

## Enhanced Iron Solubility at Low pH in Global Aerosols

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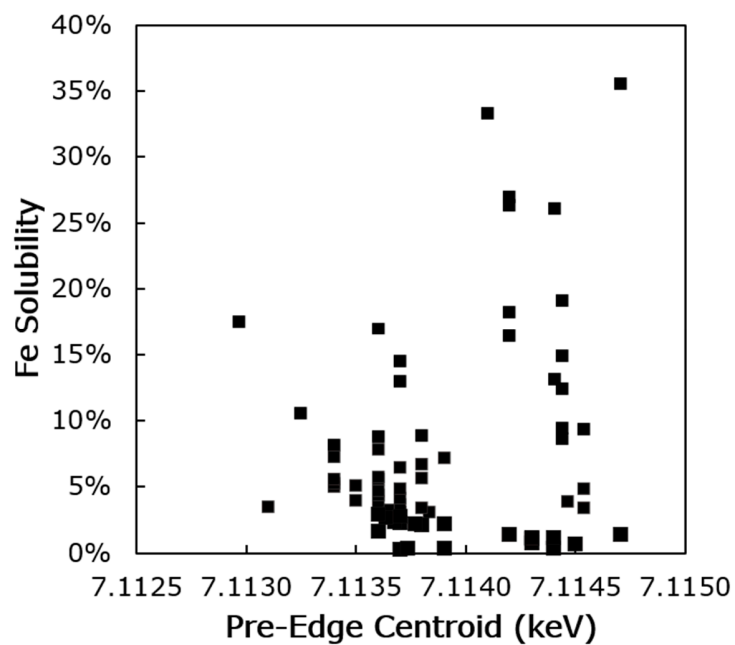
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**Figure S1.** The pre-edge centroid (keV), a reliable indicator of iron oxidation state, has higher values for more oxidized iron. The solubility of aerosol iron from all sampling locations shows no correlation with the oxidation state, suggesting that oxidation state is not an indicator of iron solubility on a global scale.

**Table S1.** Sample collection information for India, Southern Ocean, Finokalia Research Station and GEOTRACES samples.

Sample ID	Collection	Sample Deployment	Sample Collection	Predominant Source	Total Volume (m <sup>3</sup> )
SF1	India	9/16/2015	9/17/2015	mixed	1020
SF2	India	9/18/2015	9/19/2015	mixed	1020
SF3	India	9/19/2015	9/20/2015	mixed	1020
SF5	India	9/20/2015	9/21/2015	mixed	1020
SF6	India	9/21/2015	9/22/2015	mixed	1020
SF4	India	9/22/2015	9/23/2015	mixed	1020
SF7	India	9/23/2015	9/24/2015	mixed	1020
SF8	India	9/24/2015	9/25/2015	mixed	1020
SF9	India	9/25/2015	9/26/2015	mixed	1020
SF10	India	9/26/2015	9/27/2015	mixed	1020
SF11	India	9/27/2015	9/28/2015	mixed	1020
SF12	India	9/28/2015	9/29/2015	mixed	1020
SF13	India	9/29/2015	9/30/2015	mixed	1020
SF14	India	9/30/2015	10/1/2015	mixed	1020
C2	Southern Ocean	11/30/2010	12/2/2010	Open Ocean	86.4
C13	Southern Ocean	3/2/2011	3/4/2011	Open Ocean	86.4
C9	Southern Ocean	2/12/2011	2/15/2011	Open Ocean	86.4
6250	GEOTRACES	11/7/11	11/8/11	N. American	92.10
6319	GEOTRACES	11/8/11	11/9/11	N. American	74.96
6546	GEOTRACES	11/12/11	11/13/11	N. American	114.32
6964	GEOTRACES	11/20/11	11/21/11	marine	95.01
7245	GEOTRACES	11/24/11	11/26/11	marine	161.00
7587	GEOTRACES	11/30/11	12/1/11	marine	115.54
7899	GEOTRACES	12/5/11	12/6/11	N. Africa	116.01
7946	GEOTRACES	12/6/11	12/7/11	N. Africa	137.92
8004	GEOTRACES	12/7/11	12/8/11	N. Africa	108.81
BL_165	FRS	12/18/09	12/19/09	SW	45.59
BL_185	FRS	2/10/10	2/11/10	SW	38.37
BL_197	FRS	3/10/10	3/11/10	SW	48.68
BL_206	FRS	3/31/10	4/1/10	SW	51.38
BL_213	FRS	4/16/10	4/17/10	SW	56.10
BL_305	FRS	11/10/10	11/11/10	S	58.72
BL_324	FRS	12/24/10	12/25/10	S	38.87
BL_260	FRS	7/28/10	7/29/10	NW	49.32
BL_274	FRS	9/6/10	9/7/10	NE	67.16
BL_275	FRS	9/9/10	9/10/10	N	56.67
BL_277	FRS	9/13/10	9/14/10	NE	70.06
BL_330	FRS	1/7/11	1/8/11	NW	47.79
BL_333	FRS	1/14/11	1/15/11	N	45.80
BL_344	FRS	2/9/11	2/10/11	NE	43.83

**Table S2.** Sample collection information for samples collected at the Bermuda Institute of Ocean Science.

<b>Sample Deployment</b>	<b>Sample Collection</b>	<b>Predominant Source</b>	<b>Total Volume (m<sup>3</sup>)</b>
26/Jun/2011 10:19	2/Jul/2011 13:50	mixed	838.88
20/Jul/2011 13:30	26/Jul/2011 8:40	Saharan	774.29
26/Jul/2011 16:42	1/Aug/2011 9:13	mixed	715.94
1/Aug/2011 9:21	7/Aug/2011 11:00	Saharan	714.56
7/Aug/2011 11:06	13/Aug/2011 10:18	N. Am/Caribbean	784.84
13/Aug/2011 10:21	19/Aug/2011 13:22	Saharan	808.20
19/Aug/2011 13:25	25/Aug/2011 9:30	Saharan	768.70
6/Sep/2011 9:05	12/Sep/2011 8:51	Saharan	802.46
26/Sep/2011 8:30	30/Sep/2011 10:02	Saharan	542.86
30/Sep/2011 10:05	6/Oct/2011 14:38	mixed	827.92
6/Oct/2011 14:42	12/Oct/2011 10:25	N. Am	779.79
18/Oct/2011 10:12	24/Oct/2011 9:00	Saharan	795.22
5/Nov/2011 10:49	11/Nov/2011 15:50	mixed	831.69
11/Nov/2011 15:54	17/Nov/2011 14:23	N. Am	934.11
17/Nov/2011 14:27	23/Nov/2011 14:45	N. Am	806.96
23/Nov/2011 14:47	29/Nov/2011 15:33	mixed	793.56
5/Dec/2011 9:03	11/Dec/2011 9:25	North	795.74
22/Jan/2012 15:29	28/Jan/2012 10:12	mixed	769.01
28/Jan/2012 10:15	3/Feb/2012 9:20	N. Am	803.10
3/Feb/2012 9:25	9/Feb/2012 9:23	N. Am	769.74
15/Feb/2012 9:30	21/Feb/2012 8:56	N. Am	806.47
21/Feb/2012 9:00	27/Feb/2012 9:18	mixed	801.73
5/Mar/2012 8:36	10/Mar/2012 10:19	mixed	684.31
16/Mar/2012 9:54	22/Mar/2012 9:43	mixed	794.18
28/Mar/2012 9:45	3/Apr/2012 8:54	N. Am	777.83
3/Apr/2012 8:57	9/Apr/2012 9:34	N. Am	797.32
9/Apr/2012 9:36	13/Apr/2012 11:15	mixed	806.72
27/Apr/2012 9:44	3/May/2012 9:30	mixed	789.34
3/May/2012 9:36	9/May/2012 9:26	North	777.73
9/May/2012 9:30	15/May/2012 10:47	mixed	787.57
15/May/2012 10:48	21/May/2012 9:09	Saharan	795.67
27/May/2012 10:26	2/Jun/2012 9:51	mixed	802.12
2/Jun/2012 9:55	8/Jun/2012 9:10	N. Am	756.62
14/Jun/2012 9:28	20/Jun/2012 14:00	mixed	831.43
26/Jun/2012 15:18	2/Jul/2012 9:28	mixed	772.86

Table S3. Sample Collection Locations.

Collection	Latitude	Longitude
BIOS	32.24° N	64.87° W
FRS	35.32° N	25.67° E
GEOTRACES	39.71° N	69.86° W
	39.70° N	69.79° W
	38.33° N	68.87° W
	31.75° N	64.17° W
	29.70° N	56.82° W
	25.55° N	43.54° W
	22.37° N	35.62° W
	20.88° N	32.62° W
Southern Ocean	19.43° N	29.38° W
	56° S	94° E
	69° S	77° E
India	44° S	96° E
	28.5° N	77.3° E

Table S4. Bermuda Institute of Ocean Science Aerosol Data.

Sample Deployment	pH at 80% RH	Fe(II) (%)	Pre-Edge Centroid (keV)	Total Fe (ng/m <sup>3</sup> )	Soluble Fe (%)	Distance Travelled in 5 days (km)
26/Jun/2011 10:19	7.7	44.1	7.1137	80.4	4.2%	1600
2/Jul/2011 14:08	-	-	-	52.7	1.8%	1360
14/Jul/2011 9:17	-	-	-	63.1	1.0%	2771
20/Jul/2011 13:30	3.8	30.1	7.1139	238.6	2.2%	1586
26/Jul/2011 16:42	7.7	44.1	7.1137	142.6	3.1%	2136
1/Aug/2011 9:21	3.3	37.1	7.1138	414.2	2.1%	1521
7/Aug/2011 11:06	-	30.1	7.1139	108.9	7.3%	2229
13/Aug/2011 10:21	3.7	34.7	7.1138	265.9	3.1%	1950
19/Aug/2011 13:25	4.0	51.0	7.1136	521.9	1.7%	1300
25/Aug/2011 9:33	-	-	-	98.1	0.4%	2064
31/Aug/2011 11:17	-	-	-	49.4	1.3%	1857
6/Sep/2011 9:05	3.2	44.1	7.1137	128.2	2.8%	2021
12/Sep/2011 8:55	-	-	-	98.3	1.3%	2071
26/Sep/2011 8:30	4.5	44.1	7.1137	153.5	2.3%	1260
30/Sep/2011 10:05	3.8	51.0	7.1136	28.0	7.9%	2736
6/Oct/2011 14:42	4.2	51.0	7.1136	60.0	4.4%	3714
12/Oct/2011 10:32	-	-	-	55.7	2.5%	2171
18/Oct/2011 10:12	3.6	37.1	7.1138	23.7	8.9%	2221
24/Oct/2011 9:05	-	-	-	34.7	1.5%	3569
31/Oct/2011 9:48	-	-	-	21.4	8.5%	1790
5/Nov/2011 10:49	3.1	44.1	7.1137	15.4	13.0%	2814
11/Nov/2011 15:54	3.7	65.0	7.1134	25.6	8.2%	4186
17/Nov/2011 14:27	3.9	65.0	7.1134	30.6	7.4%	3886

Sample Deployment	pH at 80% RH	Fe(II) (%)	Pre-Edge Centroid (keV)	Total Fe (ng/m <sup>3</sup> )	Soluble Fe (%)	Distance Travelled in 5 days (km)
23/Nov/2011 14:47	4.6	65.0	7.1134	62.6	5.6%	4021
5/Dec/2011 9:03	-	65.0	7.1134	58.2	5.1%	3857
22/Jan/2012 15:29	7.3	44.1	7.1137	55.8	4.8%	3629
28/Jan/2012 10:15	4.8	37.1	7.1138	36.4	6.8%	4371
3/Feb/2012 9:25	3.4	75.5	7.1133	19.3	10.6%	4086
15/Feb/2012 9:30	3.4	51.0	7.1136	26.0	8.8%	4386
21/Feb/2012 9:00	3.9	58.0	7.1135	25.5	4.0%	5243
27/Feb/2012 9:20	-	-	-	35.7	1.9%	4280
5/Mar/2012 8:36	3.5	65.0	7.1134	43.3	5.3%	4520
10/Mar/2012 10:23	-	-	-	65.0	0.8%	2800
16/Mar/2012 9:54	4.4	51.0	7.1136	109.5	2.9%	3171
22/Mar/2012 9:45	-	-	-	88.4	1.9%	2336
28/Mar/2012 9:45	4.2	44.1	7.1137	84.6	3.8%	4271
3/Apr/2012 8:57	3.3	51.0	7.1136	53.3	5.6%	3607
9/Apr/2012 9:36	7.4	37.1	7.1138	44.0	5.7%	4580
13/Apr/2012 11:21	-	-	-	89.7	2.4%	3053
27/Apr/2012 9:44	7.6	51.0	7.1136	65.9	4.7%	2607
3/May/2012 9:36	-	51.0	7.1136	69.0	3.9%	3164
9/May/2012 9:30	3.6	58.0	7.1135	74.0	5.2%	2986
15/May/2012 10:48	3.3	44.1	7.1137	37.3	6.5%	1414
21/May/2012 9:15	-	-	-	46.1	1.3%	1857
27/May/2012 10:26	7.5	51.0	7.1136	40.9	5.8%	1614
2/Jun/2012 9:55	4.3	47.6	7.1137	155.1	3.3%	2814
8/Jun/2012 9:16	-	-	-	35.3	1.4%	3229
14/Jun/2012 9:28	5.8	37.1	7.1138	21.7	3.5%	1329
20/Jun/2012 14:05	-	-	-	93.3	2.1%	1686
26/Jun/2012 15:18	3.9	44.1	7.1137	392.3	14.4%	2686

**Table S5.** India, GEOTRACES, Southern Ocean and Finokalia Research Station Aerosol Data.

Sample ID	Collection	pH at 80% RH	% Fe(II)	Pre-Edge Centroid (keV)	Total Fe (ng/m <sup>3</sup> )	Soluble Fe (%)	Distance Travelled in 5 days (km)
SF1	India	-	0.0	7.1145	-	-	500
SF2	India	-	0.0	7.1144	612.5	19.2%	2000
SF3	India	-	0.0	7.1144	647.8	15.0%	1500
SF5	India	-	0.0	7.1144	937.1	12.4%	1200
SF6	India	-	0.0	7.1144	-	-	1800
SF4	India	-	0.0	7.1145	1947.6	3.9%	3000
SF7	India	-	0.0	7.1145	1018.8	9.4%	2000
SF8	India	-	0.0	7.1144	271.9	13.2%	1600
SF9	India	-	0.0	7.1144	-	-	1500
SF10	India	-	0.0	7.1145	1972.2	3.5%	1000
SF11	India	-	0.0	7.1144	1039.4	9.5%	2400
SF12	India	-	2.1	7.1143	-	-	1950
SF13	India	-	0.0	7.1145	1517.2	4.9%	2500
SF14	India	-	0.0	7.1144	1365.8	8.6%	2600
C2	Southern Ocean	-	9.1	7.1142	10.0	27.0%	3500
C13	Southern Ocean	-	0.0	7.1145	38.0	0.8%	4800
C9	Southern Ocean	-	0.0	7.1147	30.0	1.5%	1650
6250	GEOTRACES	-	44.1	7.1137	25.7	3.2%	2100
6319	GEOTRACES	-	48.7	7.1136	80.2	2.7%	3300
6546	GEOTRACES	-	39.4	7.1138	38.3	2.3%	5500
6964	GEOTRACES	-	86.0	7.1131	1.7	3.5%	4000
7245	GEOTRACES	-	95.3	7.1130	0.9	17.6%	1400
7587	GEOTRACES	-	45.8	7.1137	13.6	2.4%	3000
7899	GEOTRACES	6.0	30.1	7.1139	623.5	0.4%	2000
7946	GEOTRACES	7.1	44.1	7.1137	1580.0	0.3%	2000
8004	GEOTRACES	7.1	41.7	7.1137	2529.8	0.4%	2100
BL_165	FRS	7.1	2.1	7.1143	1537.2	1.1%	3400
BL_185	FRS	7.1	-	-	3455.3	4.3%	5500
BL_197	FRS	7.1	2.1	7.1143	734.6	0.8%	5600
BL_206	FRS	7.1	0.0	7.1144	1213.7	1.2%	3400
BL_213	FRS	7.1	0.0	7.1144	2009.0	0.4%	2700
BL_305	FRS	7.1	2.1	7.1143	323.6	1.2%	1950
BL_324	FRS	7.1	9.1	7.1142	1798.4	1.5%	2300
BL_260	FRS	7.5	9.1	7.1142	52.1	18.3%	1050
BL_274	FRS	2.4	51.0	7.1136	18.0	17.0%	1500
BL_275	FRS	1.8	9.1	7.1142	73.1	26.4%	1300
BL_277	FRS	2.3	0.0	7.1144	18.8	26.2%	1350
BL_330	FRS	3.7	9.1	7.1142	1.9	16.4%	1800
BL_333	FRS	2.8	16.1	7.1141	14.4	33.3%	2400
BL_344	FRS	3.6	0.0	7.1147	28.0	35.6%	1650

**Table S6.** Correlation values of iron solubility. Fields in purple indicate bulk data and individual particles are shown in light orange. The darkest value of each color indicates the highest value. High solubility is defined as greater than 8%, medium solubility is between 3% and 8%, and low solubility is below 3%.

Variable	High Solubility	Medium Solubility	Low Solubility	All Data
pH @ RH 80	0.042688	0.060224	0.698564	0.235733
% Fe(II)	0.083235	0.022738	0.297388	0.053377
Pre-Edge Centroid (keV)	0.092429	0.026193	0.258085	0.058321
Total Fe (ng/m <sup>3</sup> )	0.141756	0.054668	0.245822	0.027118
Distance Travelled in 5 days (km)	0.096638	0.019433	0.002188	0.050779
Iron(III) Oxide (%)	0.004280	0.074307	0.001578	0.023829
Fe(II) Sulfate (%)	0.013609	0.030661	0.082703	0.000069
Fe(II) Silicate (%)	-	-	0.011735	0.036728
Fe(III) Phosphate (%)	0.000532	0.035063	0.011735	0.000045
Fe(III) Sulfate (%)	0.004133	0.059175	0.019535	0.007003
Secondary Phases (%)	0.008470	0.014992	0.032218	0.009748
Pre-Edge Centroid (keV)	0.000086	0.000002	0.002418	0.039703
% Fe(II)	0.041357	0.000419	0.001054	0.071432
Iron(III) Oxide	0.419772	0.016852	0.288424	0.002285
Iron(II) Silicate	0.034774	0.034991	0.116536	0.038312
Fe(II) Sulfate	0.000000	0.129184	0.032342	0.049721
Fe(III) Sulfate	0.030321	0.000532	0.143433	0.028085
Iron(III) phosphate	0.037920	0.002150	0.000025	0.000472



**Table S7.** Composition of bulk aerosol and individual aerosol particles samples.

Sample Site	Type	Group ID	Group Size	Fe(III) Oxide	Fe(II) Sulfate	Fe(III) Sulfate	Fe(III) Phosphate	Ilmenite	Jarosite	Fe(II) Silicate
<b>India</b>	Bulk	1	14	87.5 ± 2.1%	12.5 ± 1.5%	-	-	-	-	-
	Individual Particle	1	34	92.6 ± 0.2%	7.4 ± 0.2%	-	-	-	-	-
		2	15	100 ± 3.9%	-	-	-	-	-	-
		3	9	25.2 ± 2.8%	-	-	-	-	-	74.8 ± 3.9%
<b>Southern Ocean</b>	Bulk	1	2	83.3 ± 9.4%	16.7 ± 0.7%	-	-	-	-	-
		2	1	53.0 ± 1.9%	-	-	-	47.0 ± 4%	-	-
	Individual Particle	1	4	-	11.7 ± 0.4%	-	88.3 ± 5.5%	-	-	-
		2	3	52.2 ± 30.5%	20.6 ± 1.7%	-	-	-	26.9 ± 60.4%	-
		3	4	-	2.7 ± 4.3%	-	-	38.6 ± 42.6%	58.6 ± 73.7%	-
<b>Finokalia Research Station</b>	Bulk	1	13	86.7 ± 0.5%	13.3 ± 0.5%	-	-	-	-	-
	Individual Particle	1	24	94.7 ± 0.3%	5.3 ± 0.3%	-	-	-	-	-
		2	2	74.8 ± 5.6%	-	-	-	25.2 ± 5.6%	-	-
<b>BIOS</b>	Bulk	1	12	81.3 ± 2%	18.7 ± 1%	-	-	-	-	-
		2	20	83.9 ± 0%	16.1 ± 0%	-	-	-	-	-
		3	5	-	38.5 ± 0.4%	61.5 ± 208%	-	-	-	-
	Individual Particle	1	6	17.5 ± 36%	-	-	-	-	-	82.5 ± 11%
		2	12	71.5 ± 1%	-	-	-	-	-	28.5 ± 3%
		3	10	84.5 ± 4%	-	-	-	-	-	15.5 ± 2%
<b>GEO-TRACES</b>	Bulk	1	2	47.2 ± 121%	-	-	10.7 ± 111%	-	-	42.1 ± 11%
		2	3	89.5 ± 0.3%	10.5 ± 4%	-	-	-	-	-
		3	4	32.9 ± 103%	28.7 ± 140%	-	38.4 ± 90%	-	-	-
	Individual Particle	1	27	-	69.2 ± 4%	-	30.8 ± 4%	-	-	-
		2	6	-	36.6 ± 2%	63.4 ± 59%	-	-	-	-
		3	1	10.8 ± 89%	26.6 ± 2%	-	62.6 ± 96%	-	-	-