

Text S1 Experimental methods and results

Text S1.1 Experimental methods

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

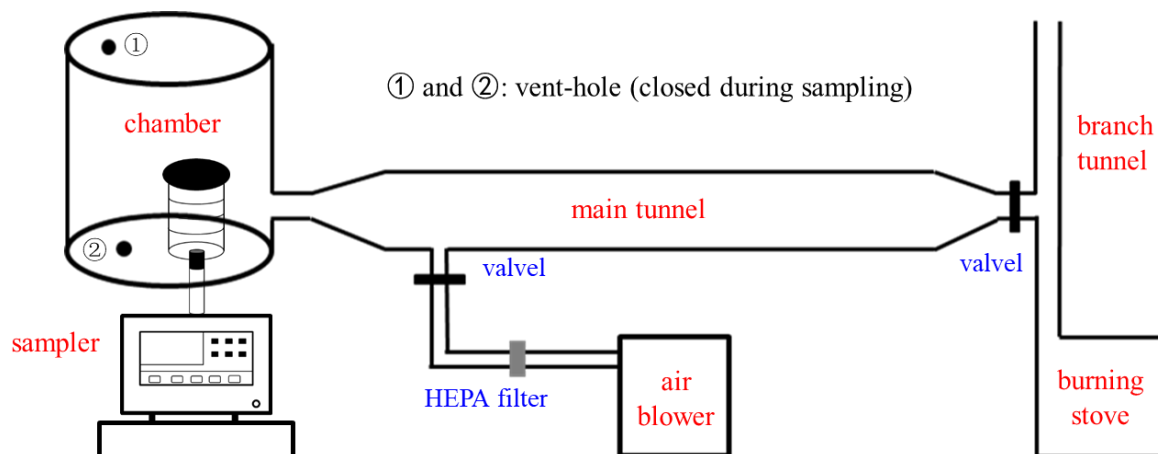


Figure S1. Schematic diagram of the apparatus used in our work to collect aerosol particles emitted by domestic coal and biofuel burning.

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

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



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

Text S1.2 Experimental results

Table S1. Fe content and solubility for power plant coal fly ash samples (each from a coal 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

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

sample	Fe content (μg/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

38 **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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42 **Table S4.** Fe content and solubility for biofuel burning aerosols examined in this work.

sample	Fe content (μg/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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Table S5. Fe content and solubility for municipal waste fly ash, oil fly ash and oil bottom ash samples examined in this work.

sample	Fe content ($\mu\text{g/g}$)	Fe content (mg/g)	Fe solubility (%)
Municipal waste fly ash	3.87×10^3	3.9	0.58
	2.26×10^4	22.6	2.41
	2.97×10^4	29.7	1.54
oil bottom ash	1.91×10^5	191.5	25.47
oil fly ash	1.83×10^4	18.3	11.70
	9.06×10^3	9.1	13.43

49 **Table S6.** Summary of Fe content (mg/g) for anthropogenic and combustion aerosol Fe determined in our present study and previous work (*n*:
50 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

52 **Table S7.** Summary of Fe solubility (%) for anthropogenic and combustion aerosol Fe determined in our present study and previous work (*n*:
53 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

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Text S2 Modeling methods and results

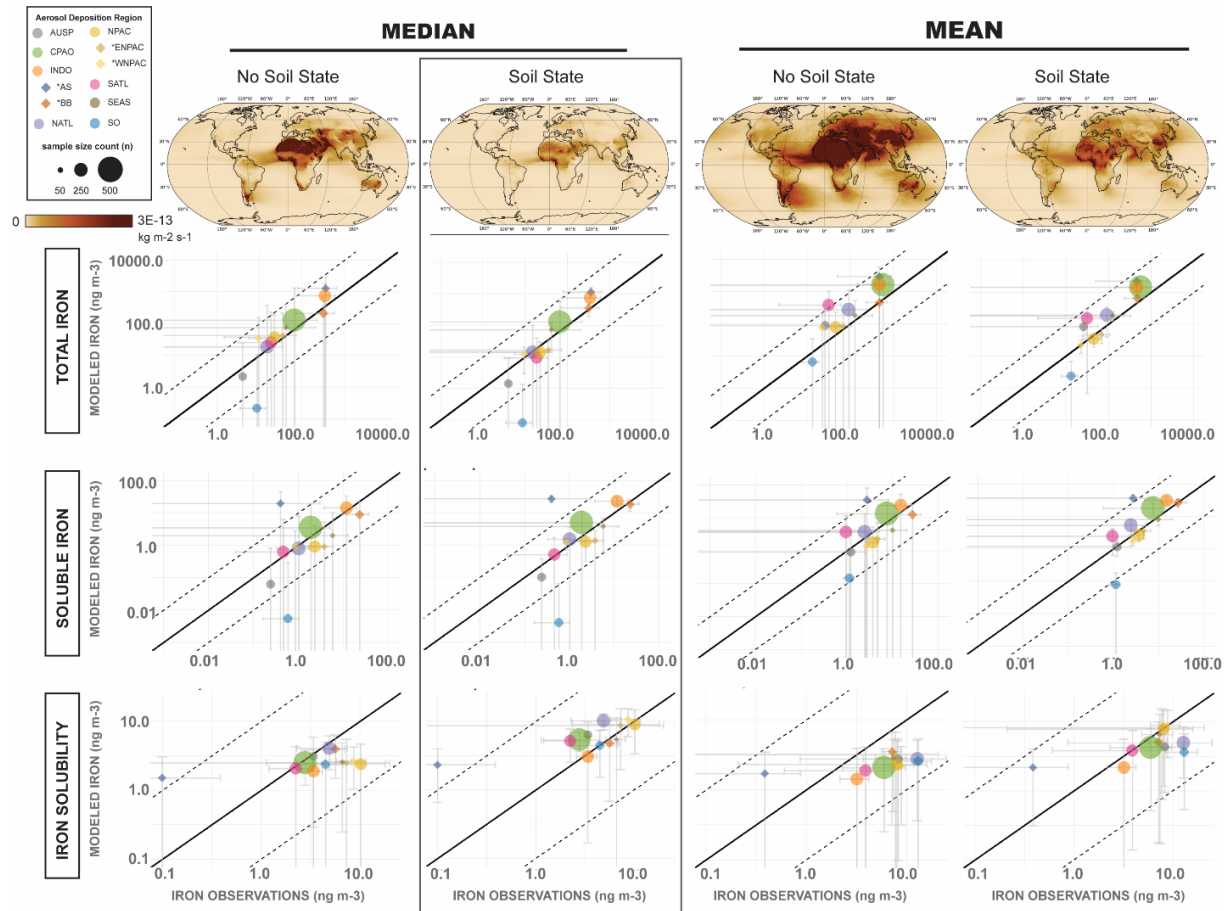


Figure S3. Model-observation comparisons of surface aerosol Fe concentrations using two different dust flux schemes (soil state submodule included versus no soil state submodule). As indicated by the black box, predictive capability for aerosol Fe concentrations by the model was best when soil state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

66 **Table S8.** Fe deposition flux budgets (Gg a^{-1}) for each key marine region. Values reported herein only include fluxes to marine systems (model
67 grid cells where ocean fraction $> 50\%$). AS = Arabian Sea, CPAO = Central Pacific and Atlantic Ocean, AUSP = Australia and South Pacific, BB
68 = Bay of Bengal, ENPAC = Eastern North Pacific, NATL = North Atlantic, SATL = South Atlantic, SEAS = Southeastern Asia, SO = Southern
69 Ocean, WNPAC = Western North Pacific.

Simulation		Region												
		Global	SATL	NATL	AS	BB	INDO	SEAS	ENPAC	WNPAC	ARCT	AUSP	SO	CPAO
Total Fe	PD-BASE	23232	1266	2217	6711	1534	8246	706	283	283	283	59	1214	597
Total Anth. Fe	PD-BASE	585.7	23.8	71.9	56.8	65.3	122.1	241.5	61.1	61.1	61.1	9.3	11.3	5.3
Soluble Fe	PD-BASE	367	17	33	76	31	107	21	11	11	11	2	18	10
	PD-RESI	400	18	38	80	35	115	35	14	14	14	2	19	10
	PD-BIOF	412	20	39	81	37	118	37	14	14	14	3	19	11
	PD-IND	409	20	38	81	37	118	37	14	14	14	2	19	11
Soluble Anth. Fe	PD-BASE	36.2	1.7	3.9	4.2	4.5	8.7	10.7	5.1	5.1	5.1	0.6	0.5	0.3
	PD-RESI	69.5	2.4	8.5	7.8	9.2	17.0	24.8	8.3	8.3	8.3	1.0	1.2	0.5
	PD-BIOF	80.8	4.4	9.4	9.3	11.0	20.3	26.7	8.6	8.6	8.6	1.1	1.4	0.7
	PD-IND	78.3	4.4	8.9	9.1	10.8	19.9	26.2	8.3	8.3	8.3	1.0	1.3	0.7

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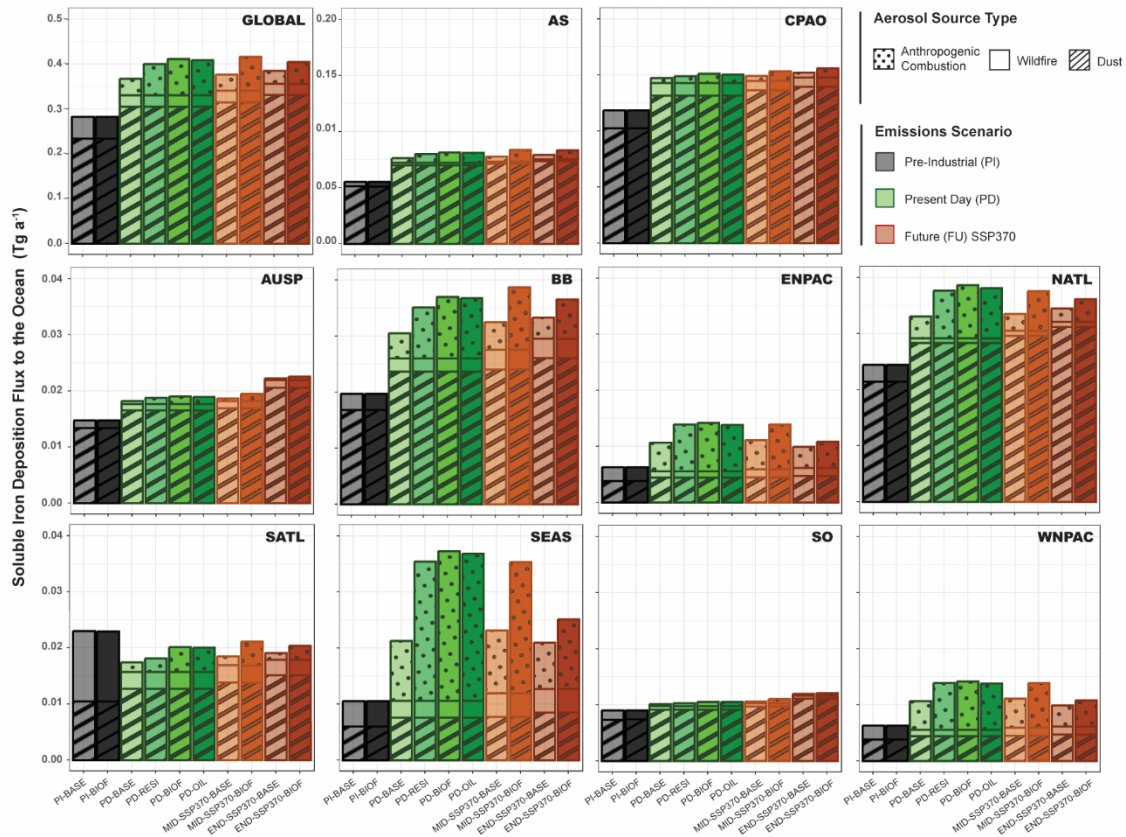


Figure S4. Deposition fluxes of soluble aerosol Fe to marine ecosystems globally and in regional grouping (see Figure 1 in the main text). Fluxes are source-apportioned (dust, biofuel burning, and anthropogenic combustion) and provided for each model simulation. AS = Arabian Sea, CPAO = Central Pacific and Atlantic Ocean, AUSP = Australia and South Pacific, BB = Bay of Bengal, ENPAC = Eastern North Pacific, NATL = North Atlantic, SATL = South Atlantic, SEAS = Southeastern Asia, SO = Southern Ocean, WNPAC = Western North Pacific. Note the difference in y-axes between the global, AS/CPAO, and remainder of regions.

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