atmosphere

Supplementary Material for Chemical Composition of PM_{10} in 16 Urban, Industrial and Background Sites in Italy†

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Table S1. Latitude and longitude of the sampling sites and mean temperature during the sampling periods.

Location	Sampling Site		24-h Mean Temperature (°C)			
			Cold Periods		Warm Periods	
City/Village	Latitude	Longitude	Min	Max	Min	Max
Chamonix (F) CH	45°55'21" N	06°52'07" E	−2	9	11	19
Monfalcone MF	45°48'22" N	13°31'57" E	1	11	18	27
Ferrara FE	44°50'56" N	11°33'53" E	1	12	16	32
Rubbiano RB	44°40'58" N	10°04'46" E	5	10	19	29
La Spezia LS	44°06'23" N	09°50'00" E	-	-	23	27
Civitavecchia CV	42°06'35" N	11°46'21" E	-	-	17	21
M.rotondo sc. MR	42°06'21" N	12°38'24" E	5	13	21	31
Guidonia GU	41°59'38" N	12°43'27" E	8	18	20	25
Rome RM	41°54'08" N	12°31'01" E	5	16	20	26
Fontechiari FC	41°40'09" N	13°40'31" E	2	17	19	34
Frosinone FR	41°37'00" N	13°16'52" E	5	15	12	20
Brindisi BR	40°33'54" N	18° 1'46" E	9	17	-	-
Viggiano VG	40°20'05" N	15°54'02" E	11	16	18	26
Strongoli m. ST	39°14'51" N	17°06'16" E	6	15	21	26
Palermo PA	38°07'01" N	13°21'49" E	-	-	14	22
Gela GE	37°04'14" N	14°14'32" E	14	18	22	26

SM1. Rationale for the choice of conversion factors

To check the correspondence between the mass of PM_{10} directly determined by gravimetry or beta attenuation and the mass reconstructed from PM components, it is necessary to take into account all PM macro-components, that is the chemical species that typically constitute more than 1% of the PM mass. In the case of PM_{10}, macro-components include a few elements (Al, Si, Fe, Na, K, Mg, Ca), inorganic ions, elemental carbon and the whole of organic species (no individual organic species can be generally considered as a macro-component). Moreover, in the case of elements, which are generally present in the form of oxides, it is necessary to account for oxygen, and, in the case of organic species, we need to take into account non-C atoms, which are not determined by the thermo-optical analysis.

In the case of elements, we considered the following oxides: Al_2O_3, SiO_2, Fe_2O_3, Na_2O, K_2O, MgO and CaO and applied the following conversion factors: Al 1.89; Si 2.14; Fe 1.42; Na_i 1.35; K_i 1.2; Mg_i 1.67; Ca_i 1.4. In the case of Na, K, Mg and Ca, we applied the conversion factor to the insoluble fraction only (indicated by the subscript "i"), calculated by subtracting the IC determination (soluble fraction) from the XRF determination (total amount) [4,9,16,22,25,26].

Carbonate, which is not directly measured, was calculated considering the soluble fractions of calcium and magnesium; these values were multiplied by 1.5 and 2.5, respectively, to account for the carbonate content in $CaCO_3$ and $MgCO_3$ [4,9,16].

To convert organic carbon to organic matter, it is necessary to account for unmeasured H, O, N, and S, which are generally present in organic molecules. The multiplier depends on the extent of organic oxidation and secondary organic aerosol formation. It was found that it may vary from 1.2 in the case of very fresh aerosol (e.g. kerbside) to 2.6 for very aged aerosol [25]. Many studies [e.g. 28, 29] suggest the use of 1.6 for urban traffic sites, 1.8 for urban background and peri-urban sites and 2.1 for remote and agricultural sites. According to this classification, we applied the factor 1.6 to LS, GU, PA, where the sampling locations were sited in urban-traffic areas, 1.8 to CH, MF, RB, CV, MR, RM, FR, BR, VG, ST, GE, which were located in residential or peri-urban areas, 2.1 to FC, which is a regional background site, located in an agricultural area.

Table S2. Loadings of PCA run on the total elemental concentrations.

	PC1	PC2	PC3	PC4	PC5	PC6
Variance %	**44.5**	**15.7**	**8.1**	**5.2**	**3.9**	**3.6**
As	0.23	0.18	0.07	0.35	0.02	0.24
Ba	0.20	−0.22	−0.19	0.15	0.31	−0.18
Bi	0.25	0.18	0.10	0.00	0.05	0.05
Cd	0.20	0.25	0.01	0.27	−0.09	0.02
Ce	0.14	−0.33	−0.28	0.02	0.16	−0.01
Co	0.15	−0.20	0.18	−0.01	−0.15	0.35
Cs	0.23	−0.08	−0.30	−0.01	0.01	0.12
Cu	0.26	0.11	−0.20	−0.20	0.05	0.07
Fe	0.28	0.04	0.11	−0.21	0.12	−0.12
La	0.16	−0.26	−0.25	0.22	0.15	0.05
Li	0.22	−0.17	0.33	−0.17	0.04	0.20
Mg	0.08	−0.32	0.09	−0.08	−0.37	0.45
Mn	0.20	0.05	0.39	−0.23	0.29	−0.25
Mo	0.26	0.20	0.00	0.00	−0.02	0.00
Ni	0.23	−0.14	0.22	0.16	−0.33	−0.26
Pb	0.24	0.27	0.01	0.22	−0.02	0.21
Rb	0.23	0.05	−0.15	−0.21	−0.19	−0.40
Sb	0.23	0.14	−0.25	−0.12	−0.02	0.09
Sn	0.27	0.22	−0.01	0.08	−0.02	0.03
Ti	0.19	−0.26	0.28	−0.27	0.19	0.08
Tl	0.11	−0.01	−0.35	−0.42	−0.40	−0.04
U	0.16	−0.32	−0.13	0.22	0.23	−0.07
V	0.12	−0.27	0.14	0.35	−0.43	−0.39

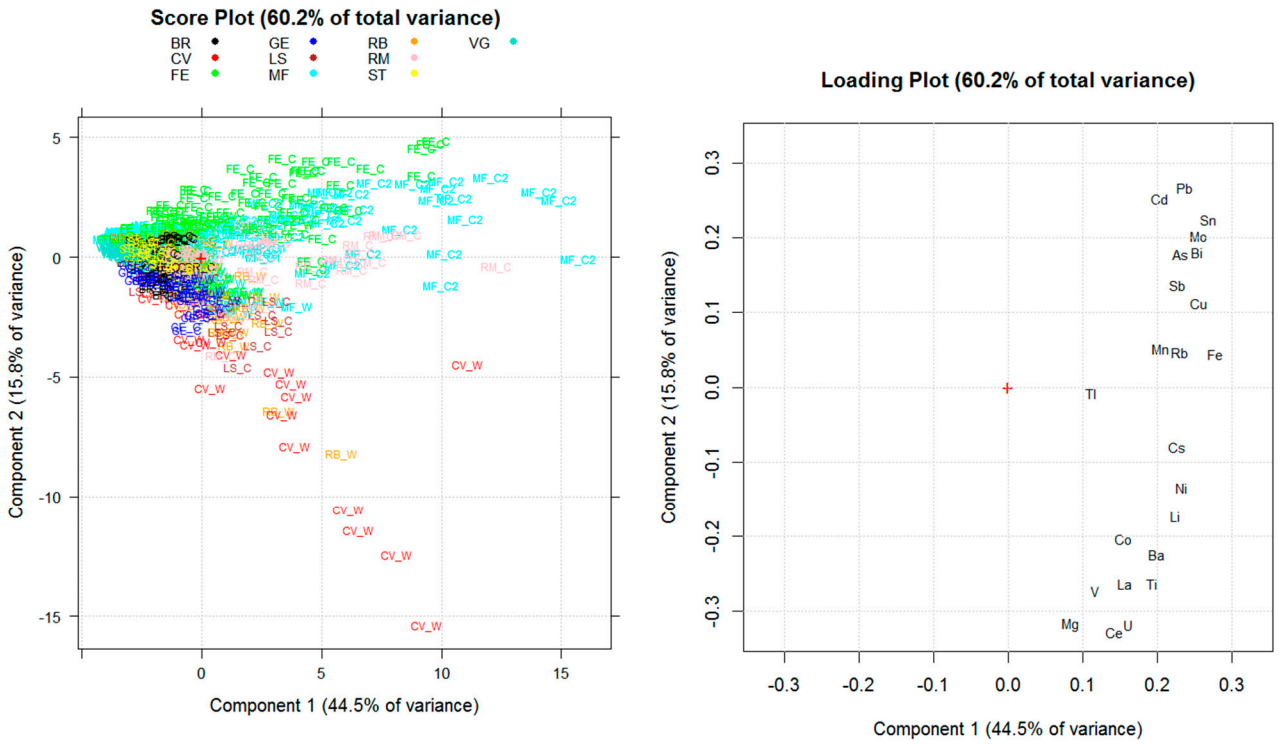

Figure S1. PCA of elemental concentration (all data): score plot (left panel) and loading plot (right panel) of components 1 and 2. _C and _W after the site code indicate the cold and warm periods, respectively. In the case of MF, the two winter periods are reported as _C1 and _C2.

Table S3. Concentration of the micro- and trace elements (ng/m^3) in the soluble fraction during the cold period.

		As	Ba	Bi	Cd	Ce	Co	Cs	Cu	Fe	La	Li	Mg	Mn	Mo	Ni	Pb	Rb	Sb	Sn	Ti	Tl	U	V
MF1 N = 53	**MEAN**	**0.46**	**1.6**	**0.031**	**0.13**	**0.010**	**0.029**	**0.020**	**3.1**	**56**	**0.005**	**0.06**	**40**	**6.4**	**0.22**	**0.56**	**1.5**	**0.7**	**0.4**	**0.25**	**0.34**	**0.020**	**0.002**	**1.1**
	MEDIAN	0.37	1.4	0.023	0.12	0.008	0.025	0.019	2.7	45	0.004	0.05	29	4.0	0.16	0.48	1.0	0.6	0.5	0.15	0.24	0.018	0.002	0.8
	0.1	0.14	0.6	0.004	0.05	0.002	0.011	0.007	1.3	16	0.001	0.02	16	2.1	0.08	0.20	0.3	0.4	0.1	0.03	0.06	0.006	<0.001	0.2
	0.9	1.07	2.6	0.058	0.25	0.021	0.052	0.035	5.3	114	0.011	0.12	86	13	0.46	1.04	3.4	1.1	0.8	0.60	0.64	0.038	0.004	2.5
MF2 N = 54	**MEAN**	**0.72**	**4.0**	**0.075**	**0.37**	**0.018**	**0.086**	**0.041**	**7.9**	**123**	**0.009**	**0.25**	**59**	**21**	**1.33**	**1.65**	**2.8**	**1.4**	**1.1**	**0.86**	**0.72**	**0.039**	**0.005**	**2.8**
	MEDIAN	0.69	3.6	0.059	0.36	0.017	0.075	0.038	6.5	103	0.008	0.20	55	13	1.24	1.43	2.8	1.3	1.0	0.61	0.37	0.039	0.005	2.6
	0.1	0.22	1.3	0.010	0.09	0.005	0.027	0.016	2.4	23	0.002	0.08	21	4.2	0.29	0.33	0.7	0.3	0.3	0.10	0.08	0.015	0.001	0.3
	0.9	1.37	7.0	0.176	0.67	0.030	0.177	0.067	17.0	283	0.017	0.45	103	47	2.92	3.43	5.3	2.7	1.9	1.97	1.91	0.065	0.009	6.0
FE N = 82	**MEAN**	**1.02**	**2.8**	**0.047**	**0.35**	**0.012**	**0.058**	**0.030**	**5.8**	**38**	**0.012**	**0.15**	**72**	**6.1**	**1.02**	**0.64**	**4.0**	**0.7**	**0.9**	**0.32**	**0.10**	**0.035**	**0.002**	**0.7**
	MEDIAN	0.96	2.4	0.034	0.26	0.010	0.044	0.026	4.9	28	0.009	0.12	64	5.6	0.88	0.57	3.3	0.6	0.8	0.20	0.09	0.029	0.002	0.5
	0.1	0.46	0.9	0.012	0.12	0.005	0.023	0.015	2.5	13	0.005	0.08	18	2.5	0.29	0.27	1.5	0.1	0.4	0.06	0.03	0.018	0.001	0.2
	0.9	1.59	5.3	0.096	0.65	0.017	0.095	0.049	10.6	86	0.025	0.23	130	9.8	1.86	1.05	7.6	1.6	1.6	0.82	0.18	0.055	0.004	1.3
RB N = 21	**MEAN**	**0.27**	**1.0**	**0.013**	**0.08**	**0.010**	**0.187**	**0.008**	**2.1**	**18**	**0.005**	**0.05**	**43**	**3.1**	**0.48**	**0.49**	**0.9**	**0.3**	**0.2**	**0.07**	**0.07**	**0.012**	**0.001**	**0.3**
	MEDIAN	0.26	0.8	0.006	0.06	0.005	0.095	0.007	1.9	13	0.003	0.05	36	2.9	0.22	0.38	0.5	0.3	0.1	0.05	0.05	0.010	0.001	0.2
	0.1	0.10	0.4	0.001	0.02	0.002	0.032	0.004	0.8	5	0.001	0.02	10	1.1	0.07	0.14	0.3	0.1	0.1	0.01	0.01	0.004	<0.001	0.0
	0.9	0.36	1.7	0.031	0.16	0.021	0.264	0.013	3.5	44	0.010	0.09	86	6.0	0.74	0.95	2.4	0.6	0.5	0.16	0.16	0.018	0.003	0.6
RM N = 42	**MEAN**	**0.30**	**4.5**	**0.012**	**0.13**	**0.025**	**0.048**	**0.041**	**10.2**	**15**	**0.012**	**0.07**	**115**	**2.8**	**0.47**	**0.53**	**0.6**	**1.5**	**1.1**	**0.16**	**0.12**	**0.080**	**0.001**	**0.5**
	MEDIAN	0.29	4.6	0.007	0.11	0.018	0.030	0.036	8.1	12	0.009	0.06	56	2.1	0.26	0.43	0.5	1.3	0.8	0.09	0.09	0.072	0.001	0.3
	0.1	0.13	2.5	0.003	0.04	0.008	0.012	0.010	3.2	6	0.004	0.03	26	1.0	0.11	0.24	0.1	0.4	0.2	0.05	0.04	0.021	<0.001	0.1
	0.9	0.43	6.5	0.015	0.22	0.045	0.068	0.081	20.7	29	0.021	0.11	301	4.9	0.81	0.79	1.1	2.7	2.8	0.19	0.18	0.154	0.003	0.8
BR N = 42	**MEAN**	**0.19**	**1.0**	**0.034**	**0.08**	**0.035**	**0.031**	**0.012**	**1.4**	**12**	**n.a.**	**0.04**	**87**	**1.8**	**n.a.**	**0.96**	**1.4**	**0.4**	**0.4**	**0.07**	**0.18**	**0.029**	**0.003**	**2.3**
	MEDIAN	0.18	1.0	0.012	0.07	0.032	0.032	0.013	1.3	11	n.a	0.04	69	1.9	n.a	0.95	1.3	0.4	0.3	0.06	0.20	0.023	0.003	2.2
	0.1	0.08	0.5	0.001	0.03	0.021	0.015	0.007	0.7	4	n.a.	0.02	40	0.7	n.a.	0.48	0.6	0.1	0.1	0.01	0.03	0.010	0.001	0.7
	0.9	0.29	1.6	0.085	0.13	0.056	0.045	0.019	2.5	21	n.a.	0.06	191	2.5	n.a.	1.44	2.2	0.7	0.6	0.15	0.31	0.068	0.006	3.9

VG N = 28	**MEAN**	**0.10**	**1.5**	**0.004**	**0.03**	**n.a.**	**0.011**	**0.005**	**0.5**	**4**	**n.a.**	**0.02**	**47**	**1.0**	**0.04**	**0.19**	**0.2**	**0.2**	**0.1**	**0.02**	**n.a.**	**0.015**	**n.a.**	**0.5**
	MEDIAN	0.04	1.6	0.002	0.02	n.a	0.010	0.005	0.4	4	n.a	0.02	47	0.9	0.03	0.15	0.2	0.1	0.1	0.00	n.a	0.007	n.a	0.4
	0.1	0.01	0.1	0.000	0.01	n.a.	0.001	0.001	0.2	2	n.a.	0.01	26	0.2	0.01	0.06	0.0	0.1	0.0	0.00	n.a.	0.002	n.a.	0.1
	0.9	0.13	2.9	0.011	0.04	n.a.	0.024	0.010	1.0	7	n.a.	0.02	67	2.2	0.08	0.35	0.3	0.2	0.2	0.06	n.a.	0.038	n.a.	0.9
ST N = 41	**MEAN**	**0.18**	**1.5**	**n.a.**	**0.07**	**0.004**	**0.004**	**0.011**	**1.4**	**16**	**0.002**	**0.06**	**163**	**4.9**	**0.05**	**0.49**	**0.7**	**0.5**	**0.3**	**0.00**	**0.02**	**n.a.**	**0.001**	**0.8**
	MEDIAN	0.17	1.5	n.a	0.07	0.003	0.002	0.009	1.2	13	0.001	0.04	156	5.1	0.05	0.28	0.5	0.5	0.2	0.00	0.01	n.a	0.001	0.4
	0.1	0.09	0.7	n.a.	0.03	0.001	0.001	0.006	0.8	5	0.000	0.01	99	0.0	0.04	0.20	0.3	0.3	0.1	0.00	0.00	n.a.	<0.001	0.3
	0.9	0.32	2.7	n.a.	0.12	0.011	0.007	0.017	2.3	31	0.006	0.09	233	7.6	0.07	0.97	1.4	0.7	0.4	0.01	0.08	n.a.	0.001	1.7
GE N = 28	**MEAN**	**0.17**	**1.5**	**0.004**	**0.05**	**0.020**	**0.055**	**0.007**	**2.2**	**6**	**0.012**	**0.07**	**180**	**3.2**	**0.38**	**0.94**	**0.7**	**0.2**	**1.0**	**0.03**	**0.09**	**0.011**	**0.004**	**2.7**
	MEDIAN	0.17	1.5	0.003	0.06	0.015	0.046	0.006	1.8	6	0.011	0.06	166	2.7	0.22	0.92	0.6	0.1	0.3	0.03	0.08	0.009	0.004	3.2
	0.1	0.12	0.9	0.002	0.04	0.009	0.025	0.003	1.1	3	0.006	0.05	106	1.8	0.14	0.63	0.3	0.1	0.2	0.02	0.05	0.005	0.002	1.4
	0.9	0.23	1.8	0.005	0.07	0.037	0.097	0.012	2.1	8	0.019	0.11	277	5.3	0.83	1.25	0.7	0.2	0.8	0.05	0.14	0.019	0.007	3.6

Table S4. Concentration of the micro- and trace elements (ng/m^3) in the soluble fraction during the warm period.

		As	Ba	Bi	Cd	Ce	Co	Cs	Cu	Fe	La	Li	Mg	Mn	Mo	Ni	Pb	Rb	Sb	Sn	Ti	Tl	U	V
MF N = 111	**MEAN**	**0.36**	**1.8**	**0.034**	**0.08**	**0.016**	**0.038**	**0.016**	**3.8**	**42**	**0.007**	**0.09**	**46**	**8.9**	**0.32**	**0.70**	**1.2**	**0.3**	**0.4**	**0.28**	**0.35**	**0.019**	**0.003**	**1.1**
	MEDIAN	0.31	1.7	0.026	0.07	0.008	0.030	0.012	3.5	35	0.005	0.05	41	6.9	0.29	0.56	1.2	0.3	0.3	0.25	0.24	0.017	0.002	0.5
	0.1	0.17	0.7	0.006	0.03	0.002	0.011	0.004	1.7	9	0.001	0.02	19	1.8	0.13	0.30	0.4	0.1	0.2	0.09	0.04	0.005	0.000	0.1
	0.9	0.64	3.1	0.083	0.15	0.043	0.075	0.032	6.3	79	0.017	0.21	83	19	0.56	1.25	2.1	0.4	0.6	0.46	0.89	0.033	0.006	2.4
FE N = 84	**MEAN**	**0.51**	**1.0**	**0.022**	**0.09**	**0.012**	**0.038**	**0.010**	**3.4**	**15**	**0.007**	**0.04**	**74**	**4.1**	**0.22**	**0.69**	**1.0**	**0.2**	**0.6**	**0.20**	**0.09**	**0.026**	**0.002**	**1.4**
	MEDIAN	0.46	0.8	0.021	0.07	0.009	0.037	0.008	3.2	13	0.006	0.04	65	3.8	0.20	0.55	0.9	0.1	0.5	0.17	0.08	0.020	0.002	1.0
	0.1	0.25	0.4	0.007	0.03	0.006	0.014	0.003	1.8	4	0.003	0.03	30	2.2	0.10	0.25	0.4	0.1	0.2	0.05	0.01	0.007	0.001	0.3
	0.9	0.81	1.7	0.040	0.17	0.023	0.064	0.018	5.8	29	0.011	0.06	138	6	0.37	1.40	1.7	0.3	1.0	0.40	0.19	0.044	0.004	3.2
RB N = 21	**MEAN**	**0.27**	**3.7**	**0.051**	**0.08**	**0.039**	**0.109**	**0.018**	**8.1**	**31**	**0.024**	**0.12**	**235**	**5.6**	**0.50**	**3.03**	**1.3**	**0.6**	**0.6**	**0.16**	**0.40**	**0.009**	**0.004**	**5.9**
	MEDIAN	0.27	3.7	0.025	0.08	0.042	0.111	0.018	7.7	33	0.026	0.11	217	6.4	0.54	3.14	1.4	0.6	0.6	0.17	0.39	0.010	0.004	6.6
	0.1	0.18	1.6	0.010	0.03	0.024	0.062	0.009	3.0	14	0.015	0.06	156	2.7	0.23	1.80	0.8	0.4	0.1	0.07	0.18	0.003	0.003	3.4
	0.9	0.37	5.8	0.107	0.11	0.052	0.148	0.025	11.8	43	0.030	0.16	361	7	0.71	3.93	1.8	0.8	0.9	0.22	0.58	0.016	0.005	7.8
LS N = 14	**MEAN**	**0.38**	**2.0**	**0.015**	**0.06**	**0.014**	**0.106**	**0.006**	**3.1**	**19**	**0.007**	**0.05**	**60**	**3.4**	**0.29**	**0.64**	**0.7**	**0.1**	**0.4**	**0.20**	**0.11**	**0.112**	**0.003**	**1.0**
	MEDIAN	0.30	2.0	0.011	0.04	0.012	0.080	0.005	3.2	18	0.006	0.04	50	3.1	0.26	0.66	0.6	0.1	0.3	0.14	0.09	0.088	0.003	1.0
	0.1	0.18	0.8	0.006	0.02	0.007	0.034	0.003	1.2	6	0.003	0.02	21	1.7	0.12	0.25	0.3	0.1	0.2	0.06	0.04	0.037	0.001	0.2
	0.9	0.59	3.3	0.026	0.12	0.023	0.173	0.009	5.0	31	0.012	0.09	105	6	0.44	0.96	1.2	0.2	0.6	0.38	0.18	0.230	0.006	1.6
CV N = 21	**MEAN**	**0.40**	**8.5**	**0.002**	**0.05**	**0.062**	**0.192**	**0.014**	**2.5**	**7**	**0.069**	**0.06**	**208**	**5.0**	**0.17**	**2.83**	**0.5**	**0.4**	**0.8**	**0.07**	**0.12**	**0.024**	**0.011**	**8.0**
	MEDIAN	0.33	6.9	0.001	0.05	0.042	0.189	0.013	2.3	5	0.038	0.06	180	4.9	0.16	2.76	0.5	0.3	0.3	0.07	0.08	0.020	0.006	5.9
	0.1	0.18	3.2	0.001	0.02	0.011	0.055	0.005	1.2	3	0.012	0.03	84	1.3	0.09	1.05	0.2	0.1	0.1	0.03	0.03	0.011	0.002	2.2
	0.9	0.73	17.7	0.004	0.08	0.139	0.265	0.024	4.0	12	0.195	0.09	380	10	0.26	4.40	0.7	0.7	1.4	0.13	0.30	0.037	0.030	16.0
RM N = 14	**MEAN**	**0.26**	**3.7**	**0.020**	**0.10**	**0.056**	**0.047**	**0.013**	**6.6**	**22**	**0.024**	**0.03**	**55**	**2.6**	**0.28**	**0.78**	**0.3**	**0.3**	**0.8**	**0.16**	**0.22**	**0.024**	**0.002**	**1.6**
	MEDIAN	0.21	3.6	0.018	0.10	0.029	0.047	0.011	5.4	17	0.014	0.03	59	2.5	0.20	0.78	0.3	0.3	0.7	0.13	0.13	0.020	0.002	1.8
	0.1	0.17	3.0	0.006	0.06	0.021	0.035	0.006	3.8	9	0.011	0.03	33	2.2	0.12	0.44	0.1	0.3	0.4	0.07	0.09	0.016	0.001	0.4
	0.9	0.43	4.5	0.034	0.16	0.136	0.058	0.020	11.0	37	0.065	0.04	81	3	0.43	1.16	0.6	0.4	1.2	0.27	0.24	0.037	0.003	2.5

	MEAN	**0.35**	**0.9**	**0.020**	**0.10**	**n.a.**	**0.062**	**0.028**	**2.9**	**18**	**n.a.**	**0.10**	**80**	**3.9**	**0.31**	**2.24**	**1.2**	**0.2**	**0.6**	**0.16**	**0.25**	**0.058**	**0.006**	**8.3**
GE	MEDIAN	0.39	0.9	0.020	0.08	n.a	0.059	0.026	3.0	18	n.a	0.09	80	3.9	0.33	2.00	1.3	0.1	0.5	0.18	0.25	0.055	0.005	7.0
N = 27	0.1	0.22	0.6	0.015	0.06	n.a.	0.049	0.013	2.2	7	n.a.	0.08	60	3.2	0.17	1.61	0.9	0.0	0.3	0.12	0.18	0.032	0.004	5.8
	0.9	0.43	1.3	0.025	0.20	n.a.	0.080	0.043	3.8	30	n.a.	0.11	103	5	0.44	3.20	1.6	0.3	0.9	0.20	0.32	0.085	0.007	11.8

Table S5. Loadings of PCA run on the soluble fraction of elements.

	PC1	PC2	PC3	PC4	PC5	PC6
Variance %	**36**	**13.6**	**7.7**	**5.9**	**5.5**	**4.3**
As	0.25	0.15	0.15	0.16	0.31	0.20
Ba	0.15	−0.26	0.41	−0.03	−0.31	−0.07
Bi	0.26	0.12	−0.17	0.08	0.06	0.03
Cd	0.24	0.18	0.14	0.04	0.24	0.12
Ce	0.10	−0.35	0.13	0.19	−0.19	0.25
Co	0.13	−0.28	−0.06	0.13	0.26	−0.14
Cs	0.21	0.09	0.35	−0.03	0.07	0.07
Cu	0.19	0.03	0.27	0.05	−0.20	−0.46
Fe	0.29	0.10	−0.15	0.00	−0.15	−0.08
La	0.09	−0.38	0.15	0.18	−0.05	0.34
Li	0.28	0.08	−0.13	−0.16	−0.08	0.10
Mg	0.04	−0.31	0.07	−0.38	0.18	−0.09
Mn	0.28	0.04	−0.30	−0.12	−0.27	0.06
Mo	0.28	0.13	0.04	0.04	0.08	−0.04
Ni	0.21	−0.33	−0.19	−0.05	0.27	−0.21
Pb	0.22	0.19	0.11	0.07	0.36	0.26
Rb	0.22	0.00	0.24	−0.38	−0.11	−0.04
Sb	0.18	0.03	0.26	0.08	−0.03	−0.33
Sn	0.30	0.11	−0.14	0.08	−0.07	−0.07
Ti	0.24	−0.01	−0.36	−0.06	−0.29	0.02
Tl	−0.02	0.00	0.07	−0.71	0.07	0.26
U	0.13	−0.30	−0.06	0.13	−0.17	0.37
V	0.13	−0.37	−0.23	−0.05	0.36	−0.24

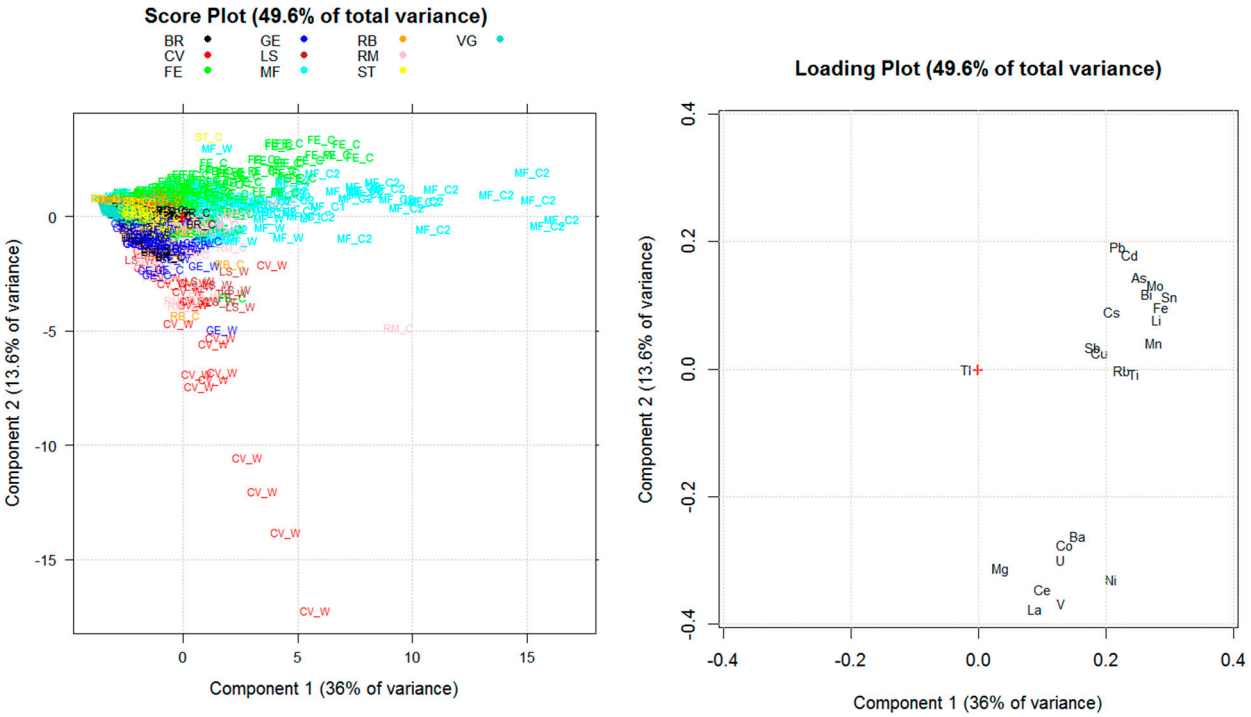

Figure S2. PCA of the soluble fraction of elements (all data): score plot (left panel) and loading plot (right panel) of Components 1 and 2.

Table S6. Percent solubility of micro- and trace elements during the cold period.

		As	Ba	Bi	Cd	Ce	Co	Cs	Cu	Fe	La	Li	Mg	Mn	Mo	Ni	Pb	Rb	Sb	Sn	Ti	Tl	U	V
	MEAN	74	42	25	79	9.0	35	69	43	18	8.9	53	61	49	40	31	29	85	50	17	9.3	87	31	58
MF1	MEDIAN	75	42	23	82	8.3	34	71	43	17	8.5	52	64	52	39	30	28	87	51	14	8.8	90	31	62
N = 53	0.1	58	30	7.0	67	4.0	26	57	29	6.1	4.2	40	44	30	22	19	12	75	26	3.6	3.5	79	16	33
	0.9	91	55	52	90	15	49	83	55	32	15	67	75	64	62	44	46	94	73	38	16	96	46	77
	MEAN	59	36	15	81	8.2	35	75	35	12	6.0	66	64	45	49	27	23	84	39	11	7.7	79	30	57
MF2	MEDIAN	60	36	14	85	8.2	36	78	35	10	6.2	67	67	46	51	27	23	89	41	11	6.6	82	29	59
N = 54	0.1	48	22	6	63	4.9	24	64	25	5.0	4.0	57	50	33	33	15	14	67	24	3.9	3.5	67	20	33
	0.9	72	49	26	93	12	45	84	46	22	7.8	76	76	58	62	37	34	94	55	19	13	88	39	77
	MEAN	68	42	13	86	7.9	33	74	37	10	6.7	65	64	49	49	26	30	69	42	6.4	4.2	84	27	47
FE	MEDIAN	68	41	11	87	7.1	33	75	38	8.4	6.1	66	66	48	50	26	30	78	41	4.6	3.8	86	26	45
N = 82	0.1	58	29	5	77	3.9	22	62	25	4.2	3.6	49	48	41	33	16	19	36	28	1.8	1.3	74	17	26
	0.9	77	89	28	89	11	48	84	54	24	6.1	78	79	58	73	37	43	89	59	18	11	91	41	73
	MEAN	60	22	9.1	74	4.2	24	42	29	6.4	4.0	26	36	35	37	13	25	58	26	3.4	1.5	78	16	32
RB	MEDIAN	63	21	7.1	74	3.3	24	41	29	4.1	3.2	20	35	34	34	9	21	57	25	2.6	1.0	84	15	27
N = 21	0.1	43	13	1.2	56	1.0	7	20	16	1.9	1.3	11	17	18	21	4	12	28	11	0.8	0.3	59	8	8
	0.9	71	29	19	92	8.7	41	68	43	15	7.2	43	55	49	49	22	40	83	42	6.2	2.3	92	24	56
	MEAN	62	75	4.4	58	3.9	27	45	33	3.0	3.9	43	58	32	23	21	9	69	29	3.4	2.0	62	8	38
RM	MEDIAN	57	73	3.4	55	2.8	25	48	32	2.5	3.1	43	55	32	20	20	8	72	29	2.6	1.4	65	8	35
N = 42	0.1	39	51	2.3	38	1.5	16	27	20	1.3	1.5	31	37	26	12	10	4	51	22	1.3	0.8	44	3	23
	0.9	99	104	6.2	77	7.3	37	61	49	4.6	6.7	53	85	41	35	30	15	80	36	6.5	4.2	75	16	56
	MEAN	62	38	22	67	18	38	47	52	8.1	n.a.	33	68	51	n.a.	50	37	64	43	11	5.9	79	32	62
BR	MEDIAN	64	39	24	67	17	39	43	58	7.7	n.a	28	73	54	n.a	52	38	63	44	9.0	5.3	80	35	62
N = 42	0.1	51	27	5.3	58	12	24	30	25	3.0	n.a.	20	46	40	n.a.	34	26	39	30	2.0	1.4	72	13	45
	0.9	72	48	35	77	27	47	66	78	13	n.a.	51	82	59	n.a.	63	48	87	56	22	11	86	49	76
	MEAN	36	31	11	81	n.a.	45	49	44	9.0	n.a.	35	75	60	80	30	24	65	56	6.9	n.a.	n.a.	n.a.	63
VG	MEDIAN	33	34	9.4	83	n.a	49	47	46	7.7	n.a	36	76	60	86	30	24	67	58	3.2	n.a	n.a	n.a	66
N = 28	0.1	15	3	2.1	65	n.a.	28	27	25	6.7	n.a.	18	57	49	67	13	12	48	33	1.2	n.a.	n.a.	n.a.	43
	0.9	60	52	22	93	n.a.	58	65	60	14	n.a.	48	87	70	94	44	36	82	75	20	n.a.	n.a.	n.a.	79
	MEAN	44	66	n.a.	68	2.9	30	49	72	21	2.5	25	77	70	33	28	60	85	44	0.6	3.9	n.a.	11	50
ST	MEDIAN	44	73	n.a	78	2.7	31	53	79	20	2.3	27	87	85	35	27	70	90	47	0.1	2.6	n.a	11	51
N = 41	0.1	20	34	n.a.	44	0.0	10	24	39	11	0.6	1	49	0	18	7	37	82	23	0.0	0.0	n.a.	0	27
	0.9	62	85	n.a.	88	5.1	42	67	87	32	4.4	48	95	91	49	47	77	95	58	2.3	8.7	n.a.	25	67
	MEAN	73	23	8.9	78	n.a.	41	35	44	3.0	n.a.	42	79	60	54	45	22	48	47	n.a.	2.3	72	29	61
GE	MEDIAN	69	22	9.1	79	n.a	41	34	43	2.7	n.a	40	79	60	56	46	21	49	49	n.a	2.3	72	31	65
N = 28	0.1	60	15	4.1	69	n.a.	35	14	37	2.0	n.a.	28	73	55	44	34	15	27	42	n.a.	1.2	63	20	52
	0.9	91	33	13	84	n.a.	47	57	49	4.3	n.a.	64	89	66	63	56	31	66	58	n.a.	3.5	81	36	70

Table S7. Percent solubility of micro- and trace elements during the warm period.

		As	Ba	Bi	Cd	Ce	Co	Cs	Cu	Fe	La	Li	Mg	Mn	Mo	Ni	Pb	Rb	Sb	Sn	Ti	Tl	U	V
	MEAN	72	41	23	87	6.3	39	55	53	13	6.2	44	60	56	51	35	40	60	54	23	6.2	85	25	53
MF	MEDIAN	70	34	15	82	6.5	39	55	45	8.1	5.9	42	64	55	51	33	32	59	50	12	3.8	84	27	56
N = 111	0.1	59	17	7	67	3.6	25	32	31	3.9	3.6	22	48	43	34	18	21	27	34	2.9	1.6	70	16	30
	0.9	87	54	32	93	12	50	80	60	19	11	72	79	67	63	51	45	84	67	29	9.0	93	37	74
	MEAN	80	23	16	70	6.3	32	34	39	6.1	5.3	26	49	53	43	33	26	37	59	15	3.2	83	22	56
FE	MEDIAN	81	22	15	72	5.4	33	32	40	5.7	5.2	24	48	54	42	34	26	35	61	15	2.5	88	22	58
N = 84	0.1	69	14	8.5	58	4.1	23	20	28	2.8	3.8	17	30	43	32	19	19	23	48	5.7	0.4	62	15	38
	0.9	91	34	23	81	7.2	43	47	50	10	7.0	34	67	64	54	49	33	56	68	24	6.2	95	29	74
	MEAN	66	31	10	73	5.2	22	28	34	5.2	4.7	17	36	33	38	21	25	31	48	10	1.6	74	25	42
RB	MEDIAN	67	31	8.1	79	4.3	23	28	36	4.9	3.8	17	36	33	38	20	25	27	46	8.7	1.7	77	26	41
N = 21	0.1	48	17	5.2	45	2.9	14	15	19	1.9	2.5	10	20	18	29	10	15	16	40	5.3	0.8	50	16	18
	0.9	81	44	19	93	8.7	30	42	51	11	7.5	26	52	48	50	33	37	51	60	15	2.9	90	37	66
	MEAN	63	40	15	74	n.a.	43	37	50	5.9	n.a.	31	70	54	54	49	44	68	46	n.a.	3.8	79	30	64
LS	MEDIAN	63	40	14	73	n.a	40	38	50	5.6	n.a	31	69	57	54	49	44	65	45	n.a	3.5	80	27	64
N = 14	0.1	63	32	10	65	n.a.	28	29	25	2.1	n.a.	22	47	42	39	39	31	42	37	n.a.	1.3	65	18	51
	0.9	63	48	18	82	n.a.	56	52	64	8.5	n.a.	53	87	65	62	65	68	78	54	n.a.	4.9	91	39	78
	MEAN	65	44	3.5	83	7.3	50	21	48	2.4	11	28	74	59	54	48	54	72	51	8.9	1.4	79	27	67
CV	MEDIAN	65	39	3.1	82	6.8	51	20	47	2.3	11	23	76	59	54	48	55	76	50	8.3	1.3	79	27	72
N = 21	0.1	49	28	2.2	74	5.2	37	14	44	1.4	4.9	12	47	52	37	42	42	55	42	4.4	0.6	66	15	50
	0.9	82	61	5.6	92	11	64	29	55	3.5	15	54	90	64	70	57	71	91	59	13	2.1	90	38	83
	MEAN	79	36	13	79	12	41	27	47	6.1	7.9	25	60	48	35	44	14	53	42	9.1	2.0	73	19	67
RM	MEDIAN	85	36	12	77	8.5	42	28	47	5.8	7.6	27	61	48	33	43	13	54	41	7.1	2.1	75	17	72
N = 14	0.1	57	29	3.6	70	4.9	35	13	34	2.8	5.0	18	46	42	21	30	5	47	31	4.1	1.2	68	10	49
	0.9	98	42	20	93	28	48	36	63	9.3	11	30	73	57	51	56	23	60	53	13	3.0	79	26	76
	MEAN	68	14	28	70	n.a.	81	47	49	7.9	n.a.	36	61	63	52	54	26	32	54	n.a.	4.0	95	36	74
GE	MEDIAN	69	13	29	69	n.a	81	47	47	7.1	n.a	34	62	63	52	54	25	29	53	n.a	4.0	95	36	75
N = 27	0.1	57	10	22	62	n.a.	78	29	43	3.3	n.a.	27	54	58	46	41	23	10	47	n.a.	2.3	93	32	65
	0.9	80	19	32	81	n.a.	85	59	58	15	n.a.	44	69	68	59	66	31	52	61	n.a.	5.2	96	41	84

Table S8. Loadings of PCA run on the solubility percentage of elements.

	PC1	PC2	PC3	PC4	PC5	PC6
Variance %	**36.8**	**13.8**	**8.1**	**6.2**	**5.5**	**4.5**
As	0.14	0.07	0.32	0.07	0.17	0.66
Ba	0.03	−0.23	−0.22	0.48	−0.15	0.31
Bi	0.28	0.05	0.28	0.07	−0.14	−0.07
Cd	0.19	−0.13	−0.07	−0.03	0.60	0.01
Ce	0.20	−0.03	0.07	−0.25	−0.32	0.08
Co	0.23	0.10	−0.21	−0.24	−0.11	0.11
Cs	0.10	−0.47	0.12	−0.05	0.08	−0.08
Cu	0.25	0.09	−0.22	0.34	−0.08	0.02
Fe	0.24	−0.18	0.14	0.31	−0.13	−0.23
La	0.18	0.00	0.03	−0.26	−0.16	0.14
Li	0.09	−0.46	0.05	−0.21	0.08	0.02
Mg	0.15	−0.18	−0.47	−0.07	−0.07	0.00
Mn	0.24	0.13	−0.22	0.14	0.04	−0.15
Mo	0.22	0.07	−0.10	−0.21	0.14	−0.41
Ni	0.23	0.20	−0.23	−0.17	−0.16	0.28
Pb	0.25	0.04	−0.20	0.29	0.12	−0.03
Rb	0.09	−0.44	−0.14	0.05	−0.13	0.02
Sb	0.24	0.25	0.09	0.11	0.10	−0.19
Sn	0.24	0.16	0.32	0.14	−0.06	−0.08
Ti	0.21	−0.21	0.29	0.04	−0.27	−0.02
Tl	0.22	−0.05	0.07	−0.05	0.47	0.15
U	0.27	−0.02	0.13	−0.24	−0.05	−0.07
V	0.26	0.09	−0.13	−0.19	−0.07	0.13

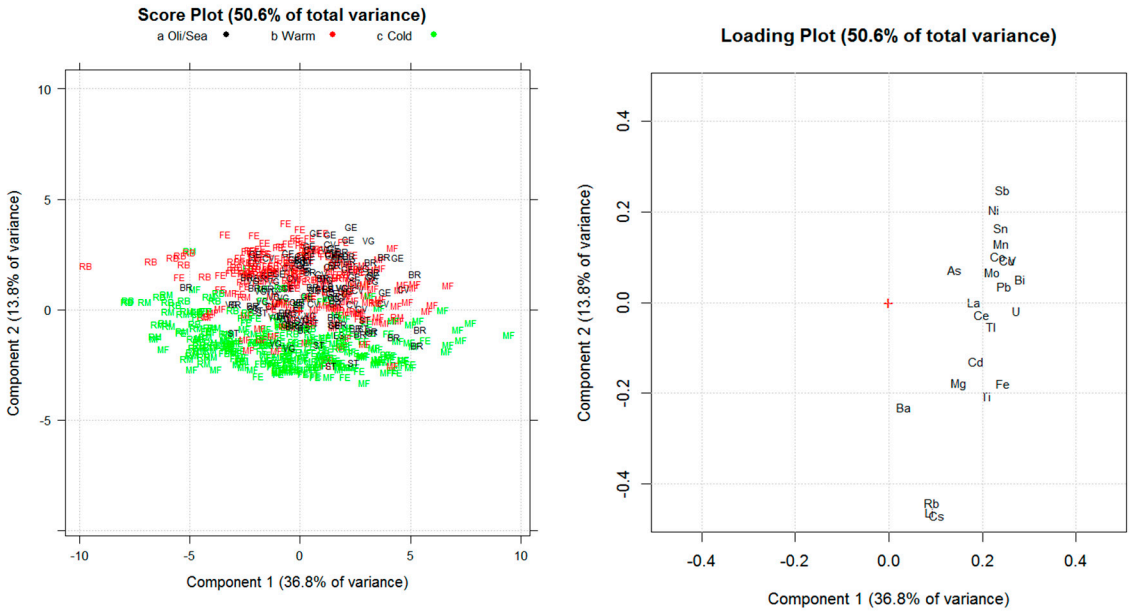

Figure S3. PCA of the solubility percentage of elements (all data): score plot (left panel).and loading plot (right panel) of components 1 and 2.