Supplementary Material for:

Shelley et al.

Regional trends in the fractional solubility of Fe and other metals from North Atlantic aerosols (GEOTRACES cruises GA01 and GA03) following a two-stage leach

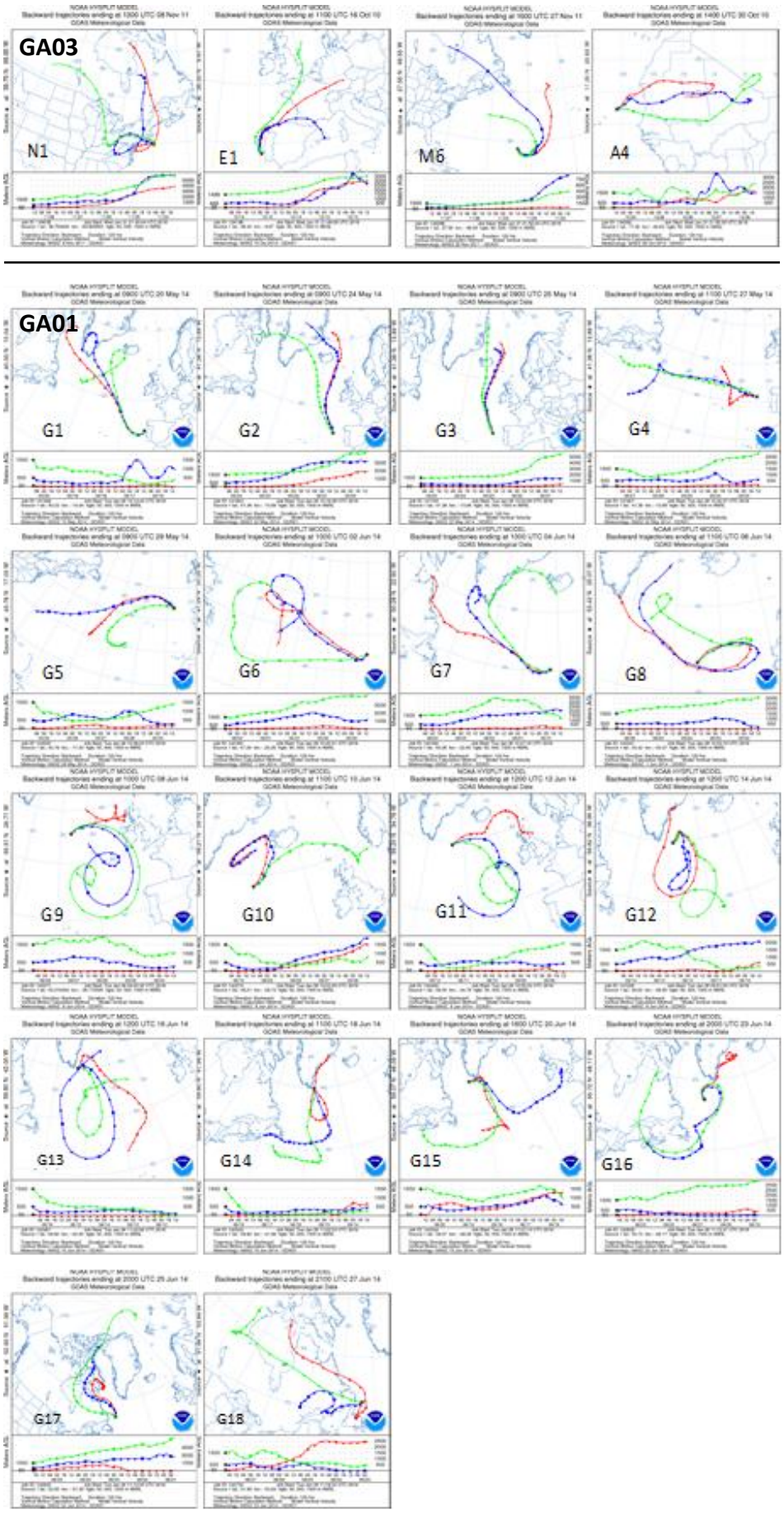
GA03
N1
E1
M6
A4
GA01
G1
G2
G3
G4
G5
G6
G7
G8
G9
G10
G11
G12
G13
G14
G15
G16
G17
G18

Figure S1

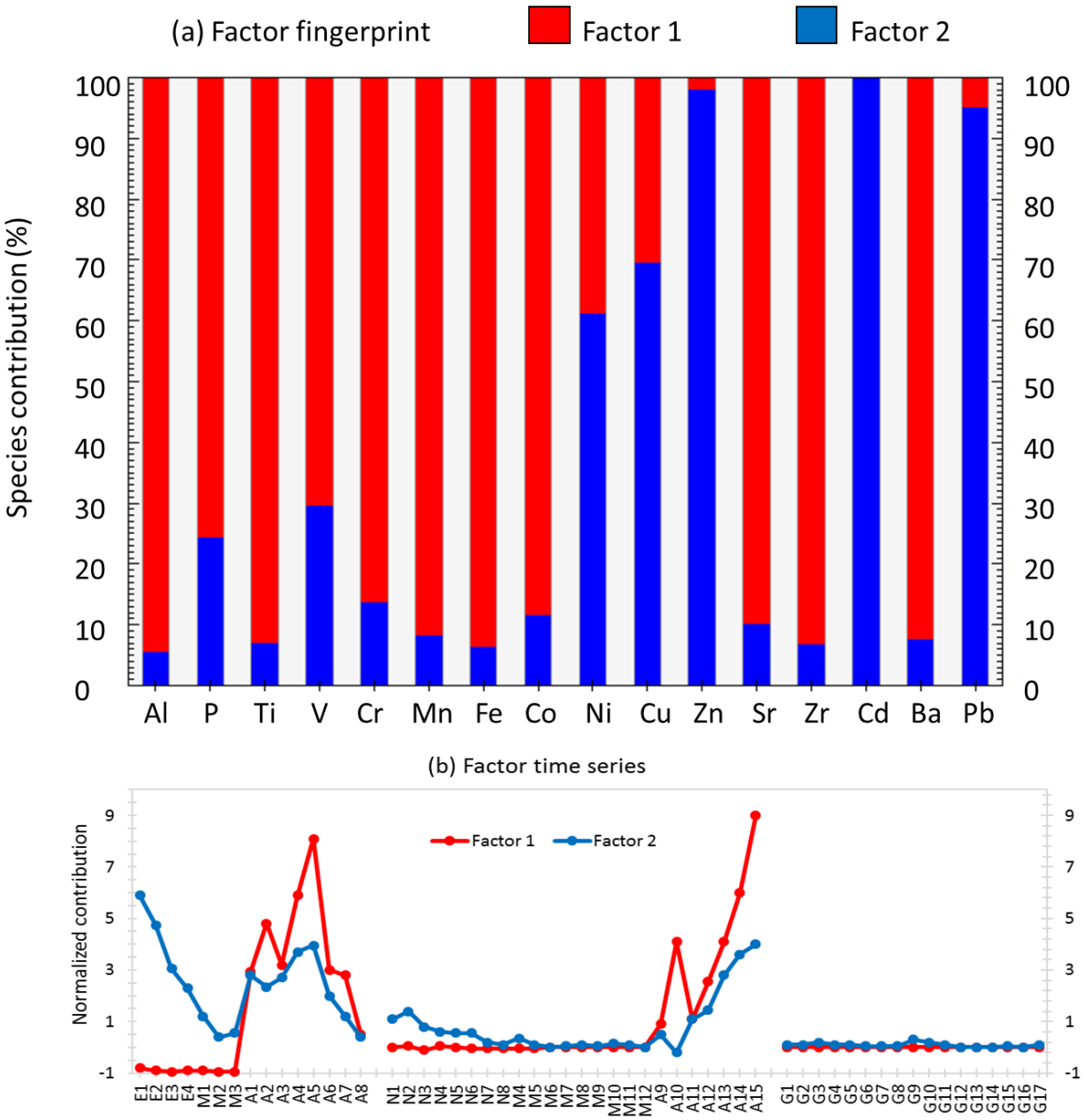
(a) Factor fingerprint
Factor 1
Factor 2
Species contribution (%)
100
90
80
70
60
50
40
30
20
10
0
Al P Ti V Cr Mn Fe Co Ni Cu Zn Sr Zr Cd Ba Pb
(b) Factor time series
Normalized contribution
Factor 1
Factor 2
9
7
5
3
1
-1
E1 E2 E3 E4 M1 M2 M3 A1 A2 A3 A4 A5 A6 A7 A8
N1 N2 N3 N4 N5 N6 N7 N8 M4 M5 M6 M7 M8 M9 M10 M11 M12 A9 A10 A11 A12 A13 A14 A15
G1 G2 G3 G4 G5 G6 G7 G8 G9 G10 G11 G12 G13 G14 G15 G16 G17

Figure S2.

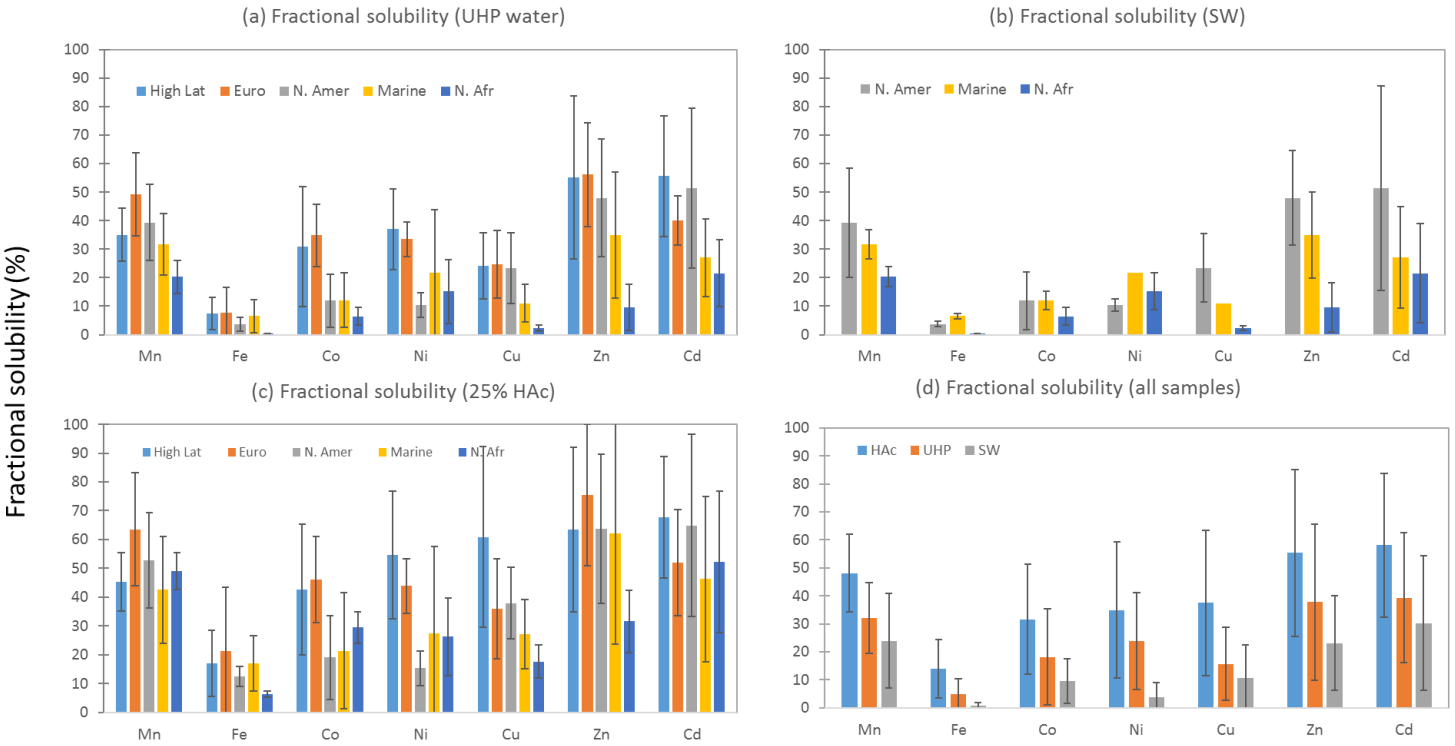
(a) Fractional solubility (UHP water)
(b) Fractional solubility (SW)
(c) Fractional solubility (25% HAc)
(d) Fractional solubility (all samples)
Fractional solubility (%)
High Lat
Euro
N. Amer
Marine
N. Afr
HAc
UHP
SW
Mn
Fe
Co
Ni
Cu
Zn
Cd

Figure S3

Table S1

(a) UHP water leach				Na	Al	Ti	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb	Cl^-	NO_3^-	SO_4^{2-}
UHP water matrix blank	ng/m3	(n=9)	**Legend**	0.035	4.6E-04	1.1E-04	4.8E-05	2.8E-04	2.3E-05	3.2E-05	8.8E-05	8.0E-05	4.6E-06	7.31E-06	7.0E-06	1.3E-05	3.4E-06
DL (3xSD)			ND = not determined	0.006	3.4E-04	5.7E-05	7.7E-06	7.4E-05	2.2E-05	7.3E-06	3.2E-05	1.2E-04	2.3E-06	3.76E-06	9.1E-06	3.3E-06	2.7E-21
			<DL = below detection														
Filter Blank	ng/m3	(n=3)	NC = not certified	0.065	5.7E-04	5.0E-04	5.1E-05	2.7E-04	7.5E-06	4.6E-05	1.7E-04	2.4E-04	5.5E-06	4.5E-05	2.8E-04	2.2E-05	1.6E-06
SD				0.006	4.6E-05	1.0E-04	1.5E-06	1.4E-05	1.5E-06	7.6E-06	1.4E-06	1.1E-04	1.4E-06	1.7E-05	5.6E-05	1.1E-05	2.8E-06
SRM	% recovery			97%	109%	NC	99%	103%	99%	100%	104%	91%	95%	100%	105%	105%	99%
Trace elements = NIST 1643e; anions = Hach SRM	SD	(n=3)		1%	1%	NC	6%	4%	6%	1%	3%	3%	1%	6%	3%	3%	5%

Sample #	Leach medium	Air mass regime (5 day AMBT)	Start date	Start lat (N)	Start Long (W)	End date	End lat (N)	End long (W)	Na ng/m3	Al ng/m3	Ti ng/m3	Mn ng/m3	Fe ng/m3	Co ng/m3	Ni ng/m3	Cu ng/m3	Zn ng/m3	Cd ng/m3	Pb ng/m3	Cl^- µg/L	NO_3^- µg/L	SO_4^{2-} µg/L
5037 (E1)	UHP water	Euro	15-Oct-10	38.32	9.66	16-Oct-10	38.33	9.67	1819	3.12	0.165	1.25	2.81	0.0273	0.865	0.724	6.64	0.0637	1.23	4649	251	697
5145 (E2)	UHP water	Euro	16-Oct-10	38.33	9.66	17-Oct-10	38.33	9.66	861	3.90	0.159	1.35	4.02	0.0365	0.854	1.15	8.55	0.0668	1.17	1189	151	290
5197 (E3)	UHP water	Euro	17-Oct-10	38.33	9.66	19-Oct-10	35.20	16.00	903	3.82	0.141	0.867	2.98	0.0220	0.673	0.623	4.11	0.0330	0.787	7835	996	1724
5299 (E4)	UHP water	Euro	19-Oct-10	35.20	16.00	20-Oct-10	34.09	17.57	744 (37)	1.89 (0.07)	0.0126 (0.0033)	0.368 (0.054)	1.43 (0.04)	0.0118 (0.0015)	0.483 (0.069)	0.280 (0.021)	1.82 (0.23)	0.0171 (0.0031)	0.485 (0.043)	530	1686	971
5330 (M1)	UHP water	Marine	20-Oct-10	34.07	17.61	21-Oct-10	31.32	21.55	1006	1.20	0.0644	0.102	0.325	0.0181	0.312	0.107	1.70	0.0390	0.279	2998	350	604
5425 (M2)	UHP water	Marine	21-Oct-10	31.11	21.86	23-Oct-10	27.53	22.01	2595	0.00651	0.0543	0.0151	0.0848	0.00092	0.0523	0.0264	0.293	0.00339	0.0214	6873	876	1416
5579 (M3)	UHP water	Marine	23-Oct-10	27.53	22.01	25-Oct-10	24.01	22.00	4013 (1147)	0.157	0.0281 (0.0251)	0.0847 (0.0260)	0.227 (0.070)	0.0061 (0.0018)	0.288 (0.074)	0.0751 (0.0204)	0.671 (0.180)	0.0055 (0.0002)	0.142 (0.0235)	853	244	254
5729 (A1)	UHP water	N. Afr.	25-Oct-10	24.01	22.00	28-Oct-10	17.35	18.25	1540	22.9	0.403	5.73	11.0	0.0504	0.574	0.0601	0.595	0.00927	0.0664	1449	3202	1795
5738 (A2)	UHP water	N. Afr.	28-Oct-10	17.36	18.25	29-Oct-10	17.36	18.25	1050	20.0	0.327	6.79	9.44	0.0535	0.582	0.0235	0.524	0.00684	0.0508	1080	4472	2697
5764 (A3)	UHP water	N. Afr.	29-Oct-10	17.36	18.25	29-Oct-10	17.36	19.06	1440	9.28	0.320	6.12	5.13	0.0486	0.621	0.0269	0.606	0.00876	0.0501	3100	15008	6423
5848 (A4)	UHP water	N. Afr.	30-Oct-10	17.35	20.02	30-Oct-10	17.35	20.83	1430	24.1	0.429	12.7	10.1	0.118	0.874	0.0562	0.947	0.0175	0.0615	3869	1989	2506
5870 (A5)	UHP water	N. Afr.	31-Oct-10	17.36	20.83	31-Oct-10	17.36	20.86	3170	23.7	0.470	15.9	9.82	0.117	1.31	0.0511	0.501	0.0168	0.0366	6609	2051	2877
5965 (A6)	UHP water	N. Afr.	01-Nov-10	17.35	20.91	01-Nov-10	17.35	22.79	6090	24.1	0.544	7.11	10.3	0.0763	0.781	0.0623	0.567	0.0131	0.0876	1790	1797	1388
5999 (A7)	UHP water	N. Afr.	01-Nov-10	17.35	22.79	01-Nov-10	17.35	23.07	8740	4.53	0.243	3.61	1.48	0.0392	0.305	0.0354	0.638	0.0111	0.102	30297	4890	7418
6070 (A8)	UHP water	N. Afr.	03-Nov-10	17.40	24.50	03-Nov-10	17.34	24.55	5105 (346)	3.46 (0.39)	0.133 (0.032)	1.29 (0.17)	1.13 (0.29)	0.0157 (0.0018)	0.153 (0.0011)	0.0041 (0.0013)	0.212 (0.010)	0.0036 (0.0009)	0.0127 (0.0060)	3232	2272	1798
6250 (N1)	UHP water	N. Amer.	07-Nov-11	39.71	69.87	08-Nov-11	39.70	69.80	1330 (198)	0.848 (0.153)	0.102 (0.012)	0.243 (0.020)	0.824 (0.160)	0.0033 (0.0000)	0.106 (0.0076)	0.142 (0.013)	0.984 (0.131)	0.0110 (0.0038)	0.206 (0.027)	2072 (336)	1059 (96)	943 (91)
6319 (N2)	UHP water	N. Amer.	08-Nov-11	39.70	69.80	09-Nov-11	39.35	69.54	733	2.44	0.139	0.911	2.19	0.0136	0.187	0.663	3.00	0.0270	0.531	527	3385	1303
6452 (N3)	UHP water	N. Amer.	09-Nov-11	39.35	69.54	11-Nov-11	38.65	69.20	1220	0.518	0.0489	0.0722	0.306	0.00455	0.0919	0.0675	0.703	0.0522	0.0600	1156	325	384
6546 (N4)	UHP water	N. Amer.	12-Nov-11	38.33	68.87	13-Nov-11	38.36	68.86	2000	0.914	0.118	0.495	0.869	0.00368	0.0534	0.114	1.05	0.0285	0.201	3881	1126	1213

6677 (N5)	UHP water	N. Amer.	13-Nov-11	38.36	68.86	15-Nov-11	37.58	68.40	3307 (323)	1.45 (0.220	0.132 (0.020)	0.241 (0.024)	0.988 (0.195)	0.0025 (0.0002)	0.0517 (0.0025)	0.0527 (0.0087)	0.543 (0.052)	0.0148 (0.0055)	0.144 (0.025)	7228 (23)	686 (23)	1491 (256)
6788 (N6)	UHP water	N. Amer.	15-Nov-11	37.62	68.38	17-Nov-11	37.62	68.38	11400	0.663	0.0505	0.160	0.253	0.00463	0.0700	0.0464	0.980	0.107	0.0818	24640	338	3187
6903 (N7)	UHP water	Marine	19-Nov-11	32.02	64.44	20-Nov-11	31.75	64.17	3250	0.841	0.00337	0.0501	0.277	0.00035	0.0175	0.0178	0.490	0.00497	0.0445	6486	251	980
6964 (N8)	UHP water	Marine	20-Nov-11	31.75	64.17	21-Nov-11	31.55	63.43	1930	0.148	0.0472	0.0318	0.0604	0.00043	0.0113	0.0251	0.147	0.00228	0.0220	3723	147	563
7156 (M4)	UHP water	Marine	21-Nov-11	31.53	63.37	24-Nov-11	29.70	56.82	1018 (74)	0.291 (0.162)	0.0356 (0.0019)	0.0401 (0.0012)	0.164 (0.022)	0.0010 (0.0002)	0.0325 (0.0001)	0.0085 (0.0012)	0.172 (0.064)	0.0038	0.0349 (0.0020)	1728 (120)	211 (32)	426 (26)
7245 (M5)	UHP water	Marine	24-Nov-11	29.70	56.82	26-Nov-11	27.58	49.63	1000	0.242	0.00527	0.0149	0.167	0.00079	0.0274	0.00875	0.0897	0.00332	0.0123	1701	150	466
7316 (M6)	UHP water	Marine	26-Nov-11	27.58	49.63	27-Nov-11	27.55	49.55	980 (49)	0.0829 (0.0124)	0.0366 (0.0049)	0.0112 (0.0017)	0.0395 (0.0123)	0.0010 (0.0001)	0.0521 (0.0032)	0.0090 (0.0029)	0.0816 (0.0113)	0.0023 (0.0006)	0.0041 (0.0014)	1886 (34)	165 (4)	457 (63)
7357 (M7)	UHP water	Marine	27-Nov-11	27.55	49.55	28-Nov-11	26.31	45.39	952	0.355	0.00174	0.0236	0.309	0.00778	0.0474	0.00461	0.0663	0.00419	0.0302			
7426 (M8)	UHP water	Marine	28-Nov-11	26.28	45.30	29-Nov-11	26.14	44.83	701	0.247	0.00439	0.0161	0.181	0.00064	0.0236	0.00907	0.102	0.00227	0.0111	1225	188	380
7509 (M9)	UHP water	Marine	29-Nov-11	26.14	44.83	30-Nov-11	25.58	43.60	1205 (49)	1.15 (0.42)	0.0331 (0.0047)	0.0946 (0.0034)	0.355 (0.078)	0.0023 (0.0000)	0.0324 (0.0048)	0.0087 (0.0021)	0.122 (0.040)	0.0044 (0.0021)	0.0115 (0.0000)	2336 (76)	202 (19)	452 (17)
7587 (M10)	UHP water	Marine	30-Nov-11	25.55	43.54	01-Dec-11	24.15	40.22	1800	1.05	0.0439	0.0814	0.323	0.00250	0.0472	0.00933	0.124	0.00400	0.0183	3471	182	620
7655 (M11)	UHP water	Marine	01-Dec-11	24.15	40.22	02-Dec-11	24.15	40.22	2150	0.491	0.0436	0.0271	0.179	0.00111	0.0304	0.00619	0.101	0.00466	0.0132	4640	200	711
7697 (M12)	UHP water	Marine	02-Dec-11	24.15	40.22	03-Dec-11	22.70	36.73	1435	0.531 (0.102)	0.0421 (0.0108)	0.0167 (0.0094)	0.101 (0.040)	0.0009 (0.0006)	0.0282 (0.0115)	0.0182 (0.0084)	0.245 (0.126)	0.0114 (0.0037)	0.0070 (0.0013)	2789 (853)	168 (72)	475 (142)
7791 (A9)	UHP water	N. Afr.	03-Dec-11	22.70	36.73	04-Dec-11	22.36	35.87	1510	3.73	0.100	1.53	1.86	0.0125	0.0890	0.0180	0.191	0.0127	0.0122	2954	523	996
7860 (A10)	UHP water	N. Afr.	04-Dec-11	22.36	35.87	05-Dec-11	22.38	35.87	3430	13.0	0.278	9.03	7.00	0.0496	0.106	0.0463	0.190	0.0108	0.0126	6931	1196	1982
7899 (A11)	UHP water	N. Afr.	05-Dec-11	22.37	35.62	06-Dec-11	20.88	32.62	2410 (113)	7.36 (0.50)	0.148 (0.040)	3.88 (0.20)	2.57 (0.69)	0.0367 (0.0022)	0.109 (0.000)	0.0331 (0.0036)	0.556 (0.026)	0.00585 (0.0004)	0.0523 (0.0002)	4734 (32)	914 (14)	1207 (10)
7946 (A12)	UHP water	N. Afr.	06-Dec-11	20.88	32.62	07-Dec-11	19.43	29.38	2310	9.87	0.243	3.56	4.94	0.0212	0.0825	0.0260	0.125	0.00591	0.0137	4642	801	1969
8004 (A13)	UHP water	N. Afr.	07-Dec-11	19.43	29.38	08-Dec-11	19.43	29.38	2350	18.6	0.371	5.36	10.6	0.0283	0.0746	0.0415	0.0844	0.00661	0.0122	4823	932	2065
8044 (A14)	UHP water	N. Afr.	08-Dec-11	19.43	29.38	09-Dec-11	18.13	26.13	1700	15.6	0.436	10.6	8.21	0.0532	0.170	0.0540	0.122	0.00693	0.0125	3246	1367	2207
8045 (A15)	UHP water	N. Afr.	09-Dec-11	17.75	25.31	09-Dec-11	17.40	24.53	1430	40.2	0.917	14.2	24.4	0.0883	0.143	0.0904	0.260	0.00405	0.0316	2664	1237	2003
geoa1 (G1)	UHP water	High Lat	19-May-14	40.33	10.04	20-May-14	40.33	10.04	ND	0.0959 (0.0795)	0.000692	0.0345 (0.0044)	0.146 (0.133)	0.00099 (0.00021)	0.0470 (0.0049)	0.0050 (0.0007)	0.138 (0.103)	0.00108 (0.00009)	0.00630 (0.00045)	22380 (454)	708 (252)	4730 (363)
geoa2 (G2)	UHP water	High Lat	23-May-14	40.33	12.22	24-May-14	41.38	13.89	ND	0.558	0.00430	0.100	0.430	0.00122	0.0197	0.0121	0.386	0.00355	0.0394	32834	1821	8803
geoa3 (G3)	UHP water	High Lat	24-May-14	41.38	13.89	25-May-14	41.38	13.89	ND	1.02	0.00667	0.143	0.801	0.00203	0.0580	0.0175	0.413	0.00530	0.0649	28138	4687	11913
geoa4 (G4)	UHP water	High Lat	25-May-14	41.38	13.89	27-May-14	41.38	13.89	ND	0.886	0.00321	0.124	0.663	0.00163	0.0557	0.0244	0.402	0.00331	0.0453	20151	3489	8485
geoa5 (G5)	UHP water	High Lat	27-May-14	41.38	13.89	29-May-14	43.78	17.03	ND	1.18	<DL	0.0819	0.483	0.00125	0.0327	0.0174	0.190	0.00150	0.0219	9714	1750	7571
geoa6 (G6)	UHP water	High Lat	30-May-14	45.05	18.51	02-Jun-14	47.29	20.26	ND	0.989 (0.340)	0.00022 (0.00025)	0.0588 (0.0011)	0.323 (0.025)	0.00154 (9.2E-05)	0.0945 (0.0053)	0.0174 (0.0021)	0.254 (0.115)	0.000933 (0.000110)	0.0145 (0.0023)	24218 (1850)	2764 (369)	10273 (623)
geoa7 (G7)	UHP water	High Lat	02-Jun-14	47.29	20.26	04-Jun-14	50.28	22.60	ND	0.405	0.00125	0.0530	0.210	0.00059	0.0177	0.0102	0.162	0.00158	0.0190	42421	827	8710
geoa8 (G8)	UHP water	High Lat	04-Jun-14	50.28	22.60	06-Jun-14	53.42	25.07	ND	0.897	0.000815	0.0651	0.425	0.00082	0.0304	0.0112	0.702	0.00117	0.0201	42177	1470	9816
geoa9 (G9)	UHP water	High Lat	06-Jun-14	53.42	25.07	08-Jun-14	55.51	26.71	ND	9.90	0.00273	0.0803	1.45	0.00308	0.0848	0.0377	1.373	0.00323	0.0660	66497	3550	22961
geoa10 (G10)	UHP water	High Lat	08-Jun-14	55.51	26.71	10-Jun-14	58.21	29.72	ND	1.50 (0.19)	0.00939 (0.00155)	0.0686 (0.0043)	1.04 (0.04)	0.00157 (6.33E-5)	0.0254 (0.0016)	0.0258 (0.0053)	0.295 (0.006)	0.00164 (0.00005)	0.0449 (0.0020)	5362 (143)	783 (70)	11314 (374)
geoa11 (G11)	UHP water	High Lat	10-Jun-14	58.21	29.72	12-Jun-14	59.20	34.78	ND	1.60	0.00157	0.0344	0.385	0.00064	0.0192	0.0192	0.711	0.00093	0.0218	47062	1096	12920
geoa12 (G12)	UHP water	High Lat	12-Jun-14	59.20	34.78	14-Jun-14	59.62	38.95	ND	0.146	<DL	0.00240	0.039	0.00013	0.00240	0.00524	0.068	0.000116	0.00292	20601	<DL	4613
geoa13 (G13)	UHP water	High Lat	14-Jun-14	59.62	38.95	16-Jun-14	59.80	42.00	ND	0.752	<DL	0.00733	0.145	0.00028	0.0122	0.00518	0.0216	0.000152	0.00696	14614	<DL	6564

geoa14 (G14)	UHP water	High Lat	16-Jun-14	59.80	42.00	18-Jun-14	59.80	41.99	ND	0.788 (0.442)	0.00036 (0.00011)	0.00960 (0.00350)	0.0710 (0.0280)	0.00047 (0.00023)	0.0186 (0.0192)	0.00363 (0.00211)	0.174 (0.115)	0.000191 (0.000105)	0.00418 (0.00075)	10671 (884)	<DL	2896 (77)
geoa15 (G15)	UHP water	High Lat	18-Jun-14	59.70	42.53	20-Jun-14	59.07	46.09	ND	0.256	0.00112	0.00669	0.131	0.00028	0.0198	0.00947	0.148	0.00065	0.0180	24743	127	5894
geoa16 (G16)	UHP water	High Lat	21-Jun-14	56.91	47.42	23-Jun-14	55.72	48.17	ND	0.065	<DL	0.00158	0.012	0.00003	0.00247	0.00133	0.0247	0.000118	0.0021	27407	<DL	5021
geoa17 (G17)	UHP water	High Lat	23-Jun-14	55.72	48.17	25-Jun-14	52.93	51.39	ND	0.756	0.00140	0.130	0.321	0.00103	0.0156	0.0243	0.423	0.00479	0.0384	44813	1568	6883
geoa18 (G18)	UHP water	High Lat	25-Jun-14	52.93	51.39	27-Jun-14	51.99	53.84	ND	0.0232	0.000651	0.0213	0.0596	0.00021	0.000168	0.00530	0.108	0.000331	0.00806	15940	640	3888

(b) Seawater leach

			Na	Al	Ti	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
Seawater matrix blank	ng/m3	(n=3)	ND	0.0149	ND	0.00169	4.3E-04	8.9E-06	8.9E-04	2.3E-04	6.4E-06	5.3E-06	4.3E-05
DL (3xSD)			ND	0.0379	ND	0.00256	6.5E-04	1.6E-05	1.0E-04	9.2E-05	0.0E+00	3.5E-06	3.9E-05
Filter blank	ng/m3	(n=5)	ND	0.0080	ND	0.00149	2.7E-04	7.7E-06	8.9E-04	3.1E-04	1.1E-04	8.1E-06	4.0E-05
SD			ND	0.0021	ND	0.00022	7.6E-05	2.2E-06	3.2E-05	4.9E-05	8.5E-05	1.9E-06	8.1E-06
SAFe D2 (extraction 8 Oct 2013)	conc (nM)		ND	0.763	ND	0.342	0.944	0.0480	8.03	2.28	7.78	1.01	0.032
	SD		ND	0.028	ND	0.093	0.031	0.0030	0.22	0.22	0.18	0.01	0.003
	consensus value (May 2013)		NC	1.03	NC	0.350	0.933	0.0457	8.63	2.28	7.43	0.99	0.029
	SD		NC	0.09	NC	0.050	0.023	0.0029	0.25	0.15	0.25	0.02	0.001
	% recovery			74%		98%	101%	105%	93%	100%	105%	103%	111%
	SD			3%		27%	3%	7%	3%	10%	2%	1%	11%

Sample #	Leach medium	Air mass regime (5 day AMBT)	Start date	Start lat (N)	Start Long (W)	End date	End lat (N)	End long (W)	Na	Al ng/m3	Ti ng/m3	Mn ng/m3	Fe ng/m3	Co ng/m3	Ni ng/m3	Cu ng/m3	Zn ng/m3	Cd ng/m3	Pb ng/m3
6070 (A8)	Seawater	N. Afr.	03-Nov-10	17.40	24.50	03-Nov-10	17.34	24.55	ND	2.42	ND	0.718	0.344	0.0160	0.0944	0.0124	0.208	0.00346	0.0411
6250 (N1)	Seawater	N. Amer.	07-Nov-11	39.71	69.87	08-Nov-11	39.70	69.80	ND	0.931 (0.035)	ND	0.296 (0.032)	0.346 (0.010)	0.00443 (0.0002)	0.0236 (0.0081)	0.133 (0.003)	1.11 (0.06)	0.0284 (0.0007)	0.317 (0.002)
6319 (N2)	Seawater	N. Amer.	08-Nov-11	39.70	69.80	09-Nov-11	39.35	69.54	ND	2.52 (0.61)	ND	1.07 (0.30)	1.24 (0.15)	0.0158 (0.0019)	0.0622 (0.0177)	0.548 (0.075)	3.04 (0.25)	0.02565 (0.0034)	0.651 (0.048)
6452 (N3)	Seawater	N. Amer.	09-Nov-11	39.35	69.54	11-Nov-11	38.65	69.20	ND	0.714	ND	0.0835	0.152	0.00609	0.0115	0.0598	0.641	0.0338	0.104
6546 (N4)	Seawater	N. Amer.	12-Nov-11	38.33	68.87	13-Nov-11	38.36	68.86	ND	0.847	ND	0.480	0.325	0.00433	<DL	0.0878	0.871	0.0149	0.273
6677 (N5)	Seawater	N. Amer.	13-Nov-11	38.36	68.86	15-Nov-11	37.58	68.40	ND	0.812	ND	0.227	0.305	0.00277	0.00367	0.0448	0.534	0.108	0.195
6788 (N6)	Seawater	N. Amer.	15-Nov-11	37.62	68.38	17-Nov-11	37.62	68.38	ND	1.43	ND	<DL	0.132	0.00481	<DL	0.00285	0.665	0.280	0.111
6903 (N7)	Seawater	Marine	19-Nov-11	32.02	64.44	20-Nov-11	31.75	64.17	ND	0.0442	ND	<DL	0.0218 (0.004)	0.00063 (0.00008)	<DL	0.0120 (0.0018)	0.183 (0.028)	0.00683 (0.0008)	0.0698 (0.0112)
6964 (N8)	Seawater	Marine	20-Nov-11	31.75	64.17	21-Nov-11	31.55	63.43	ND	1.71	ND	<DL	0.0621	0.00197	<DL	0.0177	0.0823	0.00179	0.0322
7156 (M4)	Seawater	Marine	21-Nov-11	31.53	63.37	24-Nov-11	29.70	56.82	ND	<DL	ND	<DL	0.0310	0.00085	<DL	<DL	0.161	0.00948	0.0827
7245 (M5)	Seawater	Marine	24-Nov-11	29.70	56.82	26-Nov-11	27.58	49.63	ND	<DL	ND	<DL	0.0238	0.0002209	<DL	<DL	0.0649	0.00563	0.0173
7316 (M6)	Seawater	Marine	26-Nov-11	27.58	49.63	27-Nov-11	27.55	49.55	ND	<DL	ND	<DL	0.00076	0.0004346	<DL	<DL	0.0715	0.00195	0.0101
7357 (M7)	Seawater	Marine	27-Nov-11	27.55	49.55	28-Nov-11	26.31	45.39	ND	0.176	ND	<DL	0.0485 (0.096)	0.00050 (0.00008)	<DL	<DL	0.0661 (0.006)	0.00377 (0.0022)	0.0285 (0.0028)
7426 (M8)	Seawater	Marine	28-Nov-11	26.28	45.30	29-Nov-11	26.14	44.83	ND	<DL	ND	<DL	0.0119	0.0002492	<DL	<DL	0.0644	0.00098	0.0174

7509 (M9)	Seawater	Marine	29-Nov-11	26.14	44.83	30-Nov-11	25.58	43.60	ND	0.196 (0.035)	ND	0.0206 (0.0214)	0.0562 (0.017)	0.00234 (0.00035)	<DL	<DL	0.0813 (0.014)	0.00470	0.0227 (0.0065)
7587 (M10)	Seawater	Marine	30-Nov-11	25.55	43.54	01-Dec-11	24.15	40.22	ND	0.166	ND	0.00287	0.0451	0.00209	<DL	<DL	0.100	0.000796	0.0350
7655 (M11)	Seawater	Marine	01-Dec-11	24.15	40.22	02-Dec-11	24.15	40.22	ND	0.477	ND	<DL	0.0124 (0.002)	0.00077 (0.00003)	<DL	<DL	0.104 (0.007)	0.0026 (0.0006)	0.0235 (0.0029)
7697 (M12)	Seawater	Marine	02-Dec-11	24.15	40.22	03-Dec-11	22.70	36.73	ND	<DL	ND	<DL	<DL	0.0001524	<DL	<DL	0.0992	0.000857	0.00990
7791 (A9)	Seawater	N. Afr.	03-Dec-11	22.70	36.73	04-Dec-11	22.36	35.87	ND	1.94 (0.03)	ND	1.45	0.259 (0.014)	0.0183 (0.0004)	0.0516 (0.0010)	0.00243 (0.00202)	0.260 (0.009)	0.00863 (5.91e-05)	0.0663 (0.001)
7860 (A10)	Seawater	N. Afr.	04-Dec-11	22.36	35.87	05-Dec-11	22.38	35.87	ND	3.58	ND	7.85	0.372	0.0739	0.0232	0.0305	0.269	0.0156	0.0472
7899 (A11)	Seawater	N. Afr.	05-Dec-11	22.37	35.62	06-Dec-11	20.88	32.62	ND	6.03	ND	2.92	0.786	0.0403	0.0381	0.0137	0.479	0.00546	0.112
7946 (A12)	Seawater	N. Afr.	06-Dec-11	20.88	32.62	07-Dec-11	19.43	29.38	ND	3.74 (0.22)	ND	3.28 (0.172)	0.336 (0.013)	0.0357 (0.0005)	0.0275 (0.0047)	0.0211 (0.0052)	0.243 (0.033)	0.0093 (7.79e-05)	0.0438 (0.0048)
8004 (A13)	Seawater	N. Afr.	07-Dec-11	19.43	29.38	08-Dec-11	19.43	29.38	ND	6.73	ND	7.33	0.648	0.0815	0.0535	0.0449	0.377	0.0153	0.0440
8044 (A14)	Seawater	N. Afr.	08-Dec-11	19.43	29.38	09-Dec-11	18.13	26.13	ND	7.32	ND	12.8	0.828	0.136	0.160	0.0714	0.227	0.0187	0.0398
8045 (A15)	Seawater	N. Afr.	09-Dec-11	17.75	25.31	09-Dec-11	17.40	24.53	ND	5.23	ND	18.2	0.392	0.206	<DL	<DL	0.174	0.0134	0.0240

(c) 25% acetic acid leach

			Na	Al	Ti	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
25% HAc matrix blank	ng/m3	(n=3)	ND	0.00210	0.002575	9.58E-05	0.00052	2.865E-05	1.9E-04	8.1E-04	5.3E-04	1.53E-05	6.13E-05
DL (3xSD)			ND	0.00027	0.00020	9.31E-05	0.000118	2.927E-06	9.2E-05	1.5E-04	2.4E-04	5.68E-06	3.34E-05
Filter blank	ng/m3	(n=3)	ND	0.0066	0.000575	0.000135	0.00239	3.178E-05	2.4E-04	8.1E-04	2.2E-03	1.46E-05	4.84E-05
SD			ND	0.002	6.4E-05	9.55E-06	0.00017	4.603E-08	6.5E-06	1.3E-05	6.8E-04	3.46E-06	1.18E-05

Sample #	**Leach medium**	**Air mass regime (5 day AMBT)**	**Start date**	**Start lat (N)**	**Start Long (W)**	**End date**	**End lat (N)**	**End long (W)**	**Na**	**Al** ng/m3	**Ti** ng/m3	**Mn** ng/m3	**Fe** ng/m3	**Co** ng/m3	**Ni** ng/m3	**Cu** ng/m3	**Zn** ng/m3	**Cd** ng/m3	**Pb** ng/m3
5037 (E1)	25% HAc	Euro	15-Oct-10	38.32	9.66	16-Oct-10	38.33	9.67	ND	6.52	0.24	1.52	8.63	0.035	1.07	0.98	8.09	0.073	2.02
5145 (E2)	25% HAc	Euro	16-Oct-10	38.33	9.66	17-Oct-10	38.33	9.66	ND	9.04	0.17	1.85	14.2	0.051	1.20	1.66	11.6	0.101	2.47
5197 (E3)	25% HAc	Euro	17-Oct-10	38.33	9.66	19-Oct-10	35.20	16.00	ND	7.38	0.14	1.09	7.48	0.029	0.89	1.09	5.66	0.040	1.33
5299 (E4)	25% HAc	Euro	19-Oct-10	35.20	16.00	20-Oct-10	34.09	17.57	ND	3.37 (0.73)	0.015 (0.034)	0.48 (0.07)	3.73 (0.79)	0.0153 (0.009)	0.617 (0.074)	0.362 (0.034)	2.76 (0.48)	0.021 (0.005)	0.85 (0.07)
5330 (M1)	25% HAc	Marine	20-Oct-10	34.07	17.61	21-Oct-10	31.32	21.55	ND	3.91	0.0696	0.145	2.41	0.0254	0.436	0.275	2.53	0.0550	0.696
5425 (M2)	25% HAc	Marine	21-Oct-10	31.11	21.86	23-Oct-10	27.53	22.01	ND	0.963	0.0580	0.0283	0.675	0.00134	0.0793	0.0682	1.055	0.00422	0.0893
5579 (M3)	25% HAc	Marine	23-Oct-10	27.53	22.01	25-Oct-10	24.01	22.00	ND	1.09 (0.07)	0.033 (0.026)	0.117 (0.039)	0.89 (0.09)	0.0090 (0.0030)	0.3578 (0.0115)	0.093 (0.020)	1.18 (0.42)	0.0075 (0.0033)	0.268 (0.042)
5729 (A1)	25% HAc	N.Afr	25-Oct-10	24.01	22.00	28-Oct-10	17.35	18.25	ND	239	0.403	13.99	116	0.233	0.924	0.366	2.38	0.0211	1.21
5738 (A2)	25% HAc	N.Afr	28-Oct-10	17.36	18.25	29-Oct-10	17.36	18.25	ND	273	0.775	18.2	136	0.289	1.025	0.327	2.36	0.0187	1.15
5764 (A3)	25% HAc	N.Afr	29-Oct-10	17.36	18.25	29-Oct-10	17.36	19.06	ND	190	0.789	13.8	99.2	0.198	0.975	0.267	1.455	0.0125	0.526
5848 (A4)	25% HAc	N.Afr	30-Oct-10	17.35	20.02	30-Oct-10	17.35	20.83	ND	516	1.36	28.4	266	0.483	1.51	0.911	3.46	0.0309	1.44
5870 (A5)	25% HAc	N.Afr	31-Oct-10	17.36	20.83	31-Oct-10	17.36	20.86	ND	545	1.11	34.4	270	0.516	2.03	0.626	2.18	0.0263	0.942
5965 (A6)	25% HAc	N.Afr	01-Nov-10	17.35	20.91	01-Nov-10	17.35	22.79	ND	212	0.895	12.7	109	0.202	1.08	0.412	1.55	0.0202	1.57
5999 (A7)	25% HAc	N.Afr	01-Nov-10	17.35	22.79	01-Nov-10	17.35	23.07	ND	102	0.470	6.66	53.5	0.105	0.485	0.234	1.23	0.0162	0.435

6070 (A8)	25% HAc	N.Afr	03-Nov-10	17.40	24.50	03-Nov-10	17.34	24.55	ND	29.6 (2.0)	0.182 (0.037)	2.10 (0.22)	15.6 (3.4)	0.034 (0.004)	0.205 (0.007)	0.020 (0.007)	0.776 (0.440)	0.006 (0.002)	0.088 (0.012)
6250 (N1)	25% HAc	N.Amer	07-Nov-11	39.71	69.87	08-Nov-11	39.70	69.80	ND	2.06 (0.24)	0.139 (0.032)	0.363 (0.026)	3.33 (0.20)	0.007 (0.001)	0.150 (0.180)	0.229 (0.018)	1.44 (0.38)	0.023 (0.017)	0.427 (0.117)
6319 (N2)	25% HAc	N.Amer	08-Nov-11	39.70	69.80	09-Nov-11	39.35	69.54	ND	6.39	0.153	1.20	9.29	0.0208	0.232	0.850	3.64	0.0310	0.901
6452 (N3)	25% HAc	N.Amer	09-Nov-11	39.35	69.54	11-Nov-11	38.65	69.20	ND	1.77	0.0599	0.121	2.56	0.00676	0.135	0.144	1.09	0.0628	0.162
6546 (N4)	25% HAc	N.Amer	12-Nov-11	38.33	68.87	13-Nov-11	38.36	68.86	ND	3.59	0.126	0.684	4.62	0.00624	0.0964	0.184	1.32	0.0347	0.427
6677 (N5)	25% HAc	N.Amer	13-Nov-11	38.36	68.86	15-Nov-11	37.58	68.40	ND	2.59 (0.35)	0.149 (0.206)	0.3051 (0.030)	2.25 (0.20)	0.0035 (0.003)	0.534 (0.004)	0.088 (0.015)	0.716 (0.098)	0.018 (0.007)	0.281 (0.051)
6788 (N6)	25% HAc	N.Amer	15-Nov-11	37.62	68.38	17-Nov-11	37.62	68.38	ND	2.42	0.0911	0.194	1.14	0.00530	0.0855	0.125	1.27	0.120	0.181
6903 (N7)	25% HAc	N.Amer	19-Nov-11	32.02	64.44	20-Nov-11	31.75	64.17	ND	1.59	0.00774	0.0691	0.616	0.00134	0.0470	0.0378	0.610	0.00935	0.102
6964 (N8)	25% HAc	N.Amer	20-Nov-11	31.75	64.17	21-Nov-11	31.55	63.43	ND	0.664	0.0500	0.0416	0.174	0.000426	0.0119	0.0291	0.221	0.00326	0.0598
7156 (M4)	25% HAc	Marine	21-Nov-11	31.53	63.37	24-Nov-11	29.70	56.82	ND	2.03 (1.12)	0.062 (0.010)	0.062 (0.001)	0.389 (0.039)	0.0032 (0.0002)	0.047 (0.002)	0.027 (0.019)	0.334 (0.156)	0.0074 (0.0015)	0.111 (0.005)
7245 (M5)	25% HAc	Marine	24-Nov-11	29.70	56.82	26-Nov-11	27.58	49.63	ND	2.23	0.00975	0.0194	0.374	0.00362	0.0340	0.0228	0.148	0.00485	0.0324
7316 (M6)	25% HAc	Marine	26-Nov-11	27.58	49.63	27-Nov-11	27.55	49.55	ND	1.31 (0.063)	0.039 (0.007)	0.015 (0.003)	0.108 (0.021)	0.0018 (0.0004)	0.091 (0.004)	0.015 (0.005)	0.102 (0.025)	0.0066 (0.0048)	0.020 (0.007)
7357 (M7)	25% HAc	Marine	27-Nov-11	27.55	49.55	28-Nov-11	26.31	45.39	ND	4.24	0.0132	0.0407	0.560	0.00914	0.0636	0.0195	0.107	0.0295	0.0585
7426 (M8)	25% HAc	Marine	28-Nov-11	26.28	45.30	29-Nov-11	26.14	44.83	ND	1.66	0.00713	0.0227	0.307	0.00107	0.0342	0.0173	0.230	0.00430	0.0289
7509 (M9)	25% HAc	Marine	29-Nov-11	26.14	44.83	30-Nov-11	25.58	43.60	ND	4.54 (1.16)	0.044 (0.009)	0.135 (0.006)	1.00 (0.13)	0.0039 (0.0003)	0.041 (0.005)	0.033 (0.019)	0.39 (0.17)	0.0077 (0.0025)	0.0356
7587 (M10)	25% HAc	Marine	30-Nov-11	25.55	43.54	01-Dec-11	24.15	40.22	ND	7.27	0.0879	0.124	1.12	0.00514	0.0647	0.0460	0.309	0.00670	0.0635
7655 (M11)	25% HAc	Marine	01-Dec-11	24.15	40.22	02-Dec-11	24.15	40.22	ND	1.53	0.0622	0.0384	0.447	0.00148	0.0369	0.0190	0.109	0.00581	0.0387
7697 (M12)	25% HAc	Marine	02-Dec-11	24.15	40.22	03-Dec-11	22.70	36.73	ND	2.79 (0.11)	0.049 (0.013)	0.027 (0.016)	0.42 (0.22)	0.0013 (0.0009)	0.045 (0.016)	0.043 (0.023)	0.77 (0.59)	0.038 (0.035)	0.025 (0.001)
7791 (A9)	25% HAc	N.Afr	03-Dec-11	22.70	36.73	04-Dec-11	22.36	35.87	ND	63.9	0.233	3.79	31.7	0.0543	0.161	0.0877	0.439	0.0516	0.186
7860 (A10)	25% HAc	N.Afr	04-Dec-11	22.36	35.87	05-Dec-11	22.38	35.87	ND	369	0.850	22.9	183	0.314	0.385	0.346	1.22	0.0704	0.581
7899 (A11)	25% HAc	N.Afr	05-Dec-11	22.37	35.62	06-Dec-11	20.88	32.62	ND	95.2 (2.7)	0.37 (0.04)	6.33 (0.32)	48.0 (1.1)	0.086 (0.005)	0.19 (0.01)	0.13 (0.01)	0.92 (0.08)	0.0081 (0.0006)	0.37 (0.01)
7946 (A12)	25% HAc	N.Afr	06-Dec-11	20.88	32.62	07-Dec-11	19.43	29.38	ND	223	0.518	12.89	106	0.211	0.279	0.320	1.10	0.0144	0.461
8004 (A13)	25% HAc	N.Afr	07-Dec-11	19.43	29.38	08-Dec-11	19.43	29.38	ND	438	0.909	24.3	206	0.393	0.430	0.382	1.51	0.0250	0.641
8044 (A14)	25% HAc	N.Afr	08-Dec-11	19.43	29.38	09-Dec-11	18.13	26.13	ND	543	1.14	37.2	246	0.612	0.679	0.823	1.91	0.0237	0.900
8045 (A15)	25% HAc	N.Afr	09-Dec-11	17.75	25.31	09-Dec-11	17.40	24.53	ND	644	2.03	56.5	307	0.913	0.720	0.617	1.62	0.0187	0.933
geoa1 (G1)	25% HAc	High Lat	19-May-14	40.33	10.04	20-May-14	40.33	10.04	ND	0.80 (0.48)	0.0047 (0.0011)	0.051 (0.007)	0.56 (0.16)	0.0017 (0.0003)	0.077 (0.010)	0.062 (0.064)	0.21 (0.16)	0.0015 (0.0002)	0.037 (0.004)
geoa2 (G2)	25% HAc	High Lat	23-May-14	40.33	12.22	24-May-14	41.38	13.89	ND	1.66	0.0244	0.130	1.45	0.00239	0.0370	0.0285	0.455	0.00430	0.112
geoa3 (G3)	25% HAc	High Lat	24-May-14	41.38	13.89	25-May-14	41.38	13.89	ND	2.07	0.0378	0.185	2.16	0.00296	0.0823	0.0303	0.469	0.00616	0.228
geoa4 (G4)	25% HAc	High Lat	25-May-14	41.38	13.89	27-May-14	41.38	13.89	ND	1.88	0.0188	0.151	1.69	0.00232	0.0686	0.0419	0.488	0.00372	0.106
geoa5 (G5)	25% HAc	High Lat	27-May-14	41.38	13.89	29-May-14	43.78	17.03	ND	3.20	0.0128	0.141	1.78	0.00255	0.0594	0.0379	0.299	0.0027	0.0788
geoa6 (G6)	25% HAc	High Lat	30-May-14	45.05	18.51	02-Jun-14	47.29	20.26	ND	2.0 (0.4)	0.0010 (0.0012)	0.072 (0.002)	0.92 (0.13)	0.0021 (0.0001)	0.13 (0.02)	0.043 (0.013)	0.31 (0.14)	0.0012 (0.0002)	0.037 (0.005)
geoa7 (G7)	25% HAc	High Lat	02-Jun-14	47.29	20.26	04-Jun-14	50.28	22.60	ND	1.89	0.00846	0.0704	0.801	0.00126	0.0743	0.0386	0.222	0.00182	0.0434
geoa8 (G8)	25% HAc	High Lat	04-Jun-14	50.28	22.60	06-Jun-14	53.42	25.07	ND	2.20	0.00765	0.0792	1.37	0.00112	0.0408	0.0404	0.845	0.00131	0.0686
geoa9 (G9)	25% HAc	High Lat	06-Jun-14	53.42	25.07	08-Jun-14	55.51	26.71	ND	12.71	0.0104	0.0934	2.34	0.00348	0.0999	0.0637	1.4611	0.00441	0.132
geoa10 (G10)	25% HAc	High Lat	08-Jun-14	55.51	26.71	10-Jun-14	58.21	29.72	ND	3.0 (0.5)	0.030 (0.004)	0.084 (0.005)	1.6 (0.1)	0.0021 (0.0001)	0.036 (0.004)	0.056 (0.022)	0.47 (0.10)	0.0020 (0.0008)	0.099 (0.012)
geoa11 (G11)	25% HAc	High Lat	10-Jun-14	58.21	29.72	12-Jun-14	59.20	34.78	ND	2.48	0.0133	0.0376	0.705	7.2E-04	0.0217	0.0454	0.785	1.0E-03	0.0362

geoa12 (G12)	25% HAc	High Lat	12-Jun-14	59.20	34.78	14-Jun-14	59.62	38.95	ND	0.477	0.00373	0.00240	0.104	1.6E-04	0.00367	0.0172	0.0683	2.0E-04	0.0106
geoa13 (G13)	25% HAc	High Lat	14-Jun-14	59.62	38.95	16-Jun-14	59.80	42.00	ND	1.24	<DL	0.00733	0.1742	3.1E-04	0.0189	0.00678	0.0216	2.0E-04	0.0601
geoa14 (G14)	25% HAc	High Lat	16-Jun-14	59.80	42.00	18-Jun-14	59.80	41.99	ND	1.5 (0.9)	0.0032 (0.0026)	0.012 (0.004)	0.32 (0.26)	0.00072 (0.00030)	0.026 (0.027)	0.0074 (0.0059)	0.21 (0.15)	0.00025 (0.00016)	0.010 (0.007)
geoa15 (G15)	25% HAc	High Lat	18-Jun-14	59.70	42.53	20-Jun-14	59.07	46.09	ND	0.576	0.00362	0.00801	0.2026	3.7E-04	0.0279	0.0149	0.148	7.5E-04	0.0412
geoa16 (G16)	25% HAc	High Lat	21-Jun-14	56.91	47.42	23-Jun-14	55.72	48.17	ND	0.119	<DL	0.0016	0.0125	6.8E-05	0.0025	0.00319	0.0247	1.3E-04	0.00567
geoa17 (G17)	25% HAc	High Lat	23-Jun-14	55.72	48.17	25-Jun-14	52.93	51.39	ND	2.69	0.0162	0.171	1.04	0.00146	0.0205	0.0360	0.430	0.00539	0.0828
geoa18 (G18)	25% HAc	High Lat	25-Jun-14	52.93	51.39	27-Jun-14	51.99	53.84	ND	0.935	0.00278	0.0830	0.221	0.00037	0.00113	0.0150	0.1168	4.2E-04	0.0305

Note: Matrix blanks and filter blanks are calculated by dividing the ng/leach by 100 m3 (the average volume of air filtered across each filter)

Table S2.

		High Latitude (this study)	Volcanic ash (Eyjafjallajökull) (Achterberg et al., 2013)	Icelandic tephra (Oladottir et al., 2011)	Icelandic sand (Baratoux et al., 2011)	North American (this study)	IMPROVE (Nov-Dec 2011)	European (this study)	Mace Head (1989-1994; 1996-97) (*=Huang et al., 1999; **=Spokes et al., 2001)	North African (this study)	UCC (Rudnick & Gao, 2003)
Ti/Al	min	0.007		0.11	0.05	0.07	0.002	0.039		0.076	
	max	0.098		0.29	0.22	0.44	0.454	0.071		0.087	
	mean	0.049	0.11	0.21	0.12	0.17	0.082	0.054		0.080	0.047
Mn/Al	min	0.002		0.013	0.013	0.020	0.001	0.009		0.012	
	max	0.021		0.037	0.026	0.060	1.892	0.027		0.014	
	mean	0.011	0.022	0.028	0.018	0.036	0.062	0.017	0.038*	0.013	0.010
Fe/Al	min	0.12		1.09	0.77	0.86	0.10	0.10		0.72	
	max	0.89		1.61	1.60	1.42	9.34	0.95		0.85	
	mean	0.46	0.97	1.38	1.05	1.08	1.25	0.58	0.63**	0.78	0.48
Co/Al	min	0.00004			0.00045	0.0004		0.00037		0.00028	
	max	0.00092			0.00068	0.0092		0.00086		0.00048	
	mean	0.00032			0.00055	0.0032		0.00061	0.00024**	0.00034	0.00021
Ni/Al	min	0.002			0.0006	0.013	0.002	0.015		0.0008	
	max	0.140			0.0015	0.156	1.517	0.028		0.0019	
	mean	0.016	0.0006		0.0008	0.054	0.028	0.019		0.0011	0.00058
Cu/Al	min	0.0023			0.0007	0.009	0.001	0.020		0.00056	
	max	0.0512			0.0015	0.080	0.759	0.028		0.00117	
	mean	0.0078			0.0009	0.034	0.065	0.023		0.00080	0.00034
Zn/Al	min	0.019			0.0008	0.04	0.03	0.040		0.0012	
	max	0.158			0.0020	0.44	4.24	0.122		0.0046	
	mean	0.056	0.0022		0.0012	0.17	0.54	0.081	0.15*, 0.0013**	0.0022	0.00082
Cd/Al	min	0.00008				0.0003		0.00069		0.000006	
	max	0.00102				0.0245		0.00107		0.000122	
	mean	0.00025				0.0055		0.00087		0.000031	0.0000011
Pb/Al	min	0.003				0.009	0.001	0.013		0.00050	
	max	0.384				0.052	1.41	0.032		0.00142	
	mean	0.034	0.000057			0.028	0.12	0.023	0.010*	0.00073	0.00021

Table S3

Sol %		Mn	Fe	Co	Ni	Cu	Zn	Cd
HAc	High Lat	45 ± 10	17 ± 11	43 ± 23	55 ± 22	61 ± 31	63 ± 29	68 ± 21
UHP	High Lat	35 ± 9	7.4 ± 5.6	31 ± 21	37 ± 14	24 ± 12	55 ± 29	56 ± 21
HAc	Euro	64 ± 20	21 ± 22	46 ± 15	44 ± 9	36 ± 17	75 ± 25	52 ± 18
UHP	Euro	49 ± 14	7.8 ± 8.7	35 ± 11	33 ± 6	25 ± 12	56 ± 18	40 ± 9
HAc	N. Amer	53 ± 17	13 ± 4	19 ± 14	15 ± 6	38 ± 12	64 ± 26	65 ± 32
UHP	N. Amer	39 ± 13	3.8 ± 2.4	12 ± 9	10 ± 4	23 ± 12	48 ± 21	51 ± 28

SW	N. Amer	41 ±19	1.5 ± 1.0	16 ± 10	2.4 ± 2.2	18 ± 12	36 ± 17	44 ± 36
HAc	Marine	43 ± 19	17 ± 10	21 ± 20	28 ± 30	27 ± 12	62 ± 38	46 ± 29
UHP	Marine	32 ± 11	6.5 ± 5.9	12 ± 10	22 ± 22	11 ± 7	35 ± 22	27 ± 14
SW	Marine	5 ± 5	1.1 ± 1.0	3.9 ± 3.2			23 ± 15	19 ± 18
HAc	N. Afr	49 ± 6	6.4 ± 1.0	29 ± 6	26 ± 13	18 ± 6	32 ± 11	52 ± 24
UHP	N. Afr	20 ± 6	0.4 ± 0.1	6.4 ± 3.1	15 ± 11	2.2 ± 1.2	10 ± 8	22 ± 12
SW	N. Afr	18 ± 3	0.05 ± 0.05	9.1 ± 3.1	4.7 ± 6.4	1.9 ± 0.8	10 ± 9	32 ± 17

Table S4

Sol %	Mn	Fe	Co	Ni	Cu	Zn	Cd
HAc	48 ± 14	14 ± 11	32 ± 20	35 ± 24	37 ± 26	55 ± 30	58 ± 26
UHP	32 ± 13	4.9 ± 5.5	18 ± 17	24 ± 17	16 ± 13	38 ± 28	39 ± 23
SW	24 ± 17	0.8 ± 1.1	10 ± 8.0	3.8 ± 5.3	11 ± 12	23 ± 17	30 ± 24

Table 5

Al **N American**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.464	Intercept	0.492	0.445	1.106	0.311	-0.597	1.582
R Square	0.215	X Variable 1	0.430	0.336	1.283	0.247	**-0.391**	**1.252**
Adjusted R Square	0.084							
Standard Error	0.666							
Observations	8							

Al **Marine**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.414	Intercept	1.190	0.619	1.921	0.195	-1.48	3.86
R Square	0.171	X Variable 1	-1.397	2.173	-0.643	0.586	**-10.75**	**7.95**
Adjusted R Square	-0.243							
Standard Error	0.562							
Observations	4							

Al **N African**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.458	Intercept	1.377	10.771	0.128	0.902	-24.98	27.73
R Square	0.209	X Variable 1	2.723	2.160	1.261	0.254	**-2.562**	**8.008**

Adjusted R Square	0.078
Standard Error	11.420
Observations	8

Mn **N American**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.992	Intercept	0.029	0.034	0.868	0.449	-0.078	0.137
R Square	0.984	X Variable 1	0.840	0.061	13.769	0.001	**0.646**	**1.034**
Adjusted R Square	0.979							
Standard Error	0.047							
Observations	5							

Mn **N African**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.977	Intercept	1.123	0.590	1.903	0.106	-0.321	2.568
R Square	0.954	X Variable 1	0.742	0.066	11.185	0.000	0.580	0.904
Adjusted R Square	0.947							
Standard Error	1.072							
Observations	8							

Fe **N American**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.968	Intercept	0.171	0.088	1.947	0.099	-0.044	0.387
R Square	0.937	X Variable 1	1.702	0.181	9.407	0.000	1.259	2.145
Adjusted R Square	0.926							
Standard Error	0.186							
Observations	8							

Fe **Marine**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.927	Intercept	0.073	0.028	2.639	0.039	0.005	0.141
R Square	0.859	X Variable 1	4.923	0.813	6.054	0.001	2.934	6.913
Adjusted R Square	0.836							
Standard Error	0.043							
Observations	8							

Fe **N African**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*

Multiple R	0.025	Intercept	7.190	7.407	0.971	0.369	-10.93	25.31
R Square	0.001	X Variable 1	0.830	13.777	0.060	0.954	**-32.88**	**34.54**
Adjusted R Square	-0.166							
Standard Error	8.129							
Observations	8							

Co **N American**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.992	Intercept	#####	0.0003	-1.292	0.244	-0.001	0.0004
R Square	0.984	X Variable 1	0.890	0.047	18.914	0.000001	**0.775**	**1.005**
Adjusted R Square	0.981							
Standard Error	0.001							
Observations	8							

Co **Marine**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.072	Intercept	0.002	0.001	1.363	0.222	-0.002	0.005
R Square	0.005	X Variable 1	0.209	1.177	0.178	0.865	**-2.672**	**3.090**
Adjusted R Square	-0.161							
Standard Error	0.003							
Observations	8							

Co **N African**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.941	Intercept	0.011	0.005	2.114	0.079	-0.002	0.024
R Square	0.886	X Variable 1	0.360	0.053	6.820	0.000	0.231	0.489
Adjusted R Square	0.867							
Standard Error	0.009							
Observations	8							

Ni **N American**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.986	Intercept	0.055	0.009	6.190	0.025	0.017	0.093
R Square	0.971	X Variable 1	2.154	0.262	8.231	0.014	1.028	3.281
Adjusted R Square	0.957							
Standard Error	0.012							
Observations	4							

Ni **N African**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.843	Intercept	0.072	0.014	5.091	0.004	0.035	0.108
R Square	0.711	X Variable 1	0.631	0.180	3.505	0.017	**0.168**	**1.094**
Adjusted R Square	0.653							
Standard Error	0.021							
Observations	7							

Cu **N American**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.997	Intercept	0.007	0.008	0.868	0.419	-0.012	0.026
R Square	0.994	X Variable 1	1.187	0.039	30.815	0.000	1.092	1.281
Adjusted R Square	0.993							
Standard Error	0.018							
Observations	8							

Cu **N African**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.808	Intercept	0.015	0.007	2.219	0.077	-0.002	0.033
R Square	0.653	X Variable 1	0.593	0.193	3.069	0.028	**0.096**	**1.089**
Adjusted R Square	0.584							
Standard Error	0.011							
Observations	7							

Zn **N American**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.987	Intercept	0.166	0.077	2.172	0.073	-0.021	0.354
R Square	0.974	X Variable 1	0.921	0.061	14.981	0.000	**0.771**	**1.072**
Adjusted R Square	0.970							
Standard Error	0.152							
Observations	8							

Zn **Marine**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.877	Intercept	0.030	0.018	1.668	0.146	-0.014	0.075
R Square	0.770	X Variable 1	0.862	0.193	4.478	0.004	**0.391**	**1.334**
Adjusted R Square	0.731							

Standard Error	0.017
Observations	8

Zn **N African**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.671	Intercept	-0.111	0.195	-0.570	0.599	-0.653	0.431
R Square	0.450	X Variable 1	1.104	0.610	1.809	0.145	**-0.591**	**2.799**
Adjusted R Square	0.312							
Standard Error	0.140							
Observations	6							

Cd **N American**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.847	Intercept	0.011	0.009	1.329	0.232	-0.010	0.033
R Square	0.717	X Variable 1	0.312	0.080	3.903	0.008	0.116	0.508
Adjusted R Square	0.670							
Standard Error	0.020							
Observations	8							

Cd **Marine**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.811	Intercept	0.002	0.001	2.726	0.034	0.000	0.004
R Square	0.658	X Variable 1	0.567	0.167	3.395	0.015	0.158	0.975
Adjusted R Square	0.601							
Standard Error	0.001							
Observations	8							

Cd **N African**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.476	Intercept	0.004	0.003	1.366	0.244	-0.005	0.013
R Square	0.226	X Variable 1	0.328	0.304	1.081	0.340	**-0.515**	**1.171**
Adjusted R Square	0.033							
Standard Error	0.003							
Observations	6							

Pb **N American**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.995	Intercept	-0.018	0.010	-1.841	0.115	-0.042	0.006

R Square	0.990	X Variable 1	0.818	0.034	24.350	0.000	0.735	0.900
Adjusted R Square	0.988							
Standard Error	0.018							
Observations	8							

Pb **Marine**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.804	Intercept	0.003	0.005	0.614	0.562	-0.009	0.014
R Square	0.647	X Variable 1	0.424	0.128	3.315	0.016	0.111	0.737
Adjusted R Square	0.588							
Standard Error	0.008							
Observations	8							

Pb **N African**

Regression Statistics			*Coefficients*	*Standard Error*	*t Stat*	*P-value*	*Lower 95%*	*Upper 95%*
Multiple R	0.938	Intercept	-0.013	0.007	-2.020	0.113	-0.031	0.005
R Square	0.879	X Variable 1	0.551	0.102	5.393	0.006	0.267	0.835
Adjusted R Square	0.849							
Standard Error	0.006							
Observations	6							

Captions for supplementary material

Figure S1. Air mass 5-day back trajectory simulations, with arrival heights of 50, 500 and 1500 m. The first four are representative simulations for N. American, European, Marine and Norh African samples, redrawn from the Supplemental Material of Shelley et al. (2015). The other simulations are reproduced from Shelley et al. (2017). Note that the labelling system has changed slightly from these earlier publications. In Shelley et al. (2017) the simulations were labelled geoa1-18, and here they are G1-18. The simulations in Shelley et al. (2015) were not labelled with their sample number. For reference, the sample numbers can also be found in Table S1.

Figure S2. Positive Matrix Factorisation outputs using total TE concentration data: (a) Factor fingerprint and (b) factor time series, for total aerosol TEs for cruises GA03-2010 (E1 – A8), GA03-2011 (N1 – A15), and GA01 (G1 – 17), where E = European, M = Marine, N = North American, A = North African and G = High Latitude aerosols, Factor 1 represents a mineral dust source (red, highest contributions from Al, Ti, Fe, Zr, and Ba), and Factor 2 a pollution source (blue, highest contributions from Ni, Cu, Zn, Cd, and Pb). Note that all of the variance for aerosol Cd is explained by the pollution factor. The factor time series indicates when each factor was dominant. The y-axes of the factor time series represent log normalised data. These figures were produced using the US Environmental Protection Agency's Positive Matrix Factorisation model, EPA PMF v. 5.0

Figure S3. Fractional solubility of bioactive aerosol TEs (average ± 1 SD) from different source regions following leaches with (a) UHP water, (b) seawater, and (c) 25 % acetic acid, and (d) a comparison of fractional solubility using the three leach media

Table S1. Metadata and concentrations of TEs from (a) UHP water, (b) seawater, and (c) 25% acetic acid leaches. Note that (1) replicate leaches were not performed on all samples, the values in brackets in the data columns are the standard deviation of replicate samples (n = 3), (2) a different labelling convention was used in Shelley et al. (2017) to refer to the GA01 samples. Here we use G1-18 to refer to the samples collected during GA01 (A1-18 in Shelley et al., 2017), and A1-15 to refer to the North African samples from GA03; these labels can be found in brackets after the GEOTRACES sample number in the first column, and (4) matrix blanks and filter blanks were calculated by dividing the concentration of TE (ng) per leach by 100 m^3 (the average volume of air filtered across each filter)

Table S2. Elemental mass ratios normalised to Al from literature values and this study. Literature values are not shown for N. African dust as these have been discussed in detail in Shelley et al. (2017)

Table S3. Fractional solubility of bioactive TEs (± 1 SD), corresponding to Figures S3(a-c), using Eqn. 1 (UHP water soluble) and Eqn. 2 (UHP water plus acetic acid soluble).

Table S4. Fractional solubility of bioactive TEs (± 1 SD), corresponding to Figure S3(d), using Eqn. 1 (UHP water soluble) and Eqn. 2 (UHP water plus acetic acid soluble).

Table S5. Summary statistics for the fractional solubility of TEs following UHP water and seawater instantaneous leaches. The slope is not significantly different to 1.0 where the confidence interval includes 1.0 (bold font). There were too few paired Marine samples for Mn, Ni and Cu to assess whether the slope differed significantly from 1.0.

Additional references

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