

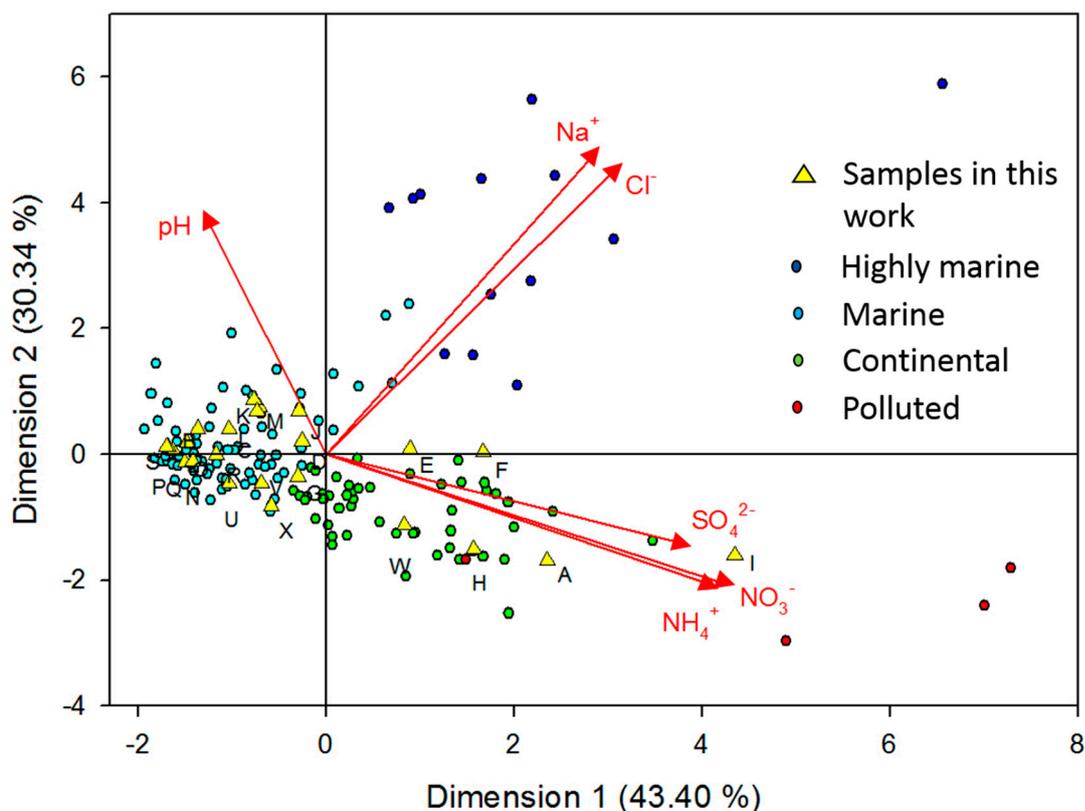
Supplementary Materials


Figure S1. Bi-plot resulting from the PCA (PCA1) and detailing: with circles, the scores of the samples of the puy de Dôme database (highly marine are represented in blue, marine in light blue, continental in green and polluted in red); with triangles, the scores of the samples presented in this work (yellow with labels); with the red vectors, the loadings of the six experimental variables pH, Cl⁻, NO₃⁻, SO₄²⁻, Na⁺ & NH₄⁺; PCA is performed using μM unit concentrations and automatic normalization of the data.

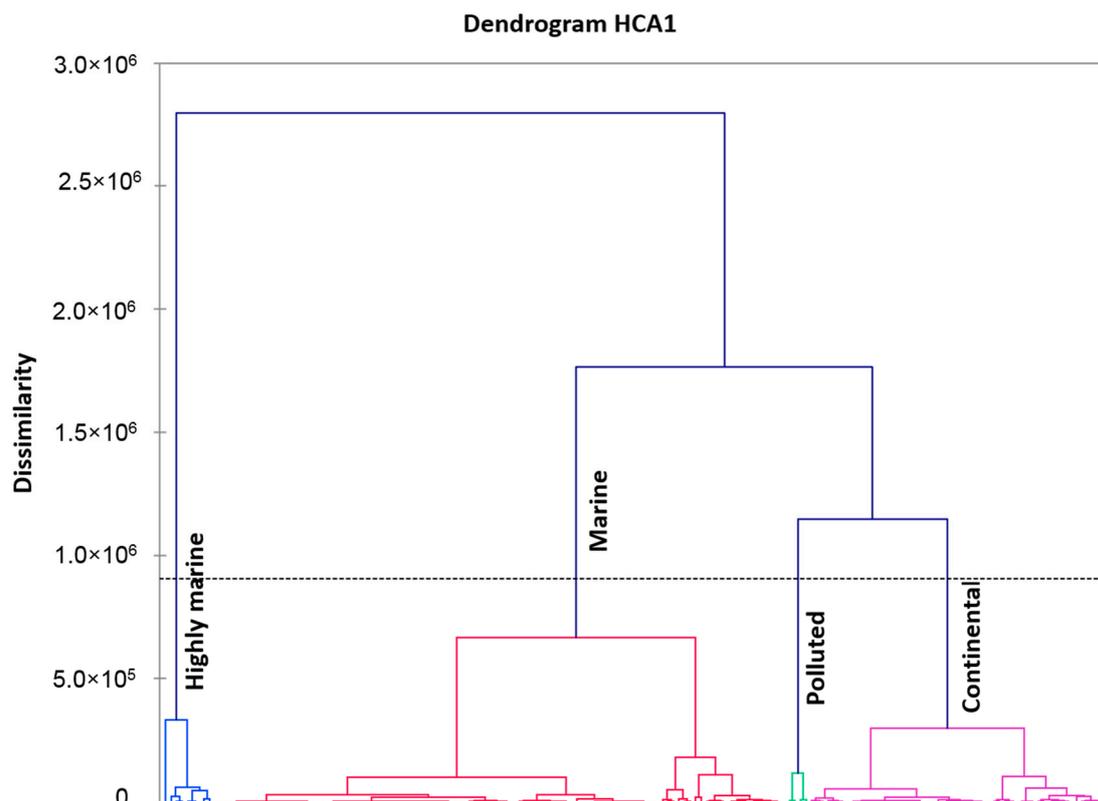


Figure S2. Dendrogram of hierarchic cluster analysis performed on the same dataset of PCA1.

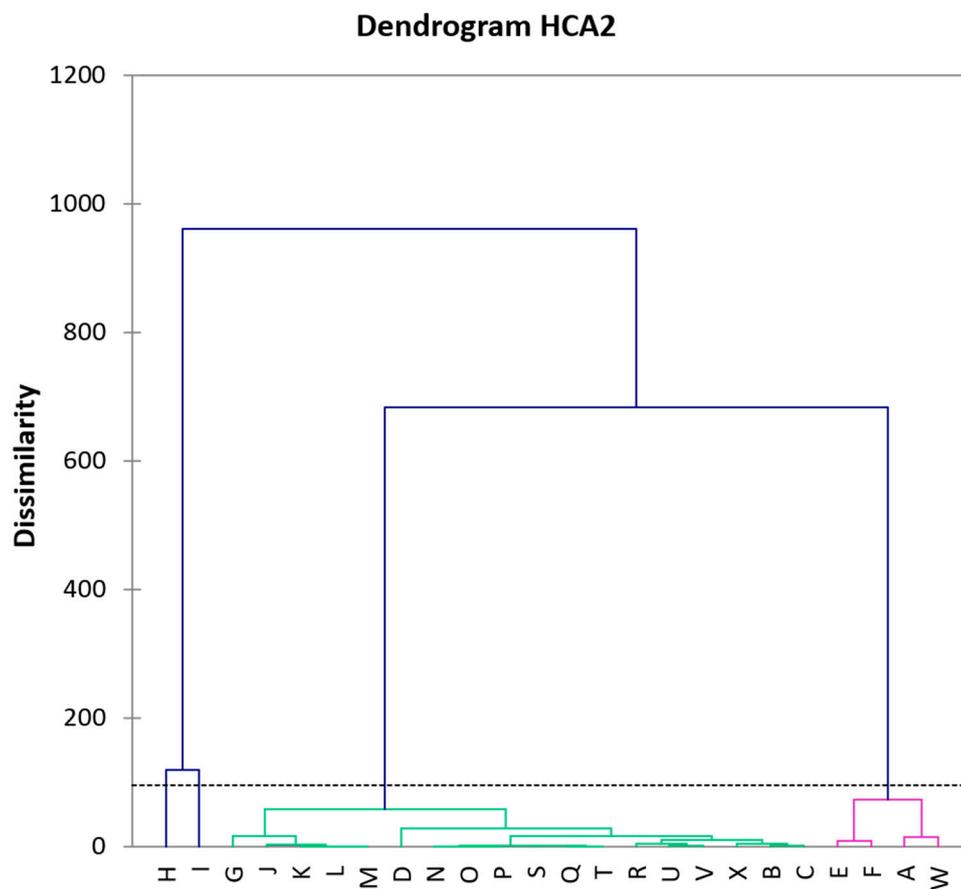


Figure S3. Dendrogram of hierarchic cluster analysis performed on the same dataset of PCA2.

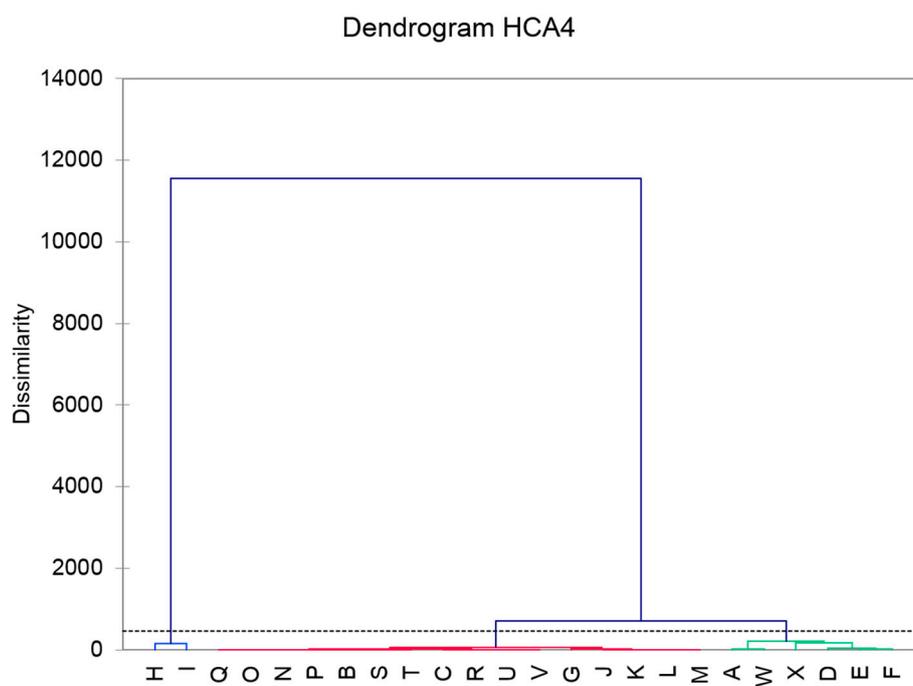


Figure S4. Dendrogram of hierarchic cluster analysis performed on the same dataset of PCA4.

Table S1. Minimum (Min), first quartile (1st Qu), median, mean, third quartile (3rd Qu) and maximum (Max) values for metal concentration of samples considered in this work. Concentrations are reported in $\mu\text{g L}^{-1}$ for all trace elements except Mg^{2+} , reported in mg L^{-1} . BLQ: below quantification limit.

Element	Min.	1st Qu.	Median	Mean	3rd Qu.	Max.	LQ	Blank
As	0.00	0.00	0.09	0.10	0.15	0.42	0.0194	BLQ
Cd	0.07	0.14	0.20	0.25	0.30	0.81	0.0153	BLQ
Co	0.00	0.00	0.00	0.02	0.02	0.17	0.0140	BLQ
Cu	0.00	1.65	5.04	10.20	8.75	55.70	1.1094	2.64
Fe	0.00	0.00	0.00	4.48	0.00	54.93	1.4154	0.80
Mg *	0.00	0.04	0.08	0.16	0.17	1.32	1.5426	0.008
Mn	0.00	0.00	0.00	1.06	0.75	9.10	0.2963	1.90
Pb	0.00	0.00	0.00	0.32	0.10	2.63	0.1360	BLQ
Rb	0.00	0.19	0.36	0.40	0.61	1.06	0.0350	BLQ
Sb	0.02	0.06	0.09	0.14	0.24	0.42	0.0120	0.03
Sr	0.34	0.69	1.07	1.58	1.57	6.15	0.1740	0.33
Ti	0.00	0.00	1.68	2.89	4.20	12.89	0.4480	BLQ
Tl	0.00	0.00	0.00	0.01	0.01	0.02	0.0011	BLQ
U	0.00	0.00	0.00	0.01	0.01	0.11	0.0033	BLQ
V	0.04	0.14	0.22	0.82	0.48	3.87	0.0153	BLQ
Zn	3.32	26.01	55.91	57.49	78.22	130.40	4.2293	7.3

Table S2. Physico-chemical parameters of sampled clouds. SO_4^{2-} , NO_3^- , Cl^- , Na^+ , NH_4^+ and Mg^{2+} are reported in mg L^{-1} ; trace metal concentrations are reported in $\mu\text{g L}^{-1}$.

Sample	Date	Start	Stop	Class	pH	SO_4^{2-}	NO_3^-	Cl^-	Na^+	NH_4^+	As	Cd	Co	Cu	Fe	Mg	Mn	Pb	Rb	Sb	Sr	Ti	Tl	U	V	Zn
A	26/03/2014	7h40	9h10	Continental	5.48	5.42	13.62	0.88	0.21	4.54	0.40	0.20	0.04	4.25	0.00	0.12	3.21	0.67	0.84	0.42	1.85	5.86	0.02	0.00	0.26	107.55
B	04/04/2014	19h45	21h45	Marine	6.24	1.3	1.24	0.68	0.11	0.71	0.00	0.22	0.02	9.79	0.00	0.04	0.28	0.00	0.38	0.06	0.92	6.86	0.01	0.00	0.47	117.82
C	04/04/2014	21h45	00h00	Marine	6.53	1.75	2.32	0.64	0.09	1.29	0.09	0.19	0.01	4.71	0.00	0.05	0.11	0.00	0.65	0.12	1.30	1.42	0.01	0.02	1.71	75.13
D	05/04/2014	1h30	4h05	Continental	6.6	2.5	4.5	1.03	0.15	2.23	0.16	0.14	0.03	5.46	0.00	0.10	0.37	0.00	0.69	0.29	0.97	1.96	0.01	0.00	3.51	85.81
E	05/04/2014	4h05	7h00	Continental	6.88	3.82	8.17	1.7	0.28	3.47	0.20	0.08	0.02	1.19	0.00	0.18	0.59	0.00	0.36	0.26	1.93	1.13	0.01	0.02	3.71	38.09
F	05/04/2014	7h35	9h40	Continental	6.8	6.48	8.25	2.66	0.4	3.83	0.25	0.12	0.03	5.64	0.00	0.34	1.97	0.00	0.68	0.37	3.84	0.00	0.01	0.02	3.91	75.69
G	05/11/2014	4h10	5h50	Marine	4.74	5.58	1.31	1.24	1.13	0	0.11	0.30	0.03	29.49	0.00	0.18	1.10	1.00	1.07	0.09	1.89	2.24	0.00	0.00	0.34	107.98
H	05/11/2014	7h00	9h10	Marine	4.06	13.38	2.46	1.46	0.96	0.71	0.00	0.14	0.01	10.64	53.04	0.08	0.00	1.10	0.20	0.04	1.16	2.88	0.00	0.00	0.15	46.78
I	05/11/2014	9h15	11h35	Marine	4.1	23.75	4.69	3.43	2.18	1.51	0.12	0.82	0.04	24.20	55.48	0.38	2.41	1.80	0.44	0.09	6.21	9.02	0.01	0.11	0.52	76.73
J	12/11/2014	15h15	17h15	Marine	5.38	3.67	1.53	2.76	1.84	0.27	0.09	0.24	0.00	1.54	0.00	0.16	0.00	0.00	0.27	0.10	1.00	5.94	0.00	0.00	0.22	66.16
K	12/11/2014	17h20	19h15	Marine	5.51	2.76	1.22	2.39	1.59	0	0.09	0.57	0.00	1.71	0.00	0.26	0.00	0.00	0.61	0.23	1.49	0.00	0.01	0.00	0.22	131.70
L	12/11/2014	19h30	21h35	Marine	5.58	2.32	1.28	2.41	1.7	0	0.14	0.52	0.00	0.00	0.00	0.16	0.00	0.00	0.37	0.07	1.26	3.70	0.00	0.00	0.15	107.94
M	12/11/2014	21h35	00h00	Marine	5.35	2.25	1.32	2.26	1.76	0	0.00	0.35	0.00	7.94	0.00	0.11	0.00	0.00	0.11	0.02	1.46	3.68	0.00	0.00	0.08	44.39
N	14/11/2014	16h45	18h50	Marine	5.51	1.08	0.38	0.21	0.34	0	0.00	0.20	0.00	1.85	0.00	0.01	0.00	0.00	0.00	0.02	0.81	0.00	0.00	0.00	0.12	67.77
O	17/11/2014	15h30	18h25	Marine	5.63	0.84	0.64	0.16	0.29	0	0.00	0.12	0.00	3.47	0.00	0.02	0.64	0.00	0.32	0.34	0.68	3.48	0.00	0.00	0.07	27.58
P	17/11/2014	18h25	21h20	Marine	5.7	0.77	0.94	0.07	0.24	0	0.00	0.31	0.00	0.03	0.00	0.00	0.00	0.00	0.09	0.08	0.86	0.77	0.00	0.01	0.04	14.36
Q	17/11/2014	21h20	00h00	Marine	5.33	1.05	1.26	0.19	0.31	0	0.08	0.51	0.00	6.50	0.00	0.03	0.00	0.00	0.16	0.07	0.34	0.00	0.00	0.00	0.08	22.33
R	18/11/2014	6h05	8h10	Marine	5.44	2.71	1.15	0.58	0.5	0	0.07	0.27	0.00	4.50	0.00	0.04	0.00	0.00	0.11	0.10	0.41	0.00	0.00	0.00	0.24	39.32
S	18/11/2014	17h10	19h10	Marine	5.7	1.58	0.84	0.42	0.4	0.04	0.00	0.07	0.01	0.95	0.00	0.02	0.00	0.00	0.23	0.04	0.71	0.00	0.00	0.01	0.14	3.74
T	18/11/2014	19h20	21h15	Marine	5.32	1.35	1.29	0.26	0.4	0	0.00	0.07	0.00	0.00	0.00	0.02	0.00	0.00	0.05	0.02	1.39	13.02	0.00	0.01	0.19	3.35
U	18/11/2014	21h20	23h30	Marine	4.85	1.92	2.43	0.63	0.39	0	0.07	0.17	0.00	8.51	0.00	0.04	0.00	0.00	0.28	0.06	0.63	6.07	0.00	0.00	0.13	34.68
V	19/11/2014	00h00	2h00	Marine	4.63	2.56	2.88	1.28	0.62	0	0.00	0.15	0.00	7.35	0.00	0.07	0.00	0.41	0.37	0.06	0.47	0.00	0.00	0.00	0.18	10.43
W	16/02/2016	10h50	14h20	Continental	5.6	2.59	13.89	0.28	0.16	2.04	0.18	0.14	0.00	51.16	0.00	0.06	5.82	2.66	0.35	0.13	0.55	0.64	0.02	0.00	0.21	20.32
X	05/06/2016	20h45	23h36	Continental	5.1	1.62	2.62	0.12	0.07	1.77	0.42	0.24	0.17	56.26	0.00	1.33	9.19	0.00	1.02	0.32	6.11	1.30	0.02	0.01	3.20	67.84

Table S3. Description of the various cloud sampling sites where trace metal concentrations were determined.

Reference	Sampling Site	State	Geographical Coordinates	Altitude (m a.s.l.)	Sampling period	Air Mass Origin	Land Use
This work	Mt. puy de Dôme	France	45°46' N 2°58' E	1465	March, April and November 2014 February and June 2016	Marine/Continental	Grassland, forest, agriculture
[37]	Mt. Elden	Arizona (USA)	35°14' N 111°35' E	2834	Summer 2005 – 2007	Continental	“Coconino national forest”
[15]	Vallombrosa	Italy	43°73' N 11°55' E	950	November 1992 – March 1995	Marine/Polluted	“Riserva statale Vallombrosa” protected forest
[17]	Kala Bagh	Pakistan	34°3' N 73°2' E	2400	19–28 July 1996	Continental	Mountainous treed region
[14]	Mt. Schmücke	Germany	50°39' N 10°47' E	605	January 2001–December 2003	Continental	Thuringian forest
[16]	Mt. Lu	China	29°35' N 115°59' E	1165	27 August–9 September, 2011	Polluted	Large, non-ferrous industries and coal-fired power plants
[38]	Mt. Tai	China	36°16' N 117°06' E	1543	20 March–20 April, 2007 15 October–15 November 2008	Polluted	Steel industries and mining activities
[6]	Mt. Brocken	Germany	51°80' N 10°67' E	1141	May–June 1997 October 1997	Continental	Grassland, forest, agriculture

Table S4. Correlation matrix obtained by the PCA3 analysis considering the concentration of Cl^- , NO_3^- , SO_4^{2-} , Na^+ and NH_4^+ and the total metal concentration. The color code depicts in blue values between 0 and 0.2, in light blue values between 0.2 and 0.4, in light orange values between 0.4 and 0.6, in orange values between 0.6 and 0.8 and in dark orange values between 0.8 and 1.

Variables	SO_4^{2-}	NO_3^-	Cl^-	Na^+	NH_4^+	Metal	pH
SO_4^{2-}	1	0.21	0.60	0.52	0.25	0.24	-0.50
NO_3^-	0.21	1	0.09	-0.24	0.85	0.10	0.22
Cl^-	0.60	0.09	1	0.83	0.18	0.19	-0.11
Na^+	0.52	-0.24	0.83	1	-0.30	0.10	-0.47
NH_4^+	0.25	0.85	0.18	-0.30	1	0.31	0.44
Metal	0.24	0.10	0.19	0.10	0.31	1	-0.14
pH	-0.50	0.22	-0.11	-0.47	0.44	-0.14	1