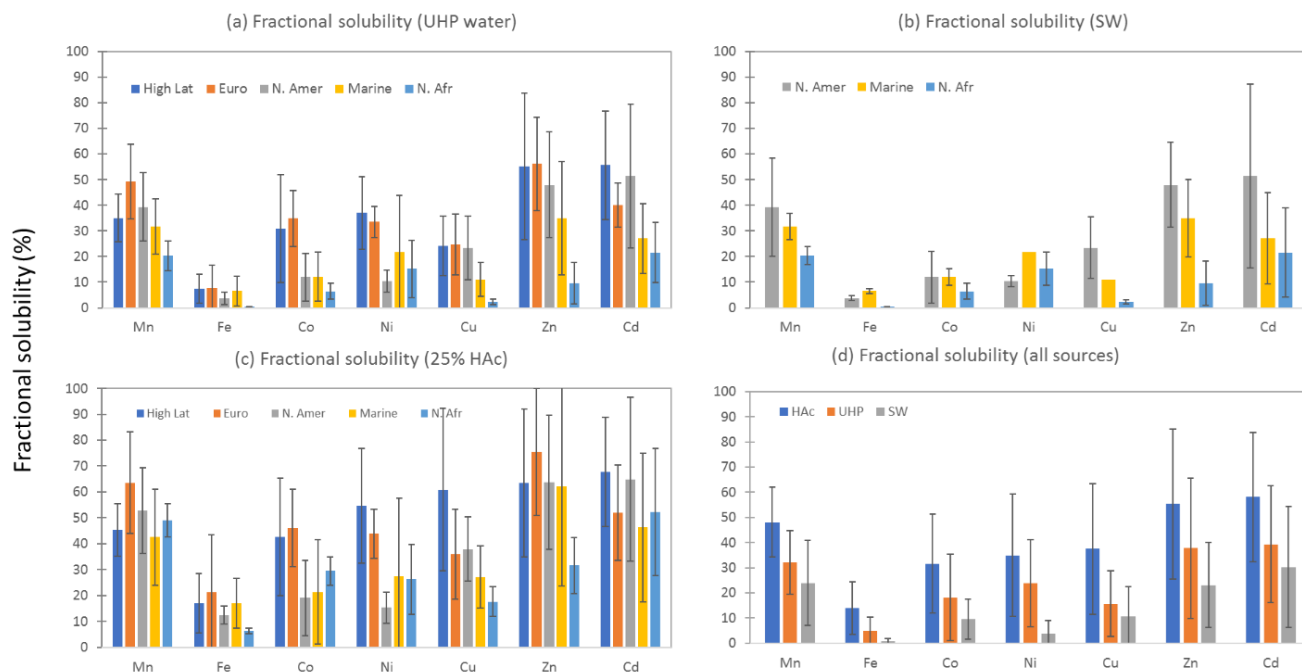


1 **Supplementary Material for: Shelley et al. Characterisation of aerosol provenance from the**
 2 **fractional solubility of Fe (Al, Ti, Mn, Co, Ni, Cu, Zn, Cd and Pb) in North Atlantic aerosols**
 3 **(GEOTRACES cruises GA01 and GA03) using a two stage leach**
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7 **Figure S1.**

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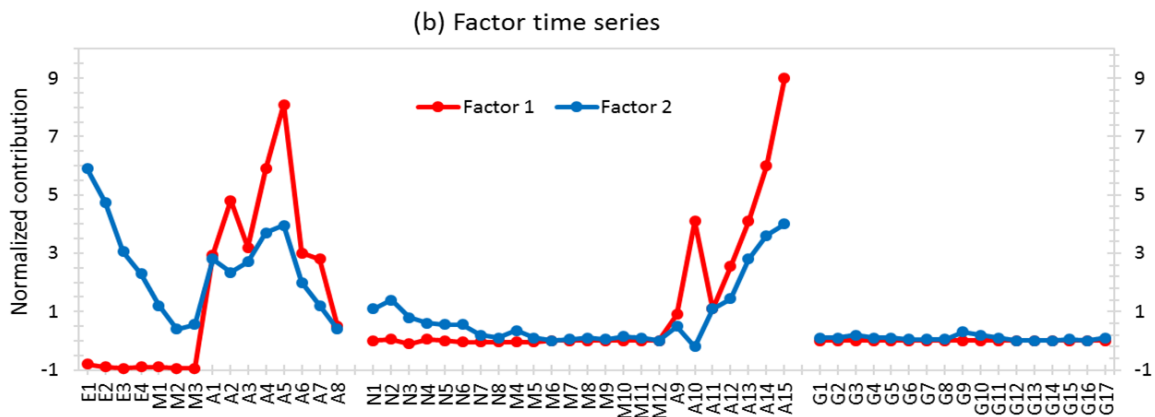
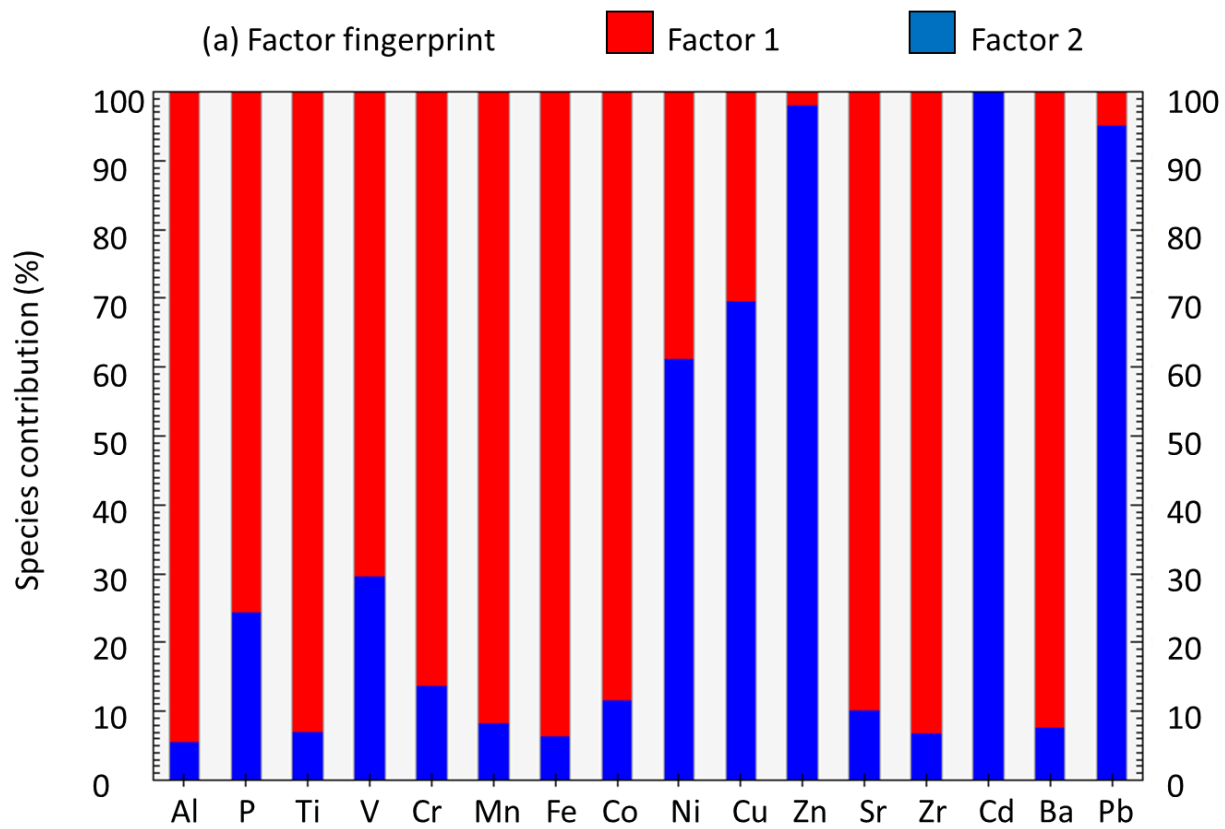


Figure S2

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Table S1

(a) UHP water leach									Na	Al	Ti	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb	Cr	NO ₃ ⁻	SO ₄ ²⁻		
UHP water matrix blank	ng/m ³	(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	7.312E-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	3.762E-06	9.1E-06	3.3E-06			2.7E-21
			<DL = below detection																					
Filter Blank	ng/m ³	(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/m ³	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	Cr µg/L	NO ₃ ⁻ µg/L	SO ₄ ²⁻ µg/L		
5037	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	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	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	UHP water	Euro	19-Oct-10	35.20	16.00	20-Oct-10	34.09	17.57	744	1.89	0.01	0.37	1.43	0.0118	0.483	0.280	1.82	0.0171	0.485	530	1686	971		
5330	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	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	UHP water	Marine	23-Oct-10	27.53	22.01	25-Oct-10	24.01	22.00	4013	0.157	0.0281	0.0847	0.23	0.0061	0.288	0.0751	0.671	0.00550	0.142	853	244	254		
5729	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	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	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	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	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	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	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	UHP water	N. Afr.	03-Nov-10	17.40	24.50	03-Nov-10	17.34	24.55	5105	3.46	0.133	1.29	1.13	0.0157	0.153	0.00413	0.21	0.00359	0.0127	3232	2272	1798		
6250	UHP water	N. Amer.	07-Nov-11	39.71	69.87	08-Nov-11	39.70	69.80	1330	0.848	0.102	0.24	0.82	0.00326	0.106	0.142	0.98	0.0110	0.206	2073	1059	943		
6319	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	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	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	UHP water	N. Amer.	13-Nov-11	38.36	68.86	15-Nov-11	37.58	68.40	3307	1.45	0.132	0.241	0.988	0.00253	0.0517	0.0527	0.54	0.0148	0.144	7228	686	1491		
6788	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	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	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	UHP water	Marine	21-Nov-11	31.53	63.37	24-Nov-11	29.70	56.82	1018	0.291	0.0356	0.0402	0.164	0.00100	0.0325	0.00847	0.17	0.0038	0.0349	1728	211	426
7245	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	UHP water	Marine	26-Nov-11	27.58	49.63	27-Nov-11	27.55	49.55	980	0.0829	0.0366	0.0112	0.0395	0.00097	0.0521	0.00897	0.08	0.00225	0.00410	1886	165	457
7357	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	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	UHP water	Marine	29-Nov-11	26.14	44.83	30-Nov-11	25.58	43.60	1205	1.15	0.0331	0.0946	0.355	0.00228	0.0324	0.00873	0.12	0.00445	0.0115	2336	202	452
7587	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	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	UHP water	Marine	02-Dec-11	24.15	40.22	03-Dec-11	22.70	36.73	1435	0.53	0.0421	0.0167	0.101	0.00095	0.0282	0.0182	0.24	0.0114	0.00701	2789	168	475
7791	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	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	UHP water	N. Afr.	05-Dec-11	22.37	35.62	06-Dec-11	20.88	32.62	2410	7.36	0.148	3.88	2.57	0.0367	0.1092	0.0331	0.56	0.00585	0.0523	4734	914	1208
7946	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	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	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	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	UHP water	High Lat	19-May-14	40.33	10.04	20-May-14	40.33	10.04	ND	0.0959	0.000692	0.0345	0.146	0.00099	0.0470	0.0050	0.138	0.00108	0.00630	22380	708	4730
geoa2	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	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	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	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	UHP water	High Lat	30-May-14	45.05	18.51	02-Jun-14	47.29	20.26	ND	0.989	0.000221	0.0588	0.323	0.00154	0.0945	0.0174	0.254	0.00093	0.0145	24218	2764	10273
geoa7	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	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	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	1.447	0.00308	0.0848	0.0377	1.373	0.00323	0.0660	66497	3550	22961
geoa10	UHP water	High Lat	08-Jun-14	55.51	26.71	10-Jun-14	58.21	29.72	ND	1.50	0.00939	0.0686	1.044	0.00157	0.0254	0.0258	0.295	0.00164	0.0449	5362	783	11314
geoa11	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	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	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	UHP water	High Lat	16-Jun-14	59.80	42.00	18-Jun-14	59.80	41.99	ND	0.788	0.000364	0.00960	0.071	0.00047	0.0186	0.00363	0.174	0.000191	0.00418	10671	<DL	2896
geoa15	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	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	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	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	Start Long	End date	End lat	End long	Na	Al	Ti	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
				(N)	(W)		(N)	(W)											
6070	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	Seawater	N. Amer.	07-Nov-11	39.71	69.87	08-Nov-11	39.70	69.80	ND	0.931	ND	0.296	0.346	0.00443	0.0236	0.133	1.11	0.0284	0.317
6319	Seawater	N. Amer.	08-Nov-11	39.70	69.80	09-Nov-11	39.35	69.54	ND	2.52	ND	1.07	1.24	0.0158	0.0622	0.548	3.04	0.0257	0.651
6452	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	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	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	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	Seawater	Marine	19-Nov-11	32.02	64.44	20-Nov-11	31.75	64.17	ND	0.0442	ND	<DL	0.0218	0.000633	<DL	0.0120	0.183	0.00683	0.0698
6964	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	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	Seawater	Marine	24-Nov-11	29.70	56.82	26-Nov-11	27.58	49.63	ND	<DL	ND	<DL	0.0238	0.000221	<DL	<DL	0.0649	0.00563	0.0173
7316	Seawater	Marine	26-Nov-11	27.58	49.63	27-Nov-11	27.55	49.55	ND	<DL	ND	<DL	0.00076	0.000435	<DL	<DL	0.0715	0.00195	0.0101
7357	Seawater	Marine	27-Nov-11	27.55	49.55	28-Nov-11	26.31	45.39	ND	0.176	ND	<DL	0.0485	0.00050	<DL	<DL	0.0661	0.00377	0.0285
7426	Seawater	Marine	28-Nov-11	26.28	45.30	29-Nov-11	26.14	44.83	ND	<DL	ND	<DL	0.0119	0.000249	<DL	<DL	0.0644	0.00098	0.0174
7509	Seawater	Marine	29-Nov-11	26.14	44.83	30-Nov-11	25.58	43.60	ND	0.196	ND	0.0206	0.0562	0.00234	<DL	<DL	0.0813	0.00474	0.0227
7587	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	Seawater	Marine	01-Dec-11	24.15	40.22	02-Dec-11	24.15	40.22	ND	0.477	ND	<DL	0.0124	0.000772	<DL	<DL	0.104	0.00261	0.0235
7697	Seawater	Marine	02-Dec-11	24.15	40.22	03-Dec-11	22.70	36.73	ND	<DL	ND	<DL	<DL	0.000152	<DL	<DL	0.0992	0.000857	0.00990
7791	Seawater	N. Afr.	03-Dec-11	22.70	36.73	04-Dec-11	22.36	35.87	ND	1.94	ND	1.45	0.259	0.0183	0.0516	0.00243	0.260	0.00863	0.0663
7860	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	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	Seawater	N. Afr.	06-Dec-11	20.88	32.62	07-Dec-11	19.43	29.38	ND	3.74	ND	3.28	0.336	0.0357	0.0275	0.0211	0.243	0.00934	0.0438
8004	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	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	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

25% HAc matrix blank	ng/m3	(n=3)							Na	Al	Ti	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
									ND	0.00210	0.0025746	9.6E-05	0.00052	2.87E-05	1.9E-04	8.1E-04	5.3E-04	1.53E-05	6.128E-05

DL (3xSD) ND 0.00027 0.00020 9.3E-05 0.000118 2.93E-06 9.2E-05 1.5E-04 2.4E-04 5.68E-06 3.344E-05

Filter blank ng/m3 (n=3) ND 0.00655094 0.0005754 0.00013 0.00239 3.18E-05 2.4E-04 8.1E-04 2.2E-03 1.46E-05 4.837E-05

SD ND 0.00196684 6.399E-05 9.6E-06 0.00017 4.6E-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	Start Long	End date	End lat	End long	Na	Al	Ti	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
				(N)	(W)		(N)	(W)		ng/m3	ng/m3	ng/m3	ng/m3	ng/m3	ng/m3	ng/m3	ng/m3	ng/m3	ng/m3
5037	25% HAc	Euro	15-Oct-10	38.32	9.66	16-Oct-10	38.33	9.67	ND	3.40	0.0707	0.268	5.82	0.00739	0.206	0.255	1.45	0.008977	0.781
5145	25% HAc	Euro	16-Oct-10	38.33	9.66	17-Oct-10	38.33	9.66	ND	5.14	0.00890	0.502	10.2	0.0142	0.344	0.509	3.07	0.0341	1.30
5197	25% HAc	Euro	17-Oct-10	38.33	9.66	19-Oct-10	35.20	16.00	ND	3.56	0.00415	0.220	4.49	0.00685	0.212	0.466	1.56	0.006912	0.543
5299	25% HAc	Euro	19-Oct-10	35.20	16.00	20-Oct-10	34.09	17.57	ND	1.48	0.00250	0.114	2.30	0.00357	0.134	0.0822	0.935	0.00410	0.365
5330	25% HAc	Marine	20-Oct-10	34.07	17.61	21-Oct-10	31.32	21.55	ND	2.71	0.00516	0.0426	2.08	0.00736	0.124	0.168	0.832	0.0160	0.417
5425	25% HAc	Marine	21-Oct-10	31.11	21.86	23-Oct-10	27.53	22.01	ND	0.957	0.00370	0.0132	0.590	0.000421	0.0271	0.0418	0.762	0.000827	0.0679
5579	25% HAc	Marine	23-Oct-10	27.53	22.01	25-Oct-10	24.01	22.00	ND	0.938	0.00507	0.0319	0.663	0.00285	0.0697	0.0179	0.505	0.002016	0.126
5729	25% HAc	N.Afr	25-Oct-10	24.01	22.00	28-Oct-10	17.35	18.25	ND	216	0.393	8.25	105	0.183	0.350	0.306	1.78	0.0119	1.14
5738	25% HAc	N.Afr	28-Oct-10	17.36	18.25	29-Oct-10	17.36	18.25	ND	253	0.448	11.4	126	0.236	0.444	0.304	1.83	0.0119	1.10
5764	25% HAc	N.Afr	29-Oct-10	17.36	18.25	29-Oct-10	17.36	19.06	ND	181	0.468	7.64	94.1	0.150	0.354	0.240	0.850	0.003776	0.476
5848	25% HAc	N.Afr	30-Oct-10	17.35	20.02	30-Oct-10	17.35	20.83	ND	491	0.934	15.7	256	0.365	0.641	0.854	2.51	0.0134	1.38
5870	25% HAc	N.Afr	31-Oct-10	17.36	20.83	31-Oct-10	17.36	20.86	ND	521	0.642	18.5	260	0.399	0.717	0.575	1.68	0.00951	0.906
5965	25% HAc	N.Afr	01-Nov-10	17.35	20.91	01-Nov-10	17.35	22.79	ND	188	0.352	5.62	99.0	0.125	0.302	0.350	0.985	0.007048	1.48
5999	25% HAc	N.Afr	01-Nov-10	17.35	22.79	01-Nov-10	17.35	23.07	ND	97.52	0.226	3.05	52.0	0.0660	0.180	0.198	0.601	0.005041	0.333
6070	25% HAc	N.Afr	03-Nov-10	17.40	24.50	03-Nov-10	17.34	24.55	ND	26.10	0.0493	0.805	14.5	0.0185	0.0519	0.0156	0.564	0.002464	0.0757
6250	25% HAc	N.Amer	07-Nov-11	39.71	69.87	08-Nov-11	39.70	69.80	ND	1.21	0.0374	0.120	2.51	0.00343	0.0437	0.0866	0.456	0.0119	0.221
6319	25% HAc	N.Amer	08-Nov-11	39.70	69.80	09-Nov-11	39.35	69.54	ND	3.96	0.0145	0.286	7.10	0.00714	0.0453	0.187	0.634	0.003988	0.371
6452	25% HAc	N.Amer	09-Nov-11	39.35	69.54	11-Nov-11	38.65	69.20	ND	1.25	0.0109	0.0486	2.26	0.00220	0.0426	0.0766	0.384	0.0107	0.102
6546	25% HAc	N.Amer	12-Nov-11	38.33	68.87	13-Nov-11	38.36	68.86	ND	2.68	0.00815	0.189	3.75	0.00255	0.0430	0.0696	0.270	0.00620	0.226
6677	25% HAc	N.Amer	13-Nov-11	38.36	68.86	15-Nov-11	37.58	68.40	ND	1.14	0.0170	0.0641	1.26	0.000994	0.0168	0.0355	0.173	0.003178	0.137
6788	25% HAc	N.Amer	15-Nov-11	37.62	68.38	17-Nov-11	37.62	68.38	ND	1.76	0.0406	0.0342	0.891	0.000664	0.0155	0.0790	0.286	0.0136	0.0995
6903	25% HAc	N.Amer	19-Nov-11	32.02	64.44	20-Nov-11	31.75	64.17	ND	0.750	0.00437	0.0190	0.339	0.000994	0.0295	0.0200	0.119	0.004381	0.0576
6964	25% HAc	N.Amer	20-Nov-11	31.75	64.17	21-Nov-11	31.55	63.43	ND	0.516	0.00283	0.00983	0.114	<DL	0.000583	0.00401	0.0745	0.000984	0.0378
7156	25% HAc	Marine	21-Nov-11	31.53	63.37	24-Nov-11	29.70	56.82	ND	1.74	0.0266	0.0221	0.225	0.00219	0.0144	0.0183	0.162	0.003612	0.0761
7245	25% HAc	Marine	24-Nov-11	29.70	56.82	26-Nov-11	27.58	49.63	ND	1.98	0.00449	0.00452	0.208	0.00283	0.00665	0.0140	0.0578	0.001526	0.0201
7316	25% HAc	Marine	26-Nov-11	27.58	49.63	27-Nov-11	27.55	49.55	ND	1.23	0.00273	0.00448	0.0687	0.000821	0.0389	0.00565	0.0206	0.004327	0.0155
7357	25% HAc	Marine	27-Nov-11	27.55	49.55	28-Nov-11	26.31	45.39	ND	3.88	0.0115	0.0171	0.251	0.00136	0.0162	0.0148	0.0409	0.0254	0.0283
7426	25% HAc	Marine	28-Nov-11	26.28	45.30	29-Nov-11	26.14	44.83	ND	1.42	0.00274	0.00660	0.125	0.000426	0.0106	0.00818	0.128	0.002025	0.0178
7509	25% HAc	Marine	29-Nov-11	26.14	44.83	30-Nov-11	25.58	43.60	ND	3.39	0.0104	0.0406	0.649	0.00155	0.00852	0.0240	0.268	0.00326	0.0241
7587	25% HAc	Marine	30-Nov-11	25.55	43.54	01-Dec-11	24.15	40.22	ND	6.21	0.0440	0.0429	0.800	0.00264	0.0175	0.0367	0.185	0.002694	0.0452
7655	25% HAc	Marine	01-Dec-11	24.15	40.22	02-Dec-11	24.15	40.22	ND	1.03	0.0186	0.0113	0.268	0.000371	0.00650	0.0128	0.0085	0.001155	0.0254
7697	25% HAc	Marine	02-Dec-11	24.15	40.22	03-Dec-11	22.70	36.73	ND	2.26	0.00663	0.0100	0.321	0.000418	0.0169	0.0247	0.521	0.0264	0.0181
7791	25% HAc	N.Afr	03-Dec-11	22.70	36.73	04-Dec-11	22.36	35.87	ND	60.13	0.133	2.25	29.9	0.0418	0.0722	0.0696	0.248	0.0389	0.174

7860	25% HAc	N.Afr	04-Dec-11	22.36	35.87	05-Dec-11	22.38	35.87	ND	356	0.572	13.9	176	0.264	0.279	0.300	1.03	0.059599	0.569
7899	25% HAc	N.Afr	05-Dec-11	22.37	35.62	06-Dec-11	20.88	32.62	ND	87.76	0.220	2.45	45.4	0.0495	0.0837	0.0922	0.362	0.002221	0.284
7946	25% HAc	N.Afr	06-Dec-11	20.88	32.62	07-Dec-11	19.43	29.38	ND	213	0.274	9.33	101	0.190	0.196	0.294	0.974	0.00851	0.447
8004	25% HAc	N.Afr	07-Dec-11	19.43	29.38	08-Dec-11	19.43	29.38	ND	419	0.539	19.0	195	0.365	0.356	0.341	1.43	0.0184	0.629
8044	25% HAc	N.Afr	08-Dec-11	19.43	29.38	09-Dec-11	18.13	26.13	ND	528	0.699	26.6	238	0.559	0.509	0.769	1.78	0.0168	0.887
8045	25% HAc	N.Afr	09-Dec-11	17.75	25.31	09-Dec-11	17.40	24.53	ND	604	1.11	42.2	282	0.825	0.577	0.526	1.36	0.0147	0.901
geoa1	25% HAc	High Lat	19-May-14	40.33	10.04	20-May-14	40.33	10.04	ND	0.709	0.00396	0.0160	0.416	0.00068	0.0299	0.0567	0.0685	0.00044	0.0305
geoa2	25% HAc	High Lat	23-May-14	40.33	12.22	24-May-14	41.38	13.89	ND	1.10	0.0201	0.0296	1.02	0.001162	0.0173	0.0165	0.0697	0.00075	0.0720952
geoa3	25% HAc	High Lat	24-May-14	41.38	13.89	25-May-14	41.38	13.89	ND	1.04	0.0311	0.0428	1.36	0.00093	0.0242	0.0128	0.056103	0.00086	0.163
geoa4	25% HAc	High Lat	25-May-14	41.38	13.89	27-May-14	41.38	13.89	ND	0.996	0.0156	0.0277	1.03	0.00070	0.0129	0.017503	0.0858	0.000415	0.0605
geoa5	25% HAc	High Lat	27-May-14	41.38	13.89	29-May-14	43.78	17.03	ND	2.02	0.0128	0.0587	1.30	0.00130	0.0268	0.0205	0.109	0.00120	0.0570
geoa6	25% HAc	High Lat	30-May-14	45.05	18.51	02-Jun-14	47.29	20.26	ND	1.01	0.00957	0.0136	0.598	0.00058	0.0355	0.0256	0.0547	0.00029	0.0224
geoa7	25% HAc	High Lat	02-Jun-14	47.29	20.26	04-Jun-14	50.28	22.60	ND	1.49	0.00721	0.0174	0.592	0.00067	0.0566	0.0283	0.0605	0.00023	0.0244
geoa8	25% HAc	High Lat	04-Jun-14	50.28	22.60	06-Jun-14	53.42	25.07	ND	1.30	0.00683	0.0142	0.942	0.00029	0.0105	0.0292	0.143	0.000144	0.0485
geoa9	25% HAc	High Lat	06-Jun-14	53.42	25.07	08-Jun-14	55.51	26.71	ND	2.81	0.00768	0.0131	0.892	0.00040	0.0151	0.0259	0.0885	0.001178	0.0664
geoa10	25% HAc	High Lat	08-Jun-14	55.51	26.71	10-Jun-14	58.21	29.72	ND	1.47	0.0204	0.0151	0.573	0.00048	0.0101	0.0302	0.178	0.000313	0.0540
geoa11	25% HAc	High Lat	10-Jun-14	58.21	29.72	12-Jun-14	59.20	34.78	ND	0.876	0.0117	0.00321	0.319	8.1E-05	0.002495	0.0263	0.0736	7.65E-05	0.0144
geoa12	25% HAc	High Lat	12-Jun-14	59.20	34.78	14-Jun-14	59.62	38.95	ND	0.332	0.00373	<DL	0.0651	3.0E-05	0.001266	0.0120	<DL	7.94E-05	0.0076711
geoa13	25% HAc	High Lat	14-Jun-14	59.62	38.95	16-Jun-14	59.80	42.00	ND	0.489	<DL	<DL	0.0290	2.5E-05	0.006757	0.00160	<DL	4.91E-05	0.0531
geoa14	25% HAc	High Lat	16-Jun-14	59.80	42.00	18-Jun-14	59.80	41.99	ND	0.753	0.00282	0.00253	0.244	2.5E-04	0.0106	0.012401	0.0668	8.93E-05	0.0220
geoa15	25% HAc	High Lat	18-Jun-14	59.70	42.53	20-Jun-14	59.07	46.09	ND	0.321	0.00251	0.00132	0.0711	8.7E-05	0.008157	0.005391	<DL	9.88E-05	0.0232
geoa16	25% HAc	High Lat	21-Jun-14	56.91	47.42	23-Jun-14	55.72	48.17	ND	0.0536	<DL	<DL	<DL	4.2E-05	<DL	0.001864	<DL	7.91E-06	0.00357
geoa17	25% HAc	High Lat	23-Jun-14	55.72	48.17	25-Jun-14	52.93	51.39	ND	1.93	0.0148	0.0410	0.715	0.00043	0.004884	0.0117	0.007572	0.00060	0.0444005
geoa18	25% HAc	High Lat	25-Jun-14	52.93	51.39	27-Jun-14	51.99	53.84	ND	0.911	0.00213	0.0616	0.161	0.00016	0.000959	0.00966	0.00870	9.25E-05	0.0225

Note: Matrix blanks and filter blanks are calculated by dividing the concentration of TE (ng) per leach by 100 m³ (the average filtered across each filter)

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Table S2

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

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Table S3

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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

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Captions for supplementary material

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34 **Figure S1. Fractional solubility of bioactive aerosol TEs (average ± 1 SD) following leaches with (a) UHP**
35 **water, (b) seawater, and (c) 25 % acetic acid, and (d) a comparison of fractional solubility using the three**
36 **leach media**

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38 **Figure S2. Positive Matrix Factorisation outputs: (a) Factor fingerprint and (b) factor time series, for total**
39 **aerosol TEs for cruises GA03-2010 (E1 – A8), GA03-2011 (N1 – A15), and GA01 (G1 – 17), where E =**
40 **European, M = Marine, N = North American, A = North African and G = High Latitude aerosols, Factor 1**
41 **represents a mineral dust source (red, highest contributions from Al, Fe and Zr), and Factor 2 a pollution**
42 **source (blue, highest contributions from Cd, Zn and Pb). Note that all of the variance for aerosol Cd is**
43 **explained by the pollution factor. The factor time series indicates which factor when each factor was**

44 **dominant. The y-axes of the factor time series represent log normalised data. These figures were produced**
45 **using the US Environmental Protection Agency's Positive Matrix Factorisation model, EPA PMF v. 5.0**

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49 **Table S1. Metadata and concentrations of TEs from (a) UHP water, (b) seawater, and (c) 25% acetic acid**
50 **leaches**

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52 **Table S2. Fractional solubility of bioactive TEs (± 1 SD), corresponding to Figures S1(a-c)**

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54 **Table S3. Fractional solubility of bioactive TEs (± 1 SD), corresponding to Figure S1(d)**

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