

1 **Supplementary materials**

2 **Table S1.** COPREM input source profiles (unconstrained parameters are marked in grey).

	Soil	Marine	Cu/Ni	Zn	Combustion
Al	0.0000813	0.0005	1	1	1
Si	0.00021	0.00002	1	1	1
S	0.00000052	0.88	1	1	1
K	0.0000259	0.378	1	1	1
Ca	0.0000363	0.398	1	1	1
Ti	0.0000044	0	1	1	1
V	0.00000011	0.000003	1	1	1
Cr	0.0000002	0.0000002	1	1	1
Mn	0.0000007	0.0000000995	1	1	1
Fe	0.0000505	0.000002	1	1	1
Ni	0.00000001	0.0000001	1	1	1
Cu	0.00000002	0.000001	1	0	1
Zn	0.00000008	0.000005	1	1	1
Ga	0.0000000015	0	1	1	1
As	0.00000001	0.00001	1	1	1
Se	0.0000000009	0.000004	1	1	1
Rb	0.000000031	0.00012	1	1	1
Sr	0.00000015	0.008	1	1	1
Zr	0.00000022	0	1	1	1
Pb	0.000000006	0.000004	1	1	1
Na <sup>+</sup>	0	10.5	1	1	1
Cl <sup>-</sup>	0.00000048	18.9	1	1	1
NH <sub>4</sub> <sup>+</sup>	0	0	1	1	1
NO <sub>3</sub> <sup>-</sup>	0	0	1	1	1
SO <sub>x</sub>	0.00000052	1.15	1	1	1
BC	0	0	1	1	1
Br	0.0000000025	0.065	1	1	1

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1 **Table S2.** Source origin of chemical species apportioned by PMF and COPREM. Species, where  
 2 the source apportionment by PMF and COPREM differ by >20%, are printed in grey.

Species	Soil		Marine		Cu/Ni		Zn		Combustion	
	PMF	COPREM	PMF	COPREM	PMF	COPREM	PMF	COPREM	PMF	COPREM
Al	48	58	10	0	22	13	9	11	10	19
Si	62	73	3	0	17	4	12	13	7	10
S	0	0	3	0	68	54	10	6	20	40
K	25	29	27	31	21	7	9	8	19	25
Ca	35	35	28	27	27	18	0	0	10	20
Ti	63	67	1	0	15	6	11	14	10	14
V	33	25	28	33	3	0	9	18	27	25
Cr	7	52	0	0	17	0	46	32	30	16
Mn	39	42	7	0	20	10	8	10	26	38
Fe	59	69	3	0	16	4	11	12	11	15
Ni	5	3	8	0	29	36	29	40	29	22
Cu	12	5	15	0	45	56	2	13	26	26
Zn	0	2	1	0	0	0	69	98	30	0
Ga	29	0	16	0	0	27	28	70	27	4
As	9	5	10	0	5	0	12	27	63	68
Se	12	0	5	0	19	22	20	37	44	41
Rb	43	11	5	3	16	31	10	25	26	31
Sr	13	12	42	44	22	12	2	0	21	32
Zr	69	76	0	0	4	0	13	17	14	7
Pb	1	1	4	0	15	0	7	14	72	86
Na <sup>+</sup>	1	0	70	91	6	0	6	0	17	9
Cl <sup>-</sup>	4	0	91	99	0	1	5	0	0	0
NH <sub>4</sub> <sup>+</sup>	1	0	0	0	65	29	20	24	15	46
NO <sub>3</sub> <sup>-</sup>	3	0	5	0	59	63	33	37	0	0
SO <sub>x</sub>	0	0	9	6	54	45	10	8	27	41
BC	4	0	5	1	54	37	11	1	26	61
Br	2	0	0	4	31	0	0	0	66	96

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