ranged from 5.8 to 918.9
471 mg/g, with mean and median values measured to be 312.6 ± 246.1 and 346.5 mg/g, respectively
472 (Table 2). As shown in Table S6, some previous studies have reported average Fe content to
473 be 358.9 (n = 1), 369.3 (n = 1), 312.2 (n = 1), and 329.1 ± 22.6 mg/g (n = 4) (Alizadeh and
474 Momeni, 2016; Silva et al., 2019; Souza et al., 2010; Vieira et al., 2013), in good agreement
475 with our results. Lower Fe content was also reported by previous work, with average values
476 being 86.0 (n = 1), 128.1 (n = 1), 150.8 (n = 1), 286.5 (n = 1), 284.6 (n = 1), 238.7 (n = 1), and
477 267.3 ± 4.8 mg/g (n = 4) (Al-Negheimish et al., 2021; Alsheyab and Khedaywi, 2016; Laforest
478 and Duchesne, 2006; Li et al., 2023; Loaiza et al., 2017; Stathopoulos et al., 2013; Xia and
479 Picklesi, 2000); in contrast, some previous studies also found the average or mean Fe content
480 to be around 400-500 mg/g (Machado et al., 2006; Patil et al., 2013; Ye et al., 2021), slightly
481 higher than our results.

482 Despite some variability in Fe content reported by our and previous studies (Table S6),
483 the mean or median Fe content are generally around 300–500 mg/g for steelwork fly ash. In a
484 recent modeling study (Rathod et al., 2020), the Fe content in steelwork fly ash was set to 440
485 mg/g (and the lower and upper bounds were set to 150 and 950 mg/g), being consistent with
486 experimental results.

487 **3.1.4 Biofuel burning aerosol**

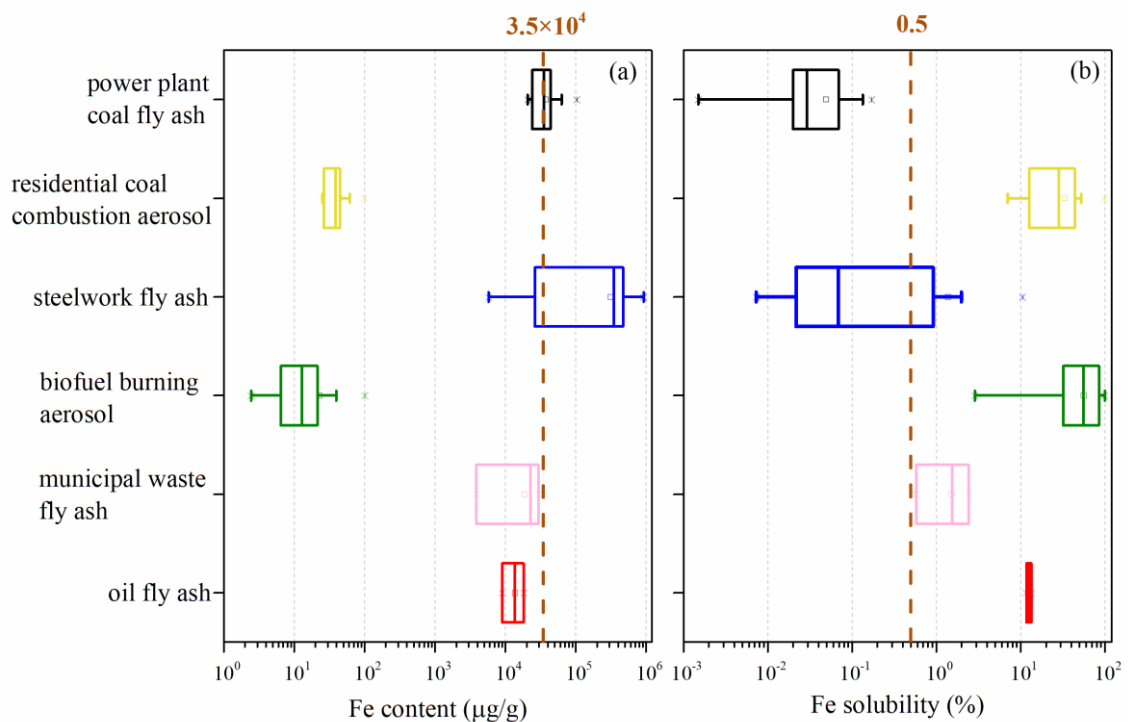
488 Our work considered biofuel burning aerosols for nine types of biofuels, including four
489 types of crop straw, one type of wild grass, and four types of wood. Fe content in biofuel
490 burning aerosols ranged from 0.002 to 0.101 mg/g (Table 2), with average and median values
491 being 0.023 ± 0.026 and 0.013 mg/g, respectively. As shown in Table S6, the average Fe
492 content was determined to be 0.024 ± 0.017 mg/g ($n = 3$) for $PM_{2.5}$ (Patil et al., 2013), very
493 close to our result; in another study (Hildemann et al., 1991), it was determined to be 0.090
494 mg/g for PM_2 ($n = 2$), higher than our result. In some other studies, average Fe content were
495 reported to be in the range of 0.162-0.440 mg/g for $PM_{2.5}$ (Alves et al., 2011; Hedberg et al.,
496 2002; Watson et al., 2001; Zhang et al., 2012) and 0.723 ± 0.661 mg/g for PM_{10} (Schmidl et
497 al., 2008), much higher than our results.

498 Fe content in biofuel burning aerosols showed large variability in different studies, likely
499 due to variations in combustion conditions and biofuel types. For example, metal content in
500 biofuel burning aerosols depended greatly on biofuel types and regions where biofuel was
501 collected (Goncalves et al., 2010), and aerosol particles emitted by wild grass combustion
502 contained larger amounts of metal than wood combustion (Jahn et al., 2021). Modeling studies
503 have used a similar distribution of Fe content between 0.2 and 0.580 mg/g for biofuel burning
504 aerosols (Luo et al., 2008; Rathod et al., 2020).

505 **3.1.5 Fe contents: comparison of anthropogenic and dust Fe**

506 Figure 2a displays Fe content for anthropogenic particles examined in our current study,
507 and the brown dashed line represents the average Fe content of desert dust (35 mg/g) (Taylor
508 and Mclennan, 1995). Steelwork fly ash has very high Fe content (median: 346.5 mg/g), about

509 one order of magnitude higher than desert dust. Power plant coal fly ash (median: 35.0 mg/g)
 510 has similar Fe content to desert dust. Compared to desert dust, Fe content were around three
 511 orders of magnitude lower for residential coal and biofuel burning aerosol (median: 0.038 and
 512 0.013 mg/g, respectively). The Fe content was much lower for residential coal and biofuel
 513 burning aerosol, likely due to lower combustion temperatures. When combustion occurs at
 514 lower temperature, the carbon content of emitted particles is higher; in addition, lower
 515 combustion temperature is not sufficient to enable Fe in the fuel to enter emitted particles via
 516 volatilization-condensation processes.



517
 518 **Figure 2.** Fe content (a) and solubility (b) measured in our work for power plant coal fly ash,
 519 residential coal combustion aerosol, steelwork fly ash, biofuel burning aerosol, municipal
 520 waste fly ash and oil fly ash. The two brown dash lines represent (a) the Fe content (3.5×10^4
 521 $\mu\text{g/g}$) and (b) Fe solubility ($\sim 0.5\%$) for desert dust, respectively.

522 **3.2 Fe solubility by fuel type**

523 **3.2.1 Power plant coal fly ash**

524 Fe solubility in acetate buffer (pH: 4.3) ranged from 0.002% to 0.17% for power plant
525 coal fly ash (Table 2), with the average and median values being $0.05 \pm 0.05\%$ and 0.03%,
526 respectively. A few previous studies measured Fe solubility of power plant coal fly ash in
527 weakly acidic or circumneutral solutions (Table S7). For example, Fe solubility was measured
528 to be 0.06% in deionized water (Oakes et al., 2012), similar to our result; it was measured to
529 be 0.2% in dilute sulfuric acid solution (pH: 4.7) (Desboeufs et al., 2005), slightly higher than
530 our result; the median Fe solubility was determined to be 0.13% in acetate buffer (pH: 4.3) and
531 0.06% in deionized water (Li et al., 2022b), both higher than the median value we obtained.
532 Overall, our work and previous studies suggest that Fe solubility is low in weakly acidic and
533 circumneutral solutions for power plant coal fly ash, with mean or median values around 0.1%.

534 Some studies also measured Fe solubility of power plant coal fly ash in highly acidic
535 solutions and found them to be much higher than those in weakly acidic and circumneutral
536 solutions. For example, Fe solubilities were found to be in the range of 20-25% at pH of 1-2
537 (Chen et al., 2012), 4.2-8.3% at pH of 2 (Fu et al., 2012), and >40% at pH of 2.1 (Baldo et al.,
538 2022). Although Fe solubility measured in strongly acidic solutions may not reflect initial Fe
539 solubility, these studies suggested that acid processing in the emission plume or wider
540 atmosphere could greatly increase Fe solubility for power plant coal fly ash.

541 **3.2.2 Residential coal combustion aerosol**

542 Fe solubility in acetate buffer (pH: 4.3) was determined to range from 7.03% to 100% for
543 residential coal combustion aerosol (Table 2), with the average and median values being 33.30

544 $\pm 27.71\%$ and 28.45% , respectively. To our knowledge, no previous study has measured Fe
545 solubility for residential coal combustion aerosol. Compared to power plant coal fly ash, Fe
546 solubility was much higher for residential coal combustion aerosol, and such difference can be
547 attributed to much higher temperature in power plant coal combustion than residential coal
548 combustion. Pyrite (FeS_2) is the major Fe-containing mineral in coal (Deng et al., 2015;
549 Oliveira et al., 2016; Rathod et al., 2020). In low-temperature combustion, pyrite is mainly
550 transformed to Fe sulfate (Bhargava et al., 2009) which has very high Fe solubility; as the
551 temperature increases to $>1000\text{ K}$, Fe sulfate is further transformed to hematite and magnetite
552 which exhibit very low solubility (Hu et al., 2006; Ram et al., 1995; Rathod et al., 2020). A
553 previous study (Rathod et al., 2020) used the relationship between combustion temperature and
554 Fe mineralogy in emitted particles to estimate Fe solubility for different combustion aerosols,
555 and Fe solubility was estimated to be as high as $\sim 32.5\%$ for residential coal combustion
556 aerosols, in good agreement with our experimental results.

557 **3.2.3 Steelwork fly ash**

558 Fe solubility in acetate buffer (pH: 4.3) was determined to range from 0.01% to 10.64%
559 for steelwork fly ash (Table 2), and the average and median values were $1.37 \pm 2.77\%$ and
560 0.07% , respectively. We note that Fe solubility was significantly higher (0.92% - 8.59%) for 8
561 samples and very low ($<0.5\%$) for the other 21 samples (Table S3), most of which showed Fe
562 solubility below 0.1% . No previous work has measured Fe solubility for steelwork fly ash. Our
563 experimental results were supported by a modeling study (Rathod et al., 2020) which suggested
564 that the major Fe-containing species in steelwork fly ash were Fe oxides with very low Fe
565 solubilities.

566 **3.2.4 Biofuel burning aerosol**

567 For biofuel burning aerosol, Fe solubility in acetate buffer (pH: 4.3) ranges from 2.86%
568 to 100% with average and median values of $56.07 \pm 30.95\%$ and 55.87%, respectively (Table
569 2). Based on the relationship between combustion temperature and Fe-containing species in
570 emitted aerosols, Fe solubility was previously estimated at 35% for wood burning (i.e., biofuel)
571 aerosol (Rathod et al., 2020), in good agreement with our experimental results.

572 The biofuel examined in our experiment was burnt in a sealed stove and contained no
573 apparent local soil contamination. As such, these results are most representative of domestic
574 biofuel combustion for which the influence of soil-derived Fe can be expected to be negligible.
575 In contrast, wildfires represent dynamic open fire systems that emit aerosol Fe in both fine and
576 coarse fractions (Hamilton et al., 2019). During wildfire combustion, not only is the biofuel
577 (biomass) consumed, but local soils are also entrained into the smoke plumes (Hamilton et al.,
578 2022; Tegler et al., 2023). These soil-derived particles are typically larger (in particle size) and
579 less soluble than their biofuel-derived counterparts (Hamilton et al., 2022), resulting in a larger
580 mass of emitted Fe, albeit with a lower overall Fe solubility. Future studies would benefit from
581 capturing emissions from open burning scenarios to better characterize the properties of
582 wildfire-emitted Fe.

583 **3.2.5 Fe solubilities: comparison of anthropogenic and dust Fe**

584 Figure 2b compares our measured Fe solubility for six types of combustion and
585 anthropogenic particles with that for desert dust. Biofuel burning aerosols (median: 55.87%)
586 and residential coal combustion aerosols (median: 28.45%) exhibited very high Fe solubility.
587 Compared to desert dust, for which Fe solubility is around 0.5% (Chuang et al., 2005; Li et al.,

588 2022b; Ooki et al., 2009; Schroth et al., 2009; Shi et al., 2011), Fe solubility was lower for
589 steelwork fly ash (median: 0.07%) and power plant coal fly ash (median: 0.03%).

590 Overall, Fe solubility in emitted particles was significantly higher for low-temperature
591 combustion (residential and biofuel burning aerosols) than high-temperature combustion
592 (steelwork fly ash and power plant coal fly ash). This is because Fe in emitted particles is
593 mainly highly soluble Fe sulfates for low **temperature** combustion (Bhargava et al., 2009;
594 Rathod et al., 2020) but Fe oxides with very low solubility for high temperature combustion
595 (Hu et al., 2006; Ram et al., 1995; Rathod et al., 2020).

596 **3.3 Modeling Results**

597 Leveraging **new measurements of** combustion Fe **solubility in residential** fuel sources as
598 reported in Sections 3.1 and 3.2, we performed a series of Earth System Model (**MIMI**)
599 simulations that **examined global Fe fluxes in response to modifying** anthropogenic Fe
600 **solubility parameters** at **their** point of emission. To pair observed solubilities (Table 2) with
601 fuel-types represented in the model, we updated Fe solubility in residential coal **burning aerosol**
602 from 0.2 to 33% **and** in residential biofuel burning **aerosol** from 10 to 56% **at the point of**
603 **emission** (Table 3) **in both the fine and coarse modes. Smelting Fe solubility was kept at 0.03%**
604 **for all simulations, since new data did not suggest an alternative solubility from what is**
605 **currently used (Table 3). We ran one additional simulation with updates to industrial sources**
606 **(PD-IND: industrial coal Fe solubility from 0.2 to 0.05% and oil from 38 to 25%), but observed**
607 **impacts to global soluble Fe fluxes following these changes were minimal and are accordingly**
608 **deferred to the Supplement.** A description of the fractional solubilities applied to each
609 anthropogenic fuel type within each model simulation is provided in Table 3.

610

611 **Table 3.** Fractional Fe solubilities applied in each model simulation to reflect experimental
 612 findings. **Bolded rows** indicate baseline simulations with no changes made to Fe solubility from
 613 previous work using MIMI. To underscore modifications between simulations, a dash (–) is
 614 provided where assigned solubility did not differ from the PD-BASE simulation.

Simulation	Fe solubility modifications by fuel-type (%)				
	Industrial Coal	Residential Coal	Oil	Residential Biofuel	Smelting
PD-BASE	0.2	0.2	38	10	0.003
PD-RESI	-	33	-	-	-
PD-BIOF	-	33	-	56	-
PD-IND	0.05	33	25	56	-
PI-BASE	NA	NA	NA	10	NA
PI-BIOF	NA	NA	NA	56	NA
MID-SSP370-BASE	0.2	0.2	38	10	0.003
MID-SSP370-BIOF	-	33	-	56	0.003
END-SSP370-BASE	0.2	0.2	38	10	0.003
END-SSP370-BIOF	-	33	-	56	0.003

615

616 3.3.1 Impacts on global soluble Fe distribution

617 By applying two new emissions inventories and new solubility parameters for residential
 618 Fe emissions within MIMI, we report a new range of soluble Fe fluxes to the ocean with
 619 regional variations. To isolate the impacts of modifications to each fuel-source, we compared
 620 each model case. When comparing PD-BASE to PD-RESI, the emissions inventory had the
 621 greatest influence on the result per sizeable differences in residential coal emissions by mass.
 622 In the high-residential inventory, global Fe emissions from residential coal were 460 Gg a⁻¹,
 623 exceeding individual emissions from all other fuel types (industrial coal: 310 Gg a⁻¹; oil: 34 Gg
 624 a⁻¹; residential biofuel: 72 Gg a⁻¹) except for smelting (1350 Gg a⁻¹; Table S8). Whereas, in the
 625 low-residential inventory, emissions were nearly two orders of magnitude less at 16 Gg a⁻¹

626 (Table S9), and lower than any other fuel source. Constraining the Fe content in residential
627 emissions is therefore a more critical knowledge gap to be addressed than constraining the
628 fractional solubility of this source.

629 Accordingly, when applying the low-residential emissions inventory and using the PD-
630 RESI solubility parameters, soluble Fe fluxes to the global ocean only increased by $< 1 \text{ Gg a}^{-1}$
631 (Figure 3; Table 4). Conversely, when applying the high-residential inventory, we saw a 92%
632 increase in anthropogenic soluble Fe fluxes ($+ 33 \text{ Gg a}^{-1}$) to the ocean, translating to a 7%
633 increase in overall soluble Fe fluxes including those from dust and wildfire (Figure 3; Table 4).
634 While the high-residential emissions inventory likely overstates residential coal burning
635 emissions by Fe mass content, the ocean regions most influenced by residential coal burning
636 become apparent, facilitating future research locations of highest interest. Anthropogenic
637 soluble Fe delivery to the Indian Ocean, eastern North Pacific, and parts of the Southern Ocean
638 increased by 300% to 400%, corresponding with a large increases in emissions from China,
639 India, Australia, and South Africa (Figure 3). This follows previous reports of relatively large
640 anthropogenic signals from these regions when compared to global averages (Rathod et al.,
641 2024; Wang et al., 2015).

642

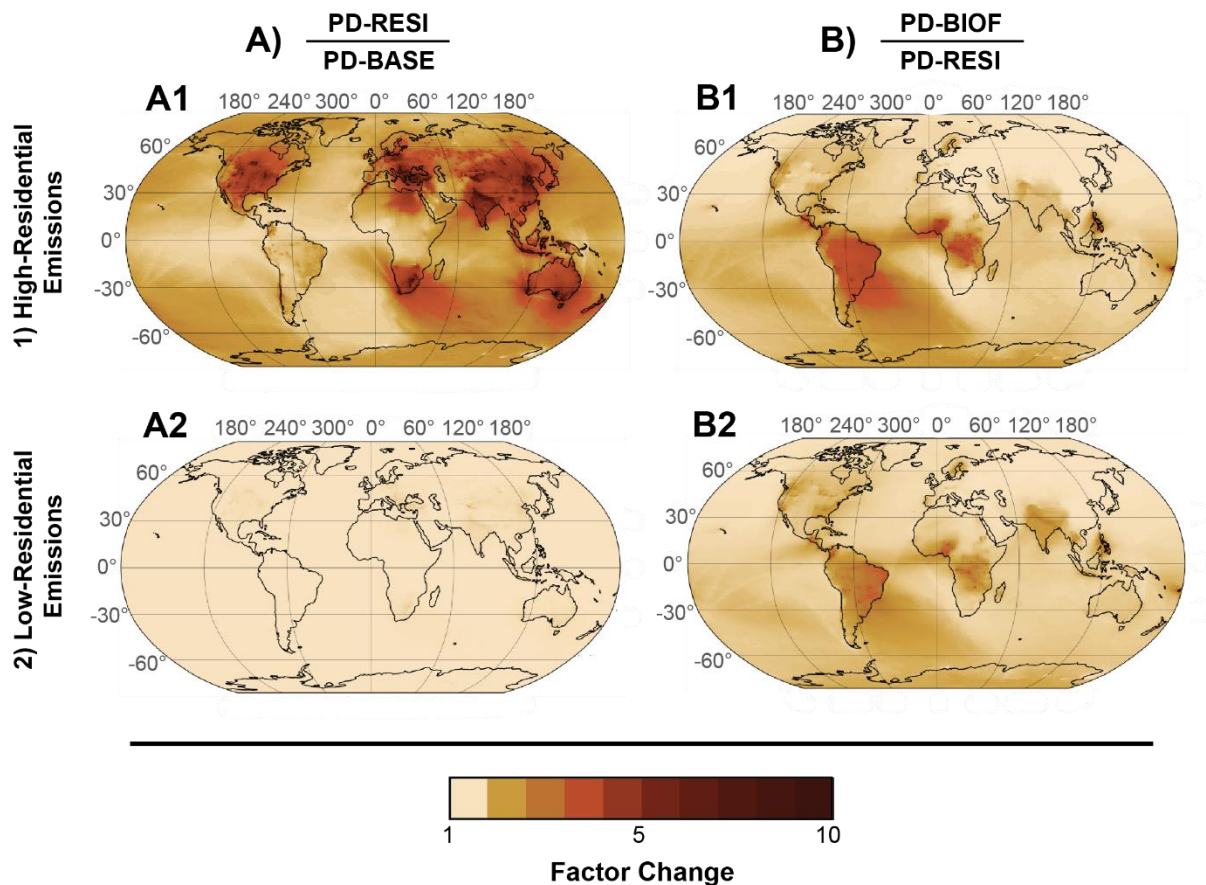
643 **Table 4.** Global soluble Fe deposition fluxes in Gg a^{-1} (relative contribution in %) to ocean
644 ecosystems for dust, wildfire, and anthropogenic combustion sources in the PD as simulated
645 by MIMI. Where ranges are reported, they reflect use of the low/high-residential emissions
646 inventories. Regional fluxes are reported in the Supplement (Table S10).

Model Simulation (case)	Dust	Wildfire	Anthropogenic Combustion	All Sources
-------------------------	------	----------	-----------------------------	-------------

PD-BASE	305 (83 %)	26 (7 %)	36 (10 %)	367 (100%)
PD-RESI	305 (76-83 %)	26 (6-7 %)	37-70 (10-17 %)	368-401 (100%)
PD-BIOF	305 (74-80 %)	26 (6-7 %)	49-81 (13-20 %)	379-412 (100%)

647

648 Despite large differences in residential coal emissions between our high- and low-
649 residential inventories, residential biofuel emissions by mass were identical between
650 inventories (72 Gg a⁻¹) because wood burning was already an isolated fuel source in our
651 inventory (Rathod et al., 2020). When using the high-residential inventory, impacts to soluble
652 Fe fluxes by biofuel were largely overshadowed by residential coal, but using the low-
653 inventory, changes to biofuel parameters in the model controlled increases in soluble Fe fluxes.
654 When we increased biofuel Fe solubility (PD-BIOF), soluble Fe fluxes to the ocean increased
655 by an additional 11 Gg a⁻¹, for a total increase of 13-44 Gg a⁻¹ from residential sources (coal +
656 biofuel) when compared to PD-BASE (Table 5). Changes to soluble Fe fluxes from biofuel
657 burning were most concentrated across the South Atlantic (Figure 3), likely due to the long
658 range transport of emissions from the Amazon rainforest and across the Congo River basin
659 where biofuel-burning in cook stoves is a common residential practice (García-López et al.,
660 2025; Stoner et al., 2021). We report a maximum change in soluble Fe fluxes using the high-
661 residential emissions inventory and PD-BIOF solubility parameters, with deposition to the
662 ocean doubling from 40 to 80 Gg a⁻¹ at the global scale (relative to PD-BASE; Table 5).



663

664 **Figure 3.** Relative changes to soluble Fe deposition fluxes using different 1-2) emission
 665 inventories and following modifications to A-B) Fe solubility parameters.

666

667 With soluble dust-Fe (310 Gg a^{-1}) and wildfire-Fe (30 Gg a^{-1}) fluxes, our different
 668 anthropogenic cases (an additional $40\text{-}80 \text{ Gg a}^{-1}$; Table 5) suggest that total soluble Fe fluxes
 669 to the ocean fall between 370 Gg a^{-1} (PD-BASE, both inventories) and 410 Gg a^{-1} (PI-BIOF,
 670 high-residential emissions inventory) at the global scale. These values fall within previous
 671 ranges of uncertainty as reported for Fe deposition fluxes to the ocean (Hamilton et al., 2023;
 672 Ito and Miyakawa, 2023), suggesting that solubility modifications tested herein align with
 673 previous Fe constraints within Earth System Models.

674

675 **Table 5.** Global anthropogenic combustion Fe emission and deposition fluxes (Gg a^{-1}) in the
 676 preindustrial (PI), present day (PD), and Future (FU), as simulated by MIMI to two significant
 677 figures. Where ranges are reported reflect use of the low/high-residential emissions inventories.

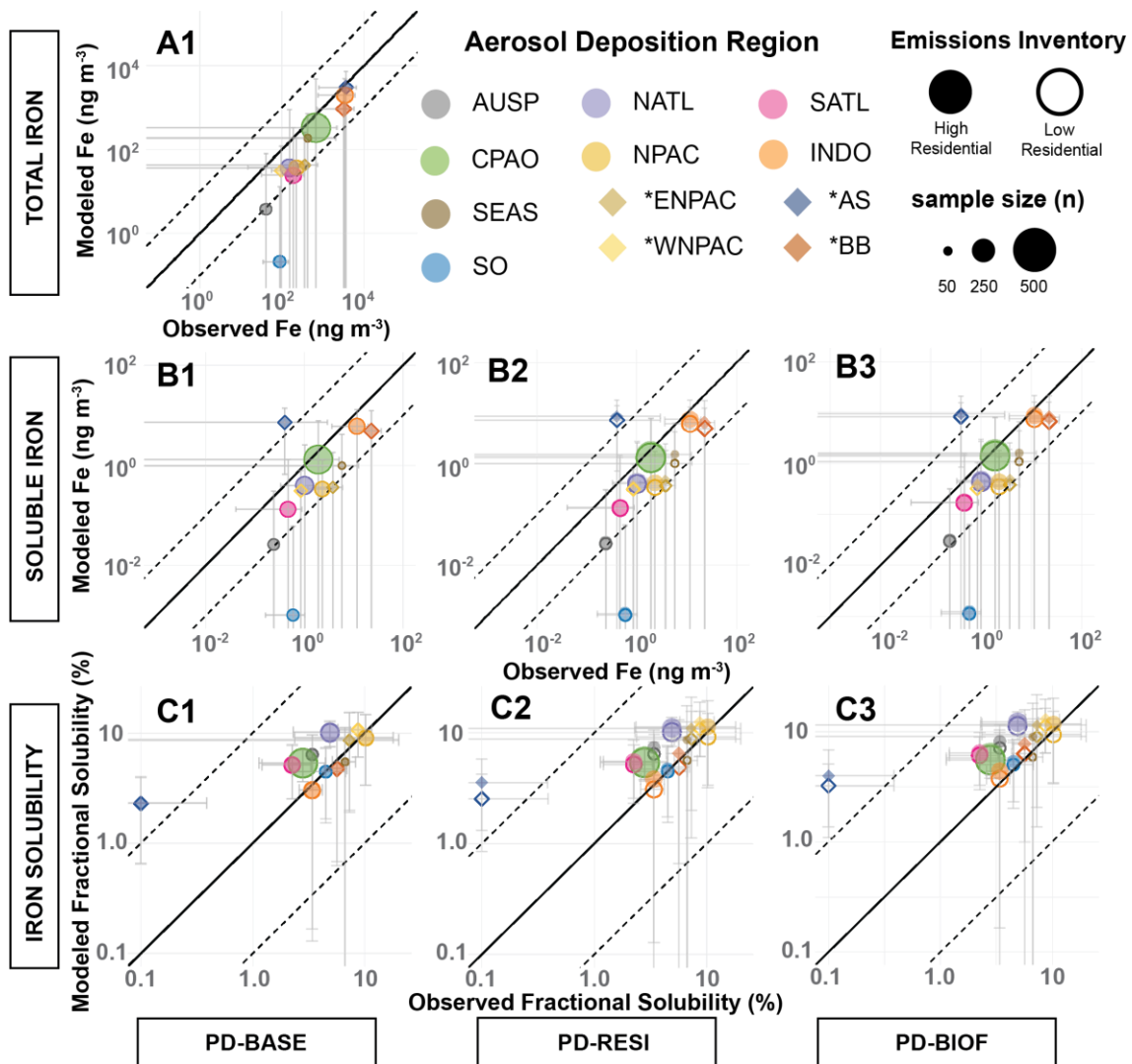
Emission Scenario	Model Simulation (case)	Fe Content	Global Emission	Global Deposition	Deposition to Ocean
PI (1750 CE)	PI-BASE	Total (Soluble)	0.80 (0.08)	0.80 (0.10)	0.30 (0.04)
	PI-BIOF	Total (Soluble)	0.80 (0.44)	0.80 (0.50)	0.30 (0.20)
PD (2010 CE)	PD-BASE	Total (Soluble)	2220 (20)	2220 (90)	590 (40)
	PD-BIOF	Total (Soluble)	2220 (60-170)	2220 (130-270)	590 (49-81)
FU (2050 CE)	MID-SSP370-BASE	Total (Soluble)	2400 (20)	2400 (90)	620 (40)
	MID-SSP370-BIOF	Total (Soluble)	2400 (180)	2400 (250)	620 (80)
FU (2100 CE)	END-SSP370-BASE	Total (Soluble)	1970 (20)	1970 (80)	510 (30)
	END-SSP370-BIOF	Total (Soluble)	1970 (90)	1970 (150)	510 (50)

678

679 3.3.2 Model-observation comparisons of total and soluble Fe concentrations

680 Comparison of modeled surface concentrations with regionally grouped, ship-based
 681 observations revealed generally good agreement between modeled and observed total and
 682 soluble aerosol Fe concentrations for all PD simulations (Figure 4). Modeled total Fe
 683 concentrations were slightly underpredicted when compared to observed values but remained
 684 well within one order of magnitude for each ocean region, with the exception of the Southern
 685 Ocean where total Fe was underestimated by several orders of magnitude. This aligns with
 686 previous efforts to model global fluxes of total and soluble aerosol Fe using MIMI v1.0 and
 687 other Earth System Models (Ito and Miyakawa, 2023; Ito et al., 2019; Liu et al., 2024). Current

688 hypotheses suggest that an Fe source, such as volcanism or mining, is not currently well
 689 represented in models, or alternatively, that limited observations are not representative of Fe
 690 conditions in the airshed of the Southern Ocean (Ito and Miyakawa, 2023; Liu et al., 2024).
 691



692
 693 **Figure 4.** Comparison of modelling and observational data: A) total Fe, B) soluble Fe, and C)
 694 Fe solubility; 1) PD-BASE, 2) PD-RESI, and 3) PD-BIOF. Data were aggregated over time
 695 and space as medians. Solid/hollow markers indicate use of the high/low-residential emissions
 696 inventories, respectively. Error bars represent spatiotemporal variance within each region. The

697 solid black line indicates a 1-to-1 relationship and the dashed lines represent deviation by ± 1
698 order of magnitude. Only PD-BASE is shown for total Fe **per consistency between cases.**

699

700 In **each** case, soluble Fe regression analyses followed a similar trend to total Fe, wherein
701 modeled averages were slightly lower than observed values and fell within one order of
702 magnitude, **apart from the** Southern Ocean and Arabian Sea (Figure 4, **Table S12**). In the PD-
703 RESI and PD-BIOF cases, soluble Fe concentrations increased **at the global scale, but the**
704 **degree varied by region.** At the global scale, **enhancing residential Fe emissions generally**
705 **improved model skill for soluble Fe concentrations (Figure 4), resulting** in an average increase
706 to modeled soluble Fe concentrations by $0.5 \pm 0.7 \text{ ng m}^{-3}$ **within each ocean region (Table S10).**
707 **For regions most influenced by residential coal burning, the improvement in model skill was**
708 **slightly higher using the high-emissions inventory, especially for Southeastern Asia (ΔRMSE**
709 **-0.5), the Bay of Bengal ($\Delta\text{RMSE} = -5.0$), and the eastern North Pacific (Figure 4; Table S12;**
710 **$\Delta\text{RMSE} = -0.1$).** For biofuel burning Fe, the emissions inventory had no effect, but **enhancing**
711 **Fe solubility most improved model skill** within the South Atlantic Ocean (Figure 4, **Table S12**
712 **$\Delta\text{RMSE} = -0.07$).** Complete summary statistics conveying impacts to model skill for soluble
713 **Fe concentrations simulated in each run are provided in the Supplement (Table S12).**

714 While the high-residential emissions inventory slightly improved estimates for soluble Fe
715 concentrations, the low-residential emissions inventory performed better when capturing
716 fractional solubility for regions influenced by residential coal (Figure 4). Despite these
717 noticeable variations between cases, ultimately, Fe solubilities calculated by the model were
718 aligned with observations **within ± 1 order of magnitude** for every region except for the Arabian

719 Sea wherein solubility was overestimated by 1-2 orders of magnitude (Figure 4). In previous
720 MIMI-validation efforts (Hamilton et al., 2019), observational data from the Arabian Sea and
721 Bay of Bengal were aggregated as the Indian Ocean and this result was not flagged. While both
722 basins receive substantial anthropogenic aerosol from India, dust from the Middle East more
723 strongly influences the Arabian Sea, and the Bay of Bengal is more strongly affected by
724 anthropogenic emissions across Southeastern Asia (Bali et al., 2019; Guieu et al., 2019).

725 In general, we found that regions with higher dust inputs more often overshot
726 measurements when compared to regions with less dust deposition and higher relative impact
727 by anthropogenic emissions (Figure 4; Table S12). In each of our modified solubility cases
728 (PD-RESI and PD-BIOF), Fe solubility for southeastern Asia, the Bay of Bengal, and the North
729 Pacific increased, but the regions heavily impacted by dust remained relatively unchanged
730 (Figure 4).

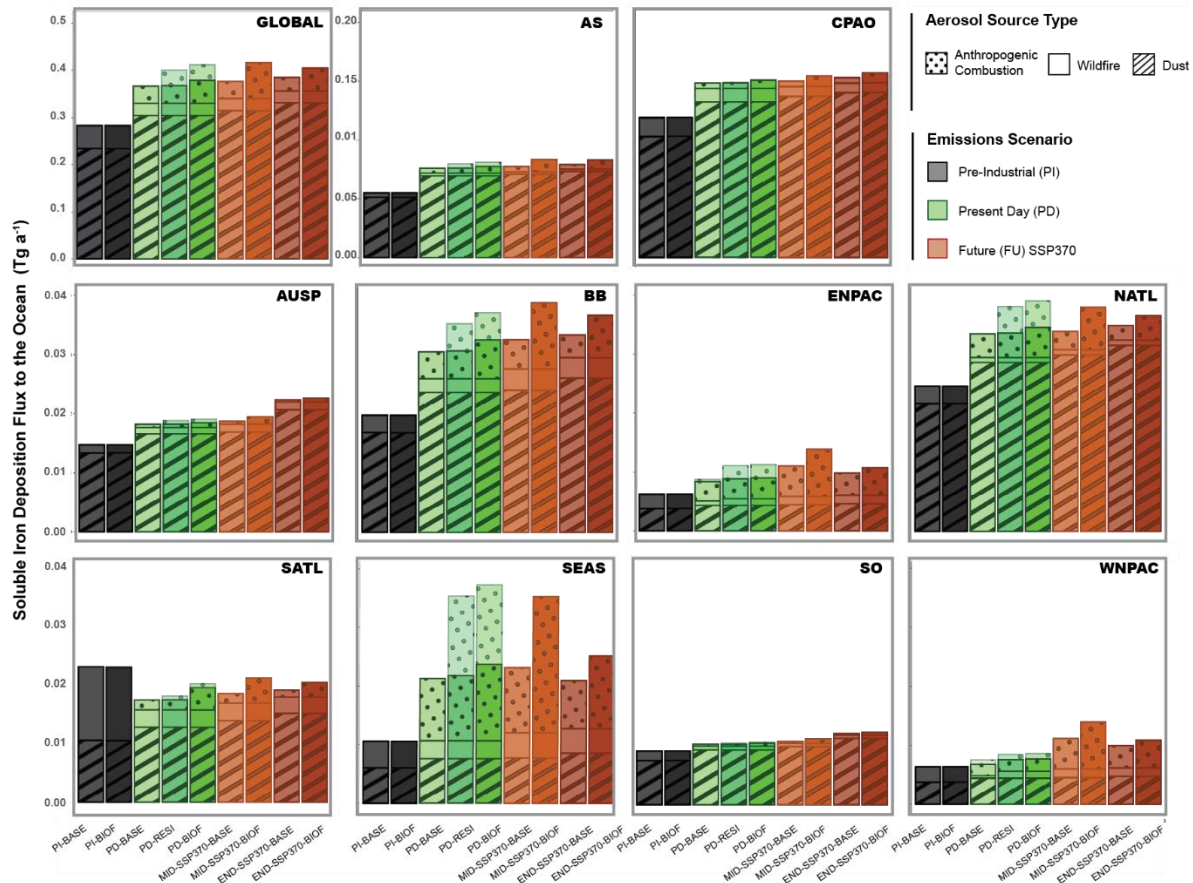
731 Withstanding source-apportioned measurements of residential coal or biofuel aerosol in
732 our observational dataset, we performed additional model-observation comparisons only using
733 measurements collected in ocean regions downwind of strong residential burning influences.
734 These regions were defined as model-resolution grid cells wherein soluble Fe fluxes increased
735 by 100% or more in the PD-BIOF simulations (Figure 3). However, this reduced median-
736 aggregated observational data points from n=990 to n=25, limiting statistical capacity to
737 constrain model fluxes. When using the smaller observational dataset, model-observational
738 comparisons for Total Fe, soluble Fe, and solubility mirrored agreement trends using the larger
739 dataset (Figure 4); those regression analyses are provided in the Supplement (Figures S4-S5)
740 with results from the PD-IND simulation.

741 Although anthropogenic soluble Fe fluxes to marine ecosystems increased by 34% to 123%
742 (<1% to 10% all sources) at the global scale and by 8% to 165% (1% to 21% all sources) for
743 individual regions (Figures 3 and 5), changes to model skill were ultimately modest (Figure 4).
744 The sizeable changes in fluxes with minimal changes to model skill further reinforce the current
745 limitations of ship-based observations in capturing representative soluble Fe fluxes,
746 particularly in the under-sampled Southern Hemisphere and in regions influenced by residential
747 coal and biofuel burning. Future efforts should prioritize expanding the spatial coverage of
748 measurements in these regions to improve model accuracy and understanding of possible
749 anthropogenic influence on remote marine biogeochemistry.

750 3.3.3 Soluble Fe under PI and FU emission scenarios

751 PI model simulations serve as a valuable reference point in understanding the specific
752 implications of anthropogenic perturbation to the Earth system. For most regions, soluble Fe
753 fluxes increased between the PI and PD eras (Figure 5), largely attributed to steadily growing
754 anthropogenic combustion emissions and industrial activities over time. Dust and wildfire Fe
755 emissions were also distinct between the PI and PD, due to climatic and land-use change
756 induced feedbacks that have altered global precipitation patterns and dust suspension
757 (Hamilton et al., 2018; Kok et al., 2023; Li et al., 2019; Mahowald et al., 2010). At the global
758 scale, we estimated that current soluble Fe fluxes to marine ecosystems exceed PI fluxes by
759 36-70 Gg a⁻¹, apart from the South Atlantic, wherein soluble Fe fluxes have decreased since
760 the PI era by 2.8-5.6 Gg a⁻¹ (Figure 5). This decrease was likely attributed to reduction in
761 wildfire burned area over past decades, particularly in sub-Saharan Africa (Andela et al., 2017;

762 Jones et al., 2022). Previous work has suggested that wildfire activity during the PI era
 763 exceeded current wildfire regimes at the global scale (Hamilton et al., 2018).



764
 765 **Figure 5.** Deposition fluxes of soluble aerosol Fe to marine ecosystems at the global and
 766 regional scale. Deposition fluxes are source-apportioned (dust, wildfire burning, and
 767 anthropogenic combustion) and provided for each case with distinct solubility parameters. For
 768 the PD, transparent bars represent the high-residential emissions inventory and opaque
 769 represent the low. Conversely, for the FU, only the high-residential inventory was applied, and
 770 the bars are opaque.

771
 772 Regardless of the inventory applied, we observed the largest increases between PI and PD
 773 soluble Fe fluxes in Southeastern Asia, the Bay of Bengal, and the North Atlantic. In

774 Southeastern Asia anthropogenic activity has specifically driven, and is projected to drive,
775 changes to future soluble Fe fluxes (Figure 5). This was the only ocean region wherein
776 anthropogenic sources were comparable (low residential inventory) or exceeded (high
777 residential inventory) dust sources of soluble Fe (Figure 5). When using the high-residential
778 inventory, anthropogenic combustion aerosol constituted up to 72% of all soluble Fe fluxes to
779 marine ecosystems in this region, a 21% increase from PD-BASE. Although dust was still the
780 largest source of Fe to the Bay of Bengal and North Atlantic, anthropogenic combustion also
781 strongly influenced soluble Fe delivery in these regions (Figure 5). When using the high-
782 residential emissions inventory with the -BIOF solubility parameters, the relative contribution
783 by anthropogenic emissions doubled in these regions (Table S10).

784 The inventory and solubility parameters used in each case revealed important implications
785 for projected trends of soluble Fe fluxes moving into the second half of the 21st century. Under
786 the SSP370 FU emissions scenario, anthropogenic Fe fluxes were projected to reach their
787 maxima by 2050 for most deposition regions and then decrease to values at or below current
788 PD conditions by 2100 (Figure 5). However, this trend did not hold for regions within the
789 Southern Hemisphere (Australia/South Pacific, Central Pacific/Atlantic, and Southern Ocean)
790 where soluble aerosol Fe fluxes were projected to continually increase through the end of the
791 century (Figure 5). This was not due to direct changes in anthropogenic combustion emissions,
792 but rather due to changes in dust emissions as the primary source of soluble Fe to the Southern
793 Hemisphere (Figure 5).

794 By the end of the century under SSP370, PD-BASE simulations suggest that soluble Fe
795 deposition to global marine systems will increase slightly from 367 to 369 Gg a⁻¹ (~1% increase)

796 by 2100. However, when considering higher residential Fe emissions with higher solubilities
797 representing new upper bound, projected changes to soluble Fe fluxes between PD/MID-
798 SSP370 and MID-SSP370/END-SSP370 was reversed at the global scale and for many regions,
799 with projected decreases by the end of the century (~2% decrease; Figure 5). The projected
800 losses in soluble Fe between the middle and end of the 21st century was especially apparent for
801 the Bay of Bengal (2-6%), the eastern and western North Pacific (12-29%), and across
802 Southeastern Asia (11-41%) (Figure 5), suggesting that various marine ecosystems could face
803 a more significant deviation from current soluble Fe supply than has previously been
804 represented in Earth System Models.

805 Projected changes in soluble Fe fluxes by 2100 under FU emission scenarios, including
806 SSP370, have strong implications for the spatiotemporal distribution of net marine primary
807 productivity, mostly in Fe limited regions. For example, although we demonstrated that
808 anthropogenic emissions most greatly influenced Fe dynamics across Southeastern Asia, it is
809 important to note that primary production in this ocean region is not typically limited by Fe
810 (Bazzani et al., 2023), so ecosystem-level effects by atmospheric Fe are less likely to be
811 observed therein. However, similar to Southeastern Asia, we found that changes to
812 anthropogenic emissions more strongly impacted soluble Fe fluxes in the eastern North Pacific
813 when compared to dust and wildfire sources. Recent work suggests that the atmospheric supply
814 of anthropogenic Fe has already shifted phytoplankton bloom dynamics in the open ocean by
815 accelerating the seasonal uptake of upwelled nitrogen in HNLC regions, including North
816 Pacific (Hawco et al., 2025). Such regions are anticipated to be especially sensitive to changes

817 in anthropogenic Fe given that they are historically limited by trace metals including Fe
818 (Bazzani et al., 2023; Moore et al., 2013; Nishioka and Obata, 2017).

819 **In addition to our findings,** diverse lines of evidence suggest that half of the soluble Fe
820 flux to the North Pacific comes from Asian anthropogenic sources (Hamilton et al., 2019;
821 Hamilton et al., 2020a; Hawco et al., 2025; Rathod et al., 2020). Li et al. (2024) found that the
822 magnitude of chlorophyll-a response to Fe deposition off the coast of China was lowered by a
823 factor of 4 during COVID-19 in March 2020 when anthropogenic emissions across East Asia
824 were substantially reduced. The authors speculated that a reduction in soluble Fe from
825 anthropogenic activities, either via the primary emission of soluble Fe or via a reduction in Fe
826 solubilization via co-emitted acidic species (e.g., SO_x), resulted in a lessened supply of soluble
827 Fe delivered during the deposition event. Moreover, using Fe isotopes to trace source origins
828 of atmospheric Fe, Hawco et al. (2025) recently showed that the springtime delivery of
829 anthropogenic Fe could be one major factor driving observed seasonal and geographic shifts to
830 the North Pacific transition zone, a highly productive boundary in the North Pacific. **Isotopic**
831 **signatures capable of distinguishing residential coal combustion from other anthropogenic**
832 **combustion sources have not yet been identified, but** our findings suggest that residential coal
833 burning is an especially important source of soluble Fe to the North Pacific and the South China
834 Sea, and across southeastern Asia. Accordingly, we find that projected losses of anthropogenic
835 emissions over the course of this century will **most** greatly influence nutrient dynamics in these
836 key marine ecosystems.

837 **4 Conclusions**

838 Anthropogenic activity has added a multitude of new aerosol Fe sources to the atmosphere.
839 Understanding how these new sources alter **the magnitude and timing of soluble Fe aerosol**
840 **fluxes to the ocean** aids understanding of **how human activity is** changing marine primary
841 productivity and ocean ecosystem **functions** within the Anthropocene. However, estimating the
842 contribution of anthropogenic emissions to soluble aerosol Fe fluxes is challenging given the
843 wide variety of sources, **each with their own distinct physicochemical profiles. Lack of**
844 **observational constraints** leads to large variation across different modeling studies on the
845 magnitude of the deposition flux **from anthropogenic sources. We address some of these**
846 **uncertainties in this study by** measuring **the** Fe content and solubility of aerosol Fe from several
847 important anthropogenic sources, including a first assessment of the contribution from **two**
848 **major** biofuels, **namely residential coal and wood. We** find that median Fe solubilities vary by
849 greater than three orders of magnitude **across fuel types**, from 0.03% for power plant coal fly
850 ash to 55.87% for biofuel burning aerosol.

851 To understand the impact of increasing **anthropogenic** source **representations** of fractional
852 Fe **solubility**, we **created two new emission inventories that distinguished residential from**
853 **industrial sources, and further** refined Fe solubility parameters for **each sources** within **MIMI**,
854 an atmospheric Fe module embedded within the CESM2.

855 **At the global scale, we** found that current (PD) **soluble Fe fluxes to the ocean from**
856 **anthropogenic sources could exceed current modeled values by 34% to 123%. This represents**
857 **an increase of over 3 orders of magnitude from the PI when biofuel sources are assumed to be**
858 **the only source of anthropogenic Fe (Hamilton et al. 2020a). Projected (FU) soluble Fe fluxes**

859 from anthropogenic sources remain similar to the PD through to the middle of the century
860 before declining by up to 38% at the end of the century under SSP370.

861 At the regional scale, including residential coal and biofuel burning sources in the model
862 resulted in the most notable impacts for the Bay of Bengal, across Southeast Asia, and
863 throughout the North Pacific and North Atlantic (i.e., regions strongly influenced by nearby
864 continental anthropogenic activity). However, these regions are generally under-sampled in
865 terms of shipborne aerosol Fe observations, and therefore, to reduce the largest source of
866 uncertainty, more measurements are needed in regions downwind of residential Fe sources to
867 better constrain the contribution of human activity on global biogeochemical cycles.

868

869 **Data availability.**

870 Experimental data can be found in the manuscript or the Supplement, or are available at
871 <https://doi.org/10.5281/zenodo.17766592> (Li, 2025). Modeling output data, coding scripts are
872 available at <https://github.com/haleyplaas/CombustionFe> and emission inventories are located
873 at <https://doi.org/10.5281/zenodo.17903186>.

874 **Competing interests.**

875 At least one of the (co-)authors is a member of the editorial board of Atmospheric
876 Chemistry and Physics.

877 **Author contribution.**

878 MT initiated this study; MT and DSH designed this study and secured funding resources;
879 RL, YZ, YC and TZ conducted experimental work; HEP, SR and DSH conducted modeling
880 work; YY provided key samples used in this work and contributed to data analysis; RL and

881 HEP analyzed the results; RL, HEP, DSH and MT wrote the manuscript; all the authors
882 reviewed and approved the manuscript.

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889

890

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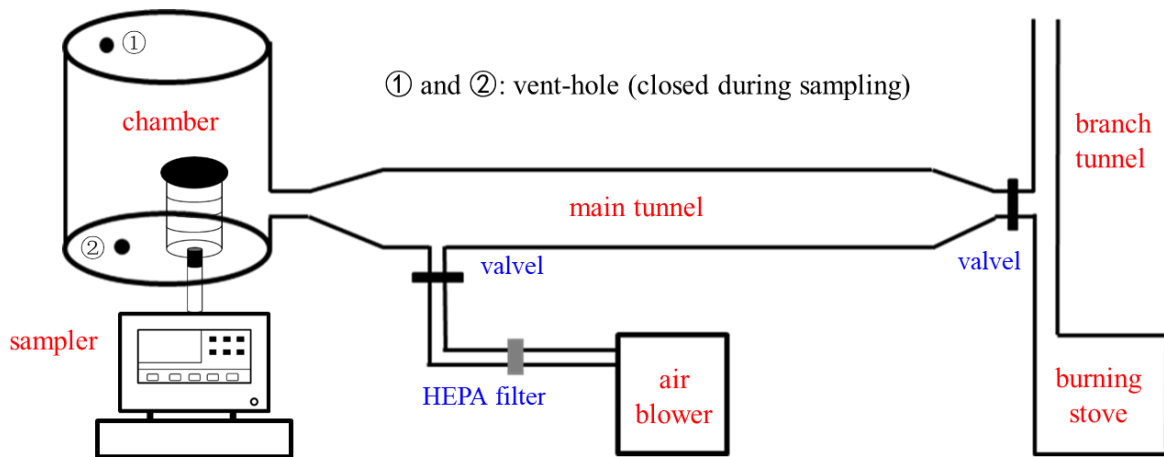
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1 **Text S1 Experimental methods and results**

2 **Text S1.1 Experimental methods**

3 Figure S1 show the schematic diagram of the apparatus used to generate and collect
4 aerosol emitted by residential coal and biofuel combustion, and Figure S2 displays one of its
5 photos taken during the sampling.



6
7 **Figure S1.** Schematic diagram of the apparatus used in our work to collect aerosol particles
8 emitted by domestic coal and biofuel burning.

9
10 As shown in Figure 1, coal and biofuel were burned in a commercial cook stove, which is
11 widely used in rural areas in China. Exhaust in the chimney, generated by coal and biofuel
12 combustion, could go directly to the ambient air, or alternatively it could enter a horizontally-
13 mounted long metal tube (inner diameter: 30 cm; length: 200 cm). After exiting the long metal
14 tube, the exhaust entered a vertically-mounted chamber (inner diameter: 45 cm; height: 50 cm),
15 and PM_{2.5} was collected onto pre-cleaned Whatman 41 (W41) cellulose filters (diameter: 88
16 mm) via a medium volume aerosol sampler (TH-150C, Tianhong Co.) operated at a flow rate
17 of 100 L/min. Filters were cleaned to reduce background using the procedure detailed
18 elsewhere (Zhang et al., 2022). Aerosol sampling was stopped automatically when the pressure

19 dropped to the threshold because of accumulation of aerosol on the filter, and sampling times
20 ranged from a few to tens of minutes, varying for fuel types. Between combustion experiments,
21 the tube was flushed to remove smoke generated from the previous combustion experiment.



22
23 **Figure S2.** A photo of the apparatus used in our work to collect aerosol particles emitted by
24 domestic coal and biofuel burning.

25
26

27 **Text S1.2 Experimental results**

28 **Text S1.2.1 Fe content of municipal waste fly ash, oil fly ash and oil bottom ash**

29 For the three municipal waste fly ash samples we investigated, Fe content ranged from
30 3.9 to 29.7 mg/g, with average and median values being 18.7 ± 13.3 and 22.6 mg/g (Table S5).
31 Several previous studies measured Fe content in municipal waste fly ash (Table S6). For
32 example, the average Fe content were measured to be 18.0 ± 13.3 mg/g ($n = 3$) and 23.1 mg/g
33 ($n = 1$) in two studies (Cobo et al., 2009; Raclavská et al., 2017), very similar to our results;
34 another four studies (Funari et al., 2017; Liu et al., 2009; Wu and Ting, 2006; Wu et al., 2012)
35 reported lower Fe content, ranging from 5.2 to 10.9 mg/g; some other studies (Bayuseno and
36 Schmahl, 2011; Lin et al., 2003; Wan et al., 2006; Zhang et al., 2011) also reported slightly
37 higher Fe content, ranging from 27.1 to 34.3 mg/g. In summary, most studies suggest that Fe
38 content in municipal waste fly ash are around 20 mg/g, and it has been set to 18.8 mg/g in a
39 modeling study (Rathod et al., 2020), being consistent with experimental results.

40 Fe content in the two oil fly ash samples we examined were measured to be 9.1 and 18.3
41 mg/g (Table S5), and the average value was determined to be 13.7 ± 4.6 mg/g. The Fe content
42 was measured to be 15.0 mg/g for one oil fly ash sample (Celo et al., 2015), close to the value
43 we reported. In another two studies (Agrawal et al., 2008; Sippula et al., 2014), the average Fe
44 content was measured to be 1.98 ± 0.35 ($n = 4$) and 1.60 ± 1.21 mg/g ($n = 14$), lower than our
45 result. In a modeling study (Rathod et al., 2020), the Fe content was set to 10 mg/g for oil fly
46 ash, being consistent with the experimental results reported by our work and Celo et al. In
47 addition, in our work the Fe content was measured to be 191 mg/g for one heavy oil bottom
48 ash sample, much higher than that for oil fly ash.

49 **Text S1.2.2 Fe solubility of municipal waste fly ash, oil fly ash and oil bottom ash**

50 Fe solubility in acetate buffer (pH: 4.3) was determined to range from 0.58% to 2.41% for
51 municipal waste fly ash (Table S5), with average and median values being $1.51 \pm 0.92\%$ and
52 1.54%, respectively. Few previous studies measured Fe solubility for municipal waste fly ash.
53 Fe solubility was estimated to be $<2\%$ for municipal waste fly ash when combustion
54 temperature exceeded 1100 K (Rathod et al., 2020), agreeing with our experimental results.

55 Fe solubility in acetate buffer (pH: 4.3) was determined to be 11.70% and 13.43% for the
56 two oil fly ash samples we examined (Table S5), with an average value of $12.56 \pm 0.87\%$. In
57 previous work, Fe solubility was measured to be 35.7% at pH of 4.7 (Desboeufs et al., 2005)
58 and 70% in deionized water (Schroth et al., 2009), both higher than our results. Although Fe
59 solubility measured in different studies showed some variations, all the studies suggested that
60 oil fly ash exhibited very high Fe solubility. Moreover, Fe solubility in acetate buffer (pH: 4.3)
61 was measured in our work to be 25.47% for one heavy oil bottom ash. Oil fly ash was emitted
62 by high temperature combustion but showed high Fe solubility. This is probably because heavy
63 oil has high sulfur content, leading to the formation of sulfuric acid in combustion that can
64 condense onto co-emitted Fe oxide particles and form highly soluble Fe sulfate (Rathod et al.,
65 2020; Sippula et al., 2009).

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67

68 **Table S1.** Fe content and solubility for power plant coal fly ash samples (each from a coal
 69 power plant located in a different province in China) examined in this work.

sample	Fe content ($\mu\text{g/g}$)	Fe content (mg/g)	Fe solubility (%)
1	3.65×10^4	36.5	0.013
2	2.07×10^4	20.7	0.038
3	4.41×10^4	44.1	0.028
4	2.65×10^4	26.5	0.029
5	3.44×10^4	34.4	0.008
6	2.50×10^4	25.0	0.029
7	3.82×10^4	38.2	0.128
8	10.4×10^4	103.8	0.014
9	2.35×10^4	23.5	0.028
10	3.86×10^4	38.6	0.002
11	5.37×10^4	53.7	0.134
12	3.77×10^4	37.7	0.029
13	2.41×10^4	24.1	0.018
14	3.19×10^4	31.9	0.008
15	2.41×10^4	24.1	0.057
16	2.58×10^4	25.8	0.021
17	4.59×10^4	45.9	0.073
18	2.69×10^4	26.9	0.044
19	5.57×10^4	55.7	0.036
20	3.95×10^4	39.5	0.132
21	3.50×10^4	35.0	0.021
22	6.35×10^4	63.5	0.041
23	3.97×10^4	39.7	0.146
24	4.42×10^4	44.2	0.091
25	2.41×10^4	24.1	0.013
26	4.17×10^4	41.7	0.032
27	2.31×10^4	23.1	0.020
28	5.33×10^4	53.3	0.172
29	2.23×10^4	22.3	0.070
30	2.76×10^4	27.6	0.024
31	2.17×10^4	21.7	0.024

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71

72 **Table S2.** Fe content and solubility for domestic coal combustion aerosols examined in this
73 work.

sample	Fe content ($\mu\text{g/g}$)	Fe solubility (%)
anthracite	32	52.04
	26	27.05
semibituminous coal	35	11.71
	45	12.66
	62	34.52
	26	100.00
bituminous coal	101	7.03
	43	43.84
	25	29.86
	42	14.26

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78 **Table S3.** Fe content and solubility for steelwork fly ash samples examined in this work.

sample	Fe content ($\mu\text{g/g}$)	Fe content (mg/g)	Fe solubility (%)
1	3.47×10^5	346.5	0.022
2	5.47×10^5	546.8	0.007
3	4.46×10^5	446.2	0.022
4	4.34×10^5	434.0	0.012
5	9.79×10^4	97.9	0.069
6	1.34×10^4	13.4	6.928
7	1.28×10^4	12.8	0.191
8	2.09×10^4	20.9	0.395
9	6.45×10^5	644.9	0.011
10	2.13×10^5	213.1	4.024
11	5.66×10^5	565.6	3.126
12	3.94×10^5	393.8	1.980
13	4.48×10^5	448.3	0.042
14	2.59×10^4	25.9	0.923
15	9.74×10^4	97.4	10.640
16	1.97×10^5	197.1	0.055
17	3.03×10^5	302.8	0.035
18	5.97×10^5	596.8	0.010
19	4.73×10^5	472.7	0.022
20	4.04×10^5	404.3	0.014
21	7.32×10^5	732.2	0.013
22	3.58×10^4	35.8	0.158
23	9.19×10^5	918.9	8.589
24	4.46×10^5	446.2	0.050
25	5.81×10^3	5.8	1.983
26	6.07×10^5	607.4	0.180
27	2.60×10^4	26.0	0.068
28	5.80×10^3	5.8	0.158
29	7.38×10^3	7.4	0.064

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82 **Table S4.** Fe content and solubility for biofuel burning aerosols examined in this work.

sample	Fe content ($\mu\text{g/g}$)	Fe solubility (%)
wheat	21	60.14
	8	100.00
	6	26.09
	12	71.87
rice	n. a.	40.52
	28	47.06
	20	4.34
corn	3	44.83
	2	100.00
	3	100.00
	15	77.52
	7	88.36
rape	70	54.34
	6	57.39
cogongrass	11	89.41
	10	85.33
	19	86.46
	71	37.56
China fir	3	5.75
	3	43.64
	12	20.01
pine	72	67.55
	101	100.00
	40	65.91
poplar	18	2.86
pine needle	13	24.01
	21	27.06
	15	41.86

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84

85 **Table S5.** Fe content and solubility for municipal waste fly ash, oil fly ash and oil bottom ash
 86 samples examined in this work. WP-2 is a fly ash sample obtained from an electrostatic
 87 precipitator in a waste incineration plant in Shanghai, China (Li et al., 2021), and BCR-176R
 88 and BCR-615 are certified reference materials provided by the Institute for Reference Materials
 89 and Measurements.

sample	Fe content ($\mu\text{g/g}$)	Fe content (mg/g)	Fe solubility (%)
municipal waste fly ash			
WP-2	3.87×10^3	3.9	0.58
BCR-176R	2.26×10^4	22.6	2.41
BCR-615	2.97×10^4	29.7	1.54
oil bottom ash	1.91×10^5	191.5	25.47
oil fly ash			
diesel oil	1.83×10^4	18.3	11.70
heavy fuel oil	9.06×10^3	9.1	13.43

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92 **Table S6.** Summary of Fe content (mg/g) for anthropogenic and combustion aerosol Fe determined in our present study and previous work (*n*:
 93 number of samples examined).

sample type	size range	<i>n</i>	range	average	median	Reference
power plant		31	20.7-103.8	37.2±16.8	35.0	This work
coal fly ash		3	16.0-52.0	33.0±18.0	31.0	Baldo et al. (2021)
		7	21.8-205.1	65.9±67.4	34.4	Goodarzi (2006)
		1		46.7		Meij (1994)
		23	18.2-112.0	57.8±22.7	52.5	Moreno et al. (2005)
		4	7.7-97.3	54.3±39.5	56.0	Jankowski et al. (2006)
		4	58.9-101.0	81.1±19.4	82.3	Dutta et al. (2009)
		4	27.0-119.0	86.0±43.0	97.5	Fu et al. (2012)
	7	38.3-98.6	62.1±26.7	43.2	Li et al. (2022)	
domestic coal	PM _{2.5}	10	0.025-0.101	0.044±0.023	0.038	This work
combustion aerosol	PM _{2.5}	3		0.048±0.035		Patil et al. (2013)
	PM _{2.5}	4		0.671±0.023		Watson et al. (2001)
	PM _{2.5}	5		0.7±0.1		Zhang et al. (2012)
	PM ₁₀	3		0.061±0.044		Patil et al. (2013)
steelwork fly ash		29	5.8-918.9	312.6±246.1	346.5	This work
		1		358.9		Souza et al. (2010)
		1		369.3		Vieira et al. (2013)
		1		312.2		Silva et al. (2019)
		4	288.2-340.3	329.1±22.6	324.4	Alizadeh and Momeni (2016)
				280-380		Hagni et al. (1991)
		1			86.0	Stathopoulos et al. (2013)
	1			128.1	Xia and Picklesi (2000)	

		1		150.8		Loaiza et al. (2017)
		1		286.5		Laforest and Duchesne (2006)
		1		284.6		Alsheyab and Khedaywi (2016)
		1		238.7		Li et al. (2023)
		4	234.1-361.1	267.3±4.8	283.6	Al-Negheimish et al. (2021)
		1		489.6		Machado et al. (2006)
		2	430-470	450±20	450	Patil et al. (2013)
		10	8.2-720			Hleis et al. (2013)
		1		515.0		Ye et al. (2021)
Biofuel burning aerosol	PM _{2.5}	27	0.002-0.101	0.023±0.026	0.013	This work
	PM _{2.5}	3		0.024±0.017		Patil et al. (2013)
	PM ₂	2		0.090	0.090	Hildemann et al. (1991)
	PM _{2.5}	3		0.167±0.259		Watson et al. (2001)
	PM _{2.5}	4		0.180±0.196		Watson et al. (2001)
	PM _{2.5}	5	0.031-0.615	0.162	0.115	Hedberg et al. (2002)
	PM _{2.5}	1		0.440		Alves et al. (2011)
	PM _{2.5}	4		0.400±0.100		Zhang et al. (2012)
	PM ₁₀	4	0.250-1.70	0.723±0.661	0.470	Schmidl et al. (2008)
municipal waste fly ash		3	3.9-29.7	18.7±13.3	22.6	This work
		3	7.8-33	18.0±13.3	13.2	Raclavská et al. (2017)
		1		23.1		Cobo et al. (2009)
		1		5.2		Wu and Ting (2006)
		1		5.5		Funari et al. (2017)
		1		10.5		Wu et al. (2012)
		1		10.9		Liu et al. (2009)
		1		29.4		Zhang et al. (2011)
		1		33.8		Wan et al. (2006)

	1		34.3		Lin et al. (2003)
	1		37.1		Bayuseno and Schmahl (2011)
oil fly ash	2	9.1-18.3	13.7±4.6	13.7	This work
	7		15.0		Celo et al. (2015)
	4	1.55-2.36	1.98±0.35	2.10	Agrawal et al. (2008)
	14	0.331-4.46	1.60±1.21	1.16	Sippula et al. (2014)
oil bottom ash	1		191	191	This work

95 **Table S7.** Summary of Fe solubility (%) for anthropogenic and combustion aerosol Fe determined in our present study and previous work (*n*:
 96 number of samples examined; pH: acidity of the leaching solution).

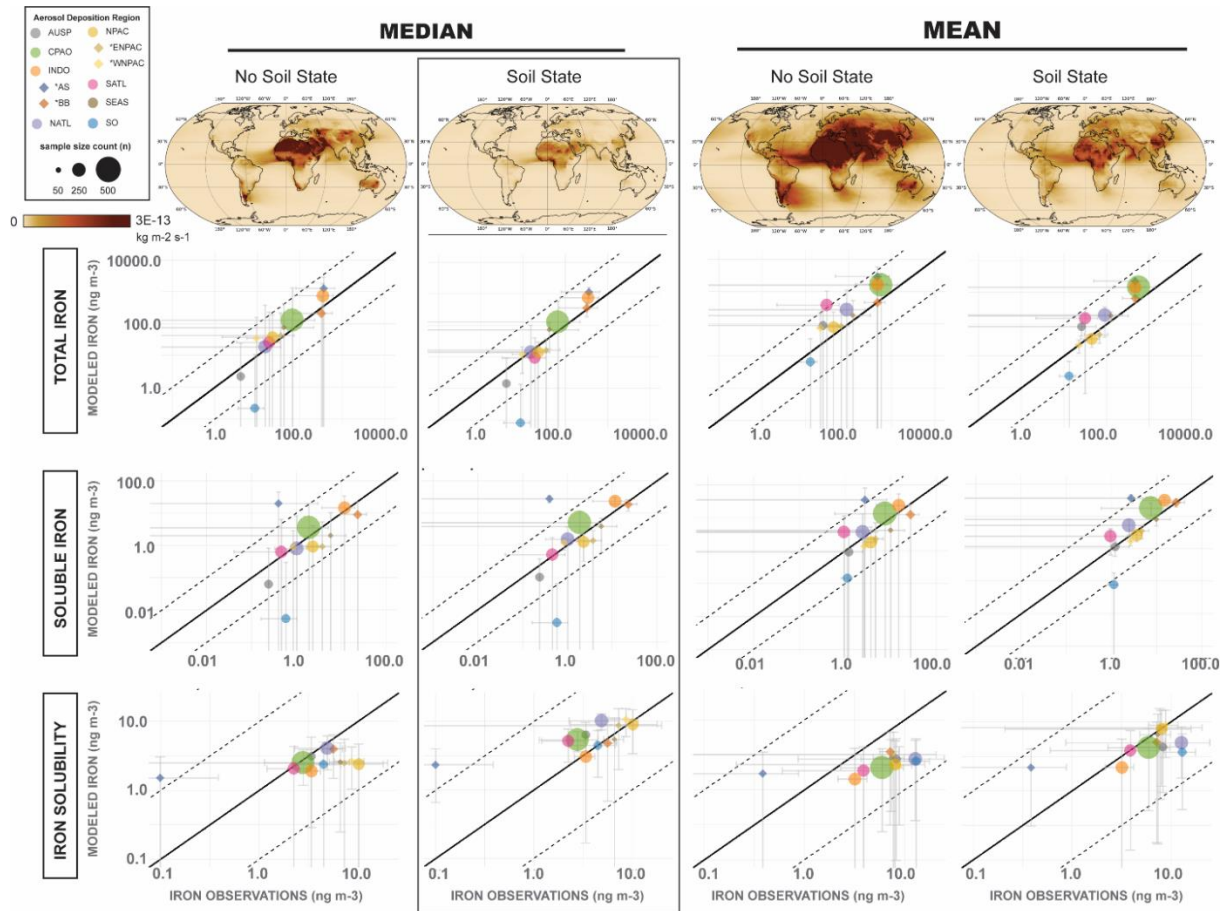
sample type	size range	<i>n</i>	pH	range	average	median	Reference
power plant		31	4.3	0.002-0.17	0.05±0.05	0.03	This work
coal fly ash		1	~6		0.06		Oakes et al. (2012)
		1	~4.7		0.2		Desboeufs et al. (2005)
		7	4.3	0.09-0.87	0.24±0.28	0.13	Li et al. (2022)
		7	~6	0.02-0.75	0.16±0.26	0.06	Li et al. (2022)
domestic coal combustion aerosol		10	4.3	7.03-100	33.30±27.71	28.45	This work
steelwork fly ash		29	4.3	0.007-10.64	1.37±2.77	0.07	This work
biofuel burning aerosol		28	4.3	2.86-100	56.07±30.95	55.87	This work
municipal waste fly ash		3	4.3	0.58-2.41	1.51±0.92	1.54	This work
oil fly ash		2	4.3	11.70-13.43	12.56±0.87	12.56	This work
		1	4.7		35.7		Desboeufs et al. (2005)
		1	~6		70%		Schroth et al. (2009)
oil bottom ash		1	4.3		25.47%		This work

97

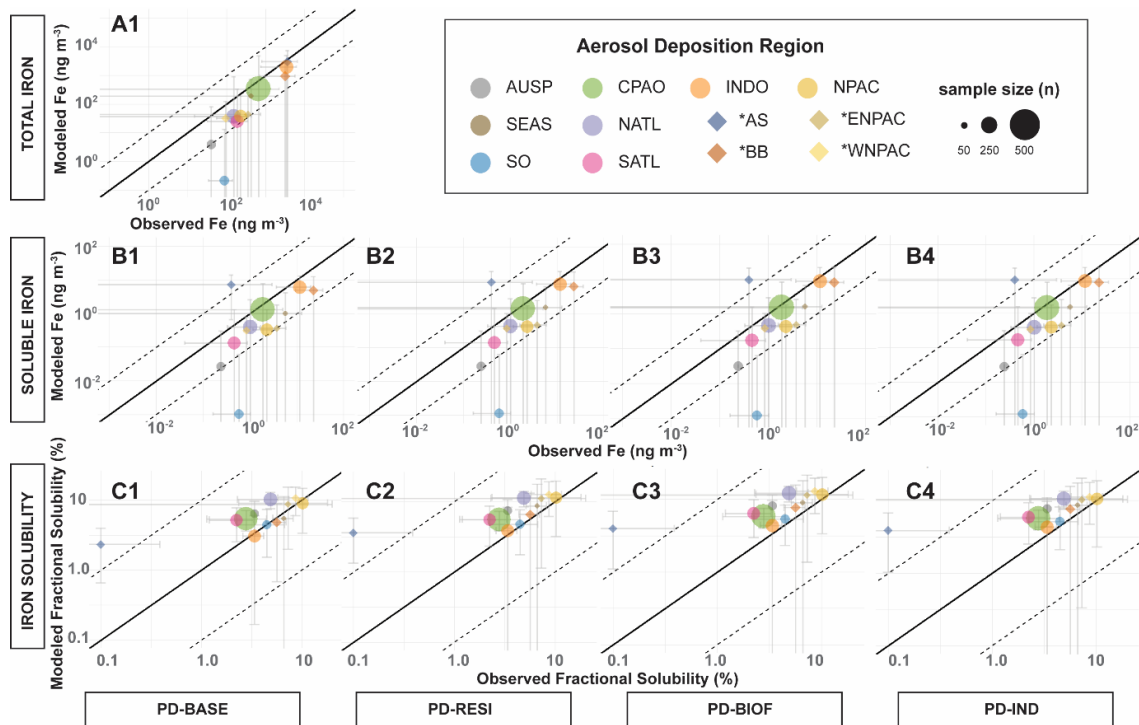
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100 **Text S2 Modeling methods and results**



101
 102 **Figure S3.** Model-observation comparisons ($n = 1624$) of surface aerosol Fe concentrations
 103 using two different dust flux schemes (soil state submodule included versus no soil state
 104 submodule). As indicated by the black box, predictive capability for aerosol Fe concentrations
 105 by the model was best when soil state was included and both modeled and observed aerosol Fe
 106 concentrations were aggregated over time and space by medians when compared to means.



107

108 **Figure S4.** Comparison of modelling and observational data including the PD-IND run, which

109 altered oil and industrial coal solubility parameters (industrial coal Fe solubility from 0.2 to

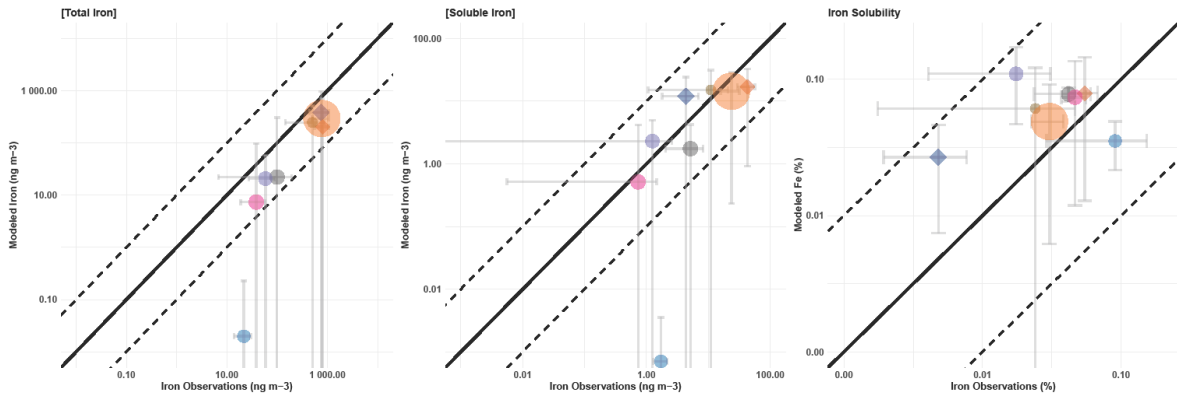
110 0.05% and oil from 38 to 25%). Error bars represent spatiotemporal variance within each region.

111 The solid black line indicates a 1-to-1 relationship and the dashed lines represent deviation by

112 ± 1 order of magnitude.

113

114



115

116 **Figure S5.** Model-observation comparisons ($n = 25$) of surface aerosol Fe concentrations only
 117 using observations co-located in regions where soluble Fe fluxes increased by a factor of 2
 118 using the PD-BIOF solubility parameters in attempt to isolate observations most representative
 119 of residential coal and biofuel aerosol.

120

121

122 **Table S8.** Fe and BC inventories as published by Rathod et al. (2020), Hoesly et al. (2018),
 123 and Bond et al. (2004) used to develop the Fe emissions inventories applied in this work.
 124 Emissions are reported in Gg a⁻¹ with three significant figures.

	Rathod “Central”-case Fe emissions		Hoesly BC Emissions		Bond BC emissions
	Sector		Sector		Sector
Fuel-Type	Industrial	Residential + Commercial	Industrial	Residential/ Commercial	Residential
Coal	254	3.90	n.d.	n.d.	*480
Wood	58.9	7.15	n.d.	n.d.	880
Total	-	-	1720	*2160	*1360
Total adj.	-	-	-	762	-

125

126

127

128 **Table S9.** Final breakdown of Fe and Black Carbon (BC) emissions by sector (fuel-type) as
 129 represented in the two new Fe emissions inventories developed and tested in this work.
 130 Emission fluxes are reported in Gg a⁻¹ with three significant figures.

Fuel-Type	Low Residential-Case		High Residential-Case	
	Iron	Black Carbon	Iron	Black Carbon
Residential Coal	16.0	2160	460	2160
Industrial Coal	750	1720	310	1720
Oil (shipping/transportation)	30.0	1250	30.0	1250
Residential Biofuel	70.0	1330	70.0	1330
Smelting	1350	n.d.	1350	n.d.
TOTAL	2220	6460	2220	6460

131

132

133

134 **Table S10a.** Fe deposition flux budgets (Gg a^{-1} ; to two significant figures) for each key marine region utilizing the high-residential emissions
 135 inventory. Values reported herein only include fluxes to marine systems (model grid cells where ocean fraction > 50%). AS: Arabian Sea, CPAO:
 136 Central Pacific and Atlantic Ocean, A USP: Australia and South Pacific, BB: Bay of Bengal, ENPAC: Eastern North Pacific, NATL: North Atlantic,
 137 SATL: South Atlantic, SEAS: Southeastern Asia, SO: Southern Ocean, WNPAC: Western North Pacific.

Simulation		Region												
		Global	SATL	NATL	AS	BB	INDO	SEAS	ENPAC	WNPAC	ARCT	A USP	SO	CPAO
Total Fe	PD-BASE	23000	1300	2200	6700	1500	8300	700	170	120	60	1200	600	8700
Total Anth. Fe	PD-BASE	590	24	72	57	65	120	240	43	19	9.3	11	5.3	39
Soluble Fe	PD-BASE	370	17	33	76	31	110	21	6.6	4.2	2.0	18	10	150
	PD-RESI	400	18	38	80	35	120	35	9.0	5.1	2.4	19	10	150
	PD-BIOF	410	20	39	81	37	120	37	9.1	5.3	3.0	19	11	150
	PD-IND	410	20	38	81	37	120	37	9.0	5.0	2.4	19	10	150
Soluble Anth. Fe	PD-BASE	36	1.7	3.9	4.2	4.5	8.7	11	3.3	1.9	0.6	0.5	0.3	4.7
	PD-RESI	70	2.4	8.5	7.8	9.2	17	25	5.6	2.9	1.0	1.2	0.5	5.9
	PD-BIOF	81	4.4	9.4	9.3	11	20	27	5.8	3.0	1.1	1.4	0.7	8.1
	PD-IND	78	4.4	8.9	9.1	11	20	26	5.6	2.8	1.0	1.3	0.7	7.6

138

139

140 **Table S10b.** Fe deposition flux budgets (Gg a^{-1} ; to two significant figures) for each key marine region utilizing the low-residential emissions
 141 inventory. Values reported herein only include fluxes to marine systems (model grid cells where ocean fraction > 50%).

	Simulation	Region												
		Global	SATL	NATL	AS	BB	INDO	SEAS	ENPAC	WNPAC	ARCT	AUSP	SO	CPAO
Total Fe	PD-BASE	23000	1300	2200	6700	1500	8200	710	170	120	59	1200	600	8600
Total Anth. Fe	PD-BASE	590	24	72	57	65	120	240	43	19	9.3	11	5.3	39
Soluble Fe	PD-BASE	370	17	33	76	30	110	21	6.6	4.2	2.0	18	10	150
	PD-RESI	370	17	33	76	31	110	22	6.7	4.2	2.0	18	10	150
	PD-BIOF	380	19	34	78	32	110	24	6.9	4.3	2.1	18	10	150
Soluble Anth. Fe	PD-BASE	36	1.7	3.9	4.2	4.5	8.7	11	3.3	1.9	0.6	0.5	0.3	4.7
	PD-RESI	37	1.7	4.1	4.3	4.7	9.0	11	3.3	1.9	0.6	0.6	0.3	4.7
	PD-BIOF	49	3.8	5.0	5.7	6.5	12	13	3.5	2.1	0.7	0.8	0.5	6.9

142

143 **Table S11.** Root Mean Square Error (RMSE) as a measure of difference between observed and
144 modeled values for total and soluble Fe (ng m⁻³) and Fe solubility, grouped by marine region.
145 Δ RMSE reflects model skill with and without the use of the new interactive soil state dust
146 emission model.

		RMSE		
		With Soil State	Without Soil State	Δ RMSE
Total Fe		495.14	4045.33	-3550.20
Soluble Fe	AS	7.67	45.24	-37.57
Solubility		0.02	0.01	0.01
Total Fe		62.93	257.65	-194.72
Soluble Fe	AUSP	2.21	1.82	0.39
Solubility		0.10	0.12	-0.02
Total Fe		463.38	238.15	225.23
Soluble Fe	BB	28.51	22.37	6.14
Solubility		0.06	0.07	-0.01
Total Fe		1211.84	3321.24	-2109.41
Soluble Fe	CPAO	18.18	20.68	-2.50
Solubility		0.15	0.15	0.00
Total Fe		97.73	86.65	11.09
Soluble Fe	ENPAC	5.90	4.86	1.03
Solubility		0.08	0.08	0.00
Total Fe		209.45	581.63	-372.17
Soluble Fe	NATL	3.93	3.68	0.25
Solubility		0.25	0.26	-0.02
Total Fe		38.59	1245.89	-1207.30
Soluble Fe	SATL	2.19	4.16	-1.97
Solubility		0.04	0.04	0.00
Total Fe		176.97	159.76	17.21
Soluble Fe	SEAS	13.88	12.11	1.77
Solubility		0.08	0.06	0.01
Total Fe		19.80	21.18	-1.38
Soluble Fe	SO	1.76	1.62	0.14
Solubility		0.22	0.23	-0.01
Total Fe		36.30	71.87	-35.57
Soluble Fe	WNPAC	4.65	4.34	0.30
Solubility		0.07	0.08	-0.01

147

148 **Table S12.** Summary statistics for model (Mod)-observation (Obs) comparisons of soluble Fe
 149 concentrations. RMSE: root mean square error, SD: standard deviation. Δ RMSE represents
 150 comparison between the base simulation (PD-BASE) and each sensitivity case using the high-
 151 residential emissions inventory as an upper estimate.

Model Case	Region	Median Surface Concentration (ng m ⁻³)		Mean Surface Concentration \pm SD (ng m ⁻³)		Δ Obs – Mod	RMSE	Δ RMSE				
		Obs	Mod	Obs	Mod							
PD-BASE			7.19		7.07 \pm 3.06	-6.79	7.67	NA				
PD-RESI	AS	0.40	8.67	2.69 \pm 4.59	10.9 \pm 9.18	-8.27	15.6	7.91				
PD-BIOF			9.36						12.4 \pm 11.7	-8.96	19.0	11.28
PD-IND			9.28						12.3 \pm 11.6	-8.89	18.8	11.12
PD-BASE			0.03						0.14 \pm 0.32	0.21	2.21	NA
PD-RESI	AUSP	0.24	0.03	1.13 \pm 2.20	0.29 \pm 0.78	0.21	2.05	-0.16				
PD-BIOF			0.03						0.36 \pm 1.02	0.21	2.16	-0.05
PD-IND			0.03						0.36 \pm 1.00	0.21	2.15	-0.06
PD-BASE			4.87						4.95 \pm 3.25	17.89	28.5	NA
PD-RESI	BB	22.76	6.50	27.03 \pm 20.1	8.06 \pm 6.70	16.26	25.0	-3.5				
PD-BIOF			7.91						9.71 \pm 8.21	14.85	23.5	-5.0
PD-IND			7.84						9.62 \pm 8.21	14.92	23.6	-4.9
PD-BASE			1.31						2.32 \pm 2.42	0.60	18.2	NA
PD-RESI	CPAO	1.91	1.43	7.28 \pm 18.1	2.38 \pm 2.42	0.48	18.2	0.0				
PD-BIOF			1.50						2.46 \pm 2.46	0.41	18.1	0.0
PD-IND			1.44						2.41 \pm 2.44	0.47	18.2	0.0
PD-BASE			0.36						0.42 \pm 0.17	3.39	5.90	NA
PD-RESI	ENPAC	3.75	0.47	4.63 \pm 4.25	0.56 \pm 0.25	3.28	5.80	-0.10				
PD-BIOF			0.49						0.57 \pm 0.26	3.26	5.79	-0.11
PD-IND			0.44						0.51 \pm 0.24	3.31	5.84	-0.06
PD-BASE			0.40						0.63 \pm 0.81	0.61	3.93	NA
PD-RESI	NATL	1.01	0.43	2.34 \pm 3.52	0.84 \pm 1.42	0.58	3.91	-0.02				
PD-BIOF			0.46						0.92 \pm 1.51	0.55	3.90	-0.03
PD-IND			0.39						0.82 \pm 1.43	0.62	3.93	0.00
PD-BASE			0.13						0.19 \pm 0.26	0.33	2.19	NA
PD-RESI	SATL	0.46	0.14	0.95 \pm 2.25	0.21 \pm 0.31	0.32	2.15	-0.04				
PD-BIOF			0.17						0.27 \pm 0.38	0.29	2.12	-0.07
PD-IND			0.17						0.26 \pm 0.37	0.30	2.13	-0.06
PD-BASE			0.98						1.09 \pm 1.01	4.79	13.9	NA
PD-RESI	SEAS	5.77	1.53	9.80 \pm 11.2	2.06 \pm 2.75	4.24	13.4	-0.5				
PD-BIOF			1.59						2.13 \pm 2.80	4.18	13.4	-0.5
PD-IND			1.52						2.04 \pm 2.73	4.24	13.4	-0.4
PD-BASE			0.001						0.01 \pm 0.01	0.58	1.76	NA
PD-RESI	SO	0.59	0.001	1.12 \pm 1.4	0.01 \pm 0.01	0.58	1.76	0.00				
PD-BIOF			0.001						0.01 \pm 0.01	0.58	1.76	0.00
PD-IND			0.001						0.01 \pm 0.01	0.58	1.76	0.00

PD-BASE			0.31		0.32 ± 0.09	0.54	4.65	NA
PD-RESI	WNPAC	0.85	0.37	2.67 ± 4.06	0.38 ± 0.11	0.47	4.62	-0.02
PD-BIOF			0.38		0.39 ± 0.12	0.46	4.62	-0.03
PD-IND			0.34		0.33 ± 0.11	0.51	4.65	0.00

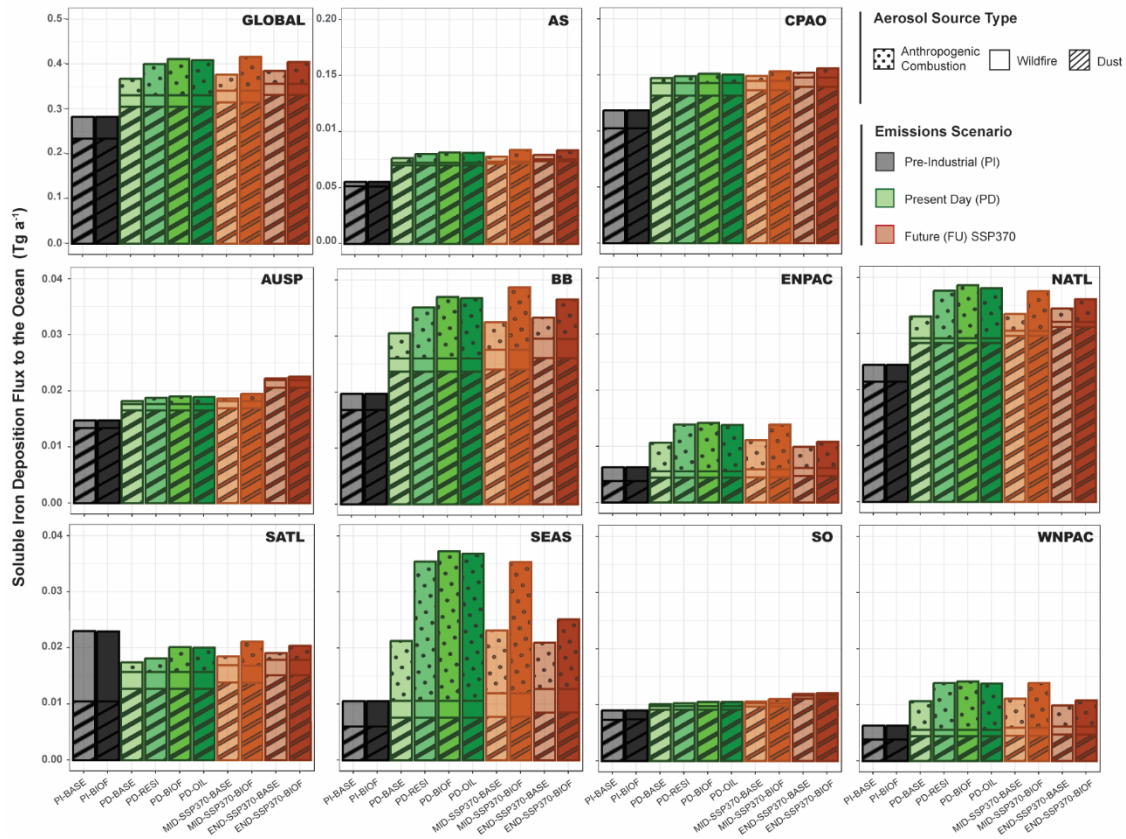
152

153 **Table S13.** Summary statistics for model (Mod)-observation (Obs) comparisons of Fe
 154 solubility. RMSE: root mean square error, SD: standard deviation. Δ RMSE represents
 155 comparison between the base simulation (PD-BASE) and each sensitivity case using the high-
 156 residential emissions inventory as an upper estimate.

Model Case	Region	Median Surface Solubility		Mean Surface Fractional Solubility \pm SD		Δ Obs – Mod median	RMSE	Δ RMSE
		Obs	Mod	Obs	Mod			
PD-BASE	AS	0.001	0.023	0.004	0.025	-0.02	0.02	NA
PD-RESI			0.040		0.035	-0.04	0.03	0.01
PD-BIOF			0.040		0.039	-0.04	0.04	0.02
PD-IND			0.039		0.039	-0.04	0.04	0.02
PD-BASE	AUSP	0.03	0.06	0.08	0.07	-0.03	0.10	NA
PD-RESI			0.07		0.08	-0.04	0.10	0.00
PD-BIOF			0.08		0.09	-0.05	0.10	0.00
PD-IND			0.08		0.08	-0.04	0.10	0.00
PD-BASE	BB	0.06	0.05	0.07	0.05	0.01	0.06	NA
PD-RESI			0.07		0.07	-0.01	0.06	0.00
PD-BIOF			0.08		0.08	-0.02	0.06	0.00
PD-IND			0.08		0.08	-0.02	0.06	0.00
PD-BASE	CPAO	0.03	0.05	0.06	0.06	-0.03	0.15	NA
PD-RESI			0.06		0.06	-0.03	0.15	0.00
PD-BIOF			0.06		0.07	-0.03	0.16	0.00
PD-IND			0.06		0.07	-0.03	0.15	0.00
PD-BASE	ENPAC	0.07	0.09	0.08	0.09	-0.01	0.08	NA
PD-RESI			0.11		0.12	-0.04	0.10	0.01
PD-BIOF			0.11		0.12	-0.04	0.10	0.01
PD-IND			0.10		0.11	-0.03	0.08	0.00
PD-BASE	NATL	0.05	0.10	0.12	0.10	-0.05	0.25	NA
PD-RESI			0.11		0.12	-0.06	0.25	0.00
PD-BIOF			0.12		0.12	-0.07	0.25	0.00
PD-IND			0.11		0.11	-0.06	0.25	0.00
PD-BASE	SATL	0.02	0.05	0.04	0.05	-0.03	0.04	NA
PD-RESI			0.06		0.05	-0.03	0.04	0.00
PD-BIOF			0.06		0.06	-0.04	0.05	0.01
PD-IND			0.06		0.06	-0.04	0.05	0.00
PD-BASE	SEAS	0.07	0.05	0.07	0.08	0.01	0.08	NA
PD-RESI			0.09		0.11	-0.02	0.08	0.00
PD-BIOF			0.09		0.11	-0.02	0.08	0.01
PD-IND			0.09		0.10	-0.02	0.07	-0.01
PD-BASE	SO	0.05	0.04	0.13	0.05	0.00	0.22	NA
PD-RESI			0.05		0.05	0.00	0.22	0.00
PD-BIOF			0.05		0.05	-0.01	0.22	0.00
PD-IND			0.05		0.05	-0.01	0.22	0.00

PD-BASE			0.11		0.12	-0.02	0.07	NA
PD-RESI	WNPAC	0.09	0.13	0.09	0.14	-0.04	0.08	0.01
PD-BIOF			0.13		0.14	-0.04	0.08	0.01
PD-IND			0.11		0.12	-0.03	0.06	-0.01

157



158

159 **Figure S6.** Deposition fluxes of soluble aerosol Fe to marine ecosystems globally and in

160 regional grouping with inclusion of the PD-IND (labeled PD-OIL in this figure) simulations.

161 Fluxes are source-apportioned (dust, biofuel burning, and anthropogenic combustion) and

162 provided for each model simulation.

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