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*Supplement of*

## **Observations of atmospheric chemical deposition to high Arctic snow**

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## S1 Snow Mixing Ratio Measurements

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### Table Notes

Sampling conditions: BS = blowing snow, DD = diamond dust

SWE = snow-water equivalent

See body of document for other definitions

Values presented as “Mixing Ratio  $\pm$  Uncertainty”

All provided mixing ratios are background subtracted

“<#” indicates values below MDL

For ICP-MS the actual analysis MDL is for the preconcentrated sample, provided are these values corrected by the typical preconcentration correction factors

Blank values indicate no measurement was completed for this sample

Only ICP-MS metals with S/N>2 are presented as those with lower S/N are considered too uncertain

Snow fluxes were calculated from presented measured concentrations, period deposition area, and period length

**Table S1: General description of collection periods**

Sample #	Date		Length (d)	Sample Collected			Average SWE (mm)	Sampling Conditions
	Start	End		SP2	IC	ICP-MS		
1	14-09-14 00:00	15-09-14 13:30	1.56	Y	Y		1.44	
2	15-09-14 13:40	23-09-14 16:30	8.12	Y	Y		0.99	
3	23-09-14 16:40	24-09-14 13:20	0.86	Y	Y		1.33	
4	24-09-14 13:30	03-10-14 14:15	9.03	Y	Y		3.38	
5	03-10-14 14:30	06-10-14 16:30	3.08	Y	Y	Y	0.35	
6	06-10-14 16:50	07-10-14 15:05	0.93	Y	Y	Y	1.43	
7	07-10-14 15:25	15-10-14 15:20	8.00	Y	Y	Y	1.99	
8	15-10-14 15:45	20-10-14 18:15	5.10	Y	Y	Y	1.72	
9	20-10-14 18:15	24-10-14 15:25	3.88	Y	Y	Y	0.35	BS
10	24-10-14 15:55	27-10-14 17:30	3.07	Y	Y	Y	0.52	
11	27-10-14 17:55	05-11-14 14:45	8.87	Y	Y	Y	1.87	BS
12	05-11-14 15:10	07-11-14 15:45	2.02	Y	Y	Y	0.21	DD
13	07-11-14 16:10	10-11-14 17:15	3.05	Y	Y	Y	0.46	
14	10-11-14 17:35	12-11-14 15:55	1.93	Y	Y	Y	1.05	
15	12-11-14 15:55	14-11-14 15:30	1.98	Y	Y	Y	0.45	Fog
16	14-11-14 15:30	17-11-14 16:20	3.03	Y	Y	Y	0.34	
17	17-11-14 16:45	19-11-14 17:05	2.01	Y	Y	Y	0.35	
18	19-11-14 17:30	26-11-14 14:55	6.89	Y	Y	Y	1.10	
19	26-11-14 15:20	05-12-14 14:45	8.98	Y	Y	Y	0.29	
20	05-12-14 15:05	12-12-14 15:15	7.01	Y	Y	Y	1.02	
21	12-12-14 15:35	15-12-14 18:00	3.10	Y	Y	Y	2.04	
22	15-12-14 18:30	17-12-14 17:25	1.95	Y	Y	Y	2.08	
23	17-12-14 17:45	22-12-14 17:00	4.97	Y	Y	Y	1.44	Blizzard
24	22-12-14 17:30	10-01-15 16:30	18.96	Y	Y	Y	0.58	Blizzard
25	10-01-15 16:50	23-01-15 15:55	12.96	Y	Y	Y	0.68	
26	23-01-15 15:55	30-01-15 16:00	7.00	Y	Y	Y	0.13	DD
27	30-01-15 16:00	02-02-15 20:15	3.18	Y	Y	Y	0.41	
28	02-02-15 20:15	13-02-15 17:30	10.89	Y	Y		0.07	BS
29	13-02-15 17:30	17-02-15 14:15	3.86	Y	Y		0.04	
30	17-02-15 14:15	18-02-15 16:50	1.11	Y	Y	Y	0.31	
31	18-02-15 16:50	23-02-15 18:45	5.08	Y	Y	Y	1.55	
32	23-02-15 19:05	04-03-15 14:20	8.80	Y	Y	Y	0.27	
33	04-03-15 14:20	06-03-15 15:15	2.04	Y	Y	Y	0.12	
34	06-03-15 15:15	09-03-15 16:55	3.07	Y	Y	Y	0.50	
35	09-03-15 16:55	11-03-15 14:15	1.89	Y	Y	Y	1.68	
36	11-03-15 14:15	13-03-15 16:55	2.11	Y	Y	Y	2.08	
37	13-03-15 16:55	16-03-15 18:15	3.06	Y	Y	Y	1.22	DD + BS
38	16-03-15 18:15	23-03-15 17:15	6.96	Y	Y	Y	0.97	
39	23-03-15 17:15	27-03-15 13:30	3.84	Y	Y	Y	0.90	
40	27-03-15 13:30	30-03-15 18:30	3.21	Y	Y	Y	1.30	
41	30-03-15 18:30	03-04-15 17:45	3.97	Y	Y	Y	1.47	
42	03-04-15 17:45	05-04-15 14:00	1.84	Y	Y	Y	1.81	
43	05-04-15 14:00	06-04-15 18:15	1.18	Y	Y		0.06	
44-45	06-04-15 18:15	08-04-15 15:45	1.90	Collection for different study				
46	08-04-15 15:45	13-04-15 19:00	5.14	Y	Y	Y	0.87	
47	13-04-15 19:00	15-04-15 14:45	1.82	Y	Y	Y	2.99	
48	15-04-15 14:45	20-04-15 18:00	5.14	Y	Y	Y	0.52	
49	20-04-15 18:00	23-04-15 14:30	2.85	Y	Y	Y	0.99	
50	23-04-15 14:30	27-04-15 15:45	4.05	Y			0.01	DD
51	27-04-15 15:45	30-04-15 15:25	2.99	Y			0.01	DD
52	30-04-15 15:25	04-05-15 19:40	4.18	Y	Y		0.06	DD
53	04-05-15 19:40	11-05-15 18:55	6.97		Y		0.47	
54	11-05-15 18:55	12-05-15 14:33	0.82	Y	Y	Y	1.55	
55	12-05-15 14:33	13-05-15 14:50	1.01	Y	Y	Y	1.53	
56	13-05-15 14:50	18-05-15 18:40	5.16	Y	Y	Y	0.42	
57	25-05-15 16:20	27-05-15 14:10	1.91	Y	Y	Y	0.40	
58	27-05-15 14:10	29-05-15 15:00	2.03	Y	Y	Y	0.51	
59	29-05-15 15:00	01-06-15 16:15	3.05	Y			0.04	Fog

**Table S2a: SP2 analysis and IC analysis – Part 1**

Sample #	Snow Mixing Ratio (ppb)								
	BC	MS	ACE	PRP	FOR	PYR	Cl <sup>-</sup>	NO <sub>2</sub> <sup>-</sup>	Br <sup>-</sup>
1	0.41 ± 0.10	3.9 ± 3.1	4 ± 11	<1.5	5.2 ± 2.5	<11	26 ± 27	<5	<5
2	1.28 ± 0.26	48.3 ± 11.7	44 ± 11	1.6 ± 1.9	9.8 ± 2.6	<11	136 ± 37	22 ± 7	6 ± 5
3	0.60 ± 0.13	7.3 ± 3.4	11 ± 7	<1.5	3.8 ± 2.0	<11	131 ± 48	8 ± 7	<5
4	0.44 ± 0.10	5.8 ± 8.0	10 ± 17	<1.5	1.9 ± 5.3	<11	142 ± 130	8 ± 18	<5
5	0.94 ± 0.10	2.2 ± 0.9	37 ± 4	6.2 ± 0.8	26.9 ± 2.7	6 ± 4	158 ± 18	<5	<5
6	1.66 ± 0.17	1.1 ± 2.9	24 ± 13	5.6 ± 3.1	23.2 ± 9.8	6 ± 16	104 ± 57	<5	<5
7	1.08 ± 0.11	4.3 ± 2.2	38 ± 10	7.7 ± 2.1	24.4 ± 5.8	11 ± 18	66 ± 25	<5	<5
8	1.28 ± 0.13	1.1 ± 3.8	10 ± 6	1.8 ± 2.9	9.3 ± 2.6	5 ± 21	117 ± 40	<5	<5
9	0.83 ± 0.09	1.4 ± 0.7	10 ± 1	<1.5	29.5 ± 1.5	3 ± 4	245 ± 13	8 ± 1	<5
10	0.85 ± 0.09	1.5 ± 0.9	12 ± 1	1.1 ± 0.7	9.0 ± 0.6	3 ± 5	291 ± 20	7 ± 1	<5
11	0.34 ± 0.04	0.8 ± 3.3	9 ± 6	1.8 ± 2.6	11.0 ± 2.7	4 ± 18	47 ± 22	<5	<5
12	0.44 ± 0.05	0.6 ± 0.4	33 ± 1	4.4 ± 0.2	28.2 ± 0.7	3 ± 2	185 ± 6	8 ± 1	<5
13	4.20 ± 0.42	1.2 ± 0.8	21 ± 2	3.0 ± 0.6	17.6 ± 1.0	2 ± 4	253 ± 16	<5	<5
14	3.75 ± 0.38	0.5 ± 2.9	10 ± 6	1.4 ± 2.3	8.6 ± 1.9	4 ± 16	457 ± 103	<5	<5
15	3.16 ± 0.32	1.3 ± 0.8	31 ± 2	3.4 ± 0.6	16.7 ± 1.0	3 ± 4	815 ± 50	16 ± 2	<5
16	2.47 ± 0.25	0.5 ± 0.7	11 ± 1	2.1 ± 0.5	13.2 ± 0.6	4 ± 4	704 ± 36	<5	<5
17	1.49 ± 0.15	0.4 ± 0.7	8 ± 1	1.6 ± 0.6	11.3 ± 0.6	3 ± 4	810 ± 46	<5	<5
18	1.83 ± 0.19	0.7 ± 1.9	7 ± 3	1.5 ± 1.5	6.4 ± 1.0	1 ± 11	1264 ± 184	<5	<5
19	3.86 ± 0.39	0.3 ± 0.9	9 ± 2	2.7 ± 0.7	11.4 ± 0.8	6 ± 5	2049 ± 142	<5	7 ± 2
20	5.51 ± 0.55	0.2 ± 1.8	24 ± 4	5.3 ± 1.0	8.8 ± 1.2	7 ± 10	245 ± 36	5 ± 3	<5
21	4.12 ± 0.41	0.4 ± 4.5	4 ± 10	4.4 ± 2.2	8.6 ± 2.9	8 ± 25	104 ± 43	6 ± 7	<5
22	1.56 ± 0.32	0.2 ± 3.6	20 ± 7	4.2 ± 1.8	12.0 ± 3.1	7 ± 20	56 ± 26	4 ± 10	<5
23	1.65 ± 0.17	0.2 ± 2.7	8 ± 5	2.4 ± 2.1	6.6 ± 1.4	2 ± 15	121 ± 29	7 ± 4	<5
24	1.89 ± 0.19	0.7 ± 1.2	14 ± 2	3.6 ± 0.9	18.6 ± 1.6	7 ± 7	4215 ± 386	<5	44 ± 4
25	2.99 ± 0.30	0.2 ± 1.3	16 ± 2	4.5 ± 0.6	6.6 ± 0.7	3 ± 7	208 ± 21	8 ± 2	6 ± 2
26	1.55 ± 0.16	0.4 ± 0.4	14 ± 1	4.0 ± 0.2	10.0 ± 0.3	3 ± 2	2226 ± 69	<5	12 ± 1
27	2.40 ± 0.24	0.3 ± 0.9	24 ± 2	6.2 ± 0.5	10.4 ± 0.6	7 ± 5	450 ± 29	8 ± 1	5 ± 2
28	13.45 ± 1.35	1.5 ± 0.2	24 ± 0	5.9 ± 0.1	19.9 ± 0.3	4 ± 1	3768 ± 66	<5	25 ± 1
29	10.18 ± 1.02	0.8 ± 0.1	27 ± 0	7.0 ± 0.1	14.5 ± 0.1	3 ± 1	1023 ± 11	<5	<5
30	5.16 ± 0.52	1.0 ± 1.0	30 ± 2	7.8 ± 0.6	10.9 ± 0.8	9 ± 5	208 ± 17	<5	<5
31	2.82 ± 0.28	0.6 ± 2.9	12 ± 5	4.3 ± 1.4	8.8 ± 1.9	12 ± 16	179 ± 42	<5	<5
32	14.79 ± 1.48	0.8 ± 0.5	25 ± 1	7.6 ± 0.3	12.5 ± 0.5	4 ± 3	898 ± 36	9 ± 1	6 ± 1
33	6.83 ± 0.68	1.0 ± 0.4	47 ± 1	12.6 ± 0.4	16.5 ± 0.5	13 ± 2	352 ± 10	6 ± 1	8 ± 1
34	5.30 ± 0.53	0.7 ± 1.0	14 ± 2	4.2 ± 0.6	13.9 ± 1.0	5 ± 6	333 ± 26	<5	<5
35	2.33 ± 0.24	0.4 ± 2.8	10 ± 4	3.6 ± 2.1	10.1 ± 2.1	7 ± 15	57 ± 20	<5	<5
36	2.26 ± 0.23	0.7 ± 3.8	14 ± 6	5.3 ± 2.0	10.8 ± 3.0	6 ± 21	312 ± 92	<5	8 ± 6
37	4.61 ± 0.46	0.7 ± 2.5	25 ± 6	8.0 ± 1.6	12.6 ± 2.3	8 ± 14	241 ± 48	5 ± 4	9 ± 4
38	4.51 ± 0.45	1.4 ± 2.2	35 ± 6	12.1 ± 1.9	14.0 ± 2.2	6 ± 12	651 ± 109	<5	14 ± 4
39	0.95 ± 0.10	0.7 ± 1.7	23 ± 4	8.8 ± 1.2	14.6 ± 1.8	8 ± 9	256 ± 34	<5	9 ± 3
40	1.23 ± 0.13	1.2 ± 2.2	28 ± 5	9.9 ± 1.6	10.4 ± 1.7	5 ± 12	297 ± 51	<5	30 ± 6
41	3.71 ± 0.37	1.1 ± 2.4	24 ± 5	10.4 ± 1.9	14.4 ± 2.5	7 ± 13	198 ± 39	<5	18 ± 5
42	1.31 ± 0.13	1.7 ± 3.4	46 ± 12	17.7 ± 4.3	13.6 ± 3.3	8 ± 19	1301 ± 335	<5	24 ± 8
43	2.50 ± 0.25	2.5 ± 0.1	51 ± 1	19.8 ± 0.3	17.1 ± 0.2	10 ± 1	623 ± 9	<5	12 ± 0
46	1.47 ± 0.15	2.0 ± 0.9	25 ± 3	12.1 ± 1.3	12.6 ± 1.3	7 ± 8	917 ± 104	<5	28 ± 4
47	0.62 ± 0.07	3.7 ± 3.6	14 ± 9	5.1 ± 2.8	10.8 ± 4.3	7 ± 30	72 ± 43	<5	7 ± 9
48	2.45 ± 0.25	18.2 ± 1.2	25 ± 2	8.5 ± 0.6	12.8 ± 0.8	9 ± 5	553 ± 35	<5	22 ± 2
49	4.18 ± 0.42	10.9 ± 1.7	11 ± 3	<1.5	14.2 ± 1.7	6 ± 9	236 ± 32	<5	8 ± 3
50	10.84 ± 1.09								
51	2.40 ± 0.24								
52	7.55 ± 0.76	75.5 ± 1.1	26 ± 0	8.3 ± 0.1	16.2 ± 0.2	12 ± 1	266 ± 4	<5	14 ± 0
53		5.2 ± 0.5	28 ± 2	3.2 ± 0.4	68.8 ± 3.9	<11	127 ± 8	<5	<5
54	0.36 ± 0.04	<1.9	10 ± 4	0.0 ± 2.0	30.1 ± 6.1	<11	123 ± 29	<5	12 ± 5
55	0.46 ± 0.05	1.5 ± 2.8	8 ± 4	<1.5	14.2 ± 3.2	<11	540 ± 117	<5	25 ± 7
56	5.20 ± 0.52	2.3 ± 0.5	175 ± 10	<1.5	487.5 ± 27.9	<11	179 ± 11	<5	12 ± 1
57	1.50 ± 0.15	19.9 ± 1.1	88 ± 5	1.4 ± 0.5	42.5 ± 2.2	<11	225 ± 12	<5	24 ± 2
58	0.94 ± 0.10	16.8 ± 1.3	21 ± 2	<1.5	14.5 ± 1.1	<11	79 ± 8	<5	7 ± 2
59	7.37 ± 0.74								

**Table S2b: IC analysis – Part 2**

Sample #	Snow Mixing Ratio (ppb)								
	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	C <sub>2</sub> O <sub>4</sub> <sup>2-</sup>	Na <sup>+</sup>	NH <sub>4</sub> <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>	H <sup>+</sup>
1	47 ± 18	134 ± 53	<18	11 ± 43	8 ± 8	12 ± 8	13 ± 43	102 ± 319	1.27 ± 0.44
2	76 ± 19	5136 ± 1222	4 ± 30	128 ± 35	17 ± 6	16 ± 6	177 ± 46	1167 ± 307	22.56 ± 5.37
3	31 ± 12	1138 ± 365	6 ± 40	77 ± 34	12 ± 8	19 ± 9	80 ± 35	402 ± 219	3.34 ± 1.07
4	107 ± 88	1318 ± 1070	<18	64 ± 80	10 ± 19	17 ± 22	51 ± 73	330 ± 523	10.55 ± 8.55
5	520 ± 54	557 ± 58	23 ± 8	93 ± 12	11 ± 2	65 ± 7	25 ± 8	287 ± 65	
6	240 ± 106	538 ± 240	18 ± 34	56 ± 41	13 ± 11	26 ± 15	27 ± 35	235 ± 266	
7	93 ± 24	870 ± 217	12 ± 31	46 ± 22	13 ± 6	11 ± 6	22 ± 19	189 ± 146	3.42 ± 0.85
8	33 ± 11	327 ± 96	15 ± 36	63 ± 28	10 ± 7	8 ± 6	12 ± 36	41 ± 264	5.67 ± 1.62
9	86 ± 5	344 ± 18	22 ± 4	126 ± 8	9 ± 1	14 ± 1	53 ± 5	628 ± 44	0.52 ± 0.03
10	93 ± 6	2055 ± 138	13 ± 8	154 ± 12	10 ± 2	23 ± 2	44 ± 6	352 ± 44	24.18 ± 1.63
11	55 ± 15	99 ± 31	10 ± 32	19 ± 19	<5	5 ± 5	9 ± 32	159 ± 145	4.50 ± 1.13
12	97 ± 3	635 ± 18	21 ± 2	100 ± 3	12 ± 1	10 ± 1	43 ± 2	461 ± 20	1.08 ± 0.03
13	100 ± 6	480 ± 29	39 ± 5	122 ± 9	11 ± 1	39 ± 3	184 ± 12	1663 ± 106	0.64 ± 0.04
14	49 ± 12	243 ± 56	16 ± 28	229 ± 54	12 ± 5	22 ± 7	93 ± 27	570 ± 177	0.54 ± 0.12
15	128 ± 8	588 ± 36	12 ± 8	432 ± 27	44 ± 3	22 ± 2	64 ± 6	134 ± 35	6.07 ± 0.37
16	119 ± 6	271 ± 14	18 ± 4	301 ± 16	12 ± 1	18 ± 1	77 ± 5	336 ± 33	1.42 ± 0.07
17	43 ± 3	171 ± 11	17 ± 7	352 ± 20	8 ± 1	18 ± 2	61 ± 5	134 ± 32	2.65 ± 0.15
18	154 ± 23	440 ± 65	33 ± 12	561 ± 82	10 ± 3	26 ± 5	133 ± 22	442 ± 103	1.63 ± 0.24
19	100 ± 7	1289 ± 90	20 ± 5	991 ± 69	27 ± 2	44 ± 3	222 ± 16	494 ± 52	3.83 ± 0.27
20	453 ± 63	306 ± 44	13 ± 17	68 ± 14	12 ± 3	9 ± 3	34 ± 12	213 ± 83	6.51 ± 0.91
21	452 ± 153	348 ± 120	21 ± 26	43 ± 29	14 ± 9	14 ± 9	19 ± 26	150 ± 194	9.19 ± 3.11
22	268 ± 74	197 ± 58	11 ± 34	8 ± 34	6 ± 6	4 ± 10	3 ± 34	33 ± 255	6.97 ± 1.92
23	640 ± 131	242 ± 52	<18	24 ± 16	6 ± 4	5 ± 4	7 ± 25	38 ± 189	9.63 ± 1.96
24	223 ± 20	1267 ± 116	102 ± 12	1601 ± 147	37 ± 4	128 ± 12	511 ± 47	582 ± 74	3.92 ± 0.36
25	93 ± 9	287 ± 28	18 ± 7	60 ± 9	7 ± 2	14 ± 2	38 ± 8	135 ± 54	4.40 ± 0.42
26	258 ± 8	544 ± 17	39 ± 3	951 ± 30	17 ± 1	46 ± 2	252 ± 8	895 ± 33	1.18 ± 0.04
27	85 ± 6	257 ± 17	13 ± 8	134 ± 10	7 ± 1	10 ± 1	60 ± 6	197 ± 38	2.65 ± 0.17
28	660 ± 12	1764 ± 31	100 ± 2	1489 ± 26	58 ± 1	106 ± 2	647 ± 11	6035 ± 106	0.07 ± 0.00
29	639 ± 7	882 ± 9	64 ± 1	327 ± 3	34 ± 0	37 ± 0	162 ± 2	1135 ± 13	0.13 ± 0.00
30	188 ± 14	232 ± 18	18 ± 6	51 ± 7	9 ± 2	10 ± 2	40 ± 6	290 ± 47	4.11 ± 0.31
31	191 ± 42	192 ± 45	8 ± 27	48 ± 19	7 ± 5	21 ± 6	17 ± 27	83 ± 202	4.40 ± 0.96
32	319 ± 13	645 ± 26	27 ± 3	342 ± 14	34 ± 2	34 ± 2	117 ± 5	241 ± 24	6.21 ± 0.25
33	170 ± 5	517 ± 15	9 ± 4	146 ± 5	16 ± 1	6 ± 1	22 ± 2	31 ± 27	9.63 ± 0.28
34	166 ± 13	238 ± 19	13 ± 10	108 ± 10	16 ± 2	7 ± 2	22 ± 6	5 ± 71	10.80 ± 0.83
35	37 ± 9	130 ± 32	7 ± 26	25 ± 17	11 ± 5	2 ± 7	9 ± 26	<133	5.94 ± 1.25
36	50 ± 16	191 ± 59	13 ± 36	123 ± 41	12 ± 7	4 ± 10	24 ± 23	4 ± 266	5.29 ± 1.52
37	38 ± 8	340 ± 67	11 ± 24	98 ± 24	14 ± 5	6 ± 4	28 ± 15	59 ± 177	6.21 ± 1.19
38	151 ± 25	282 ± 48	23 ± 13	239 ± 41	19 ± 5	14 ± 4	70 ± 17	66 ± 153	6.51 ± 1.08
39	80 ± 11	109 ± 17	12 ± 16	70 ± 13	12 ± 3	6 ± 3	34 ± 11	16 ± 119	4.50 ± 0.58
40	97 ± 16	178 ± 32	6 ± 21	77 ± 18	12 ± 4	4 ± 6	30 ± 13	12 ± 154	6.97 ± 1.16
41	<5	286 ± 54	9 ± 23	51 ± 17	13 ± 4	4 ± 6	18 ± 23	9 ± 169	4.71 ± 0.86
42	25 ± 8	269 ± 72	10 ± 32	346 ± 91	13 ± 6	8 ± 6	64 ± 25	46 ± 237	14.91 ± 3.83
43	211 ± 3	414 ± 6	23 ± 1	208 ± 3	15 ± 0	8 ± 0	43 ± 1	54 ± 14	7.30 ± 0.11
46	458 ± 52	202 ± 24	22 ± 9	291 ± 34	15 ± 3	14 ± 3	78 ± 12	121 ± 104	8.58 ± 0.97
47	<5	56 ± 38	9 ± 52	51 ± 37	12 ± 10	2 ± 14	10 ± 52	11 ± 381	1.56 ± 0.64
48	209 ± 13	426 ± 27	21 ± 5	236 ± 15	16 ± 2	12 ± 1	70 ± 6	458 ± 45	1.06 ± 0.07
49	201 ± 26	407 ± 53	19 ± 10	114 ± 18	16 ± 3	11 ± 3	99 ± 16	1332 ± 186	0.42 ± 0.05
50									
51									
52	1152 ± 16	989 ± 14	28 ± 1	133 ± 2	87 ± 1	45 ± 1	53 ± 1	412 ± 10	12.98 ± 0.18
53	736 ± 41	194 ± 12	9 ± 7	51 ± 5	46 ± 3	46 ± 3	44 ± 5	390 ± 38	1.56 ± 0.09
54	87 ± 18	46 ± 18	<18	55 ± 19	10 ± 5	23 ± 6	17 ± 25	107 ± 185	0.86 ± 0.17
55	167 ± 36	96 ± 26	<18	274 ± 61	15 ± 5	24 ± 7	51 ± 19	84 ± 198	2.78 ± 0.59
56	319 ± 18	255 ± 15	15 ± 7	76 ± 6	70 ± 4	21 ± 2	457 ± 27	3468 ± 201	0.42 ± 0.02
57	342 ± 18	331 ± 18	7 ± 7	134 ± 8	41 ± 2	55 ± 3	38 ± 4	320 ± 34	1.27 ± 0.07
58	418 ± 30	210 ± 16	<18	36 ± 6	22 ± 2	24 ± 2	15 ± 9	141 ± 41	1.06 ± 0.07
59									

**Table S3a: ICP-MS analysis – Insoluble (IS) metals Part 1**

Sample #	Snow Mixing Ratio (ppb)								
	IS Al	IS Fe	IS Ti	IS Mn	IS Mg	IS K	IS Ca	IS Pb	IS Zn
1									
2									
3									
4									
5	<3	7 ± 6	<0.3	0.05 ± 0.03	5 ± 4	4 ± 2	<9	0.08 ± 0.03	<1.7
6	<3	15 ± 7	<0.3	0.17 ± 0.04	9 ± 4	3.5 ± 2.1	<9	0.22 ± 0.04	<1.7
7	8 ± 3	14 ± 3	<0.3	0.09 ± 0.02	6 ± 2	4.0 ± 0.9	<9	0.12 ± 0.02	<1.7
8	<3	6 ± 3	<0.3	0.06 ± 0.01	<2	1.6 ± 0.7	<9	0.02 ± 0.01	<1.7
9	31 ± 3	76 ± 8	0.6 ± 0.2	0.93 ± 0.09	54 ± 5	12.0 ± 1.3	98 ± 10	0.32 ± 0.03	2.3 ± 0.9
10	7 ± 2	10 ± 3	<0.3	0.05 ± 0.01	<2	4.2 ± 0.8	<9	0.05 ± 0.01	<1.7
11	<3	5 ± 2	<0.3	0.05 ± 0.01	3 ± 1	2.0 ± 0.6	<9	0.04 ± 0.01	1.9 ± 1.2
12	5 ± 1	8 ± 1	<0.3	0.05 ± 0.01	2 ± 1	2.4 ± 0.4	<9	0.08 ± 0.01	<1.7
13	97 ± 10	219 ± 22	1.1 ± 0.2	3.19 ± 0.32	124 ± 13	44.2 ± 4.4	334 ± 34	0.37 ± 0.04	8.2 ± 1.2
14	38 ± 4	65 ± 7	0.8 ± 0.3	0.63 ± 0.06	29 ± 3	21.6 ± 2.2	48 ± 8	0.18 ± 0.02	<1.7
15	<3	<3	<0.3	0.03 ± 0.02	<2	1.4 ± 0.8	<9	0.05 ± 0.01	<1.7
16	18 ± 2	31 ± 3	<0.3	0.35 ± 0.04	17 ± 2	8.8 ± 1.0	38 ± 6	0.11 ± 0.01	<1.7
17	7 ± 3	<3	<0.3	0.15 ± 0.02	9 ± 2	3.4 ± 0.8	26 ± 9	0.09 ± 0.02	<1.7
18	49 ± 5	92 ± 9	1.5 ± 0.2	1.02 ± 0.10	40 ± 4	19.9 ± 2.0	77 ± 8	0.16 ± 0.02	<1.7
19	33 ± 5	49 ± 7	<0.3	0.37 ± 0.05	19 ± 4	18.8 ± 2.3	<9	0.18 ± 0.03	<1.7
20	17 ± 2	23 ± 3	0.4 ± 0.2	0.20 ± 0.02	9 ± 1	6.8 ± 0.8	<9	0.15 ± 0.02	<1.7
21	33 ± 4	34 ± 4	<0.3	0.29 ± 0.03	9 ± 2	14.5 ± 1.5	<9	0.37 ± 0.04	<1.7
22	<3	<3	<0.3	<0.02	<2	<1.0	<9	0.02 ± 0.01	<1.7
23	<3	<3	<0.3	<0.02	<2	<1.0	<9	0.08 ± 0.01	<1.7
24	84 ± 9	153 ± 15	0.7 ± 0.2	2.38 ± 0.24	70 ± 7	24.9 ± 2.5	146 ± 15	0.47 ± 0.05	7.1 ± 1.0
25	13 ± 2	22 ± 3	<0.3	0.21 ± 0.02	10 ± 1	6.7 ± 0.8	18 ± 6	0.08 ± 0.01	<1.7
26	58 ± 6	86 ± 9	1.3 ± 0.2	0.82 ± 0.08	43 ± 5	27.2 ± 2.8	84 ± 9	0.16 ± 0.02	<1.7
27	34 ± 4	31 ± 4	4.4 ± 0.6	0.19 ± 0.02	10 ± 2	15.5 ± 1.7	<9	0.05 ± 0.01	<1.7
28									
29									
30	12 ± 2	11 ± 2	<0.3	0.10 ± 0.01	6 ± 1	3.6 ± 0.6	<9	0.04 ± 0.01	<1.7
31	3 ± 2	4 ± 2	<0.3	0.04 ± 0.01	<2	1.4 ± 0.7	<9	0.02 ± 0.01	<1.7
32	7 ± 2	10 ± 2	<0.3	0.10 ± 0.01	4 ± 1	3.3 ± 0.6	<9	0.08 ± 0.01	<1.7
33	6 ± 2	7 ± 2	<0.3	0.06 ± 0.01	3 ± 1	2.2 ± 0.5	<9	0.08 ± 0.01	<1.7
34	<3	<3	<0.3	<0.02	<2	<1.0	<9	0.02 ± 0.01	<1.7
35	<3	<3	<0.3	0.05 ± 0.01	<2	1.5 ± 0.8	<9	0.03 ± 0.01	<1.7
36	<3	<3	<0.3	0.03 ± 0.02	<2	<1.0	<9	0.12 ± 0.03	<1.7
37	<3	6 ± 2	<0.3	0.09 ± 0.01	3 ± 1	<1.0	<9	0.20 ± 0.02	<1.7
38	13 ± 2	24 ± 3	<0.3	0.25 ± 0.03	6 ± 1	5.7 ± 0.7	<9	0.14 ± 0.02	<1.7
39	3 ± 1	4 ± 2	<0.3	0.06 ± 0.01	<2	1.7 ± 0.5	<9	0.04 ± 0.01	<1.7
40	<3	3 ± 2	<0.3	0.04 ± 0.01	<2	1.3 ± 0.6	<9	0.04 ± 0.01	<1.7
41	4 ± 2	<3	<0.3	0.05 ± 0.01	<2	1.9 ± 0.5	<9	0.05 ± 0.01	<1.7
42	5 ± 2	6 ± 2	<0.3	0.07 ± 0.01	<2	2.3 ± 0.5	<9	0.03 ± 0.01	<1.7
43									
46	10 ± 2	12 ± 2	0.9 ± 0.2	0.13 ± 0.02	4 ± 1	4.8 ± 0.7	<9	0.11 ± 0.01	<1.7
47	<3	<3	<0.3	0.03 ± 0.01	<2	0.9 ± 0.6	<9	0.02 ± 0.01	<1.7
48	12 ± 2	23 ± 3	<0.3	0.13 ± 0.02	11 ± 2	5.3 ± 0.7	<9	0.07 ± 0.01	<1.7
49	57 ± 6	126 ± 13	0.6 ± 0.2	1.39 ± 0.14	70 ± 7	27.9 ± 2.8	129 ± 13	0.25 ± 0.03	3.2 ± 0.7
50									
51									
52									
53									
54	<3	<3	<0.3	0.05 ± 0.02	<2	2.0 ± 1.0	<9	<0.02	<1.7
55	3 ± 1	<3	<0.3	0.04 ± 0.01	<2	1.1 ± 0.5	<9	<0.02	<1.7
56	247 ± 25	442 ± 44	2.7 ± 0.4	5.76 ± 0.58	447 ± 45	99.7 ± 10.0	1295 ± 130	0.51 ± 0.05	14.2 ± 1.8
57	8 ± 7	<3	<0.3	0.09 ± 0.05	7 ± 6	<1.0	<9	<0.02	<1.7
58	3 ± 1	4 ± 1	<0.3	0.04 ± 0.01	<2	1.4 ± 0.4	<9	<0.02	<1.7
59									

**Table S3b: ICP-MS analysis – Insoluble (IS) metals Part 2**

Sample #	Snow Mixing Ratio (ppb)			Snow Mixing Ratio(parts per trillion mass/mass, i.e., ppt)					
	IS Se	IS Cu	IS Ba	IS As	IS V	IS Cd	IS Tl	IS Co	IS Sb
1									
2									
3									
4									
5	0.02 ± 0.01	0.08 ± 0.04	<1.5	8.5 ± 2.5	6.0 ± 4.8	4.0 ± 1.6	<0.11	<2.4	10.2 ± 3.0
6	<0.01	0.31 ± 0.05	<1.5	<1.1	13.2 ± 4.9	3.5 ± 1.8	<0.11	<2.4	29.2 ± 3.7
7	<0.01	0.06 ± 0.03	9.9 ± 1.6	15.5 ± 1.8	12.2 ± 2.7	<0.9	<0.11	5.4 ± 2.6	15.6 ± 1.9
8	<0.01	<0.02	<1.5	5.2 ± 0.8	5.1 ± 1.7	<0.9	<0.11	3.7 ± 1.8	6.3 ± 1.0
9	0.14 ± 0.01	0.47 ± 0.05	3.1 ± 0.7	57.2 ± 5.7	48.2 ± 5.0	28.3 ± 2.8	0.57 ± 0.07	38.7 ± 4.0	19.6 ± 2.0
10	0.03 ± 0.01	<0.02	<1.5	8.5 ± 1.1	9.0 ± 2.2	1.7 ± 0.8	0.14 ± 0.09	4.1 ± 2.1	22.9 ± 2.5
11	<0.01	0.03 ± 0.02	<1.5	7.9 ± 1.0	4.9 ± 1.6	<0.9	<0.11	<2.4	<1.4
12	0.02 ± 0.00	0.07 ± 0.01	<1.5	12.4 ± 1.3	6.6 ± 1.1	1.6 ± 0.3	0.09 ± 0.04	3.3 ± 0.8	12.6 ± 1.3
13	0.40 ± 0.04	0.75 ± 0.08	6.2 ± 1.0	114.7 ± 11.5	173.4 ± 17.4	<0.9	2.02 ± 0.21	99.1 ± 10.0	29.0 ± 3.0
14	0.12 ± 0.01	0.27 ± 0.03	2.2 ± 1.2	50.2 ± 5.1	72.5 ± 7.5	6.6 ± 0.9	0.79 ± 0.10	23.6 ± 2.8	41.5 ± 4.2
15	<0.01	<0.02	<1.5	8.5 ± 1.1	<2.9	<0.9	<0.11	<2.4	4.8 ± 1.0
16	0.06 ± 0.01	0.33 ± 0.03	3.6 ± 0.8	23.2 ± 2.4	30.7 ± 3.3	2.8 ± 0.5	0.36 ± 0.06	12.8 ± 1.6	10.8 ± 1.2
17	0.02 ± 0.01	0.16 ± 0.02	<1.5	14.8 ± 1.7	10.6 ± 2.4	2.7 ± 0.7	<0.11	4.0 ± 2.3	20.5 ± 2.3
18	0.15 ± 0.01	0.28 ± 0.03	2.5 ± 0.6	64.4 ± 6.5	91.3 ± 9.2	2.9 ± 0.4	0.95 ± 0.10	53.2 ± 5.4	13.9 ± 1.5
19	0.12 ± 0.02	0.09 ± 0.04	<1.5	60.8 ± 6.3	60.9 ± 7.3	<0.9	0.79 ± 0.16	20.0 ± 3.8	9.8 ± 2.1
20	0.05 ± 0.01	0.07 ± 0.01	2.1 ± 0.9	38.7 ± 3.9	28.0 ± 3.1	<0.9	0.38 ± 0.06	10.2 ± 1.5	13.7 ± 1.5
21	0.07 ± 0.01	0.18 ± 0.02	<1.5	58.0 ± 5.8	68.4 ± 7.0	2.1 ± 0.5	0.69 ± 0.09	10.9 ± 1.6	24.7 ± 2.6
22	<0.01	0.03 ± 0.01	3.0 ± 0.8	2.1 ± 0.5	<2.9	<0.9	<0.11	<2.4	9.6 ± 1.1
23	<0.01	0.11 ± 0.02	<1.5	<1.1	<2.9	<0.9	<0.11	<2.4	<1.4
24	0.23 ± 0.02	0.56 ± 0.06	3.7 ± 0.8	92.7 ± 9.3	110.2 ± 11.1	12.6 ± 1.3	1.21 ± 0.13	82.5 ± 8.3	20.6 ± 2.1
25	<0.01	0.05 ± 0.01	3.4 ± 0.8	16.2 ± 1.7	21.5 ± 2.5	<0.9	0.28 ± 0.05	10.4 ± 1.5	2.8 ± 0.8
26	0.10 ± 0.01	0.12 ± 0.02	<1.5	32.2 ± 3.3	105.4 ± 10.6	1.9 ± 0.4	1.18 ± 0.13	41.2 ± 4.3	3.6 ± 0.7
27	<0.01	0.04 ± 0.02	<1.5	14.5 ± 1.6	89.4 ± 9.1	<0.9	<0.11	10.6 ± 1.8	4.7 ± 1.0
28									
29									
30	<0.01	<0.02	<1.5	13.6 ± 1.5	14.4 ± 2.0	<0.9	0.16 ± 0.05	4.3 ± 1.3	2.7 ± 0.9
31	0.01 ± 0.01	<0.02	<1.5	10.7 ± 1.2	6.7 ± 1.6	1.4 ± 0.6	<0.11	<2.4	3.3 ± 0.8
32	0.03 ± 0.01	0.03 ± 0.02	<1.5	24.2 ± 2.5	13.4 ± 2.0	<0.9	0.18 ± 0.06	5.5 ± 1.3	6.6 ± 1.0
33	<0.01	0.06 ± 0.01	<1.5	12.1 ± 1.3	9.7 ± 1.6	14.3 ± 1.5	0.14 ± 0.06	3.4 ± 1.4	32.4 ± 3.3
34	<0.01	<0.02	<1.5	1.9 ± 0.8	<2.9	1.9 ± 0.5	<0.11	<2.4	1.9 ± 0.9
35	<0.01	0.05 ± 0.02	1.8 ± 1.1	3.4 ± 0.7	<2.9	1044 ± 104	<0.11	<2.4	23.2 ± 2.5
36	<0.01	0.05 ± 0.02	<1.5	39.2 ± 7.9	<2.9	5.4 ± 1.3	<0.11	<2.4	16.9 ± 3.6
37	<0.01	0.05 ± 0.01	<1.5	91.2 ± 9.1	12.3 ± 1.8	<0.9	<0.11	4.6 ± 1.1	31.7 ± 3.2
38	0.04 ± 0.01	0.03 ± 0.01	2.0 ± 0.9	25.0 ± 2.5	18.8 ± 2.2	<0.9	0.30 ± 0.05	10.2 ± 1.4	6.3 ± 0.9
39	<0.01	<0.02	1.7 ± 0.8	10.1 ± 1.1	8.6 ± 1.5	<0.9	0.13 ± 0.06	2.3 ± 1.3	8.1 ± 1.0
40	<0.01	0.02 ± 0.01	<1.5	5.3 ± 0.7	4.6 ± 1.3	<0.9	<0.11	<2.4	3.3 ± 0.7
41	0.02 ± 0.00	0.04 ± 0.01	<1.5	6.2 ± 0.8	7.6 ± 1.5	<0.9	0.17 ± 0.05	2.2 ± 1.4	3.7 ± 0.7
42	0.02 ± 0.00	<0.02	8.4 ± 1.1	6.8 ± 0.9	9.1 ± 1.6	<0.9	0.17 ± 0.05	3.7 ± 1.5	2.4 ± 0.8
43									
46	0.02 ± 0.00	0.04 ± 0.01	2.5 ± 0.7	35.9 ± 3.6	17.2 ± 2.1	1.2 ± 0.5	0.24 ± 0.05	4.5 ± 1.1	7.6 ± 1.0
47	<0.01	<0.02	<1.5	3.5 ± 0.6	<2.9	1.2 ± 0.5	<0.11	<2.4	2.8 ± 0.7
48	0.04 ± 0.01	0.07 ± 0.01	<1.5	30.2 ± 3.1	17.2 ± 2.2	1.1 ± 0.5	0.24 ± 0.05	8.9 ± 1.4	12.6 ± 1.4
49	0.20 ± 0.02	0.30 ± 0.03	2.1 ± 0.6	82.0 ± 8.2	91.4 ± 9.2	3.4 ± 0.5	1.04 ± 0.11	56.8 ± 5.8	7.1 ± 0.9
50									
51									
52									
53									
54	<0.01	<0.02	<1.5	2.2 ± 1.1	5.9 ± 3.0	<0.9	<0.11	<2.4	2.3 ± 1.4
55	<0.01	0.03 ± 0.01	<1.5	2.5 ± 0.5	5.1 ± 1.3	<0.9	<0.11	<2.4	2.5 ± 0.8
56	0.73 ± 0.07	0.53 ± 0.06	5.9 ± 1.2	167.1 ± 16.7	344.7 ± 34.5	10.6 ± 1.2	3.69 ± 0.38	192.9 ± 19.4	22.0 ± 2.4
57	<0.01	2.38 ± 0.48	<1.5	6.8 ± 2.8	9.9 ± 6.7	<0.9	<0.11	<2.4	16.8 ± 4.5
58	<0.01	0.03 ± 0.01	<1.5	3.9 ± 0.5	5.1 ± 1.1	<0.9	<0.11	<2.4	4.8 ± 0.7
59									

**Table S4: ICP-MS analysis – Soluble (S) metals**

Sample #	Snow Mixing Ratio (ppb)								Mixing Ratio (ppt)	
	S Ti	S Mn	S Mg	S K	S Ca	S Na	S Cu	S Ni	S As	S Cd
1										
2										
3										
4										
5	<0.01	0.50 ± 0.17	13.0 ± 8.5	16.5 ± 5.5	121 ± 79	44 ± 17	<0.07	<0.19	<3	67 ± 24
6	<0.01	0.75 ± 0.10	15.3 ± 4.3	37.6 ± 4.3	165 ± 51	29 ± 10	<0.07	<0.19	<3	55 ± 12
7	<0.01	0.74 ± 0.10	16.2 ± 4.3	12.4 ± 3.1	207 ± 43	29 ± 10	0.84 ± 0.43	<0.19	<3	39 ± 14
8	<0.01	0.25 ± 0.07	10.0 ± 4.1	6.5 ± 2.8	<8	53 ± 9	<0.07	<0.19	<3	<2
9	0.56 ± 0.09	2.98 ± 0.30	55.6 ± 6.9	42.7 ± 4.8	582 ± 69	215 ± 23	<0.07	<0.19	30 ± 15	149 ± 18
10	<0.01	0.55 ± 0.09	24.6 ± 4.7	13.9 ± 2.6	<8	122 ± 14	<0.07	8.01 ± 1.21	<3	57 ± 12
11	<0.01	<0.01	<0.9	<0.5	<8	<2	<0.07	<0.19	<3	<2
12	<0.01	0.78 ± 0.10	17.8 ± 4.4	16.1 ± 2.7	<8	59 ± 9	<0.07	<0.19	47 ± 16	72 ± 12
13	<0.01	4.49 ± 0.45	54.8 ± 6.8	17.4 ± 2.8	792 ± 88	61 ± 10	<0.07	<0.19	<3	<2
14	<0.01	1.41 ± 0.16	47.6 ± 6.2	22.2 ± 3.1	228 ± 44	210 ± 22	<0.07	<0.19	<3	37 ± 14
15	<0.01	0.18 ± 0.07	59.1 ± 7.1	24.4 ± 3.3	<8	457 ± 46	<0.07	<0.19	58 ± 17	<2
16	<0.01	1.04 ± 0.12	50.1 ± 6.4	20.2 ± 3.0	201 ± 53	252 ± 26	<0.07	<0.19	<3	32 ± 13
17	<0.01	0.41 ± 0.08	37.7 ± 5.5	19.7 ± 2.9	107 ± 49	223 ± 24	<0.07	<0.19	<3	31 ± 13
18	0.19 ± 0.09	1.66 ± 0.18	70.2 ± 8.1	24.4 ± 3.3	252 ± 45	333 ± 34	<0.07	1.91 ± 1.16	<3	<2
19	0.49 ± 0.09	2.24 ± 0.23	240.7 ± 24.4	65.4 ± 6.9	420 ± 56	1281 ± 128	1.94 ± 0.48	<0.19	83 ± 15	87 ± 13
20	0.13 ± 0.08	0.84 ± 0.11	26.8 ± 4.8	11.1 ± 3.0	120 ± 49	120 ± 14	<0.07	<0.19	48 ± 16	23 ± 13
21	<0.01	1.27 ± 0.14	13.6 ± 4.3	12.3 ± 3.1	80 ± 48	45 ± 9	<0.07	<0.19	55 ± 16	40 ± 14
22	<0.01	<0.01	<0.9	<0.5	<8	<2	<0.07	2.45 ± 1.17	<3	<2
23	<0.01	0.23 ± 0.07	<0.9	7.6 ± 2.9	<8	24 ± 10	2.45 ± 0.41	6.45 ± 1.11	<3	47 ± 14
24	0.46 ± 0.08	4.16 ± 0.42	318.0 ± 32.1	94.8 ± 9.7	633 ± 74	1009 ± 101	<0.07	<0.19	42 ± 16	29 ± 13
25	<0.01	0.87 ± 0.11	28.7 ± 5.0	7.5 ± 2.9	146 ± 50	79 ± 11	<0.07	<0.19	43 ± 16	<2
26	<0.01	2.54 ± 0.26	146.7 ± 15.2	39.4 ± 4.5	584 ± 69	689 ± 69	<0.07	<0.19	<3	<2
27	<0.01	0.49 ± 0.08	45.4 ± 6.1	15.6 ± 2.7	<8	121 ± 14	<0.07	<0.19	40 ± 16	<2
28										
29										
30	<0.01	0.50 ± 0.08	22.5 ± 4.6	9.6 ± 3.0	<8	49 ± 9	<0.07	1.83 ± 1.16	25 ± 15	<2
31	<0.01	0.16 ± 0.07	9.7 ± 5.2	<0.5	<8	35 ± 8	<0.07	<0.19	35 ± 16	<2
32	<0.01	0.60 ± 0.09	63.2 ± 7.5	19.4 ± 2.9	96 ± 48	192 ± 21	<0.07	<0.19	63 ± 14	<2
33	<0.01	0.27 ± 0.07	17.8 ± 4.4	12.9 ± 3.2	<8	114 ± 14	<0.07	<0.19	48 ± 16	566 ± 57
34	<0.01	<0.01	18.8 ± 4.4	9.4 ± 3.0	<8	90 ± 12	<0.07	<0.19	<3	38 ± 14
35	<0.01	<0.01	<0.9	8.1 ± 2.9	<8	23 ± 10	<0.07	<0.19	<3	354 ± 37
36	<0.01	<0.01	11.8 ± 8.4	9.6 ± 4.8	<8	71 ± 20	<0.07	<0.19	265 ± 58	102 ± 29
37	<0.01	0.35 ± 0.07	17.5 ± 4.4	12.9 ± 3.2	<8	67 ± 10	<0.07	<0.19	314 ± 34	50 ± 14
38	<0.01	0.91 ± 0.23	53.1 ± 13.3	18.8 ± 5.8	<8	141 ± 32	<0.07	2.48 ± 1.88	47 ± 26	<2
39	<0.01	0.14 ± 0.07	21.9 ± 4.6	10.0 ± 3.0	<8	46 ± 9	<0.07	<0.19	38 ± 16	<2
40	<0.01	<0.01	24.0 ± 4.7	10.4 ± 3.0	<8	56 ± 9	<0.07	<0.19	<3	<2
41	<0.01	0.28 ± 0.07	8.1 ± 5.1	5.9 ± 2.8	<8	36 ± 8	<0.07	<0.19	<3	<2
42	<0.01	<0.01	61.7 ± 7.4	20.4 ± 3.0	<8	322 ± 33	<0.07	<0.19	<3	<2
43										
46	<0.01	0.81 ± 0.10	67.0 ± 7.8	21.9 ± 3.1	96 ± 48	242 ± 25	<0.07	<0.19	119 ± 17	23 ± 13
47	<0.01	<0.01	<0.9	<0.5	<8	26 ± 10	<0.07	<0.19	<3	<2
48	0.21 ± 0.09	0.88 ± 0.11	57.7 ± 7.0	22.5 ± 3.1	248 ± 45	246 ± 26	<0.07	8.54 ± 1.25	<3	37 ± 14
49	0.69 ± 0.10	3.56 ± 0.36	61.3 ± 7.3	24.2 ± 3.3	785 ± 87	84 ± 11	<0.07	2.03 ± 1.16	<3	<2
50										
51										
52										
53										
54	<0.01	<0.01	<0.9	8.8 ± 2.9	<8	42 ± 9	<0.07	<0.19	<3	<2
55	<0.01	0.15 ± 0.07	43.0 ± 5.9	19.4 ± 2.9	<8	258 ± 27	<0.07	<0.19	<3	<2
56	1.25 ± 0.16	5.40 ± 0.54	41.2 ± 5.8	66.3 ± 7.0	1772 ± 181	74 ± 10	<0.07	<0.19	<3	<2
57	<0.01	0.65 ± 0.19	21.6 ± 9.1	45.6 ± 10.1	156 ± 81	111 ± 27	0.73 ± 0.68	<0.19	<3	39 ± 22
58	<0.01	0.25 ± 0.07	<0.9	9.2 ± 3.0	<8	<2	<0.07	<0.19	<3	21 ± 13
59										



**Table S5: Overview of Arctic snow measurements by others for comparison with measured median**

Analysis	Chemical Species	This Study Median (ppb)	Typical Range Reported by Others (ppb)		
			Lower	Upper	References
SP2	BC	2.3	0.5	60	Noone and Clarke, 1988; Peters et al., 1995; Slater et al., 2002; Hagler et al., 2007; Ming et al., 2009; Doherty et al., 2010; Hegg et al., 2010; Forsström et al., 2013
IC	MS	1.1	0.5	20	Ross and Granat, 1986; Davidson et al., 1993; Li and Winchester, 1993; Legrand and Angelis, 1995; Osada et al., 1996; Toom-Sauntry and Barrie, 2002; Dibb et al., 2007; Hegg et al., 2010
	ACE	21	50	300	
	PRP	4	<10 (few measurements)		
	FOR	13	10	80	
	PYR	5*	<10 (few measurements)		
	Cl <sup>-</sup>	245	50	5000	
	Br <sup>-</sup>	2	1	15	
	NO <sub>3</sub> <sup>-</sup>	140	20	300	
	SO <sub>4</sub> <sup>2-</sup>	316	10	700	
	C <sub>2</sub> O <sub>4</sub> <sup>2-</sup>	13*	5	30	
	Na <sup>+</sup>	111	10	3000	
	NH <sub>4</sub> <sup>+</sup>	12	1	60	
	K <sup>+</sup>	14	10	100	
Mg <sup>2+</sup>	43	10	300		
Ca <sup>2+</sup>	193	10	400		
pH	H <sup>+</sup>	4.3	1	20	
ICP-MS (insoluble)	Al	7	1	25	Ross and Granat, 1986; Wolff and David, 1988; Gorzelska, 1989; Boutron et al., 1993; Davidson et al., 1993; Osada et al., 1996; Cheam et al., 1998; Hegg et al., 2009
	Fe	10	1	20	
	Mn	0.1	0.1	5	
	Pb	0.08	0.01	5	
	Cu	0.05	0.01	5	
	V	0.01	0.01	1	
	Cd	0.001*	0.001	0.01	
	Tl	0.0001	0.0001	0.001	

**Notes:** Metals measured by ICP-MS in this study are described as their insoluble median as this was the most common metric measured by others.

\* Indicates an analyte defined as weak or bad in this study (i.e., signal-to-noise < 1).

## **S2 Additional Details on Snow Measurement Methodology**

### **S2.1 Snow Analysis**

Through the cooperation of several academic and research organizations, the collected replicate snow samples were used to measure a broad suite of analytes for each snowfall: carbon species, major ions, and metals. All snow samples were kept frozen prior to analysis, throughout storage and shipping. Preparation and analysis procedures are described in the following sections.

#### **S2.1.1 Single-Particle Soot Photometry**

Black carbon analysis was completed via single-particle soot photometry (SP2) at the Desert Research Institute, Reno, Nevada, as per McConnell et al. (2007). Briefly, snow samples collected in 50 mL PYREX bottles were melted rapidly the morning of analysis, and a 50 mL aliquot of each sample was separated into polypropylene vials. Samples were sonicated for 1 minute immediately prior to analysis and atomized via Apex-Q nebulizer for transfer into the SP2. This analysis quantifies refractory black carbon, hereafter referred to as BC. An analysis blank was analyzed for every batch of 17 samples.

Only particles within a defined size range were quantified by this approach. Particles smaller than 0.02 fg or larger than 50 fg BC were not quantified by the instrument (i.e., about 30 nm and 380 nm diameters, assuming 1.7 g/cm<sup>3</sup> density of BC and spherical). By fitting a lognormal distribution to the sample size distribution, the mass outside the detected range was deemed to be negligible. The average particle size varied from 3 to 11 fg over the snow samples; however, given that samples were thawed and sonicated prior to analysis, this particle size is not expected to represent that which would have been found in the fresh snow.

#### **S2.1.2 Ion Chromatography**

Major ions and organic acids were measured by ion chromatography (IC) at Climate Chemistry Measurements and Research, ECCC, in Toronto, as per Toom-Sauntry and Barrie (2002). Briefly, samples collected in 250 mL high-density polyethylene (HDPE) bottles were melted in a warm water bath. Sample aliquots were analyzed for pH up to 3 hours after melting to minimize gas exchange that could potentially affect the results of these low ionic strength samples. The sample pH was measured using a Sartorius PP-2 pH meter with a Hach Platinum series Flat End electrode and temperature compensation. Anions and cations were measured using a Dionex DX600 Dual Ion Chromatograph and an ICS2000 IC for organic acids, all using a 200 µL sample loop. Anions were analyzed using a Dionex Ionpac AG4A 4mm Guard column and AS4A 4 mm separatory column followed by an ASRS suppressor and eluted with a 1.8 mM Na<sub>2</sub>CO<sub>3</sub> / 1.6 mM NaHCO<sub>3</sub> solution at a flow rate of 2.0 mL/min. Cations were measured using Dionex CG12A/CS12A 4 mm columns and CSRS suppressor with a 20 mM methanesulphonate (MS) eluent solution and a flow rate of 1.0 mL/min.. For organic acids, the analysis used AG18/AS18 columns, ASRS suppressor with an EG40 Eluent Generator to produce a 0.2mM/ 100mM KOH gradient at a flow of 1.0 mL/min. Samples were analyzed for organic acids right after pH measurements, and then stored in the refrigerator at 5o C, ready for inorganic analysis.

#### **S2.1.3 Inductively Coupled Plasma Mass Spectrometry**

Metals analysis was completed via inductively coupled plasma mass spectrometry (ICP-MS) at the University of Toronto. Samples collected in 500 mL HDPE bottles were melted rapidly in a microwave oven and immediately filtered with a 13 mm diameter 0.45 µm cellulose acetate filter (Whatman) using a reusable polytetrafluoroethylene filter holder (Sartorius) and a polyethylene syringe (BD). The full collected volume was filtered for each sample, ranging from 40 to 200 mL meltwater. For every four samples, a preparation blank was prepared by filtering 40 to 50 mL DIW . In all subsequent steps, these preparation blanks were treated identically to the snow samples. Both filtrate and filter were kept for analysis, to quantify soluble and

insoluble metals, respectively. It should be noted that the adopted terminology is not exact; fine particles may not be retained by the filter and thus be quantified as part of the “insoluble” portion.

To enable detection by ICP-MS, elements must be separated from any organic matrix by the addition of a digestion reagent, in this procedure nitric acid (HNO<sub>3</sub>). A 10 mL aliquot of filtrate was transferred to a polypropylene tube (Falcon) and ultra-trace grade 70% HNO<sub>3</sub> (SCP Science PlasmaPure) added to each filtrate aliquot to bring the solution to 2% HNO<sub>3</sub> mass/mass (m/m). Metals in coarser particles retained during filtration require an augmented digestion procedure to enable quantification. It has been shown that microwave digestion significantly improves the recovery of insoluble metals without the need for more hazardous and difficult to handle digestion reagents, such as hydrofluoric acid (Ödman et al. 1999; Swami et al. 2001; Pekney and Davidson 2005; US EPA 2007). Each cellulose filter was placed in a polytetrafluoroethylene microwave digestion tube (CEM MARSPress) with 0.4 mL of ultra-trace grade 70% HNO<sub>3</sub>. Filtrate and filter were sealed and left overnight at room temperature to digest. After a 10-15 hr digestion time, DIW was added to each filter vessel to dilute the solution to 2% HNO<sub>3</sub>. The filter samples were then placed in a microwave digester (CEM MARS 6) and digested using the US Environmental Protection Agency (US EPA) 3051 procedure for trace metal analysis (2007). The digested filter solutions were transferred to polypropylene tubes and centrifuged to separate any undigested residue. An average sample mass closure of ±1% was observed over the digestion procedure (i.e., sample mass was measured at all stages of analysis to confirm significant sample was not lost during handling). Preliminary trials suggested that microwave digestion improved insoluble metal detection by 96% on average.

Filter and filtrate samples were analyzed by ICP-MS (Thermo Scientific iCAP Q) under kinetic energy discrimination mode. Dwell times were set to 0.02 s for analytes with an ionization potential less than 9 eV and 0.10 s for analytes with a greater ionization potential. Wash time was set to 75 s and uptake time to 50 s for all analyses (as per Henry and Wills 2012; Wang and Miao 2016). The full 4.6-245 mass/charge ratio (m/z) range was scanned and, where necessary, interference corrections were applied (as per Henry and Wills 2012; Wang and Miao 2016). It should be noted that this preparation and analysis procedure is not expected to retain highly volatile metals, such as Hg.

All apparatus components were thoroughly cleaned before and between uses. The filter holder, syringes, sample tubes, and tweezers for filter handling were each rinsed with ultra-trace grade HNO<sub>3</sub> and twice with DIW prior to each use. Microwave digestion vessels underwent a full digestion cycle with 15 mL 70% HNO<sub>3</sub> and then were rinsed with ultra-trace grade HNO<sub>3</sub> and DIW prior to each use. All sample preparation was completed in class 100 vertical laminar flow cabinet (AirClean Systems AC 632) to minimize contamination. Prior to sample analysis, three brands of cellulose filters were tested: Millipore nitrocellulose filter (manufacturer number: HAWP01300), Whatman cellulose acetate filter (10404001), and Whatman nitrocellulose filter (7184-001). All three brands showed similar contamination levels; however, the Whatman cellulose acetate filter was found to give the most consistent background.

## **S2.2 Quality Assurance and Control**

Analysis instruments were calibrated before and between sample analyses as discussed in the body of the text. The SP2 analysis was calibrated using Cab-O-Jet 200 Black Colourant (Lot # 1115709) each day. The calibration solution was reanalyzed every batch of 17 samples as a quality control.

The IC analysis was calibrated daily. Organic acids used Alltech Analytical calibration solution for MS, Fluka Analytical's TraceCert solution for the organic acids acetate, formate, propionate, oxalate and sodium pyruvate salt from Sigma-Aldridge. High purity salts were used to make yearly 0.1N 1L stock solutions. These were used to make inorganic calibration standards with the exception of Mg and Ca. A separate calibration was made for these using SPEX CertiPrep Ca and Mg in 2% HNO<sub>3</sub> solutions. For quality control, the stock solutions were used to make a yearly batch of four internal quality controls. Dilutions of

Instrument Check Standard 2 (anions) and Instrument Check Standard 3 (cations), both from SPEX CertiPrep, were used as external quality controls and were made fresh with every run. Blanks and sample repeats were used for approximately every ten samples.

For ICP-MS analysis, an instrument performance test was completed at the beginning of each day and a calibration curve was run prior to each analysis using SCP Science Quality Control - Standard 4. Furthermore, the calibration curve was reanalyzed as a quality control check every ten samples to quantify instrument drift. These checks were found to agree with the original calibration on average by  $\pm 14\%$  across all analytes above detection limits ( $\pm 6\%$  agreement or better for analytes in Table 1). All concentrated standards were followed by an extra DIW blank to prevent extensive carry-over from standard to samples. Additional test were completed to confirm that two DIW blanks following each calibration curve was sufficient to limit any possible carry over to less than detection limits. Calibration quality was assured through analysis of Environment Canada's certified reference material TMRAIN-04 with an agreement of  $\pm 3-19\%$  across all analytes within the reference ( $\pm 10\%$  agreement or better for analytes in Table 1). An internal standard was included in the ICP-MS analysis to quantify and correct for any instrument drift or inter-sample variability. The SCP Science Int. Std. Mix 1 was selected so as to minimize interference with measured analytes while covering the full analyzed spectrum of mass to charge ratios. This internal standard was added to the digested solution as they were sampled for analysis.

As an additional quality assurance, consistency between the IC and ICP-MS measurements was considered. Although analyte measurements are expected to differ across instruments due to inherent differences to each measurement technique, the comparison of measurements by two different instruments can help support the validity of both measurement procedures. Four metals were measure by both IC and ICP-MS: Na, K, Mg, and Ca. The IC and ICP-MS measurements of these analytes were fit via linear least squares regression to assess constancy. To allow for differences in instrument detection limit, only measurements above both MDLs were included in the comparison. Thus, the Na, K, Mg, and Ca correlations included 18, 34, 31, and 29 coincident measurements, respectively. Measured IC and ICP-MS Na, K, Mg, and Ca concentrations were found to have a Pearson's correlation coefficients of 0.93, 0.62, 0.97, and 0.98, with slopes of 0.80, 1.2, 0.90, and 0.80, respectively. Thus, the two instruments showed good agreement.

### S2.3 Measurement Uncertainty

The uncertainty of each measurement was estimated based on analysis detection limits and reproducibility as follows (Reff, Eberly, and Bhave, 2007; Norris et al., 2014):

$$u_{ij} = \sqrt{(EF_j x_{ij})^2 + \left(\frac{1}{2} MDL_j\right)^2} \quad , \quad \text{if } x_{ij} \geq MDL_j \quad (S1)$$

$$u_{ij} = \frac{5}{6} MDL_j \quad , \quad \text{if } x_{ij} < MDL_j$$

where  $x_{ij}$  is the  $i^{\text{th}}$  measured value of analyte  $j$ ,  $u_{ij}$  is the uncertainty associated with this measurement,  $EF_j$  is the error fraction for this analyte, and  $MDL_j$  is the method detection limit for this analyte, as summarized in Table S6. The error fraction of each analyte was calculated as double the standard error of replicate measurements for each analysis, with a minimum of 10% imposed. The method detection limit of each analyte was calculated as three standard deviations of analyzed blank samples. The ICP-MS method analysed pre-concentrated samples; thus, the actual method detection limits are applicable only to this pre-concentrated solution. The values presented above are the approximate method detection limits at snow concentrations (i.e., the average MDL when pre-concentrated limits are adjusted to match snow concentrations). The uncertainty for any sample with known preparation concerns was doubled (e.g., partial sample melt in transit or poor mass closure over preparation); however, less than 7% of samples were noted as having potential preparation concerns.

**Table S6: Arctic snow measurement error fractions, detection limits, and signal-to-noise**

Analysis	Chemical Species	Error Fraction EF <sub>j</sub>	Method Detection Limit MDL <sub>j</sub> (ppb)	Signal-to-Noise S/N <sub>j</sub>
SP2	BC	10%	0.042	8.4
IC	MS	12%	1.9	1.2
	ACE	12%	4.4	4.1
	PRP	12%	1.5	2.1
	FOR	12%	1.2	6.2
	PYR	12%	11	0.0
	Cl <sup>-</sup>	12%	18	6.4
	Br <sup>-</sup>	12%	5	1.6
	NO <sub>3</sub> <sup>-</sup>	12%	5	6.7
	SO <sub>4</sub> <sup>2-</sup>	12%	18	6.7
	C <sub>2</sub> O <sub>4</sub> <sup>2-</sup>	12%	18	0.8
	Na <sup>+</sup>	12%	18	5.1
	NH <sub>4</sub> <sup>+</sup>	12%	5	3.6
	K <sup>+</sup>	12%	5	3.6
	Mg <sup>2+</sup>	12%	18	3.2
	Ca <sup>2+</sup>	12%	133	2.3
H <sup>+</sup>	12%	2.9	7.3	
ICP-MS (insoluble)	Al	10%	3.2	3.5
	Fe	10%	3.4	3.8
	Mn	10%	0.02	5.1
	Pb	10%	0.02	4.8
	Cu	10%	0.02	3.3
	V	10%	0.003	4.7
	Cd	10%	0.001	2.3
	Tl	10%	0.0001	2.6

The signal-to-noise (S/N) of each analyte was calculated as per Norris et al., 2014:

$$S/N_j = \frac{1}{n} \sum_{i=1}^n d_{ij} \quad ; \quad (S2)$$

$$d_{ij} = \frac{x_{ij} - u_{ij}}{u_{ij}} \quad , \quad \text{if } x_{ij} > u_{ij}$$

$$d_{ij} = 0 \quad , \quad \text{if } x_{ij} < u_{ij}$$

where S/N<sub>j</sub> is the signal-to-noise of analyte j, n is the total number of samples, and d<sub>ij</sub> is a measure of the difference between the measured value and its uncertainty for the i<sup>th</sup> measurement of this analyte (all other variables are defined as in Eq. S1). The S/N for each analyte is provided in Table S6. A S/N over 1 was considered to be strong, between 0.5 and 1 considered weak, and below 0.5 considered bad (Norris et al., 2014). The low S/N values calculated for MS and Br<sup>-</sup> are in part the result of the seasonal trend expected for these analytes with large periods of very low (<MDL) concentrations.

## S3 Ambient Atmospheric Measurements and Methodology Details

### S3.1 Ambient Atmospheric Measurements

Table S7a: Measured atmospheric concentrations – Part 1(measurement ± uncertainty)

Sample #	Date		Length (d)	Atmospheric Concentration (ng/m <sup>3</sup> )				
	Start	End		SP2 BC	MS	Cl <sup>-</sup>	Hi-Vol Br <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>
1	16-09-14 23:48	22-09-14 19:15	5.81	1.8 ± 0.4	8.1 ± 1.0	<7.3	<0.3	10.8 ± 4.8
2	22-09-14 19:30	29-09-14 14:28	6.79	2.1 ± 0.4	6.1 ± 0.8	<7.3	<0.3	9.5 ± 4.8
3	29-09-14 14:35	06-10-14 16:55	7.10	5.5 ± 1.1	5.2 ± 0.7	<7.3	0.4 ± 0.2	12.3 ± 4.9
4	06-10-14 17:06	13-10-14 17:20	7.01	2.5 ± 0.5	3.3 ± 0.5	<7.3	<0.3	21 ± 5
5	13-10-14 17:30	20-10-14 17:55	7.02	2.7 ± 0.5	1.4 ± 0.3	9.4 ± 3.8	<0.3	9.3 ± 4.8
6	20-10-14 18:05	27-10-14 15:30	6.89	3.8 ± 0.8	2.1 ± 0.4	60 ± 8	0.7 ± 0.2	21 ± 5
7	27-10-14 13:39	03-11-14 19:59	7.26	5.0 ± 1.1	2.4 ± 0.4	11.2 ± 3.9	<0.3	36 ± 6
8	03-11-14 20:10	10-11-14 18:21	6.92	15.8 ± 2.9	2.1 ± 0.4	28 ± 5	0.4 ± 0.2	35 ± 6
9	10-11-14 18:24	17-11-14 14:46	6.85	11.0 ± 2.1	1.9 ± 0.3	609 ± 73	5.6 ± 0.7	63 ± 9
10	17-11-14 14:52	24-11-14 14:59	7.00	28.1 ± 4.6	1.4 ± 0.3	1456 ± 175	12.9 ± 1.6	133 ± 17
11	24-11-14 15:05	02-12-14 15:36	8.02	17.7 ± 3.2	1.1 ± 0.3	307 ± 37	3.7 ± 0.5	73 ± 10
12	02-12-14 15:46	08-12-14 14:44	5.96	18.4 ± 3.3	1.3 ± 0.3	279 ± 34	8.6 ± 1.0	70 ± 10
13	08-12-14 14:51	15-12-14 15:46	7.04	74.9 ± 13.0	2.1 ± 0.4	15 ± 4	2.1 ± 0.3	109 ± 14
14	15-12-14 15:54	22-12-14 16:10	7.01	44.9 ± 8.0	1.4 ± 0.3	49 ± 7	5.1 ± 0.6	79 ± 11
15	22-12-14 16:17	29-12-14 16:42	7.02	25.4 ± 4.4	1.0 ± 0.3	26 ± 5	1.9 ± 0.3	51 ± 8
16	29-12-14 16:52	05-01-15 19:30	7.11	28.3 ± 5.0	0.9 ± 0.3	199 ± 24	26 ± 3	57 ± 8
17	05-01-15 19:38	12-01-15 15:46	6.84	23.8 ± 3.7	0.6 ± 0.3	1740 ± 209	51 ± 6	135 ± 17
18	12-01-15 15:51	19-01-15 16:23	7.02	5.0 ± 0.9	<0.5	57 ± 8	8.6 ± 1.0	36 ± 6
19	19-01-15 16:28	26-01-15 16:31	7.00	58.9 ± 9.8	1.1 ± 0.3	48 ± 7	3.3 ± 0.4	63 ± 9
20	26-01-15 16:58	02-02-15 18:25	7.06	34.8 ± 6.1	<0.5	505 ± 61	5.3 ± 0.7	86 ± 11
21	02-02-15 18:29	09-02-15 16:33	6.92	40.5 ± 7.1	1.1 ± 0.3	948 ± 114	9.5 ± 1.1	96 ± 12
22	09-02-15 16:39	16-02-15 16:10	6.98	66.4 ± 11.4	1.6 ± 0.3	643 ± 77	5.5 ± 0.7	163 ± 20
23	16-02-15 16:15	23-02-15 20:00	7.16	60.9 ± 10.8	<0.5	184 ± 22	4.1 ± 0.5	138 ± 17
24	23-02-15 20:09	02-03-15 17:31	6.89	55.8 ± 9.7	<0.5	905 ± 109	8.3 ± 1.0	196 ± 24
25	02-03-15 17:35	09-03-15 16:24	6.95	42.5 ± 7.4	1.3 ± 0.3	517 ± 62	5.9 ± 0.7	118 ± 15
26	09-03-15 16:30	16-03-15 17:33	7.04	40.9 ± 7.1	1.1 ± 0.3	610 ± 73	14.0 ± 1.7	144 ± 18
27	16-03-15 17:38	23-03-15 13:13	6.82	33.5 ± 6.0	1.5 ± 0.3	166 ± 20	10.8 ± 1.3	84 ± 11
28	23-03-15 13:21	30-03-15 17:47	7.18	28.8 ± 5.4	2.4 ± 0.4	18 ± 4	13.8 ± 1.7	63 ± 9
29	30-03-15 17:53	06-04-15 13:06	6.80	27.8 ± 5.2	2.6 ± 0.4	646 ± 78	13.2 ± 1.6	114 ± 14
30	06-04-15 13:26	13-04-15 13:39	7.01	26.8 ± 5.2	3.1 ± 0.5	571 ± 69	23 ± 3	142 ± 18
31	13-04-15 15:41	20-04-15 19:22	7.15	27.6 ± 5.3	11.5 ± 1.4	396 ± 48	14.6 ± 1.8	122 ± 15
32	20-04-15 19:43	27-04-15 17:03	6.89	19.9 ± 4.1	<0.5	52 ± 7	5.4 ± 0.7	121 ± 15
33	27-04-15 17:08	04-05-15 17:23	7.01	7.5 ± 1.6	<0.5	<7.3	7.4 ± 0.9	100 ± 13
34	04-05-15 17:29	11-05-15 17:56	7.02	6.0 ± 1.3	12.2 ± 1.5	<7.3	4.7 ± 0.6	72 ± 10
35	11-05-15 18:03	18-05-15 19:37	7.07	4.3 ± 0.9	14.6 ± 1.8	482 ± 58	20 ± 2	119 ± 15

Notes: “<#” indicates values below MDL

**Table S7b: Measured atmospheric concentrations – Part 2 (measurement ± uncertainty)**

Sample #	Atmospheric Concentration (ng/m <sup>3</sup> )						
	SO <sub>4</sub> <sup>2-</sup>	C <sub>2</sub> O <sub>4</sub> <sup>2-</sup>	Na <sup>+</sup>	Hi-Vol NH <sub>4</sub> <sup>+</sup>	K <sup>+</sup>	Mg <sup>2+</sup>	Ca <sup>2+</sup>
1	789 ± 95	1.6 ± 0.6	20 ± 3	<13.8	1.1 ± 0.3	7.4 ± 1.1	42 ± 6
2	1290 ± 155	2.5 ± 0.7	18 ± 2	<13.8	1.0 ± 0.3	9.3 ± 1.3	55 ± 8
3	1324 ± 159	<1.2	41 ± 5	<13.8	1.7 ± 0.4	7.5 ± 1.1	30 ± 5
4	906 ± 109	1.9 ± 0.6	7.8 ± 1.3	62 ± 10	2.0 ± 0.4	3.0 ± 0.8	18 ± 4
5	741 ± 89	<1.2	48 ± 6	62 ± 10	4.0 ± 0.6	7.3 ± 1.1	11.8 ± 4.1
6	906 ± 109	4.2 ± 0.8	137 ± 16	75 ± 11	8.6 ± 1.1	23 ± 3	37 ± 6
7	985 ± 118	6.0 ± 0.9	74 ± 9	72 ± 11	6.6 ± 0.9	18 ± 2	78 ± 10
8	779 ± 94	7.2 ± 1.0	91 ± 11	33 ± 8	8.0 ± 1.0	22 ± 3	72 ± 9
9	620 ± 75	5.1 ± 0.8	434 ± 52	30 ± 8	20 ± 2	72 ± 9	57 ± 8
10	806 ± 97	6.1 ± 0.9	921 ± 111	30 ± 8	43 ± 5	143 ± 17	84 ± 11
11	1218 ± 146	8.0 ± 1.1	478 ± 57	28 ± 8	24 ± 3	87 ± 10	91 ± 12
12	1617 ± 194	5.6 ± 0.9	464 ± 56	59 ± 10	23 ± 3	63 ± 8	51 ± 7
13	1520 ± 182	12.7 ± 1.6	118 ± 14	132 ± 17	25 ± 3	18 ± 2	29 ± 5
14	1474 ± 177	7.2 ± 1.0	151 ± 18	85 ± 12	18 ± 2	21 ± 3	16 ± 4
15	898 ± 108	3.9 ± 0.7	77 ± 9	36 ± 8	8.0 ± 1.0	10.0 ± 1.4	11.3 ± 4.1
16	1549 ± 186	7.3 ± 1.1	419 ± 50	49 ± 9	26 ± 3	80 ± 10	48 ± 7
17	1421 ± 171	13.5 ± 1.7	941 ± 113	40 ± 8	93 ± 11	334 ± 40	155 ± 19
18	645 ± 78	4.0 ± 0.8	123 ± 15	<13.8	7.9 ± 1.0	35 ± 4	46 ± 7
19	1057 ± 127	6.8 ± 1.0	127 ± 15	83 ± 12	20 ± 2	28 ± 3	30 ± 5
20	781 ± 94	7.2 ± 1.0	309 ± 37	52 ± 9	30 ± 4	94 ± 11	105 ± 13
21	882 ± 106	16 ± 2	442 ± 53	72 ± 11	45 ± 5	162 ± 20	197 ± 24
22	1150 ± 138	21 ± 3	378 ± 45	115 ± 15	48 ± 6	137 ± 16	313 ± 38
23	1312 ± 158	16 ± 2	197 ± 24	167 ± 21	34 ± 4	68 ± 8	75 ± 10
24	1013 ± 122	10.4 ± 1.4	503 ± 60	180 ± 23	54 ± 7	143 ± 17	70 ± 9
25	1216 ± 146	8.0 ± 1.1	415 ± 50	106 ± 14	35 ± 4	92 ± 11	40 ± 6
26	963 ± 116	11.0 ± 1.4	436 ± 52	131 ± 17	38 ± 5	104 ± 12	55 ± 8
27	940 ± 113	12.2 ± 1.6	155 ± 19	133 ± 17	22 ± 3	42 ± 5	34 ± 6
28	1175 ± 141	14.8 ± 1.9	92 ± 11	105 ± 14	18 ± 2	21 ± 3	18 ± 4
29	1299 ± 156	17 ± 2	495 ± 59	127 ± 17	39 ± 5	98 ± 12	52 ± 7
30	1250 ± 150	17 ± 2	496 ± 59	109 ± 15	38 ± 5	100 ± 12	56 ± 8
31	1850 ± 222	21 ± 3	585 ± 70	105 ± 14	36 ± 4	108 ± 13	136 ± 17
32	1303 ± 156	17 ± 2	170 ± 20	98 ± 14	18 ± 2	54 ± 6	204 ± 25
33	454 ± 55	5.3 ± 0.9	40 ± 5	45 ± 9	5.7 ± 0.7	9.6 ± 1.3	28 ± 5
34	276 ± 34	5.0 ± 0.8	11.1 ± 1.6	32 ± 8	3.7 ± 0.5	5.6 ± 1.0	36 ± 6
35	424 ± 51	9.3 ± 1.3	308 ± 37	42 ± 9	16 ± 2	59 ± 7	252 ± 31

### S3.2 Ambient Atmospheric Methodology

Ground-level atmospheric monitoring data from Alert were provided conducted by Environment Canada at Alert Observatory, as part of the World Meteorological Organization Global Atmospheric Watch program. The aerosol intake is at a height of 10 m above the ground, and the particles are pulled down a 20 cm diameter vertical tube at a flow rate of 1000 L/min. The sampling stream is split into stainless steel 0.5 to 2.5 cm lines, heated, as needed, to maintain a relative humidity of no more than 40%.

Atmospheric BC was monitored by SP2. The operating principles of the SP2 have been described extensively in previous publications (Stephens et al., 2003; Baumgardner, 2004; Schwarz et al., 2006), and we will only give a short overview in this work. Ambient air was sampled by the SP2 at an average flow rate of ~120 cm<sup>3</sup>/min. The sampled air passes through an intracavity Nd:YAG laser operating at 1064 nm, which heats BC-containing particles to high temperatures and causes them to incandescence. The peak intensity of the emitted incandescence light is proportional to the mass of refractory material in the particle (Moteki and Kondo, 2007; Slowik et al., 2007), which can be quantified using an external standard.

The SP2 used here had a detection range of approximately 0.50 – 40 fg of refractory BC which corresponds to BC particles with volume-equivalent diameters from 80 to 350 nm assuming a BC density of 1.8 g/cm<sup>3</sup> (Bond and Bergstrom, 2006). To estimate the mass of BC outside of this detected range, a lognormal distribution was fit to the ambient measurements. In general, the

detected range represented greater than 90% of the total estimated BC mass. To account for the small amount of BC mass outside the detection range, ambient BC measurements were multiplied by a correction factor which ranged from 1.05 to 1.66.

Major ions by IC of high-volume filters of total suspended particles (Hi-Vol) (as per Sirois and Barrie 1999). Aerosol samples were collected on Whatman 41 filters, 20 by 25 cm, which were pre-loaded into Teflon-coated cartridges and secured in plastic bags within an aerosol clean bench. Sampling used a high-volume sampler with a face velocity of 50 cm/s and an approximate collection efficiency of 95% over a period of six to eight days per filter. Sample volumes were calculated using the average flow rate measured by a critical orifice plate at the beginning and end of each collection. A field blank was prepared monthly by briefly placing a clean filter in the sampler before immediately returning it to the protective bag. Sampled filters and blanks were divided into eight aliquots and stored in a dark room temperature location for up to a year prior. An aliquot of each sample was extracted into 60 mL of water and analysed via liquid IC (Dionex). The IC instrument was calibrated as above for snow IC analysis.

### S3.3 Quality Assurance and Uncertainty

Quality assurance/control and uncertainty for ambient measurements was similar to that of the snow measurements as discussed above. Mass calibration for the atmospheric SP2 used in this work was done using Aquadag particles (Acheson Industries), which were size-selected by a differential mobility analyzer (DMA). Aquadag mobility diameters were converted to BC mass using the size-dependent effective densities reported by Gysel et al. (2011). Recent studies have shown that the SP2 is more sensitive to Aquadag than it is to ambient BC particles (Laborde et al., 2012; Moteki and Kondo, 2010). To account for this, the slope of the Aquadag calibration curve was scaled by a factor of  $0.70 \pm 0.05$ , as derived from Figure 5 in Laborde et al. (2012). The uncertainty in the 1-hour BC mass concentration is approximately 20%. This arises from uncertainty in the fit of the Aquadag calibration curve, uncertainty in the single lognormal fit to the mass distribution (used to calculate what portion of the ambient BC mass is outside of the detection range), and uncertainty in the correction factor correcting for the SP2 detection bias towards Aquadag. It should be noted that the ambient and snow SP2 instruments were calibrated using different solutions, Aquadag and Cab-O-Jet, respectively. However, these two solutions have previously been shown to both provide high confidence BC measurements (Wendl et al., 2014).

The ambient IC analysis was calibrated and checked as described for snow measurements. Uncertainty of the ambient IC measurements was quantified via Eq. S1 above using values provided in Table S8. Diagnostic variables for atmospheric SP2 and Hi-Vol measurements are also provided.

**Table S8: Arctic atmospheric measurement error fractions, detection limits, and signal-to-noise**

Analysis	Chemical Species	Error Fraction EF <sub>j</sub>	Method Detection Limit MDL <sub>j</sub> (ng/m <sup>3</sup> )	Signal-to-Noise S/N <sub>j</sub>
SP2	BC	20%	0.001	4.3
IC Hi-Vol	MS	12%	0.5	5.9
	Cl <sup>-</sup>	12%	7.3	6.0
	Br <sup>-</sup>	12%	0.3	6.2
	NO <sub>3</sub> <sup>-</sup>	12%	9	7.2
	SO <sub>4</sub> <sup>2-</sup>	12%	10	7.3
	C <sub>2</sub> O <sub>4</sub> <sup>2-</sup>	12%	1.2	6.8
	Na <sup>+</sup>	12%	1.7	7.3
	NH <sub>4</sub> <sup>+</sup>	12%	14	6.4
	K <sup>+</sup>	12%	0.6	7.3
	Mg <sup>2+</sup>	12%	1.3	7.3
Ca <sup>2+</sup>	12%	8	7.2	



## S4 Collection Campaign Details

### S4.1 Collection Site

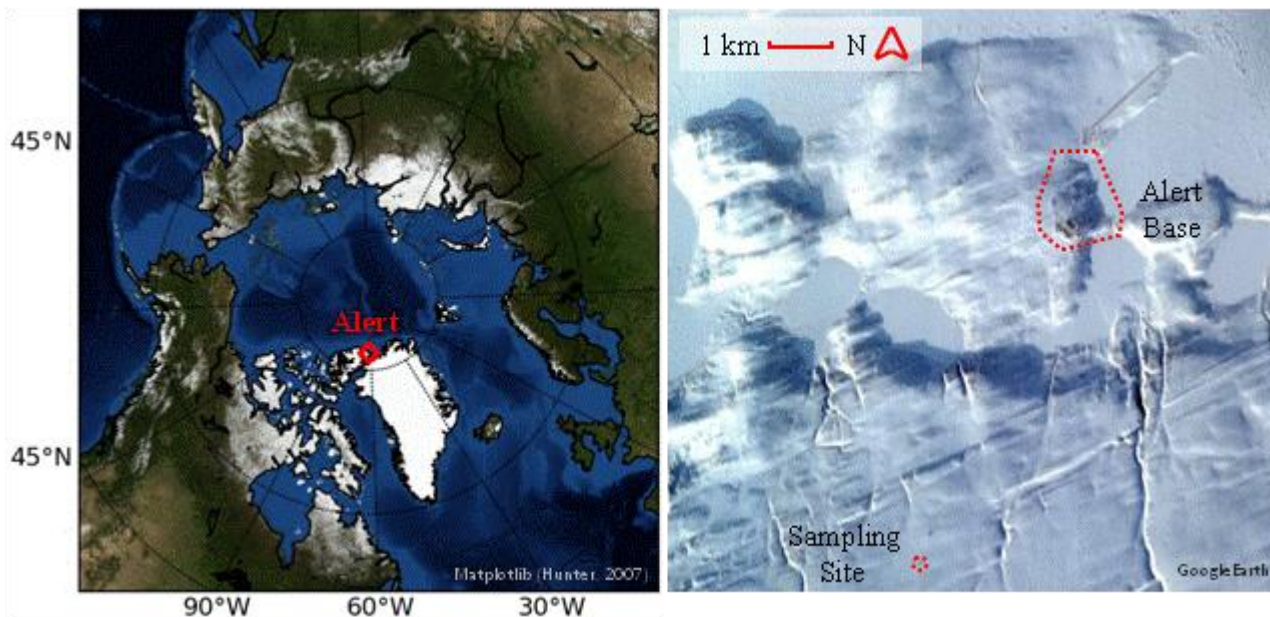


Figure S1: Alert location and base camp map

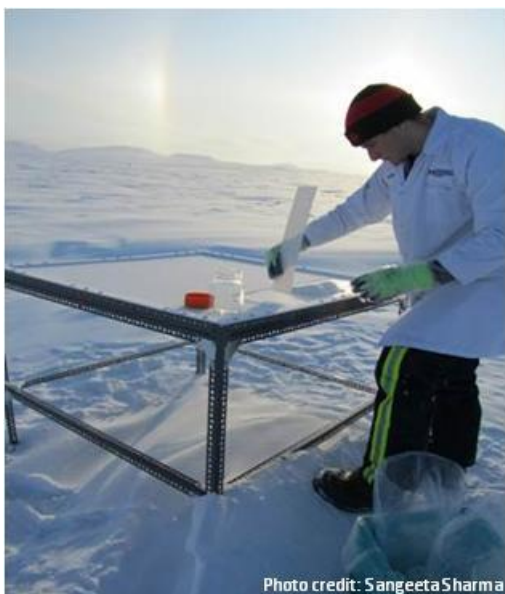


Figure S2: Alert snow table

### S4.2 Meteorological Conditions

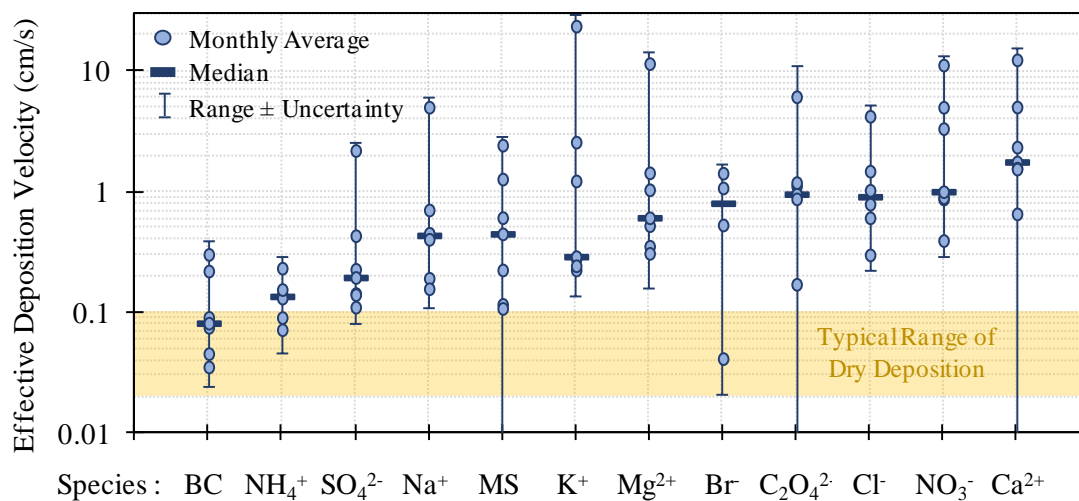
Ground-level meteorological data was collected at the Alert Base station, approximately 6 km NNE of the collection site and 50 m lower elevation (Climate IDs 2400306, 2400305, and 2400302) (retrieved Nov. 2015 from climate.weather.gc.ca). Hourly weather observations and daily snow depths at the meteorological stations showed notable disagreement with records of the collection station operators, particularly in terms of the time and extent of snow falls and other notable weather conditions such as fog and high winds. Monthly meteorological data are summarized for the campaign below (Table S9).

**Table S9: Overview of campaign meteorological conditions**

Year	Month	Temperature (°C)		Relative Humidity (%)		Wind Speed (km/h)		Pressure (kPa)		Total Precipitation (mm/month)
2014	September	-10	[-16, -5]	89	[66, 95]	9	[0, 36]	100.3	[99, 101]	16.8
	October	-17	[-29, -5]	85	[54, 92]	5	[0, 30]	101.4	[100, 103]	11.6
	November	-25	[-32, -9]	78	[56, 89]	6	[0, 54]	100.4	[98, 103]	7.4
	December	-30	[-39, -7]	74	[64, 93]	4	[0, 70]	100.6	[99, 102]	19.6
2015	January	-28	[-39, -10]	75	[52, 90]	7	[0, 67]	100.2	[98, 102]	12.4
	February	-30	[-41, -17]	72	[53, 84]	9	[0, 72]	100.1	[98, 102]	17.6
	March	-31	[-41, -22]	72	[62, 81]	5	[0, 45]	100.3	[98, 103]	25.4
	April	-21	[-35, -8]	74	[52, 87]	5	[0, 77]	100.6	[98, 104]	21.4
	May	-14	[-23, -6]	82	[54, 95]	6	[0, 34]	100.9	[99, 103]	6.4
Data Coverage		99%		99%		99%		99%		98%

**Notes:** Data are reported as: Median [Minimum, Maximum].  
Data include only the portion of each month covered by snow collection campaign.

The meteorological station snow depths differ from those measured at the snow table. While this might be indicative of undercatch by the snow table, it was also observed that meteorological conditions did differ significantly between the snow table and meteorological station 6 km away and 50 m lower in elevation. Records of weather at the two sites showed many discrepancies over the campaign in terms of both intensity and type of weather activity. Local observations have found high spatial variability in weather patterns to be common over previous seasons. Thus, the difference in snow depth at the two locations cannot be explained with certainty as an indication of collection difficulties. However, to provide greater confidence in the analysis, the calculation of effective deposition velocities was repeated using the meteorological station monthly snow depths. Figure S3 below shows a revised version of Figure 3 in the manuscript. While the absolute magnitudes of the deposition velocities do change with the use of the meteorological station depths, the general interpretation of the data does not change significantly.



Median: 0.08 0.13 0.19 0.43 0.44 0.28 0.60 0.79 0.94 0.89 0.98 1.73

**Figure S3: Effective deposition velocities using meteorological snow depth.**

Profile measurements from radiosondes, launched twice daily in 2015 and four times daily in 2014 from the Alert airport, were also obtained. The radiosonde data were used to estimate mixing heights and cloud heights over the campaign. Mixing height was taken as the lowest altitude, below 3 km, where the temperature gradient with altitude changed sign from negative to positive. This corresponds to a sharp increase in the potential temperature gradient, which typically coincides with a peak in the

relative humidity profile (e.g., sample provided in Figure S4). If no such change in gradient was found below 3 km altitude then no mixing height was taken to exist. A more conventional definition of mixing height, the lowest altitude above ground where the potential temperature matched that at ground-level, was judged unsuitable for our analysis as such a layer was found in only a small minority of profiles. Specifically, the latter was found in 22% September, October, and May (S/O/My) profiles and only 6% of November to April (N/D/Mr/A) profiles, while the former was found in 52% of S/O/My profiles and 30% of N/D/Mr/A profiles. A distinct change at this gradient-defined mixing height in the relative humidity profile and (where ozonesonde data was available) the ozone profile, suggests that this is a practical definition of mixing height. Colder months typically showed a much stronger gradient of potential temperature in the lowest kilometre above the surface (e.g., Figure S5), which would tend to suppress mixing.

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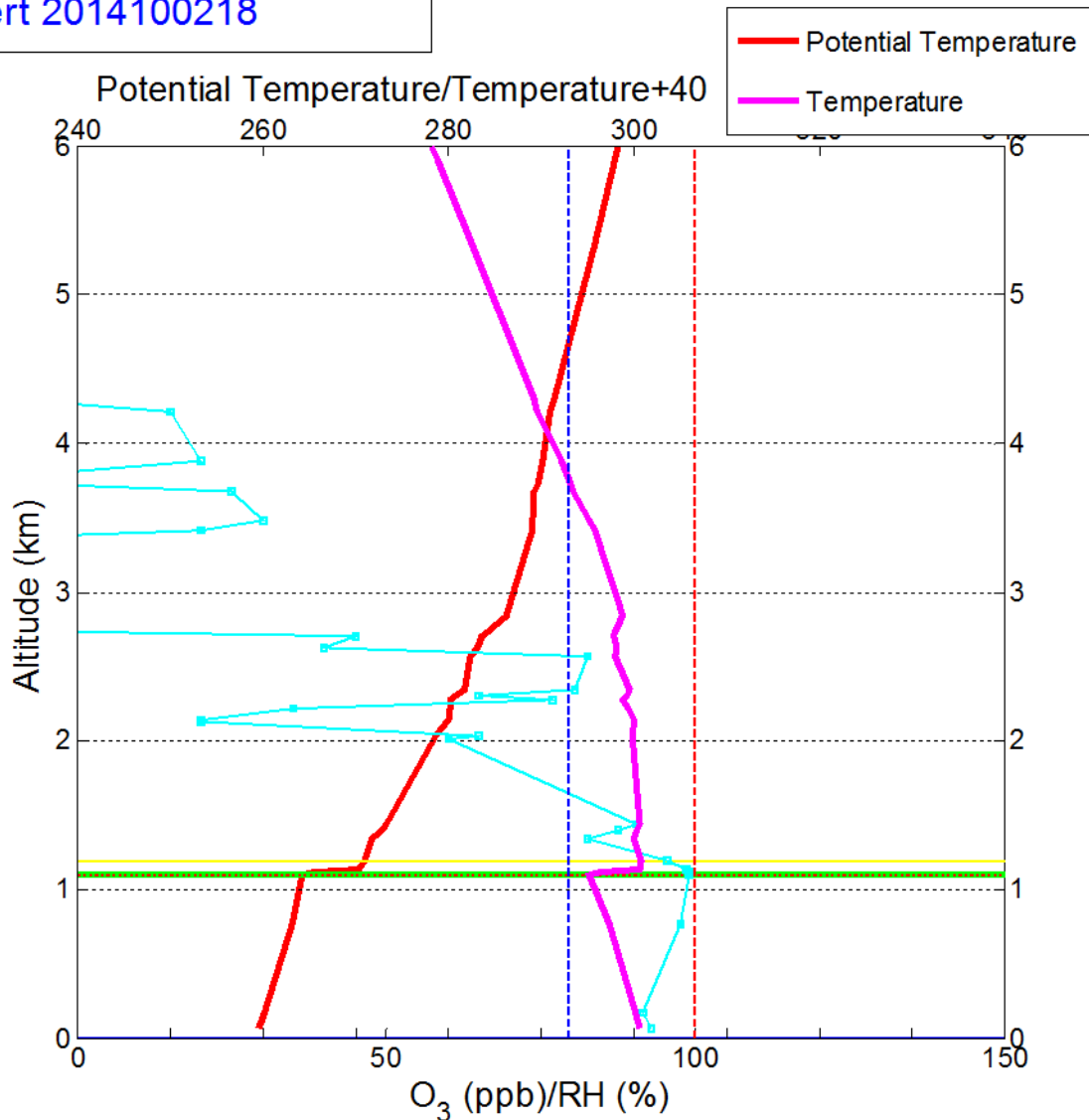


Figure S4: Meteorological profiles of temperature (magenta), potential temperature (red), and relative humidity (cyan) for October 2, 2014 at 18UT from Alert airport. The mixing height is indicated as a green horizontal line, the cloud height as a dotted red line, and temperature inversion as the yellow line. The vertical dashed lines indicate 100% relative humidity (red) and -20 C (blue). Temperatures within the cloud layer are warm enough for mixed-phase clouds to exist.

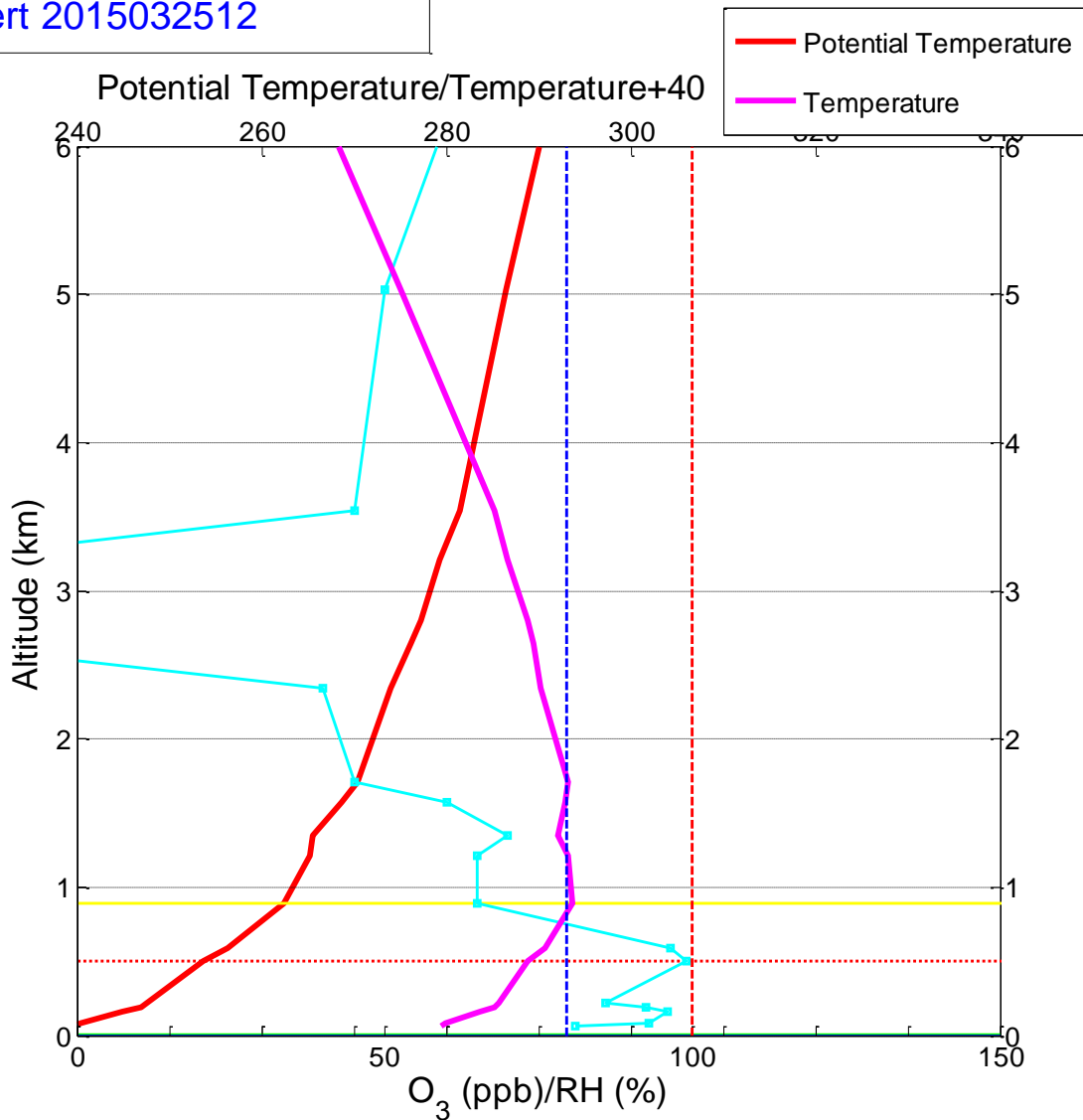
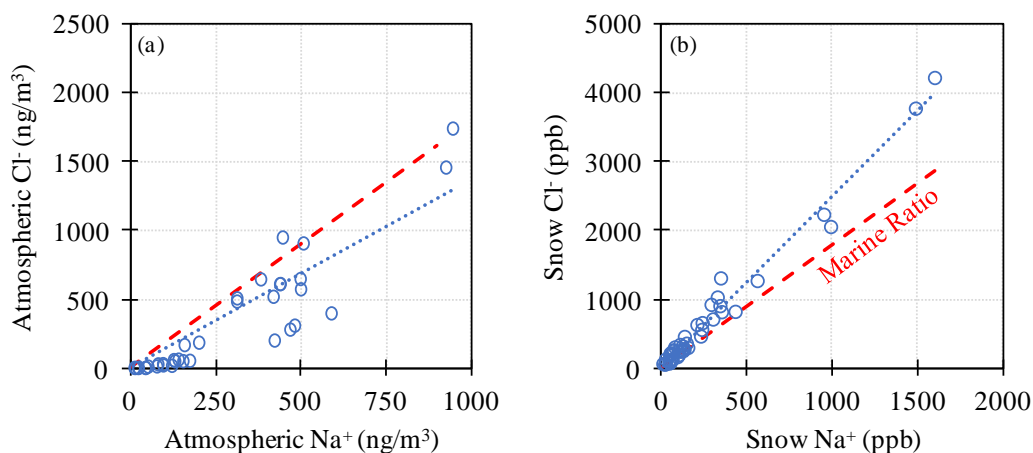


Figure S5: Meteorological profiles of temperature, potential temperature, and relative humidity for March 25, 2015 at 12UT from Alert airport. No mixing height is found, and the increase of potential temperature with height is large below the temperature inversion (indicated as a horizontal yellow line). The cloud height is indicated as a dotted horizontal red line. The vertical dashed lines indicate 100% relative humidity (red) and -20 C (blue). Temperatures within the cloud layer are likely too cold for mixed-phase clouds to exist.

Cloud height was estimated as the lowest altitude, below 3 km, where the relative humidity reached 100%. To allow for up to 5% error in the humidity sensor, this criterion was relaxed for profiles that did not show 100% relative humidity. In this case the peak of relative humidity, as long as it exceeded 95%, was taken. Different values of this criterion, up to and including 100% (no error) made no substantial difference to the monthly averaged results.

## S5 Arctic Sea Salt Aerosol Chloride Deficit

Due to the nature of the Hi-Vol analysis, the measured atmospheric concentrations include primarily the particle-phase ambient species; however, snow can scavenge both the particle and gas phases, and thus snow samples provide a composite measurement of both. The measurement of sea salt species was used as a case study to illustrate the impact of particle/gas-phase partitioning. The two dominant components of sea salt, Na and Cl, were measured both in snow samples (IC) and in the atmosphere (Hi-Vol). Previous studies have found Na and Cl observed within the Arctic to be predominantly marine in origin (Sirois and Barrie, 1999; Nguyen et al., 2013). This marine origin is supported by the excellent correlation of Na and Cl ions measured in snow (Pearson's correlation of 0.99), indicating a common source. The ratio of Na and Cl in sea salt is well established and consistent globally (Pytkowicz and Kester, 1971). So, the measured relationships of these chemical species in the atmosphere and snow were compared to the expected marine ratio (Figure S6).



**Figure S6: Atmospheric (a) and snow (b) sea salt measurements. The marine ratio of Cl<sup>-</sup>/Na<sup>+</sup>, shown in red (1.795 mass/mass), is compared to the measured average ratios, shown in blue as the line of best fit, for atmospheric (slope=1.36, correlation=0.92) and snow measurements (slope=2.49, correlation=0.99).**

It was observed that while atmospheric particulate monitoring showed a deficit in Cl, snow samples showed an enhancement in Cl relative its marine ratio with Na. This discrepancy may be explained by partitioning of Cl from the particle to the gas phase, and subsequent gas-phase scavenging by snow. It has been suggested that Cl within Arctic aerosol will partition to the gas phase during the winter, likely in the form of HCl via acid displacement reactions; however, there is no similar evidence for Na being involved in analogous chemistry (Barrie and Hoff, 1985; Toom-Sauntry and Barrie, 2002; Quinn et al., 2009). The gas-phase portion of Cl would not be captured by atmospheric particulate monitoring, causing a perceived deficit and reducing the correlation of Na and Cl if the gas-phase partitioning were non-linear. In contrast, gas-phase Cl deposited to the snowpack would be measured during snow analysis, leading to an increased Cl/Na ratio and an improved agreement between Cl and Na. The proportion of Cl in the atmosphere which partitioned to the gas phase was estimated from the observed atmospheric deficit below the marine ratio to be approximately 24% by mass on average over the 2014-15 winter season (assuming a common marine origin of Na and Cl and that Na remains fully in the particle phase). In addition, the enhancement of Cl in snow above the marine ratio demonstrates that gas-phase Cl was preferentially deposited to the snow over the particle phase. Snow Cl scavenged from the particle phase was estimated from snow Na using the observed atmospheric Cl/Na ratio (1.36 mass/mass) and the remaining Cl in snow was considered to be scavenged from the gas phase. Based on these estimated gas and particle-phase proportions in the snow and atmosphere, median effective deposition velocities were calculated as 0.16 and 0.40 cm/s for the particle and gas phases, respectively. Comparison of these velocities indicates an 86% enhancement in gas-phase deposition relative to that of the particle phase.

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