

Supplement of Biogeosciences, 15, 2271–2288, 2018
<https://doi.org/10.5194/bg-15-2271-2018-supplement>
© Author(s) 2018. This work is distributed under
the Creative Commons Attribution 4.0 License.



Supplement of

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

Rachel U. Shelley et al.

Correspondence to: Rachel U. Shelley (rshelley@fsu.edu)

The copyright of individual parts of the supplement might differ from the CC BY 4.0 License.

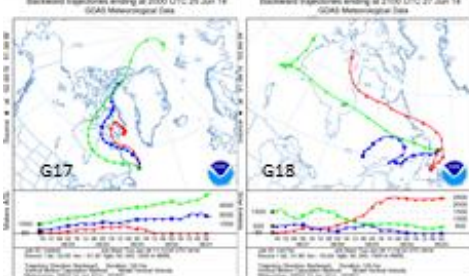
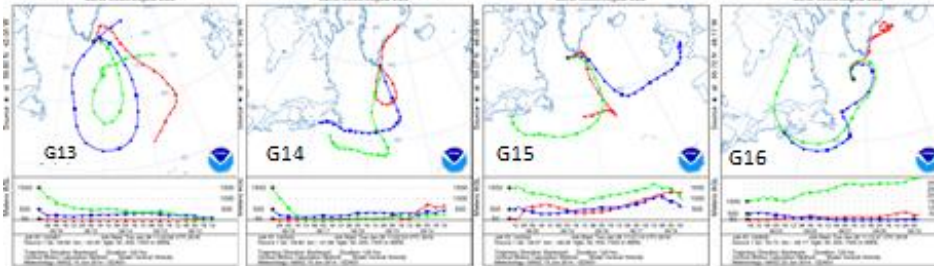
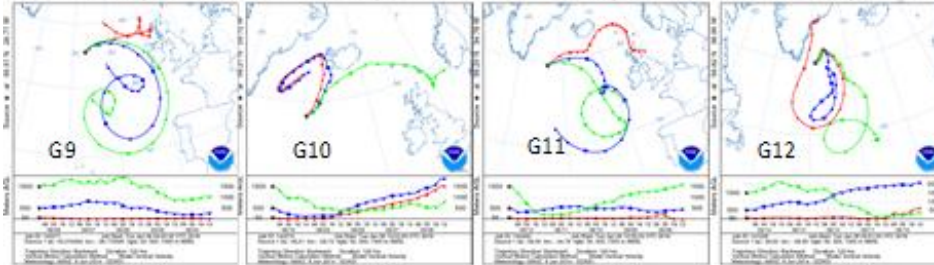
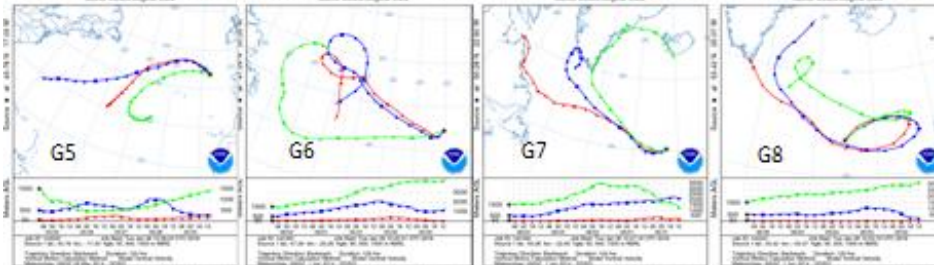
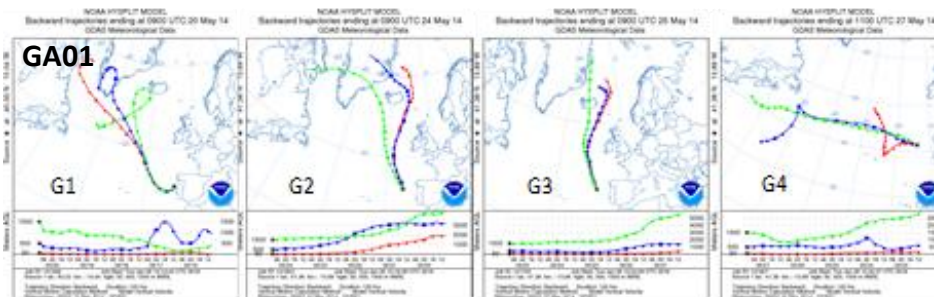
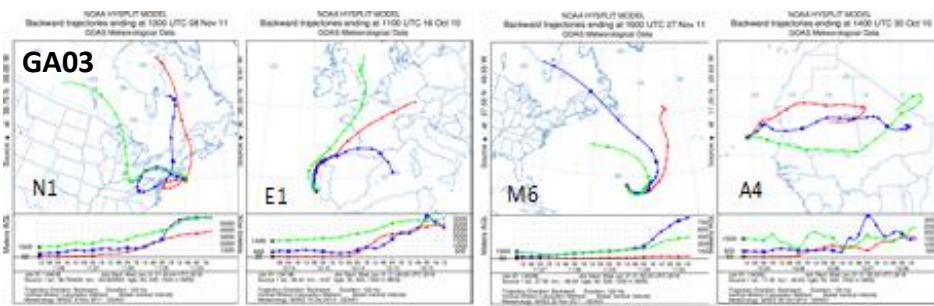
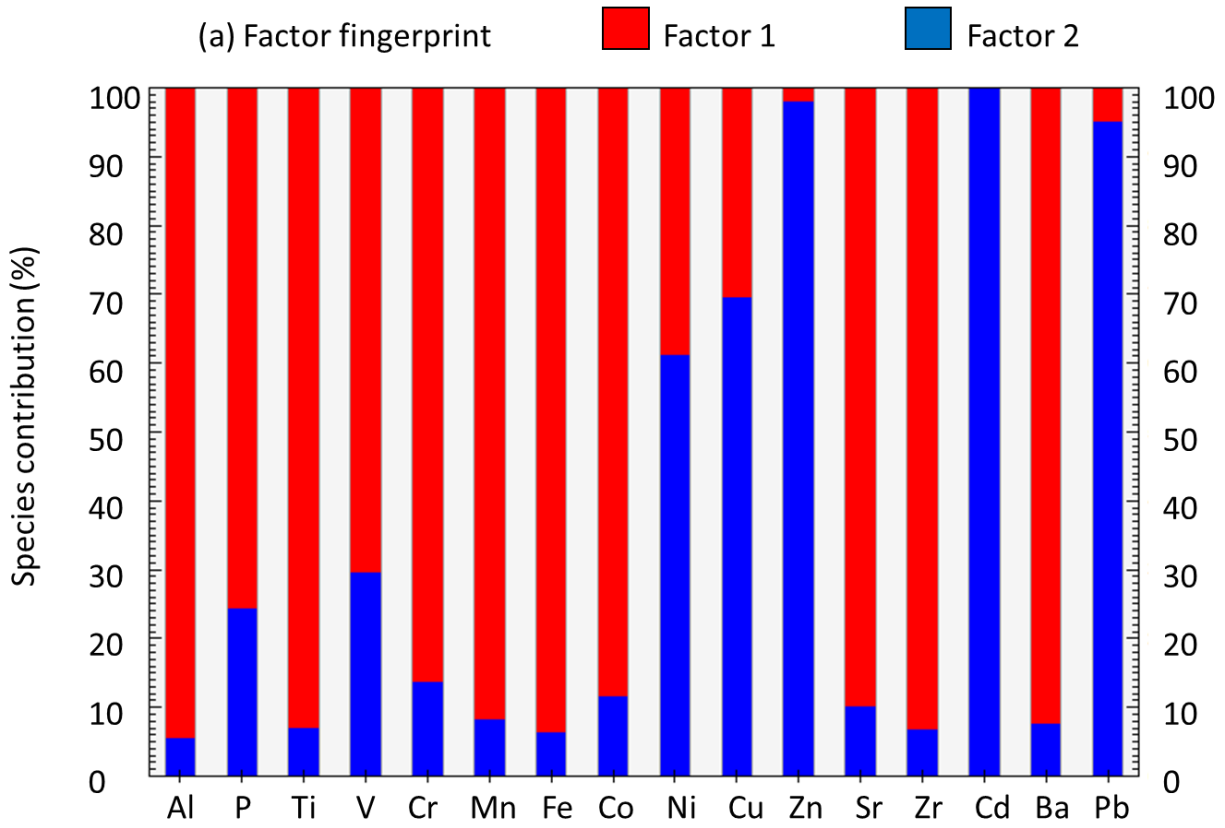
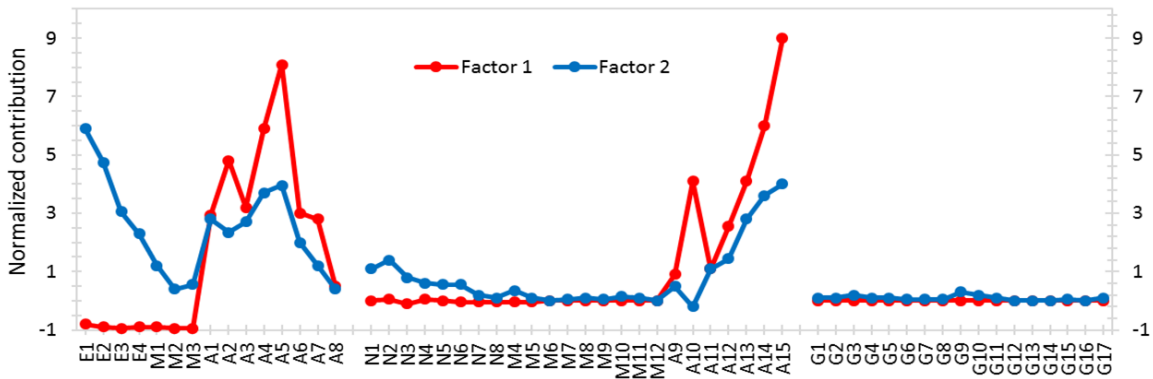


Figure S1



(b) Factor time series



11

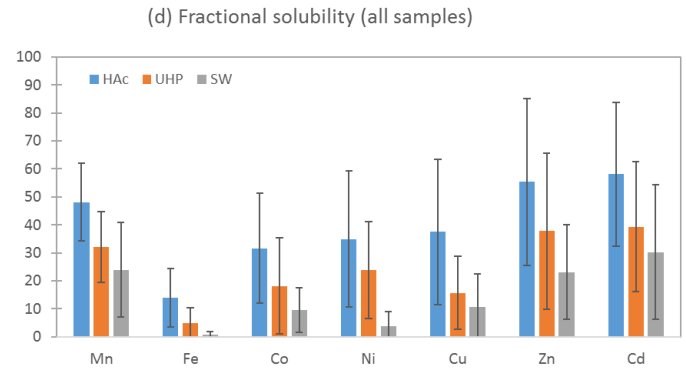
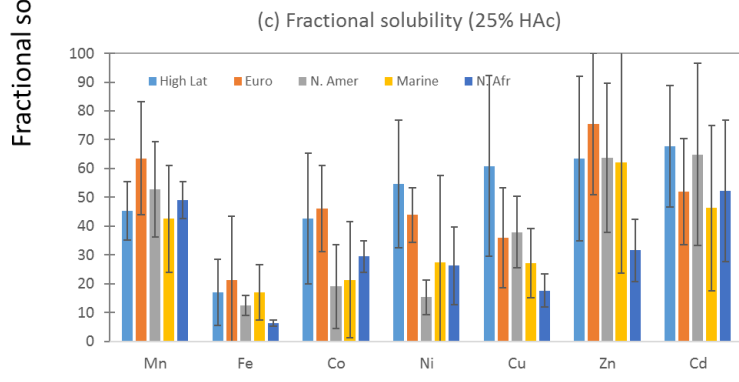
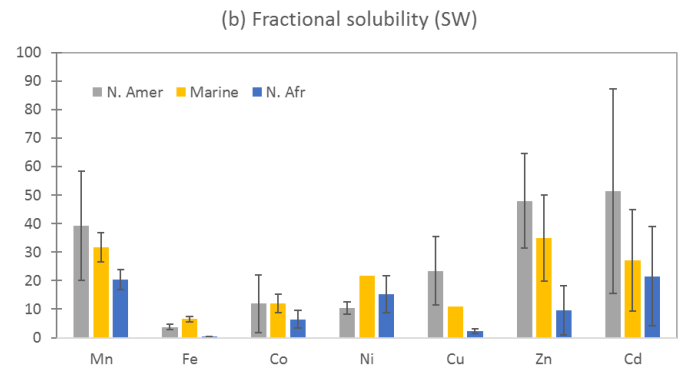
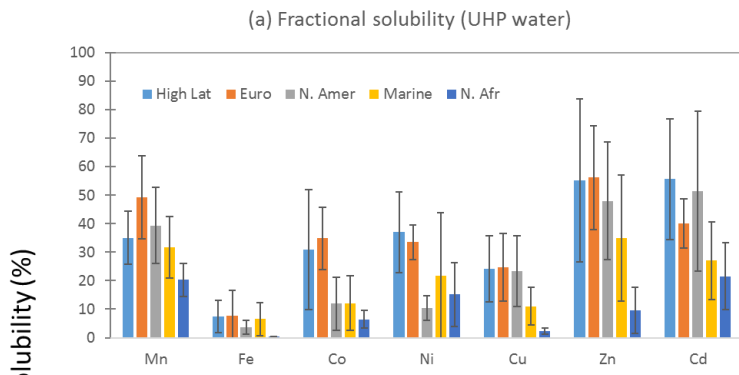
12

13 **Figure S2.**

14

15

16

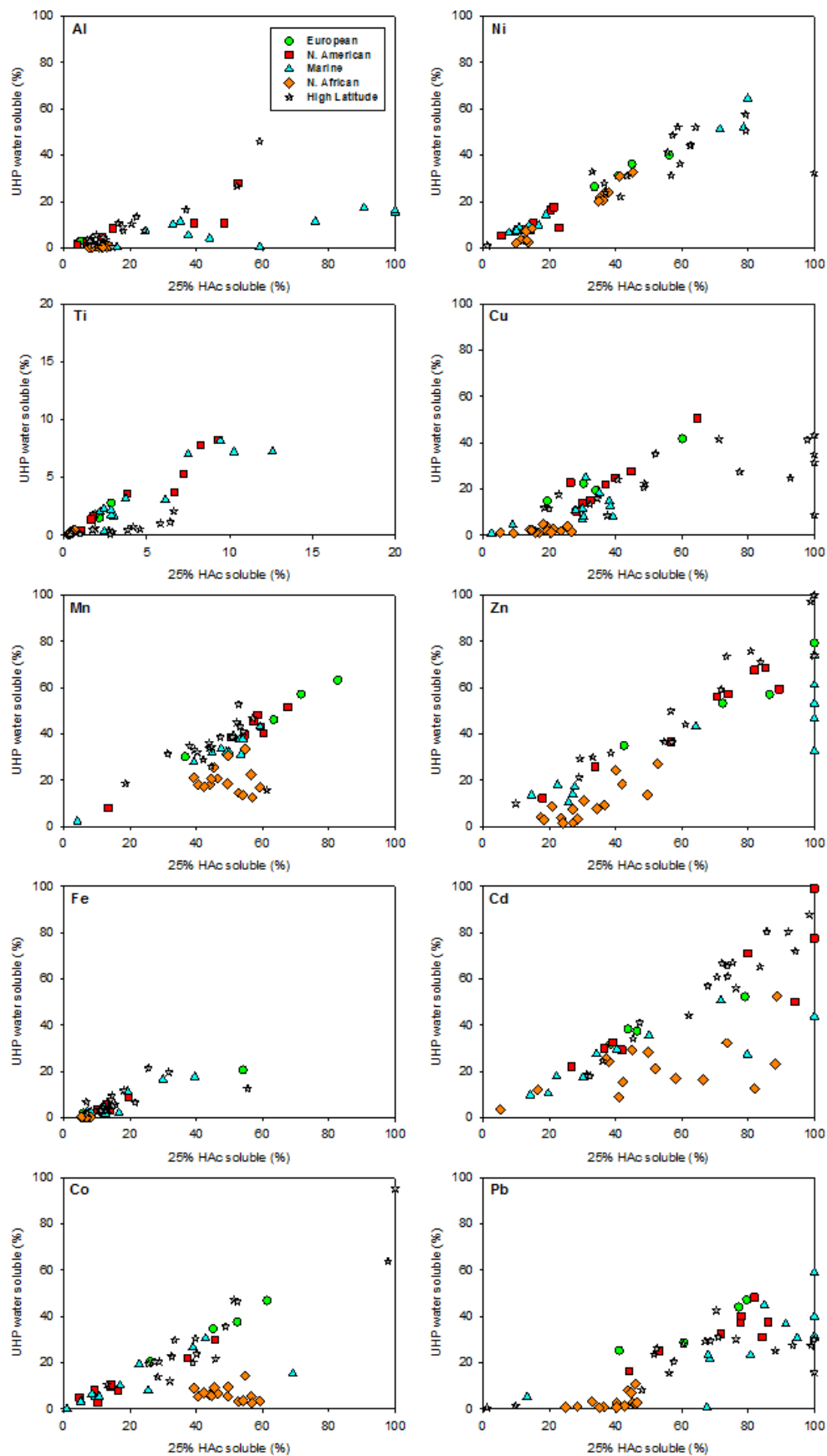


17

18

19 **Figure S3**

20



21

22 **Figure S4**

Table S1

(a) UHP water leach									Al	Ti	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
UHP water matrix blank	ng/m ³	(n=9)	Legend						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						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-06
			<DL = below detection																		
Filter Blank	ng/m ³	(n=3)	NC = not certified						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									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								109%	NC	99%	103%	99%	100%	104%	91%	95%	100%	105%	105%	99%
Trace elements = NIST 1643e; anions = Hach SRM	SD	(n=3)							1%	NC	6%	4%	6%	1%	3%	3%	1%	6%	3%	3%	5%

Sample #	Leach	Air mass regime	Start date	Start lat	Start Long	End date	End lat	End long	Al	Ti	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
	medium	(5 day AMBT)		(N)	(W)		(N)	(W)	ng/m ³	ng/m ³	ng/m ³	ng/m ³	ng/m ³	ng/m ³	ng/m ³	ng/m ³	ng/m ³	ng/m ³	μg/L	μg/L	μg/L
5037 (E1)	UHP water	Euro	15-Oct-10	38.32	9.66	16-Oct-10	38.33	9.67	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	0.256	0.00112	0.00669	0.131	0.00028	0.0198	0.00947	0.148	0.00065	0.0180	24743	127	5894

geo16 (G16)	UHP water	High Lat	21-Jun-14	56.91	47.42	23-Jun-14	55.72	48.17	0.065	<DL	0.00158	0.012	0.00003	0.00247	0.00133	0.0247	0.000118	0.0021	27407	<DL	5021
geo17 (G17)	UHP water	High Lat	23-Jun-14	55.72	48.17	25-Jun-14	52.93	51.39	0.756	0.00140	0.130	0.321	0.00103	0.0156	0.0243	0.423	0.00479	0.0384	44813	1568	6883
geo18 (G18)	UHP water	High Lat	25-Jun-14	52.93	51.39	27-Jun-14	51.99	53.84	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

			Al	Ti	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
Seawater matrix blank	ng/m3	(n=3)	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)			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)	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			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)		0.763	ND	0.342	0.944	0.0480	8.03	2.28	7.78	1.01	0.032
	SD		0.028	ND	0.093	0.031	0.0030	0.22	0.22	0.18	0.01	0.003
	consensus value (May 2013)		1.03	NC	0.350	0.933	0.0457	8.63	2.28	7.43	0.99	0.029
	SD		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)	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	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	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	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	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	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	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	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	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	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	<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	<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	<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	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	<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	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	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	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	<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	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	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	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	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	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	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	5.23	ND	18.2	0.392	0.206	<DL	<DL	0.174	0.0134	0.0240

(c) 25% acetic acid leach

									Al	Ti	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
25% HAc matrix blank	ng/m ³	(n=3)							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)									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/m ³	(n=3)							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									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)	Al ng/m ³	Ti ng/m ³	Mn ng/m ³	Fe ng/m ³	Co ng/m ³	Ni ng/m ³	Cu ng/m ³	Zn ng/m ³	Cd ng/m ³	Pb ng/m ³
5037 (E1)	25% HAc	Euro	15-Oct-10	38.32	9.66	16-Oct-10	38.33	9.67	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	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	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	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	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	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	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	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	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	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	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	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	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	102.05	0.470	6.66	53.5	0.105	0.485	0.234	1.239	0.0162	0.435
6070 (A8)	25% HAc	N.Afr	03-Nov-10	17.40	24.50	03-Nov-10	17.34	24.55	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	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	1.24	<DL	0.00733	0.1742	3.1E-04	0.0189	0.00678	0.0216	2.0E-04	0.0601

geo14 (G14)	25% HAc	High Lat	16-Jun-14	59.80	42.00	18-Jun-14	59.80	41.99	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)
geo15 (G15)	25% HAc	High Lat	18-Jun-14	59.70	42.53	20-Jun-14	59.07	46.09	0.576	0.00362	0.00801	0.2026	3.7E-04	0.0279	0.0149	0.148	7.5E-04	0.0412
geo16 (G16)	25% HAc	High Lat	21-Jun-14	56.91	47.42	23-Jun-14	55.72	48.17	0.119	<DL	0.0016	0.0125	6.8E-05	0.0025	0.00319	0.0247	1.3E-04	0.00567
geo17 (G17)	25% HAc	High Lat	23-Jun-14	55.72	48.17	25-Jun-14	52.93	51.39	2.69	0.0162	0.171	1.04	0.00146	0.0205	0.0360	0.430	0.00539	0.0828
geo18 (G18)	25% HAc	High Lat	25-Jun-14	52.93	51.39	27-Jun-14	51.99	53.84	0.935	0.00278	0.0830	0.221	0.00037	0.00113	0.0150	0.1168	4.2E-04	0.0305

(d) Total TEs (Shelley et al., 2015; 2017)

				Al	Ti	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb					
UHP water matrix blank	ng/m3	(n=9)	Legend	44.4	9.66	0.47	7.29	0.114	1.42	4.52	10.8	0.0530	0.0895					
DL (3xSD)			ND = not determined	133.9	11.9	0.20	7.66	0.121	0.32	3.21	15.8	0.0247	0.0302					
			<DL = below detection															
Filter Blank	ng/m3	(n=4)		266	211	1.18	46.5	0.131	2.16	4.33	15.9	0.0638	0.2875					
SD				57	67	0.19	5.5	0.038	0.29	1.23	4.03	0.0172	0.0697					
SRM	% recovery			90%	90%	101%	97%	100%	116%	104%	98%	118%	96%					
Buffalo River Sediment NIST 2704	SD	(n=4)		2%	3%	4%	2%	4%	12%	4%	12%	15%	4%					
5037 (E1)	Total	Euro	15-Oct-10	38.32	9.66	16-Oct-10	38.33	9.67	155	11.0	4.15	147	0.133	2.38	3.23	19.0	0.166	4.90
5145 (E2)	Total	Euro	16-Oct-10	38.33	9.66	17-Oct-10	38.33	9.66	141 (33)	8.89	2.92	110 (4)	0.0969	2.13 (0.23)	2.76	16.1	0.128	4.07
5197 (E3)	Total	Euro	17-Oct-10	38.33	9.66	19-Oct-10	35.20	16.00	129 (24)	5.07 (1.08)	1.52 (0.25)	61.9 (7.9)	0.0471 (0.005)	2.16 (0.24)	3.19 (1.27)	6.35 (1.65)	0.104	1.67 (0.20)
5299 (E4)	Total	Euro	19-Oct-10	35.20	16.00	20-Oct-10	34.09	17.57	65.8 (2.6)	2.76 (0.03)	0.583 (0.011)	6.89	0.0340 (0.0075)	1.83 (0.24)	1.87 (0.16)	3.18 (0.06)	0.0457 (0.0098)	1.10 (0.067)
5330 (M1)	Total	Marine	20-Oct-10	34.07	17.61	21-Oct-10	31.32	21.55	11.9 (0.9)	3.19 (0.18)	0.369 (0.129)	19.1 (4.2)	0.0594 (0.0006)	0.609 (0.228)	0.722 (0.099)	3.94 (1.59)	0.0334	0.761 (0.133)
5425 (M2)	Total	Marine	21-Oct-10	31.11	21.86	23-Oct-10	27.53	22.01	1.63	2.39	0.69	4.05 (0.66)	0.0258 (0.0074)	0.373 (0.385)	0.542 (0.422)	0.898	0.0123 (0.0076)	0.0695 (0.0107)
5579 (M3)	Total	Marine	23-Oct-10	27.53	22.01	25-Oct-10	24.01	22.00	5.96	0.889	0.197	7.79	0.0230	0.447	0.300	1.10	0.0186	0.316
5729 (A1)	Total	N.Afr	25-Oct-10	24.01	22.00	28-Oct-10	17.35	18.25	2540 (156)	197 (10)	31.7 (1.9)	2150 (219)	0.830 (0.070)	2.61 (0.19)	2.58 (0.575)	6.47	0.0550 (0.0372)	2.68 (0.19)
5738 (A2)	Total	N.Afr	28-Oct-10	17.36	18.25	29-Oct-10	17.36	18.25	2930	245	36.7	2100	0.971	2.83	1.94	6.84	0.0443	2.83
5764 (A3)	Total	N.Afr	29-Oct-10	17.36	18.25	29-Oct-10	17.36	19.06	2610	205	34.0	1920	0.909	2.62	2.89	6.96	0.240	1.60
5848 (A4)	Total	N.Afr	30-Oct-10	17.35	20.02	30-Oct-10	17.35	20.83	4840	374	61.2	3770	1.77	4.34	3.41	12.72	0.0621	3.13
5870 (A5)	Total	N.Afr	31-Oct-10	17.36	20.83	31-Oct-10	17.36	20.86	6500	496	77.1	4910	2.03	5.52	3.94	12.43	0.0692	3.31
5965 (A6)	Total	N.Afr	01-Nov-10	17.35	20.91	01-Nov-10	17.35	22.79	2370	185	28.1	1800	0.825	2.40	1.61	5.09	0.0449	3.37
5999 (A7)	Total	N.Afr	01-Nov-10	17.35	22.79	01-Nov-10	17.35	23.07	1330	105	17.0	1030	0.438	1.28	1.56	2.35	0.0435	0.945
6070 (A8)	Total	N.Afr	03-Nov-10	17.40	24.50	03-Nov-10	17.34	24.55	340	27.8	4.23	268	0.164	0.496	0.379	1.56	0.00682	0.212
6250 (N1)	Total	N.Amer	07-Nov-11	39.71	69.87	08-Nov-11	39.70	69.80	18.1	1.93	0.602	25.7	0.0407	0.990	0.573	8.05	0.0142	0.549
6319 (N2)	Total	N.Amer	08-Nov-11	39.70	69.80	09-Nov-11	39.35	69.54	88.9	8.69	1.77	80.2	0.0455	1.14	1.31	4.44	0.0272	1.10
6452 (N3)	Total	N.Amer	09-Nov-11	39.35	69.54	11-Nov-11	38.65	69.20	42.7 (5.1)	3.57 (0.57)	0.898 (0.083)	36.6 (5.5)	0.0478	1.16	0.445 (0.005)	1.91 (0.06)	0.160 (0.072)	0.367 (0.018)
6546 (N4)	Total	N.Amer	12-Nov-11	38.33	68.87	13-Nov-11	38.36	68.86	44.1	3.28	1.15	38.3	0.0167	0.672	0.411	1.87	0.130	0.803

6677 (N5)	Total	N.Amer	13-Nov-11	38.36	68.86	15-Nov-11	37.58	68.40	17.6	1.60	0.531	17.5	0.0246	0.630	0.239	2.11	0.0490	0.361
6788 (N6)	Total	N.Amer	15-Nov-11	37.62	68.38	17-Nov-11	37.62	68.38	6.16	1.36	0.332	8.17	0.0567	0.399	0.445	1.71	0.151	0.252
6903 (N7)	Total	N.Amer	19-Nov-11	32.02	64.44	20-Nov-11	31.75	64.17	3.02	0.770	0.127	3.15	0.0130	0.204	0.126	0.715	0.00992	0.119
6964 (N8)	Total	N.Amer	20-Nov-11	31.75	64.17	21-Nov-11	31.55	63.43	1.37 (0.02)	0.606	0.0824 (0.0028)	1.71 (0.49)	0.00891 (0.00370)	0.214 (0.035)	0.110 (0.002)	0.248 (0.058)	0.00779 (0.00465)	0.0710 (0.0064)
7156 (M4)	Total	Marine	21-Nov-11	31.53	63.37	24-Nov-11	29.70	56.82	4.35	0.493	0.125	3.23	0.0126	0.338	1.04	1.23	0.0171	0.162
7245 (M5)	Total	Marine	24-Nov-11	29.70	56.82	26-Nov-11	27.58	49.63	1.59	0.321	0.0380	0.946	0.00524	0.312	0.0759	0.121	0.0339	0.0300
7316 (M6)	Total	Marine	26-Nov-11	27.58	49.63	27-Nov-11	27.55	49.55	0.761	0.524	0.0351	0.900	0.0191	0.540	0.125	0.454	0.00824	0.0131
7357 (M7)	Total	Marine	27-Nov-11	27.55	49.55	28-Nov-11	26.31	45.39	2.22	0.544	0.0761	1.87	0.0402	0.334	0.0650	0.386	< DL	0.0514
7426 (M8)	Total	Marine	28-Nov-11	26.28	45.30	29-Nov-11	26.14	44.83	2.19 (0.23)	0.249 (0.011)	0.0428 (0.0174)	1.59 (0.07)	0.0210 (0.0089)	0.365	0.0707	0.192 (0.004)	0.00907	0.123 (0.134)
7509 (M9)	Total	Marine	29-Nov-11	26.14	44.83	30-Nov-11	25.58	43.60	19.6	1.53	0.250	14.2	0.0226	0.420	0.108	0.262	0.0255	0.0375
7587 (M10)	Total	Marine	30-Nov-11	25.55	43.54	01-Dec-11	24.15	40.22	19.3	1.43	0.253	13.6	0.0481	0.639	0.117	1.20	<DL	0.0786
7655 (M11)	Total	Marine	01-Dec-11	24.15	40.22	02-Dec-11	24.15	40.22	4.33	0.606	0.0808	3.70	0.0174	0.471	0.0493	0.744	0.0261	0.0569
7697 (M12)	Total	Marine	02-Dec-11	24.15	40.22	03-Dec-11	22.70	36.73	3.08	0.516	0.215	1.34	0.0463	0.729	0.175	0.247	0.0262	0.0176
7791 (A9)	Total	N.Afr	03-Dec-11	22.70	36.73	04-Dec-11	22.36	35.87	638	50.5	8.95	494	0.179	1.23	0.457	1.05	0.0776	0.461
7860 (A10)	Total	N.Afr	04-Dec-11	22.36	35.87	05-Dec-11	22.38	35.87	2820 (28)	237 (1)	40.5 (0.3)	2250 (7)	0.909 (0.049)	3.04 (0.002)	1.64 (0.04)	5.16 (0.30)	0.0606 (0.0357)	1.59 (0.02)
7899 (A11)	Total	N.Afr	05-Dec-11	22.37	35.62	06-Dec-11	20.88	32.62	806	69.9	11.6	623	0.256	1.30	0.692	2.29	0.0488	0.751
7946 (A12)	Total	N.Afr	06-Dec-11	20.88	32.62	07-Dec-11	19.43	29.38	1950	153	24.4	1580	0.680	2.45	1.35	3.85	0.0277	1.08
8004 (A13)	Total	N.Afr	07-Dec-11	19.43	29.38	08-Dec-11	19.43	29.38	3140	243	42.8	2530	1.10	3.14	1.98	5.59	0.0284	1.59
8044 (A14)	Total	N.Afr	08-Dec-11	19.43	29.38	09-Dec-11	18.13	26.13	4610 (127)	377 (4)	62.8 (1.9)	3600 (120)	1.58 (0.11)	5.12 (0.60)	3.98 (0.81)	7.91 (0.05)	0.0408	2.56 (0.01)
8045 (A15)	Total	N.Afr	09-Dec-11	17.75	25.31	09-Dec-11	17.40	24.53	7485 (191)	599 (11)	104 (2)	5650 (162)	2.53 (0.10)	7.15 (0.055)	4.22 (0.29)	8.75 (0.10)	0.275 (0.324)	3.74 (0.44)
geoa1 (G1)	Total	High Lat	19-May-14	40.33	10.04	20-May-14	40.33	10.04	9.03 (0.12)	0.885 (0.392)	0.120 (0.002)	5.08 (0.12)	0.00417 (0.00014)	0.129 (0.005)	0.0567 (0.0264)	0.378 (0.132)	0.00244 (0.00100)	0.0767 (0.0263)
geoa2 (G2)	Total	High Lat	23-May-14	40.33	12.22	24-May-14	41.38	13.89	21.1	1.45	0.291	12.7	0.00613	0.0892	0.0589	0.544	0.00583	0.193
geoa3 (G3)	Total	High Lat	24-May-14	41.38	13.89	25-May-14	41.38	13.89	28.9	1.90	0.423	18.7	0.00908	0.131	0.152	0.828	0.00873	0.258
geoa4 (G4)	Total	High Lat	25-May-14	41.38	13.89	27-May-14	41.38	13.89	23.0	1.44	0.320	14.3	0.00713	0.107	0.0588	0.679	0.00504	0.154
geoa5 (G5)	Total	High Lat	27-May-14	41.38	13.89	29-May-14	43.78	17.03	27.1 (1.5)	1.40 (0.008)	0.316 (0.013)	14.3 (0.9)	0.00897 (0.00369)	0.105 (0.005)	0.110 (0.018)	0.524 (0.134)	0.00834 (0.00299)	0.140 (0.011)
geoa6 (G6)	Total	High Lat	30-May-14	45.05	18.51	02-Jun-14	47.29	20.26	11.3	0.544	0.136	6.27	0.00434	0.164	0.0402	0.801	0.00130	0.0483
geoa7 (G7)	Total	High Lat	02-Jun-14	47.29	20.26	04-Jun-14	50.28	22.60	17.9	0.682	0.130	6.01	0.00275	0.0546	0.0415	0.767	0.001971	0.0611
geoa8 (G8)	Total	High Lat	04-Jun-14	50.28	22.60	06-Jun-14	53.42	25.07	12.2	0.666	0.139	6.37	0.00406	0.0734	0.0356	0.496	0.001741	0.0734
geoa9 (G9)	Total	High Lat	06-Jun-14	53.42	25.07	08-Jun-14	55.51	26.71	21.5 (2.0)	0.586 (0.082)	0.179 (0.018)	7.39 (0.81)	0.00664 (0.00039)	0.175 (0.010)	0.156 (0.029)	1.81 (0.55)	0.00578 (0.00140)	0.252 (0.009)
geoa10 (G10)	Total	High Lat	08-Jun-14	55.51	26.71	10-Jun-14	58.21	29.72	14.5	1.12	0.191	8.95	0.00515	0.0817	0.115	0.400	0.002883	0.192
geoa11 (G11)	Total	High Lat	10-Jun-14	58.21	29.72	12-Jun-14	59.20	34.78	15.0	0.350	0.0990	4.99	0.00215	0.0369	0.0465	2.37	0.00139	0.0514
geoa12 (G12)	Total	High Lat	12-Jun-14	59.20	34.78	14-Jun-14	59.62	38.95	1.96	0.131	0.0129	0.674	0.00125	0.274	0.0150	0.0930	0.000622	0.0107
geoa13 (G13)	Total	High Lat	14-Jun-14	59.62	38.95	16-Jun-14	59.80	42.00	5.65 (0.92)	0.135 (0.059)	0.0139 (0.0047)	0.681 (0.193)	0.00056 (0.00014)	0.0512 (0.0136)	0.0296 (0.0061)	0.219 (0.034)	0.00044 (0.00007)	0.0439 (0.0196)
geoa14 (G14)	Total	High Lat	16-Jun-14	59.80	42.00	18-Jun-14	59.80	41.99	2.95	0.0817	0.0301	0.568	0.0007376	0.0368	0.0427	0.394	0.000777	0.259

geoa15 (G15)	Total	High Lat	18-Jun-14	59.70	42.53	20-Jun-14	59.07	46.09	1.56	0.0716	0.0203	1.40	0.00143	0.0446	0.0799	0.124	0.001589	0.0615
geoa16 (G16)	Total	High Lat	21-Jun-14	56.91	47.42	23-Jun-14	55.72	48.17	1.21	0.00843	0.005041	0.185	0.0002119	0.00750	0.00988	0.0844	0.000147	0.465
geoa17 (G17)	Total	High Lat	23-Jun-14	55.72	48.17	25-Jun-14	52.93	51.39	21.1 (2.4)	0.952 (0.113)	0.337 (0.032)	9.71 (0.942)	0.00509 (0.00068)	0.0559 (0.0065)	0.0691 (0.0070)	0.435 (0.064)	0.00547 (0.00030)	0.137 (0.014)
geoa18 (G18)	Total	High Lat	25-Jun-14	52.93	51.39	27-Jun-14	51.99	53.84	6.38	0.302	0.135	3.14	0.0002239	<DL	0.0193	<DL	0.000507	0.0268

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

25

26

Table S3

28

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
	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

29

30 **Table S4**

31

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

32

33

34 **Table S5****AI N American**

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

AI Marine

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

AI N African

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

Adjusted R Square	0.078
Standard Error	11.420
Observations	8

Mn N American

<i>Regression Statistics</i>		<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>	
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

<i>Regression Statistics</i>		<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>	
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

<i>Regression Statistics</i>		<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>	
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

<i>Regression Statistics</i>		<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>	
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

<i>Regression Statistics</i>		<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
------------------------------	--	---------------------	-----------------------	---------------	----------------	------------------	------------------

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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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

<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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								
<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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								
<i>Regression Statistics</i>			<i>Coefficients</i>	<i>Standard Error</i>	<i>t Stat</i>	<i>P-value</i>	<i>Lower 95%</i>	<i>Upper 95%</i>
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							

35

36

37

38

39 Captions for supplementary material

40

41 Figure S1. Air mass 5-day back trajectory simulations, with arrival heights of 50, 500 and 1500 m. The first four
 42 are representative simulations for N. American, European, Marine and North African samples, redrawn from the
 43 Supplemental Material of Shelley et al. (2015). The other simulations are reproduced from Shelley et al. (2017).
 44 Note that the labelling system has changed slightly from these earlier publications. In Shelley et al. (2017) the
 45 simulations were labelled geo1-18, and here they are G1-18. The simulations in Shelley et al. (2015) were not
 46 labelled with their sample number. For reference, the sample numbers can also be found in Table S1.

47

48 Figure S2. Positive Matrix Factorisation outputs using total TE concentration data: (a) Factor fingerprint and (b)
 49 factor time series, for total aerosol TEs for cruises GA03-2010 (E1 – A8), GA03-2011 (N1 – A15), and GA01 (G1
 50 – 17), where E = European, M = Marine, N = North American, A = North African and G = High Latitude aerosols,
 51 Factor 1 represents a mineral dust source (red, highest contributions from Al, Ti, Fe, Zr, and Ba), and Factor 2 a
 52 pollution source (blue, highest contributions from Ni, Cu, Zn, Cd, and Pb). Note that all of the variance for aerosol

53 Cd is explained by the pollution factor. The factor time series indicates when each factor was dominant. The y-axes
54 of the factor time series represent log normalised data. These figures were produced using the US Environmental
55 Protection Agency's Positive Matrix Factorisation model, EPA PMF v. 5.0

56

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

60

61

62 Table S1. Metadata and concentrations of TEs from (a) UHP water, (b) seawater, (c) 25% acetic acid leaches and
63 (d) total TEs from Shelley et al. (2015; 2017). Note that (1) replicate leaches were not performed on all samples, the
64 values in brackets in the data columns are the standard deviation of replicate samples ($n = 3$), (2) a different
65 labelling convention was used in Shelley et al. (2017) to refer to the GA01 samples. Here we use G1-18 to refer to
66 the samples collected during GA01 (A1-18 in Shelley et al., 2017), and A1-15 to refer to the North African samples
67 from GA03; these labels can be found in brackets after the GEOTRACES sample number in the first column, and
68 (4) matrix blanks and filter blanks were calculated by dividing the concentration of TE (ng) per leach by 100 m^3
69 (the average volume of air filtered across each filter)

70

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

73

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

76

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

79

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

83

84 **Additional references**

85 Huang, S., K. A. Rahn, R. Arimoto, W. C. Graustein, and K. K. Turekian Semiannual cycles of pollution at
86 Bermuda, *J. Geophys. Res.*, 104, 309-317, 10.1029/1999JD900801, 1999.

87 Spokes, L., Jickells, T. and Jarvis, K.: Atmospheric inputs of trace metals to the northeast Atlantic Ocean: the
88 importance of southeasterly flow., *Marine Chemistry.*, 76, 319-330., 2001.