Supplementary Table S1. Experimental conditions, limits of detection (LOD) and sample blanks (SB) of the analytes of interest with the technique used for their determination after direct digestion.

	SF-ICP-MS					ICP-OES		
	Isotope	Resolution	LOD (ng/L)	SB (µg/L)		λ (nm)	LOD (µg/L)	SB (µg/L)
Al	27	LR-MR	400	0.63	Ca	317.93	3.1	260
As	75	MR	42	< LOD	K	769.90	1.5	8.8
Ba	135-137-138	LR-MR	300	62	Mg	285.21	1.2	< LOD
Cd	111-112-114	LR	2.3	< LOD	Na	589.59	11	210
Co	59	LR-MR	1.9	< LOD				
Cr	53	MR	10	< LOD				
Cu	63-65	LR-MR	11	< LOD				
Fe	56-57	MR	400	3.2				
Mn	55	LR-MR	8.6	0.04				
Ni	60-61-62	LR-MR	45	0.5				
Pb	206-207-208	LR-MR	40	0.23				
Sb	121-123	MR	23	< LOD				
Ti	46-47-48	LR-MR	750	< LOD				
V	51	LR-MR	58	< LOD				
Zn	66-68	LR-MR	190	4.5				

LR = low resolution; MR = medium resolution

Supplementary Table S2. Experimental conditions, limits of detection (LOD) and sample blanks (SB) of the analytes of interest with the technique used for their determination in fraction I and fraction II.

	Fraction I SF-ICP-MS					Fraction II SF-ICP-MS					Fraction I ICP-OES		
	Isotope	Resolution	LOD (ng/L)	SB (µg/L)		Isotope	Resolution	LOD (ng/L)	SB (µg/L)		λ (nm)	LOD (µg/L)	SB (µg/L)
Al	27	LR-MR	250	8.2	Al	27	LR-MR	400	40	Ca	317.93	8.5	60
As	75	MR	3.4	0.02	As	75	MR	42	< LOD	K	769.90	4.5	17
Ba	135-137-138	LR-MR	120	0.2	Ba	135-137-138	LR-MR	300	1.5	Mg	285.21	3.0	8.7
Cd	111-112-114	LR	12	< LOD	Cd	111-112-114	LR	2.3	0.03	Na	589.59	3.3	126
Co	59	LR-MR	8	< LOD	Co	59	LR-MR	1.9	0.02	Zn	213.86	0.7	9.4
Cr	53	MR	30	0.08	Cr	53	MR	10	0.05				
Cu	63-65	LR-MR	42	0.21	Cu	63-65	LR-MR	11	0.35				
Fe	56-57	MR	430	2.3	Fe	56-57	MR	400	34		**Fraction II ICP-OES**		
Mn	55	LR-MR	45	0.13	Mn	55	LR-MR	8.6	0.7		λ (nm)	LOD (µg/L)	SB (µg/L)
Ni	60-61-62	LR-MR	83	1.3	Ni	60-61-62	LR-MR	45	0.5				
Pb	206-207-208	LR-MR	24	0.62	Pb	206-207-208	LR-MR	40	0.35	Ca	317.93	3.1	168
Sb	121-123	MR	23	0.13	Sb	121-123	MR	23	< LOD	K	769.90	1.5	15
Ti	46-47-48	LR-MR	50	0.20	Ti	46-47-48	LR-MR	750	1.2	Mg	285.21	1.2	12
V	51	LR-MR	10	< LOD	V	51	LR-MR	58	< LOD	Na	589.59	11	80
Zn	66-68	LR-MR	160	9.4	Zn	66-68	LR-MR	190	5.2	Zn	213.86	1.0	5.2

LR = low resolution; MR = medium resolution

Supplementary Table S3. Major element concentrations in Arctic PM_{10} samples collected in 2015. All the values are expressed in ng/m^3.

Sampling start date	Al	Ca	Fe	K	Mg	Na	Sampling start date	Al	Ca	Fe	K	Mg	Na
28th February	6.96 ± 0.05	26.6 ± 0.6	5.8 ± 0.1	19.0 ± 0.5	70.6 ± 0.6	478 ± 5	24th June	2.08 ± 0.02	10.9 ± 0.2	2.55 ± 0.04	4.50 ± 0.09	10.3 ± 0.2	74 ± 1
4th March	0.36 ± 0.01	6.96 ± 0.02	0.67 ± 0.02	5.78 ± 0.09	21.8 ± 0.1	168 ± 1	28th June	2.08 ± 0.08	12.9 ± 0.4	2.17 ±0.06	6.0 ± 0.2	18.9 ± 0.3	144 ± 1
8th March	1.77 ± 0.05	28.5 ± 0.7	2.4 ± 0.1	21.1 ± 0.1	79 ± 1	599 ± 9	2nd July	0.64 ± 0.02	13.9 ± 0.2	1.02 ± 0.03	8.72 ± 0.08	26 ± 1	208 ± 2
12th March	5.3 ± 0.2	38.3 ± 0.9	8.06 ± 0.09	33 ±1	109 ± 2	858 ± 27	6th July	3.56 ± 0.04	13.4 ± 0.2	3.83 ± 0.06	5.1 ± 0.2	12.4 ± 0.1	88.8 ± 0.8
16th March	7.7 ± 0.2	39.8 ± 0.9	5.61 ± 0.06	27.4 ± 0.8	90 ± 2	573 ± 9	10th July	27.1 ± 0.3	17.0 ± 0.2	7.6 ± 0.1	19.8 ± 0.1	11.0 ± 0.3	63.8 ± 0.5
20th March	8.2 ± 0.1	9.5 ± 0.3	5.7 ± 0.1	5.1 ± 0.1	12.7 ± 0.1	71.0 ± 0.8	14th July	4.28 ± 0.09	9.37 ± 0.04	2.21 ± 0.06	< 1.98	4.16 ± 0.04	24.6 ± 0.3
24th March	4.5 ± 0.1	13.7 ± 0.3	3.94 ± 0.05	12.2 ± 0.2	40.3 ± 0.2	342 ± 4	18th July	1.19 ± 0.01	5.3 ± 0.1	0.63 ± 0.01	2.94 ± 0.02	2.31 ± 0.04	31.5 ± 0.5
28th March	3.3 ± 0.1	18.8 ± 0.7	2.9 ± 0.1	12.3 ± 0.1	46.7 ± 0.9	325 ± 4	22nd July	1.27 ± 0.02	12.33 ± 0.08	1.63 ± 0.03	5.83 ± 0.01	20.0 ± 0.1	171 ± 3
1st April	30.8 ± 0.4	28.6 ± 0.9	19.5 ± 0.1	18.5 ± 0.5	42 ± 1	238 ± 3	26th July	1.15 ± 0.02	10.01 ± 0.07	1.98 ± 0.03	4.4 ± 0.1	14.3 ± 0.3	104 ± 1
5th April	28.1 ± 0.8	49.7 ± 0.6	22.3 ± 0.1	19.1 ± 0.1	41.3 ± 0.6	220 ± 3	30th July	7.45 ± 0.05	9.7 ± 0.1	5.16 ± 0.09	3.94 ± 0.07	5.06 ± 0.09	27.3 ± 0.1
9th April	5.46 ± 0.09	8.1 ± 0.1	5.3 ± 0.1	6.0 ± 0.2	9.3 ± 0.3	61 ± 1	3rd August	5.55 ± 0.08	15.8 ± 0.2	4.4 ± 0.1	10.8 ± 0.1	23.3 ± 0.3	171 ± 1
13th April	9.1 ± 0.2	27.2 ± 0.4	8.0 ± 0.1	21.0 ± 0.1	66 ± 2	500 ± 1	7th August	1.38 ± 0.01	6.1 ± 0.3	1.13 ± 0.02	2.89 ± 0.07	4.91 ± 0.05	41.8 ± 0.3
17th April	2.5 ± 0.1	3.85 ± 0.06	1.8 ± 0.1	2.71 ± 0.02	8.0 ± 0.1	59.8 ± 0.2	11th August	3.0 ± 0.1	10.5 ± 0.3	2.18 ± 0.08	3.56 ± 0.03	8.71 ± 0.09	55 ± 1
21st April	3.3 ± 0.1	6.1 ± 0.1	2.50 ± 0.08	3.7 ± 0.1	12.8 ± 0.3	84.6 ± 0.1	15th August	2.04 ± 0.05	10.5 ± 0.1	2.01 ± 0.04	7.4 ± 0.1	21.9 ± 0.5	187 ± 2
25th April	4.46 ± 0.07	11.57 ± 0.08	3.22 ± 0.05	4.08 ± 0.07	11.7 ± 0.1	75 ± 1	19th August	0.95 ± 0.01	10.3 ± 0.3	1.55 ± 0.03	6.7 ± 0.1	22.6 ± 0.4	195 ± 1
29th April	6.63 ± 0.06	16.7 ± 0.2	4.97 ± 0.08	4.9 ±0.1	10.9 ± 0.1	61 ± 1	23rd August	1.09 ± 0.05	5.21 ± 0.08	1.18 ± 0.04	2.99 ± 0.05	5.5 ± 0.1	47 ± 1
3rd May	4.59 ± 0.07	15.0 ± 0.2	3.47 ± 0.05	5.4 ± 0.2	17.9 ± 0.2	125 ± 1	27th August	0.90 ± 0.01	3.64 ± 0.07	0.78 ± 0.01	2.44 ± 0.08	3.4 ± 0.1	32.3 ± 0.3
7th May	9.3 ± 0.1	14.69 ± 0.06	5.0 ± 0.1	5.85 ± 0.06	16.3 ± 0.1	112 ± 1	31st August	1.06 ± 0.03	8.0 ± 0.1	0.99 ± 0.03	2.39 ± 0.03	9.04 ± 0.09	63 ± 1
11th May	1.86 ± 0.07	8.6 ± 0.1	1.43 ± 0.06	2.92 ± 0.05	7.0 ± 0.1	45.2 ± 0.7	4th September	0.95 ± 0.02	10.1 ± 0.2	1.14 ± 0.02	4.5 ± 0.1	14.5 ± 0.2	112 ± 1
15th May	1.02 ± 0.02	7.01 ± 0.08	0.91 ± 0.01	2.66 ± 0.03	9.45 ± 0.05	71.5 ± 0.5	8th September	9.8 ± 0.1	28.0 ± 0.5	7.5 ± 0.1	14.1 ± 0.2	50.4 ± 0.4	370 ± 3
19th May	3.62 ± 0.06	17.4 ± 0.1	2.80 ± 0.05	10.8 ± 0.1	36.0 ± 0.1	270 ± 3	12th September	2.39 ± 0.04	14.1 ± 0.1	2.42 ± 0.05	9.5 ± 0.1	27.8 ± 0.4	233 ± 1
23rd May	4.42 ± 0.05	17.3 ± 0.6	3.32 ± 0.04	8.80 ± 0.08	29.3 ± 0.2	213 ± 3	16th September	18.9 ± 0.5	69 ± 2	16.9 ± 0.4	52.8 ± 0.3	162 ± 3	1242 ± 1
27th May	6.9 ± 0.2	12.57 ± 0.06	4.57 ± 0.07	5.81 ± 0.07	15.5 ± 0.2	118 ± 2	20th September	5.96 ± 0.04	14.3 ± 0.2	4.75 ± 0.06	7.4 ± 0.2	19.1 ± 0.6	141 ± 1
31st May	7.2 ± 0.2	10.5 ± 0.2	7.6 ± 0.4	5.7 ± 0.1	11.9 ± 0.2	74 ±1	24th September	0.59 ± 0.01	7.04 ± 0.06	0.33 ± 0.01	2.56 ± 0.02	10.4 ± 0.1	75.9 ±0.4
4th June	2.04 ± 0.04	14.68 ± 0.07	2.34 ± 0.05	9.1 ± 0.2	31.8 ± 0.7	243 ± 1	28th September	4.5 ± 0.1	40.6 ± 0.9	4.03 ± 0.09	28.3 ± 0.1	95 ± 1	838 ± 7
8th June	8.6 ± 0.1	20.5 ± 0.3	7.1 ± 0.1	7.8 ± 0.2	21.8 ± 0.8	139 ± 1	2nd October	11.6 ± 0.2	19.9 ± 0.8	9.3 ± 0.2	7.1 ± 0.3	15.1 ± 0.4	73.8 ± 0.8
12th June	20.6 ± 0.1	55 ± 2	18.4 ± 0.5	14.4 ± 0.2	40.5 ± 0.8	184 ± 4	6th October	1.54 ± 0.08	26.0 ± 0.9	1.57 ± 0.06	20.2 ± 0.4	73.9 ± 0.6	619 ± 1
16th June	4.9 ± 0.1	12.5 ± 0.1	2.28 ± 0.05	7.4 ± 0.1	24.5 ± 0.3	189 ± 2	14th October	7.7 ± 0.2	19.5 ± 0.5	7.5 ± 0.1	12.0 ± 0.4	37.5 ± 0.4	306 ± 6
20th June	0.88 ± 0.01	13.9 ± 0.3	1.45 ± 0.02	4.27 ± 0.08	12.2 ± 0.3	83 ±1	18th October	2.05 ± 0.06	9.34 ± 0.03	1.93 ± 0.03	4.6 ± 0.1	14.0 ± 0.1	107 ± 2

Supplementary Table S4. Minor and trace element concentrations in Arctic PM$_{10}$ samples collected in 2015. All the values are expressed in pg/m³.

Sampling start date	As	Ba	Cd	Co	Cr	Cu	Mn	Ni	Pb	Sb	Ti	V	Zn
28th February	33 ± 1	74 ± 3	5.5 ± 0.4	7.20 ± 0.07	284 ± 9	92 ± 3	214 ± 4	170 ± 10	146 ± 4	12.7 ± 0.7	220 ± 10	27.3 ± 0.8	310 ± 10
4th March	8.4 ± 0.2	13.1 ± 0.8	0.96 ± 0.01	0.70 ± 0.03	19.2 ±0.7	11 ± 1	19.2 ± 0.3	25 ± 1	38.5 ± 0.9	2.61 ± 0.04	29.2 ± 0.7	3.5 ± 0.1	35 ± 2
8th March	42 ± 1	29 ± 1	4.1 ± 0.3	1.6 ± 0.1	76 ± 4	35 ± 1	56 ± 1	37 ± 1	90 ± 2	12.7 ± 0.2	650 ± 20	14.4 ± 0.7	350 ± 20
12th March	149 ± 7	81 ± 2	27 ± 1	5.7 ± 0.2	84 ± 2	113 ± 2	240 ± 10	59 ± 3	500 ± 20	29.3 ± 0.9	298 ± 9	32.6 ± 0.3	1,040 ± 30
16th March	75 ± 4	118 ± 3	12.6 ± 0.5	6.5 ± 0.2	35 ± 1	90 ± 2	208 ± 5	52 ± 3	342 ± 5	24.5 ± 0.4	365 ± 7	27.4 ± 0.3	790 ± 10
20th March	47.0 ± 0.6	124 ± 4	8.3 ± 0.2	4.8 ± 0.2	51 ± 1	40 ± 1	184 ± 4	36 ± 1	268 ± 5	19.9 ± 0.4	370 ± 10	27.1 ± 0.8	351 ± 8
24th March	31 ± 2	92 ± 2	5.4 ± 0.2	3.3 ± 0.1	32 ± 2	52 ± 2	140 ± 9	48 ± 1	202 ± 6	19.1 ± 0.3	300 ± 10	32.0 ± 0.5	330 ± 10
28th March	22.0 ± 0.6	62 ± 2	3.26 ± 0.09	1.97 ± 0.08	22 ± 1	30 ± 1	103 ± 3	18 ± 1	100 ± 2	13.3 ± 0.2	175 ± 3	20 ± 1	160 ± 5
1st April	73 ± 7	336 ± 6	10.7 ± 0.6	12.8 ± 0.8	85 ± 2	103 ± 3	627 ± 9	69 ± 5	320 ± 6	24.1 ± 0.8	1,250 ±10	75 ± 2	600 ± 20
5th April	369 ± 11	323 ± 6	45 ± 1	17 ± 1	134 ± 7	280 ± 10	870 ± 20	133 ± 5	1,160 ± 20	56 ± 1	1,190 ± 10	127 ± 3	2,600 ± 100
9th April	84 ± 4	77 ± 3	13.7 ± 0.4	4.2 ± 0.1	103 ± 4	121 ± 3	165 ± 2	66 ± 4	485 ± 4	31.4 ± 0.5	256 ± 7	36.6 ± 0.5	690 ± 30
13th April	21 ± 1	96 ± 3	2.7 ± 0.2	9.8 ± 0.2	92 ± 3	49 ± 1	1,270 ± 50	149 ± 4	87 ± 2	12.5 ± 0.3	410 ± 10	28 ± 1	190 ± 6
17th April	13.9 ± 0.3	28 ± 1	2.1 ± 0.1	0.84 ± 0.02	20.7 ± 0.9	7.9 ± 0.4	53 ± 2	24 ± 1	56 ± 1	9.2 ± 0.3	102 ± 6	7.1 ± 0.3	105 ± 4
21st April	13.5 ± 0.7	46 ± 1	2.3 ± 0.1	1.6 ± 0.1	11.0 ± 0.6	16.0 ± 0.8	69 ± 1	13.2 ± 0.9	43 ± 1	9.8 ± 0.2	195 ± 5	8.0 ± 0.5	101 ± 4
25th April	11.9 ± 0.6	49 ± 1	2.14 ± 0.08	2.0 ± 0.1	21.6 ± 0.7	13.9 ± 0.4	86.9 ± 0.5	22 ± 1	53.1 ± 0.5	6.2 ± 0.1	192 ± 3	10.6 ± 0.5	85 ± 3
29th April	14.7 ± 0.3	71 ± 3	2.1 ± 0.2	2.7 ± 0.2	17 ± 1	17 ± 1	143 ± 3	18.6 ± 0.7	59 ± 2	6.4 ± 0.2	230 ± 10	20.9 ± 0.7	127 ± 8
3rd May	11.9 ± 0.6	53 ± 1	1.69 ± 0.08	1.5 ± 0.1	17.1 ± 0.3	15.8 ± 0.9	116 ± 5	17.4 ± 0.7	43 ± 1	6.3 ± 0.2	213 ± 4	14.1 ± 0.1	132 ± 4
7th May	10.7 ± 0.8	64 ± 2	1.56 ± 0.07	3.26 ± 0.01	39 ± 1	17.4 ± 0.9	155 ± 1	38 ± 3	37 ± 1	3.4 ± 0.1	380 ± 7	18.5 ± 0.7	113 ± 4
11th May	4.7 ± 0.2	32 ± 2	1.14 ± 0.06	0.89 ± 0.05	1.30 ± 0.06	3.8 ± 0.3	53 ± 2	6.2 ± 0.2	25.4 ± 0.8	3.3 ± 0.2	80 ± 2	5.7 ± 0.2	37 ± 2
15th May	3.7 ± 0.4	14.2 ± 0.5	0.60 ± 0.01	0.50 ± 0.02	7.6 ± 0.2	249 ± 4	26.5 ± 0.5	7.7 ± 0.3	10.2 ± 0.2	2.6 ± 0.1	69.0 ± 0.8	2.39 ± 0.09	46 ± 2
19th May	10.9 ± 0.6	41 ± 1	1.54 ± 0.07	1.51 ± 0.06	19.7 ± 0.4	9.6 ± 0.6	83 ± 2	17 ± 1	40 ± 1	5.3 ± 0.2	167 ± 7	12.3 ± 0.4	89 ± 3
23rd May	15.7 ± 0.4	62 ± 2	2.41 ± 0.02	2.1 ± 0.1	39 ± 2	12.2 ± 0.3	124 ± 4	24 ± 2	51.3 ± 0.6	8.7 ± 0.3	233 ± 6	11.5 ± 0.4	149 ± 8
27th May	11.3 ± 0.9	67 ± 4	2.06 ± 0.05	2.7 ± 0.2	6.8 ± 0.4	9.1 ± 0.3	139 ± 5	22 ± 1	40 ± 2	6.01 ± 0.07	333 ± 6	15.0 ± 0.6	117 ± 5
31st May	8.5 ± 0.6	105 ± 3	0.94 ± 0.08	6.1 ± 0.2	172 ± 8	18.9 ± 0.9	360 ± 20	95 ± 5	25.2 ± 0.5	4.0 ± 0.2	330 ± 10	16.2 ± 0.6	61 ± 6
4th June	3.6 ± 0.2	36 ± 2	< 0.37	1.2 ± 0.1	n.a.	40 ± 3	113 ± 1	10.7 ± 0.7	8.6 ± 0.3	n.a.	107 ± 6	3.4 ± 0.2	19 ± 1
8th June	7.5 ± 0.4	102 ± 4	1.08 ± 0.05	4.7 ± 0.2	12.1 ± 0.7	9.1 ± 0.6	251 ± 6	19 ± 1	16.5 ± 0.3	2.34 ± 0.05	393 ± 6	20.3 ± 0.5	50 ± 1
12th June	10.1 ± 0.5	237 ± 7	0.380 ± 0.005	9.2 ± 0.5	33 ± 1	43 ± 2	530 ± 20	46 ± 3	23.0 ± 0.5	2.8 ± 0.2	790 ± 20	36.8 ± 0.9	51 ± 2
16th June	5.9 ± 0.2	32 ± 2	1.00 ± 0.06	2.17 ± 0.07	29 ± 2	46 ± 3	95 ± 2	48 ± 1	18 ± 1	3.15 ± 0.05	203 ± 2	12.2 ± 0.3	37 ± 2
20th June	10.1 ± 0.7	24 ± 1	0.56 ± 0.03	1.02 ± 0.07	13.6 ± 0.6	122 ± 2	44 ± 1	85 ± 4	29.9 ± 0.5	3.5 ± 0.1	< 7.9	5.5 ± 0.4	440 ± 20

Sampling start date	As	Ba	Cd	Co	Cr	Cu	Mn	Ni	Pb	Sb	Ti	V	Zn
24th June	4.98 ± 0.08	32 ± 1	0.39 ± 0.01	1.9 ± 0.2	41 ± 2	19.2 ± 0.6	93 ± 3	29 ± 1	12.9 ± 0.4	2.89 ± 0.09	66 ± 2	17 ± 1	70 ± 4
28th June	4.8 ± 0.3	29 ± 1	0.47 ± 0.02	1.24 ± 0.07	55 ± 2	65 ± 2	49 ± 1	13.6 ± 0.7	10.7 ± 0.3	3.05 ± 0.09	102 ± 1	6.8 ± 0.4	67 ± 4
2nd July	3.73 ± 0.06	12.6 ± 0.3	0.58 ± 0.04	0.80 ± 0.03	15.7 ± 0.7	2.5 ± 0.2	27.2 ± 0.1	13.9 ± 0.4	4.63 ± 0.06	2.26 ± 0.06	53.8 ± 0.8	3.7 ± 0.2	30 ± 2
6th July	6.2 ± 0.2	45 ± 2	0.57 ± 0.03	3.6 ± 0.3	15.1 ± 0.5	66 ± 1	184 ± 9	15.7 ± 0.7	10.1 ± 0.2	2.22 ± 0.08	179 ± 6	7.8 ± 0.3	76 ± 3
10th July	33 ± 2	156 ± 4	12.5 ± 0.4	4.59 ± 0.09	69 ± 2	72 ± 1	440 ± 10	97 ± 4	38 ± 1	3.95 ± 0.09	470 ± 10	90 ± 1	1,160 ± 20
14th July	3.74 ± 0.07	53 ± 2	< 0.35	1.29 ± 0.08	33 ± 1	8.7 ± 0.3	81 ± 2	44 ± 3	12.6 ± 0.4	2.6 ± 0.1	141 ± 8	5.4 ± 0.3	40 ± 2
18th July	1.43 ± 0.01	16 ± 1	0.55 ± 0.02	0.51 ± 0.04	4.7 ± 0.2	0.03 ± 0.01	20.2 ± 0.6	12.9 ± 0.7	1.24 ± 0.06	2.6 ± 0.1	66 ± 3	0.74 ± 0.06	43 ± 2
22nd July	3.9 ± 0.2	16.9 ± 0.8	0.78 ± 0.01	1.15 ± 0.06	20.0 ± 0.5	5.1 ± 0.2	55 ± 2	25 ± 1	4.8 ± 0.2	2.01 ± 0.05	118 ± 4	1.87 ± 0.03	69 ± 3
26th July	6.94 ± 0.02	20 ± 1	< 0.36	0.80 ± 0.07	128 ± 3	10.1 ± 0.6	30 ± 1	16.9 ± 0.7	10.3 ± 0.3	3.6 ± 0.1	54 ± 2	10.3 ± 0.2	50 ± 3
30th July	6.7 ± 0.2	113 ± 3	2.0 ± 0.1	4.4 ± 0.4	46 ± 3	36 ± 2	168 ± 3	33 ± 1	13.9 ± 0.3	1.62 ± 0.08	118 ± 4	25 ± 2	320 ± 10
3rd August	10.8 ± 0.2	124 ± 2	2.2 ± 0.1	1.91 ± 0.01	68 ± 4	21 ± 1	115 ± 2	29 ± 2	23.1 ± 0.5	3.8 ± 0.2	197 ± 7	14.8 ± 0.8	187 ± 7
7th August	6.1 ± 0.3	21 ± 1	0.42 ± 0.03	0.82 ± 0.03	7.1 ± 0.2	2.86 ± 0.08	32 ±2	17 ± 1	6.7 ± 0.2	2.30 ± 0.07	61 ± 4	4.2 ± 0.2	41 ± 2
11th August	5.5 ± 0.3	26 ± 2	< 0.35	1.7 ± 0.1	9.2 ± 0.5	8.2 ± 0.3	74 ± 2	13.7 ± 0.8	13.6 ± 0.4	1.99 ± 0.06	340 ± 20	9.1 ± 0.6	24 ± 1
15th August	17 ± 1	36 ± 1	0.71 ± 0.03	0.82 ± 0.04	56 ± 1	33 ± 1	61 ± 2	81 ± 3	17.7 ± 0.3	2.59 ± 0.09	114 ± 4	14.6 ± 0.4	67 ± 3
19th August	5.0 ± 0.1	10.1 ± 0.7	< 0.37	0.44 ± 0.01	86 ± 2	3.8 ± 0.2	28.4 ± 0.7	40 ± 2	4.0 ± 0.1	1.08 ± 0.06	85 ± 3	4.4 ± 0.3	< 26.59
23th August	6.9 ± 0.2	18.7 ± 1	0.59 ± 0.03	1.68 ±0.06	14.3 ± 0.3	11.3 ± 0.6	32.3 ± 0.5	22 ± 1	8.9 ± 0.5	5.1 ± 0.2	320 ± 10	7.3 ± 0.5	46 ± 3
27th August	3.2 ± 0.2	56 ± 3	0.49 ± 0.05	2.0 ± 0.1	21.8 ± 0.8	6.0 ± 0.4	290 ± 10	52 ± 2	12.6 ± 0.5	1.09 ± 0.06	85 ± 2	0.90 ± 0.08	33 ± 2
31st August	1.76 ± 0.06	15.8 ± 0.3	< 0.36	0.63 ± 0.06	20.8 ± 0.7	0.52 ± 0.03	41 ± 2	21 ± 1	3.9 ± 0.1	2.8 ± 0.1	44 ± 1	1.8 ± 0.1	18.6 ± 0.7
4th September	12.7 ± 0.5	18.0 ± 0.3	1.5 ± 0.1	0.68 ± 0.06	15.2 ± 0.4	155 ± 2	36 ± 1	107 ± 7	10.3 ± 0.2	2.38 ± 0.08	< 8.2	3.10 ± 0.07	43 ± 1
8th September	8.9 ± 0.4	111 ± 4	< 0.55	4.4 ± 0.2	84 ± 2	25 ± 1	167 ± 9	91 ± 4	12.6 ± 0.4	5.6 ± 0.1	530 ± 20	18.4 ± 0.8	144 ± 9
12th September	6.35 ± 0.09	24 ± 1	0.57 ± 0.02	2.4 ± 0.1	48 ± 1	18.0 ± 0.7	58 ± 2	37 ± 2	26.4 ± 0.7	4.2 ± 0.3	58.0 ± 0.4	16.6 ± 0.5	73 ± 4
16th September	14.4 ± 0.9	210 ± 2	0.53 ± 0.04	10.8 ± 0.4	71 ± 2	32 ± 1	324 ± 4	67 ± 2	28.2 ± 0.7	2.25 ± 0.04	1,280 ± 40	41.4 ± 0.9	92 ± 4
20th September	6.3 ± 0.4	58 ± 4	1.59 ± 0.09	3.26 ± 0.09	18 ± 1	35 ± 1	120 ± 1	56 ± 2	13.1 ± 0.4	2.01 ± 0.09	260 ± 10	14.2 ± 0.7	104 ± 6
24th September	3.03 ± 0.01	103 ± 3	< 0.37	< 0.37	5.0 ± 0.2	55 ± 2	14.3 ± 0.8	12.2 ± 0.7	177 ± 3	7.7 ± 0.1	16.9 ± 0.8	0.96 ± 0.08	46 ± 3
28th September	4.6 ± 0.2	43 ± 2	< 0.33	2.37 ± 0.06	48 ± 1	147 ± 1	97 ± 3	35 ± 1	8.4 ± 0.3	3.28 ± 0.05	340 ± 10	11.6 ± 0.6	58 ± 3
2nd October	4.2 ± 0.1	107 ± 3	< 0.33	4.5 ± 0.3	134 ± 1	10.8 ± 0.5	185 ± 5	24 ± 1	15.9 ± 0.4	2.33 ± 0.08	320 ± 10	19.6 ± 0.6	37 ± 1
6th October	7.6 ± 0.7	26 ± 1	< 0.43	0.92 ± 0.08	14.9 ± 0.6	37 ± 3	34.4 ± 0.6	24 ± 1	11.3 ± 0.3	3.1 ± 0.2	139 ± 5	12.9 ± 0.9	55 ± 2
14th October	4.30 ± 0.04	81 ± 3	< 0.32	4.0 ± 0.1	39.8 ± 0.3	26.9 ± 0.5	253 ± 5	21.7 ± 0.6	11.4 ± 0.3	2.68 ± 0.06	289 ± 2	20.3 ± 0.6	48 ± 2
18th October	2.25 ± 0.01	21 ± 1	< 0.34	1.6 ± 0.1	47 ± 2	16.5 ± 0.6	42 ± 1	22 ± 1	5.27 ± 0.08	1.28 ± 0.04	98 ±4	2.8 ± 0.2	30 ± 1

Supplementary Table S5. Extraction percentages obtained for fraction I on the samples subjected to sequential extraction.

Sampling start date	Al	As	Ba	Ca	Cd	Co	Cr	Cu	Fe	K	Mg	Mn	Na	Ni	Pb	Sb	Ti	V	Zn
28th February	85	94	39	78	100	63	27	84	29	90	96	86	97	64	96	90	16	80	96
8th March	44	96	26	79	39	26	16	72	28	96	97	34	98	65	93	85	11	80	97
16th March	17	94	42	77	93	56	16	74	25	88	96	67	97	51	94	77	6	68	95
24th March	26	93	50	78	100	60	37	76	35	90	96	69	98	76	96	88	14	78	97
1st April	18	87	56	79	86	60	13	79	24	71	89	77	96	54	95	83	6	59	90
9th April	89	91	44	60	100	72	15	88	52	83	87	78	94	70	97	86	20	82	98
17th April	33	87	27	66	64	53	8	70	25	80	79	68	89	46	86	82	10	64	96
25th April	28	90	35	74	100	50	7	56	27	77	85	67	91	66	89	82	10	58	96
3rd May	20	84	31	70	100	60	8	68	24	77	87	13	94	77	84	66	5	54	94
11th May	52	88	29	69	100	55	7	60	33	78	74	65	84	28	93	84	14	56	98
23rd May	25	86	42	75	60	47	17	60	23	82	90	48	95	51	84	69	7	61	93
31st May	19	79	30	69	100	65	8	58	27	60	85	81	94	59	89	80	10	39	91
12nd June	12	64	27	88	69	50	7	68	26	39	86	68	93	40	75	66	7	27	96
20th June	39	87	34	70	100	57	9	74	36	80	88	68	94	54	95	85	15	68	93
2nd July	14	83	13	63	29	27	15	56	27	87	91	21	95	58	82	67	7	50	95
22nd July	8	75	19	54	65	35	4	67	22	81	81	51	87	46	72	62	9	46	93
3rd August	43	92	45	74	100	60	19	65	38	86	87	77	94	76	90	82	17	72	96
11th August	21	85	27	70	29	26	8	67	31	81	86	66	92	61	85	69	16	66	95
19th August	79	91	26	72	100	59	17	70	49	91	94	60	97	54	84	77	45	71	91
31st August	22	84	18	66	54	38	6	59	20	81	86	17	92	40	81	64	5	38	96
8th September	39	85	31	77	100	57	16	65	36	87	94	68	98	58	87	83	15	43	94
20th September	18	75	26	73	84	51	16	56	25	78	91	44	96	71	76	42	9	32	94
28th September	54	85	32	87	100	61	18	72	45	95	98	73	99	54	89	84	48	60	94
14th October	14	67	32	78	35	15	13	68	23	84	94	33	98	47	79	62	7	47	90
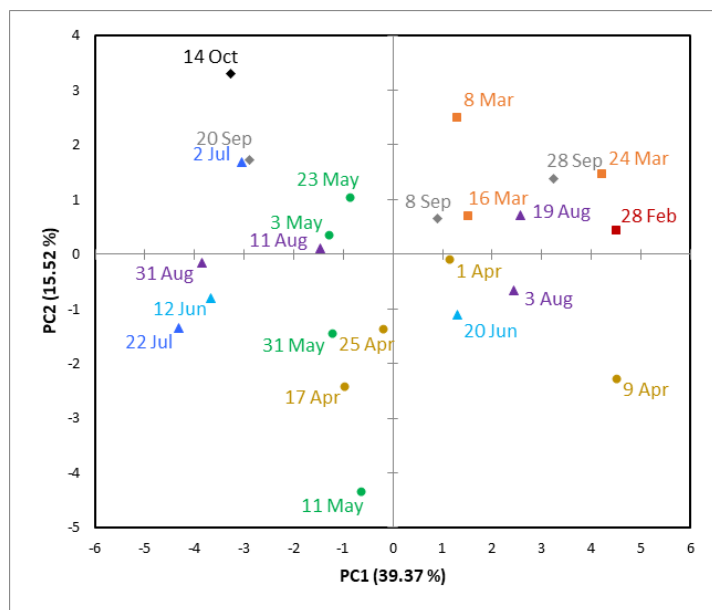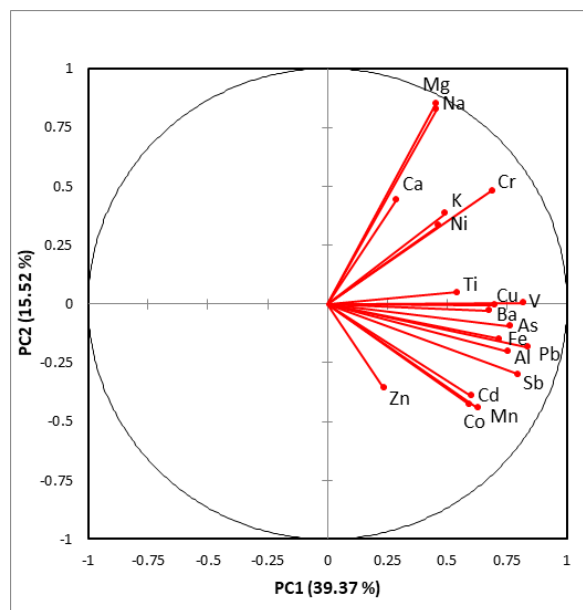
(**a**)   (**b**)

Supplementary Figure S1. Principal Component Analysis performed on the results of the sequential extraction: (**a**) score and (**b**) loading plot.