Supporting Information for:

Transboundary and local air pollutants in western Japan distinguished on the basis of ratios of metallic elements in size-segregated aerosols

Yuta Taniguchi¹, Kojiro Shimada², Akinori Takami³, Neng-Huei Lin^{2,4}, Chak K. Chan⁵, Yong Pyo Kim^{2,6}, Shiro Hatakeyama^{1,2,7}*

¹Graduate School of Agriculture, Tokyo University of Agriculture and Technology, Fuchu, Tokyo 183-8509, Japan
²Global Innovation Research Organization, Tokyo University of Agriculture and Technology, Fuchu, Tokyo 183-8538, Japan
³National Institute for Environmental Studies, Tsukuba, Ibaraki 305-0053, Japan
⁴ Department of Atmospheric Science and Department of Chemistry, National Central University, Chung-Li, Taoyuan 32001, Taiwan
⁵School of Energy and Environment, City University of Hong Kong, Hong Kong, China
⁶Department of Chemical Engineering & Materials Science and Department of Environmental Science & Engineering, Ewha Womans University, Seoul 03760, Republic of Korea
⁷Center for Environmental Science in Saitama, Kazo 347-0115, Saitama, Japan

The Supporting Information contains 2 Table and 4 Figures. Supplementary Figures and Table are submitted as ppt files.

Tables

Table S1. Enrichment factors of 16 metallic elements by particle size in each season at

Kumamoto.

Table S2. Enrichment factors of 16 metallic elements by particle size in each season at

Cape Hedo.

Figures

Fig. S1. Backward trajectory of air masses arriving at Kumamoto during sampling periods in spring (a), summer (b), autumn (c), and winter (d). Four trajectories per day at 6-h intervals were calculated for each observation period.

Fig. S2. Average Pb/Cu ratio (a) and V/As ratio (b) in each season at Kumamoto and Cape Hedo. Concentrations of Pb, V, and As were measured in the $0.5-1 \mu m$ size fraction, and Cu concentration was measured in the $2.5-10 \mu m$ size fraction.

Fig. S3. Results of backward trajectory starting from Kumamoto on 16 October 2014 (a) and on 31 October 2015 (b) and from Cape Hedo on 17 October 2014 (c) and 31 October 2015 (d). The initial altitude and calculation time were set to 500 m and 72 h, respectively.

Fig. S4. Correlation between the mass proportions of 16 elements in (a) coarse particles and (b) fine particles to the total mass of these elements at Kumamoto and Cape Hedo.

EF (sea salt)	Size	Na	Mg	Al	K	V	Mn	Fe	Cu	As	Se	\mathbf{Sr}	Cd	\mathbf{Sb}	Ba	Pb	Bi
Spring	< 0.1	1.0E+00	2.4E+00	2.8E+07	4.8E+01	3.5E+04	8.8E+05	2.9E+06	1.4E+07	1.7E+04	3.5E+05	6.0E+00	7.6E+05	1.0E+05	9.5E+03	7.1E+07	2.7E+08
	0.5 - 1	1.0E+00	2.6E+00	2.3E+07	7.3E+01	5.9E+04	3.7E+06	6.5E+06	4.1E+06	8.2E+04	1.5E+06	7.3E+00	2.3E+06	2.9E+05	1.2E+04	3.8E+08	1.9E+09
	1 - 2.5	1.0E+00	3.7E+00	2.5E+07	1.9E+01	1.4E+04	1.5E+06	4.0E+06	1.2E+06	1.8E+04	2.3E+05	9.8E+00	5.3E+05	8.3E+04	1.1E+04	6.6E+07	3.3E+08
	2.5 - 10	1.0E+00	3.0E+00	1.7E+07	9.4E+00	6.5E+03	7.4E+05	2.7E+06	5.4E+05	2.7E+03	3.4E+04	6.8E+00	6.3E+04	2.4E+04	6.4E+03	8.1E+06	2.7E+07
	10<	1.0E+00	3.3E+00	2.1E+07	9.0E+00	8.4E+03	1.0E+06	4.2E+06	7.3E+05	2.6E+03	-	7.0E+00	7.7E+04	1.7E+04	7.1E+03	7.9E+06	5.1E+07
Summer	< 0.1	1.0E+00	3.2E+00	9.1E+07	1.2E+02	7.3E+05	5.4E+06	1.7E+07	1.3E+07	-	-	2.5E+01	-	-	3.2E+04	-	3.5E+09
	0.5 - 1	1.0E+00	1.7E+00	9.0E+06	8.5E+01	4.5E+05	2.6E+06	2.1E+06	9.0E+06	9.2E+04	1.7E+06	2.9E+01	2.5E+06	3.6E+05	3.0E+04	3.4E+08	2.1E+09
	1-2.5	1.0E+00	1.1E+00	2.5E+06	3.2E+00	5.7E+03	1.7E+05	4.7E+05	2.5E+05	-	4.5E+04	2.5E+00	1.6E+05	1.2E+04	2.0E+03	1.3E+07	5.7E+07
	2.5 - 10	1.0E+00	1.1E+00	2.6E+06	2.1E+00	1.7E+03	1.1E+05	4.5E+05	1.3E+05	-	1.4E+04	1.5E+00	-	-	1.1E+03	2.3E+06	9.6E+06
	10<	1.0E+00	1.8E+00	1.7E+07	5.4E+00	7.3E+03	5.0E+05	2.2E+06	7.0E+05	-	-	4.0E+00	-	-	4.1E+03	-	-
Autumn	< 0.1	1.0E+00	4.9E+00	8.2E+07	1.0E+02	2.1E+05	6.7E+06	1.3E+07	1.7E+05	9.6E+04	1.0E+06	1.6E+01	3.7E+06	5.1E+05	1.9E+04	2.5E+08	1.9E+09
	0.5 - 1	1.0E+00	1.8E+00	1.5E+07	6.4E+01	6.9E+04	4.1E+06	4.2E+06	6.2E+05	8.1E+04	1.1E+06	6.6E+00	2.2E+06	3.3E+05	7.3E+03	2.4E+08	1.7E+09
	1-2.5	1.0E+00	2.1E+00	1.4E+07	1.0E+01	9.1E+03	1.1E+06	3.1E+06	4.3E+05	1.5E+04	9.6E+04	5.7E+00	4.0E+05	6.6E+04	8.1E+03	4.4E+07	2.4E+08
	2.5 - 10	1.0E+00	2.0E+00	1.3E+07	6.4E+00	5.2E+03	6.7E+05	2.6E+06	5.1E+05	1.9E+03	1.4E+04	4.7E+00	5.7E+04	2.1E+04	5.4E+03	6.9E + 06	2.6E+07
	10<	1.0E+00	2.0E+00	1.3E+07	4.4E+00	4.5E+03	5.4E+05	2.3E+06	8.8E+04	1.5E+03	8.3E+03	3.9E+00	1.0E+05	1.4E+04	3.2E+03	9.6E+06	2.1E+07
Winter	< 0.1	1.0E+00	1.3E+00	2.2E+07	6.3E+01	1.6E+04	5.9E+05	6.1E+06	1.7E+06	3.7E+04	5.4E+05	8.5E+00	2.3E+06	2.4E+05	-	2.7E+07	4.6E+08
	0.5 - 1	1.0E+00	1.6E+00	1.8E+07	5.2E+01	3.3E+04	3.1E+06	3.7E+06	2.5E+06	8.9E+04	1.2E+06	5.7E+00	2.4E+06	3.4E+05	-	4.2E+08	1.5E+09
	1-2.5	1.0E+00	1.7E+00	1.1E+07	9.7E+00	8.5E+03	9.5E + 05	1.9E+06	1.0E+06	1.8E+04	1.9E+05	4.2E+00	5.5E+05	6.1E+04	1.1E+04	8.2E+07	2.9E+08
	$2.5 \cdot 10$	1.0E+00	1.6E+00	8.7E+06	3.5E+00	3.9E+03	3.5E+05	1.4E+06	4.2E+05	2.1E+03	2.3E+04	3.1E+00	6.9E+04	1.8E+04	5.0E+03	5.6E + 06	3.5E+07
	10<	1.0E+00	1.5E+00	5.6E + 06	2.6E+00	2.3E+03	2.0E+05	7.9E+05	1.8E+05	-	-	1.9E+00	-	8.4E+03	-	1.8E+06	2.7E+07
EF (soil)	Size	Na	Mg	Al	K	V	Mn	Fe	Cu	As	Se	Sr	Cd	Sb	Ва	Pb	Bi
EF (soil) Spring	Size <0.1	Na 2.6E+00	Mg 1.6E+00	Al 1.0E+00	K 3.8E+00	V 9.4E+00	Mn 2.1E+00	Fe 1.6E+00	Cu 4.3E+02	As 1.5E+02	Se 5.7E+00	Sr 9.6E-01	Cd 5.4E+02	Sb 8.7E+02	Ba 1.8E+00	Pb 8.0E+01	Bi 3.4E+02
EF (soil) Spring	Size <0.1 0.5-1	Na 2.6E+00 2.9E+00	Mg 1.6E+00 1.8E+00	Al 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00	V 9.4E+00 1.9E+01	Mn 2.1E+00 1.1E+01	Fe 1.6E+00 2.4E+00	Cu 4.3E+02 9.8E+01	As 1.5E+02 7.6E+02	Se 5.7E+00 2.5E+01	Sr 9.6E-01 1.4E+00	Cd 5.4E+02 2.1E+03	Sb 8.7E+02 3.3E+03	Ba 1.8E+00 3.3E+00	Pb 8.0E+01 4.8E+02	Bi 3.4E+02 2.7E+03
EF (soil) Spring	Size <0.1 0.5-1 1-2.5	Na 2.6E+00 2.9E+00 4.2E+00	Mg 1.6E+00 1.8E+00 2.6E+00	Al 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00	V 9.4E+00 1.9E+01 4.1E+00	Mn 2.1E+00 1.1E+01 3.2E+00	Fe 1.6E+00 2.4E+00 1.6E+00	Cu 4.3E+02 9.8E+01 2.6E+01	As 1.5E+02 7.6E+02 1.0E+02	Se 5.7E+00 2.5E+01 1.5E+00	Sr 9.6E-01 1.4E+00 1.3E+00	Cd 5.4E+02 2.1E+03 3.4E+02	Sb 8.7E+02 3.3E+03 4.6E+02	Ba 1.8E+00 3.3E+00 2.3E+00	Pb 8.0E+01 4.8E+02 6.9E+01	Bi 3.4E+02 2.7E+03 3.1E+02
EF (soil) Spring	Size <0.1 0.5-1 1-2.5 2.5-10	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00	Mg 1.6E+00 1.8E+00 2.6E+00 4.1E+00	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 1.9E+00	V 9.4E+00 1.9E+01 4.1E+00 2.6E+00	Mn 2.1E+00 1.1E+01 3.2E+00 2.3E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00	Cu 4.3E+02 9.8E+01 2.6E+01 1.5E+01	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01	Se 5.7E+00 2.5E+01 1.5E+00 2.8E-01	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02	Ba 1.8E+00 3.3E+00 2.3E+00 1.8E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01
EF (soil) Spring	Size <0.1 0.5-1 1-2.5 2.5-10 10<	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01	Mg 1.6E+00 1.8E+00 2.6E+00 4.1E+00 5.7E+00	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 1.9E+00 3.0E+00	V 9.4E+00 1.9E+01 4.1E+00 2.6E+00 2.3E+00	Mn 2.1E+00 1.1E+01 3.2E+00 2.3E+00 2.5E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00	Cu 4.3E+02 9.8E+01 2.6E+01 1.5E+01 2.7E+01	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01	Se 5.7E+00 2.5E+01 1.5E+00 2.8E-01	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+02	Ba 1.8E+00 3.3E+00 2.3E+00 1.8E+00 2.2E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01
EF (soil) Spring Summer	Size <0.1 0.5-1 1-2.5 2.5-10 10< <0.1	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E-01	Mg 1.6E+00 1.8E+00 2.6E+00 4.1E+00 5.7E+00 5.5E-01	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 1.9E+00 3.0E+00 2.9E+00	V 9.4E+00 1.9E+01 4.1E+00 2.6E+00 2.3E+00 5.3E+01	Mn 2.1E+00 1.1E+01 3.2E+00 2.3E+00 2.5E+00 3.1E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00	Cu 4.3E+02 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01	Se 5.7E+00 2.5E+01 1.5E+00 2.8E-01	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.0E+00	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+02	Ba 1.8E+00 3.3E+00 2.3E+00 1.8E+00 2.2E+00 2.0E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02
EF (soil) Spring Summer	Size <0.1 0.5-1 1-2.5 2.5-10 10< <0.1 0.5-1	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E-01 6.6E+00	Mg 1.6E+00 1.8E+00 2.6E+00 4.1E+00 5.7E+00 5.5E-01 3.0E+00	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01	V 9.4E+00 1.9E+01 4.1E+00 2.6E+00 2.3E+00 5.3E+01 3.4E+02	Mn 2.1E+00 1.1E+01 3.2E+00 2.3E+00 2.5E+00 3.1E+00 1.5E+01	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 2.2E+00	Cu 4.3E+02 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01 6.5E+02	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01 - 1.6E+03	Se 5.7E+00 2.5E+01 1.5E+00 2.8E-01 - - 5.1E+01	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.0E+00 1.2E+01	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 - 4.5E+03	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+02 7.8E+03	Ba 1.8E+00 2.3E+00 1.8E+00 2.2E+00 2.0E+00 1.9E+01	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 - 9.2E+02	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02 5.9E+03
EF (soil) Spring Summer	Size <0.1 0.5-1 1-2.5 2.5-10 10< <0.1 0.5-1 1-2.5	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E-01 6.6E+00 2.4E+01	Mg 1.6E+00 1.8E+00 2.6E+00 4.1E+00 5.7E+00 5.5E-01 3.0E+00 7.0E+00	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01 2.8E+00	V 9.4E+00 1.9E+01 4.1E+00 2.6E+00 2.3E+00 5.3E+01 3.4E+02 1.5E+01	Mn 2.1E+00 1.1E+01 3.2E+00 2.3E+00 2.5E+00 3.1E+00 1.5E+01 3.6E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 2.2E+00 1.7E+00	Cu 4.3E+02 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01 6.5E+02 6.3E+01	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01 1.6E+03	Se 5.7E+00 2.5E+01 1.5E+00 2.8E-01 - 5.1E+01 4.8E+00	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.0E+00 1.2E+01 3.6E+00	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 4.5E+03 1.1E+03	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+02 7.8E+03 9.3E+02	Ba 1.8E+00 3.3E+00 2.3E+00 1.8E+00 2.2E+00 2.0E+00 1.9E+01 4.6E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 - 9.2E+02 1.3E+02	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02 5.9E+03 5.8E+02
EF (soil) Spring Summer	Size <0.1 0.5-1 1-2.5 2.5-10 10< <0.1 0.5-1 1-2.5 2.5-10	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E-01 6.6E+00 2.4E+01 2.3E+01	Mg 1.6E+00 1.8E+00 2.6E+00 4.1E+00 5.7E+00 5.5E-01 3.0E+00 6.5E+00	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01 2.8E+00 1.8E+00	V 9.4E+00 1.9E+01 4.1E+00 2.6E+00 2.3E+00 5.3E+01 3.4E+02 1.5E+01 4.4E+00	Mn 2.1E+00 1.1E+01 3.2E+00 2.3E+00 2.5E+00 3.1E+00 1.5E+01 3.6E+00 2.4E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 1.7E+00 1.6E+00	Cu 4.3E+02 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01 6.5E+02 6.3E+01 3.2E+01	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01 1.6E+03	Se 5.7E+00 2.5E+01 1.5E+00 2.8E+01 - 5.1E+01 4.8E+00 1.5E+00	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.0E+00 1.2E+01 3.6E+00 2.1E+00	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 4.5E+03 1.1E+03	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+02 7.8E+03 9.3E+02	Ba 1.8E+00 2.3E+00 2.3E+00 1.8E+00 2.2E+00 2.0E+00 1.9E+01 4.6E+00 2.5E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 - 9.2E+02 1.3E+02 2.1E+01	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02 5.9E+03 5.8E+02 9.4E+01
EF (soil) Spring Summer	Size <0.1 0.5-1 1-2.5 2.5-10 10< <0.1 0.5-1 1-2.5 2.5-10 10<	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E-01 6.6E+00 2.4E+01 2.3E+01 3.4E+00	Mg 1.6E+00 1.8E+00 2.6E+00 4.1E+00 5.7E+00 5.5E-01 3.0E+00 7.0E+00 6.5E+00 1.7E+00	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01 2.8E+00 6.8E-01	V 9.4E+00 1.9E+01 4.1E+00 2.6E+00 2.3E+00 5.3E+01 3.4E+02 1.5E+01 4.4E+00 2.8E+00	Mn 2.1E+00 1.1E+01 3.2E+00 2.3E+00 2.5E+00 3.1E+00 1.5E+01 3.6E+00 1.5E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 1.7E+00 1.6E+00 1.2E+00	Cu 4.3E+02 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01 6.5E+02 6.3E+01 3.2E+01 2.6E+01	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01 1.6E+03	Se 5.7E+00 2.5E+01 1.5E+00 2.8E+01 - 5.1E+01 4.8E+00 1.5E+00	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.0E+00 1.2E+01 3.6E+00 2.1E+00 8.5E-01	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 4.5E+03 1.1E+03	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+02 7.8E+03 9.3E+02	Ba 1.8E+00 2.3E+00 2.3E+00 1.8E+00 2.2E+00 2.0E+00 1.9E+01 4.6E+00 2.5E+00 1.4E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 9.2E+02 1.3E+02 2.1E+01	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02 5.9E+03 5.8E+02 9.4E+01
EF (soil) Spring Summer	Size <0.1 0.5-1 1-2.5 2.5-10 10< <0.1 0.5-1 1-2.5 2.5-10 10< <0.1	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E-01 6.6E+00 2.4E+01 2.3E+01 3.4E+00 7.3E-01	Mg 1.6E+00 2.6E+00 4.1E+00 5.7E+00 5.5E-01 3.0E+00 7.0E+00 6.5E+00 1.7E+00 9.0E-01	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01 2.8E+00 1.8E+00 6.8E-01 2.2E+00	V 9.4E+00 1.9E+01 4.1E+00 2.6E+00 2.3E+00 5.3E+01 3.4E+02 1.5E+01 4.4E+00 2.8E+00 1.4E+01	Mn 2.1E+00 3.2E+00 2.3E+00 2.5E+00 3.1E+00 1.5E+01 3.6E+00 2.4E+00 4.5E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 1.7E+00 1.6E+00 1.2E+00 6.5E+00	Cu 4.3E+02 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01 6.5E+02 6.3E+01 3.2E+01 2.6E+01 1.4E+00	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01	Se 5.7E+00 2.5E+01 1.5E+00 2.8E+01 - 5.1E+01 4.8E+00 1.5E+00 -	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.2E+01 3.6E+00 2.1E+00 8.5E-01 8.5E-01	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 3.2E+01 4.5E+03 1.1E+03 	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+02 7.8E+03 9.3E+02 1.6E+03	Ba 1.8E+00 3.3E+00 2.3E+00 1.8E+00 2.2E+00 2.0E+00 1.9E+01 4.6E+00 2.5E+00 1.4E+00 1.5E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 - 9.2E+02 1.3E+02 2.1E+01 8.4E+01	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02 5.9E+03 5.8E+02 9.4E+01
EF (soil) Spring Summer	Size <0.1	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E-01 6.6E+00 2.4E+01 2.3E+01 3.4E+00 7.3E-01 4.0E+00	Mg 1.6E+00 2.6E+00 4.1E+00 5.7E+00 5.5E-01 3.0E+00 7.0E+00 6.5E+00 1.7E+00 9.0E-01 1.7E+00	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01 2.8E+00 1.8E+00 6.8E-01 2.2E+00 7.7E+00	V 9.4E+00 1.9E+01 4.1E+00 2.6E+00 2.3E+00 5.3E+01 3.4E+02 1.5E+01 4.4E+00 2.8E+00 1.4E+01 2.6E+01	Mn 2.1E+00 3.2E+00 2.3E+00 2.5E+00 3.1E+00 1.5E+01 3.6E+00 2.4E+00 1.5E+00 1.5E+01	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 1.7E+00 1.6E+00 1.6E+00 1.6E+00 1.6E+00 1.2E+00 6.5E+00 2.6E+00	Cu 4.3E+02 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01 6.5E+02 6.3E+01 3.2E+01 2.6E+01 1.4E+00 2.6E+01	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01	Se 5.7E+00 2.5E+01 1.5E+00 2.8E+01 - 5.1E+01 4.8E+00 1.5E+00 1.5E+00 1.9E+01	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.2E+01 3.6E+00 2.1E+00 8.5E-01 1.5E+00	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 3.2E+01 1.1E+03 1.1E+03 2.9E+03	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+02 - 7.8E+03 9.3E+02 - 1.6E+03 5.4E+03	Ba 1.8E+00 3.3E+00 2.3E+00 1.8E+00 2.2E+00 2.0E+00 1.9E+01 4.6E+00 2.5E+00 1.4E+00 3.3E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 - 9.2E+02 1.3E+02 2.1E+01 - 8.4E+01 5.5E+02	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02 5.9E+03 5.8E+02 9.4E+01 5.7E+02 3.4E+03
EF (soil) Spring Summer	Size <0.1	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E-01 6.6E+00 2.4E+01 2.3E+01 3.4E+00 7.3E-01 4.0E+00 4.6E+00	Mg 1.6E+00 2.6E+00 4.1E+00 5.7E+00 5.5E-01 3.0E+00 7.0E+00 6.5E+00 1.7E+00 9.0E-01 1.7E+00 2.4E+00	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01 2.8E+00 1.8E+00 6.8E-01 2.2E+00 7.7E+00 1.6E+00	V 9.4E+00 1.9E+01 2.6E+00 2.3E+00 5.3E+01 3.4E+02 1.5E+01 4.4E+00 2.8E+00 1.4E+01 2.6E+01 4.5E+00	Mn 2.1E+00 3.2E+00 2.3E+00 2.5E+00 3.1E+00 1.5E+01 3.6E+00 2.4E+00 1.5E+00 1.5E+01 4.4E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 1.7E+00 1.6E+00 1.6E+00 2.2E+00 2.2E+00 2.6E+00 2.6E+00 2.0E+00	Cu 4.3E+02 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01 6.5E+02 6.3E+01 3.2E+01 2.6E+01 2.6E+01 2.6E+01 2.7E+01	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01	Se 5.7E+00 2.5E+01 1.5E+00 2.8E+01 - 5.1E+01 4.8E+00 1.5E+00 - 1.9E+01 1.9E+00	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.2E+01 3.6E+00 2.1E+00 8.5E-01 1.5E+00 1.5E+00	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 3.2E+01 4.5E+03 1.1E+03 - 1.0E+03 2.9E+03 4.9E+02	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+02 - 7.8E+03 9.3E+02 - 1.6E+03 5.4E+03 9.2E+02	Ba 1.8E+00 3.3E+00 2.3E+00 1.8E+00 2.2E+00 2.0E+00 1.9E+01 4.6E+00 2.5E+00 1.4E+00 3.3E+00 3.4E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 - 9.2E+02 1.3E+02 2.1E+01 - 8.4E+01 5.5E+02 7.4E+01	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02 5.9E+03 5.8E+02 9.4E+01 5.7E+02 3.4E+03 4.6E+02
EF (soil) Spring Summer	Size <0.1	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E+00 2.4E+01 2.3E+01 3.4E+00 7.3E-01 4.0E+00 4.6E+00 5.0E+00	Mg 1.6E+00 1.8E+00 2.6E+00 4.1E+00 5.7E+00 5.5E-01 3.0E+00 7.0E+00 6.5E+00 1.7E+00 9.0E-01 1.7E+00 2.4E+00 2.5E+00	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01 2.8E+00 1.8E+00 6.8E-01 2.2E+00 7.7E+00 1.6E+00 1.1E+00	V 9.4E+00 1.9E+01 2.6E+00 2.3E+00 5.3E+01 3.4E+02 1.5E+01 4.4E+00 2.8E+00 1.4E+01 2.6E+01 4.5E+00 2.7E+00	Mn 2.1E+00 3.2E+00 2.3E+00 2.5E+00 3.1E+00 1.5E+01 3.6E+00 1.5E+00 1.5E+00 1.5E+01 4.4E+00 2.8E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 1.7E+00 1.6E+00 1.6E+00 2.2E+00 2.2E+00 2.6E+00 2.6E+00 2.0E+00 1.9E+00	Cu 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01 6.5E+02 6.3E+01 3.2E+01 2.6E+01 2.6E+01 2.6E+01 2.7E+01 2.5E+01	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01	Se 5.7E+00 2.5E+01 1.5E+00 2.8E+01 - 5.1E+01 4.8E+00 1.5E+00 - 1.5E+00 1.9E+01 1.9E+00 2.6E-01	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.2E+01 3.6E+00 2.1E+00 8.5E-01 1.5E+00 1.5E+00 1.3E+00	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 4.5E+03 1.1E+03 2.9E+03 4.9E+02 7.0E+01	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+02 - 7.8E+03 9.3E+02 - 1.6E+03 5.4E+03 9.2E+02 3.2E+02	Ba 1.8E+00 3.3E+00 2.3E+00 1.8E+00 2.2E+00 2.0E+00 1.9E+01 4.6E+00 2.5E+00 1.4E+00 3.3E+00 3.4E+00 2.4E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 9.2E+02 1.3E+02 2.1E+01 5.5E+02 7.4E+01 1.2E+01	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02 5.9E+03 5.8E+02 9.4E+01 5.7E+02 3.4E+03 4.6E+02 5.4E+01
EF (soil) Spring Summer	Size <0.1 0.5-1 1-2.5 2.5-10 10< <0.1 0.5-1 1-2.5 2.5-10 10< <0.1 0.5-1 1-2.5 2.5-10 10< 0.5-1 1-2.5 2.5-10 10	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E+00 2.4E+01 2.3E+01 3.4E+00 7.3E-01 4.0E+00 5.0E+00 4.6E+00	$\begin{array}{c} Mg \\ 1.6E+00 \\ 1.8E+00 \\ 2.6E+00 \\ 4.1E+00 \\ 5.7E+00 \\ \hline 5.5E-01 \\ 3.0E+00 \\ 7.0E+00 \\ 6.5E+00 \\ 1.7E+00 \\ 9.0E-01 \\ 1.7E+00 \\ 2.4E+00 \\ 2.5E+00 \\ 2.4E+00 \\ \hline 2.4E+00 \\ \hline \end{array}$	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01 2.8E+00 1.8E+00 6.8E-01 2.2E+00 7.7E+00 1.6E+00 1.1E+00 7.4E-01	V 9.4E+00 1.9E+01 4.1E+00 2.6E+00 2.3E+00 5.3E+01 3.4E+02 1.5E+01 4.4E+00 2.8E+00 1.4E+01 2.6E+01 4.5E+00 2.7E+00 2.3E+00	Mn 2.1E+00 3.2E+00 2.3E+00 2.5E+00 3.1E+00 1.5E+01 3.6E+00 2.4E+00 1.5E+00 1.5E+01 4.4E+00 2.8E+00 2.2E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 1.7E+00 1.6E+00 1.6E+00 2.2E+00 2.2E+00 1.6E+00 2.6E+00 2.6E+00 2.0E+00 1.9E+00 1.7E+00	Cu 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01 6.5E+02 6.3E+01 3.2E+01 2.6E+01 2.6E+01 2.7E+01 2.5E+01 4.2E+00	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01 - 1.6E+03 - - 1.9E+02 1.0E+03 1.7E+02 3.2E+01 1.8E+01	Se 5.7E+00 2.5E+01 1.5E+00 2.8E+01 - 5.1E+01 4.8E+00 1.5E+00 - 1.5E+00 1.9E+01 1.9E+01 2.6E-01 1.6E-01	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.2E+01 3.6E+00 2.1E+00 8.5E-01 1.5E+00 1.5E+00 1.3E+00 1.3E+00 1.1E+00	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 4.5E+03 1.1E+03 2.9E+03 4.9E+02 7.0E+01 1.3E+02	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+02 7.8E+03 9.3E+02 1.6E+03 5.4E+03 9.2E+02 3.2E+02 2.0E+02	Ba 1.8E+00 3.3E+00 2.3E+00 1.8E+00 2.2E+00 2.0E+00 1.9E+01 4.6E+00 2.5E+00 1.4E+00 3.3E+00 3.4E+00 2.4E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 9.2E+02 1.3E+02 2.1E+01 5.5E+02 7.4E+01 1.2E+01 1.9E+01	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02 5.9E+03 5.8E+02 9.4E+01 5.7E+02 3.4E+03 4.6E+02 5.4E+01 4.1E+01
EF (soil) Spring Summer Autumn Winter	Size <0.1	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E+00 2.4E+01 2.3E+01 3.4E+00 7.3E-01 4.0E+00 5.0E+00 4.6E+00 2.7E+00	Mg 1.6E+00 1.8E+00 2.6E+00 4.1E+00 5.7E+00 5.5E-01 3.0E+00 7.0E+00 6.5E+00 1.7E+00 9.0E-01 1.7E+00 2.4E+00 2.5E+00 9.4E+00	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01 2.8E+00 1.8E+00 6.8E-01 2.2E+00 7.7E+00 1.6E+00 1.1E+00 7.4E-01	V 9.4E+00 1.9E+01 2.6E+00 2.3E+00 5.3E+01 3.4E+02 1.5E+01 4.4E+00 2.8E+00 1.4E+01 2.6E+01 4.5E+00 2.7E+00 2.3E+00 4.8E+00	Mn 2.1E+00 3.2E+00 2.3E+00 2.5E+00 3.1E+00 1.5E+01 3.6E+00 1.5E+00 1.5E+00 1.5E+01 4.4E+00 2.8E+00 2.2E+00 1.5E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 1.7E+00 1.6E+00 1.6E+00 2.2E+00 2.2E+00 1.6E+00 2.6E+00 2.0E+00 1.9E+00 1.7E+00 2.6E+00 2.6E+00 2.6E+00	Cu 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01 6.5E+02 6.3E+01 3.2E+01 2.6E+01 2.6E+01 2.7E+01 2.5E+01 4.2E+00 5.0E+01	As 1.5E+02 7.6E+02 2.2E+01 1.9E+01 1.6E+03	Se 5.7E+00 2.5E+01 1.5E+00 2.8E+01 - 5.1E+01 4.8E+00 1.5E+00 1.5E+00 1.9E+01 1.9E+01 1.6E-01 6.6E+00	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.2E+01 3.6E+00 2.1E+00 8.5E-01 1.5E+00 1.5E+00 1.3E+00 1.4E+00	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 4.5E+03 1.1E+03 2.9E+03 4.9E+02 7.0E+01 1.3E+02 1.7E+03	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+03 9.3E+02 - 1.6E+03 5.4E+03 9.2E+02 3.2E+02 2.0E+02 2.2E+03	Ba 1.8E+00 3.3E+00 2.3E+00 1.8E+00 2.2E+00 2.0E+00 1.9E+01 4.6E+00 2.5E+00 1.4E+00 3.3E+00 3.4E+00 2.4E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 1.3E+02 2.1E+01 5.5E+02 7.4E+01 1.2E+01 1.9E+01 3.0E+01	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02 5.9E+03 5.8E+02 9.4E+01 3.4E+03 4.6E+02 5.4E+01 4.1E+01 5.4E+02
EF (soil) Spring Summer Autumn Winter	Size <0.1	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E+00 2.4E+01 2.3E+01 3.4E+00 7.3E-01 4.0E+00 5.0E+00 4.6E+00 5.0E+00 3.3E+00	$\begin{array}{c} Mg \\ 1.6E+00 \\ 2.6E+00 \\ 4.1E+00 \\ 5.7E+00 \\ 5.7E+00 \\ 5.5E-01 \\ 3.0E+00 \\ 7.0E+00 \\ 6.5E+00 \\ 1.7E+00 \\ 9.0E-01 \\ 1.7E+00 \\ 2.4E+00 \\ 2.5E+00 \\ 2.4E+00 \\ 9.4E-01 \\ 1.4E+00 \\ \end{array}$	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01 2.8E+00 1.8E+00 6.8E-01 2.2E+00 7.7E+00 1.6E+00 1.1E+00 7.4E-01 6.3E+00	V 9.4E+00 1.9E+01 2.6E+00 2.3E+00 5.3E+01 3.4E+02 1.5E+01 4.4E+00 2.8E+00 1.4E+01 4.5E+00 2.7E+00 2.3E+00 4.8E+00 1.2E+01	Mn 2.1E+00 1.1E+01 3.2E+00 2.3E+00 2.5E+00 1.5E+01 3.6E+00 2.4E+00 1.5E+00 1.5E+01 4.4E+00 2.8E+00 2.2E+00 1.5E+00 9.2E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 2.2E+00 1.7E+00 1.6E+00 1.6E+00 2.6E+00 2.0E+00 1.9E+00 1.7E+00 1.9E+00 1.9E+00 1.7E+00	Cu 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01 6.5E+02 6.3E+01 3.2E+01 2.6E+01 2.6E+01 2.7E+01 2.5E+01 4.2E+00 5.0E+01 9.0E+01	As 1.5E+02 7.6E+02 2.2E+01 1.9E+01 1.6E+03 1.6E+03 1.9E+02 1.0E+03 1.7E+02 3.2E+01 1.8E+01 2.7E+02 7.9E+02	Se 5.7E+00 2.5E+01 1.5E+00 2.8E+01 - 5.1E+01 4.8E+00 1.5E+00 1.5E+00 1.9E+01 1.9E+01 1.6E-01 6.6E+00 1.8E+01	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.2E+01 3.6E+00 2.1E+00 8.5E-01 1.5E+00 1.3E+00 1.4E+00 1.4E+00 1.4E+00 1.4E+00 1.2E+01	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 4.5E+03 1.1E+03 2.9E+03 4.9E+02 7.0E+01 1.3E+02 1.7E+03 2.1E+03	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+03 9.3E+02 - 7.8E+03 9.3E+02 - 3.6E+03 9.2E+02 2.2E+03 3.6E+03	Ba 1.8E+00 3.3E+00 2.3E+00 2.2E+00 2.0E+00 1.9E+01 4.6E+00 2.5E+00 1.4E+00 3.3E+00 3.4E+00 2.4E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 9.2E+02 1.3E+02 2.1E+01 5.5E+02 7.4E+01 1.2E+01 1.9E+01 3.0E+01 5.5E+02	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02 5.9E+03 5.8E+02 9.4E+01 5.7E+02 3.4E+03 4.6E+02 5.4E+01 4.1E+01 5.4E+02 2.1E+03
EF (soil) Spring Summer Autumn Winter	$\begin{array}{r c c c c c c c c c c c c c c c c c c c$	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E+00 2.4E+01 2.3E+01 3.4E+00 7.3E-01 4.0E+00 5.0E+00 4.6E+00 5.0E+00 3.3E+00 5.5E+00	Mg 1.6E+00 1.8E+00 2.6E+00 4.1E+00 5.7E+00 5.5E-01 3.0E+00 7.0E+00 6.5E+00 1.7E+00 9.0E-01 1.7E+00 2.4E+00 2.5E+00 2.4E+00 2.4E+00 2.4E+00 2.4E+00 2.4E+00 2.4E+00 2.4E+00 2.4E+00 2.4E+00 2.4E+00	Al 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01 2.8E+00 1.8E+00 6.8E-01 2.2E+00 7.7E+00 1.6E+00 1.1E+00 7.4E-01 6.3E+00 1.9E+00	V 9.4E+00 1.9E+01 4.1E+00 2.6E+00 2.3E+00 5.3E+01 3.4E+02 1.5E+01 4.4E+00 2.8E+00 1.4E+01 2.6E+01 4.5E+00 2.7E+00 2.3E+00 4.8E+00 1.2E+01 5.2E+00	Mn 2.1E+00 3.2E+00 2.3E+00 2.5E+00 3.1E+00 1.5E+01 3.6E+00 2.4E+00 1.5E+01 4.4E+00 2.8E+00 2.2E+00 1.5E+01 9.2E+00 4.7E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 2.2E+00 1.7E+00 1.6E+00 1.6E+00 2.6E+00 2.0E+00 1.9E+00 1.7E+00 1.6E+00 1.9E+00 1.9E+00 1.7E+00 1.6E+00	Cu 4.3E+02 9.8E+01 2.6E+01 1.5E+01 2.7E+01 6.5E+02 6.3E+01 3.2E+01 2.6E+01 2.6E+01 2.7E+01 2.5E+01 4.2E+00 5.0E+01 9.0E+01 6.0E+01	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01 1.6E+03	Se 5.7E+00 2.5E+01 1.5E+00 2.8E-01 - 5.1E+01 4.8E+00 1.5E+00 1.5E+00 1.9E+01 1.9E+01 1.6E-01 6.6E+00 1.8E+01 4.5E+00	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.2E+01 3.6E+00 2.1E+00 8.5E-01 1.5E+00 1.3E+00 1.4E+00 1.4E+00 1.4E+00 1.4E+00 1.4E+00	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 4.5E+03 1.1E+03 1.1E+03 2.9E+03 4.9E+02 7.0E+01 1.3E+02 1.7E+03 2.1E+03 8.2E+02	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+003 9.3E+02 - 7.8E+03 9.3E+02 - 2.2E+03 3.6E+03 1.1E+03	Ba 1.8E+00 2.3E+00 2.3E+00 2.2E+00 2.0E+00 1.9E+01 4.6E+00 2.5E+00 1.4E+00 3.3E+00 3.4E+00 2.4E+00 1.4E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 9.2E+02 1.3E+02 2.1E+01 5.5E+02 7.4E+01 1.2E+01 1.9E+01 3.0E+01 5.5E+02 1.8E+02	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 9.7E+02 5.9E+03 5.8E+02 9.4E+01 5.7E+02 3.4E+03 4.6E+02 5.4E+01 4.1E+01 5.4E+02 2.1E+03 6.7E+02
EF (soil) Spring Summer Autumn Winter	$\begin{array}{r c c c c c c c c c c c c c c c c c c c$	Na 2.6E+00 2.9E+00 4.2E+00 9.9E+00 1.7E+01 6.6E+00 2.4E+01 2.3E+01 3.4E+00 7.3E-01 4.0E+00 4.6E+00 5.0E+00 4.6E+00 5.0E+00 3.3E+00 5.5E+00 6.9E+00	Mg 1.6E+00 1.8E+00 2.6E+00 4.1E+00 5.7E+00 5.5E-01 3.0E+00 7.0E+00 6.5E+00 1.7E+00 9.0E-01 1.7E+00 2.4E+00 2.5E+00 2.4E+00 2.5E+00 2.4E+00 2.5E+00 2.4E+00 9.4E-01 1.4E+00 2.5E+00 2.5E+00	Al 1.0E+00	K 3.8E+00 7.8E+00 1.9E+00 3.0E+00 2.9E+00 2.1E+01 2.8E+00 1.8E+00 6.8E-01 2.2E+00 7.7E+00 1.6E+00 1.1E+00 7.4E-01 6.3E+00 6.3E+00 6.3E+00 6.3E+00 6.3E+00 6.3E+00 6.3E+00 6.3E+00 6.3E+00 6.3E+00	V 9.4E+00 1.9E+01 4.1E+00 2.6E+00 3.4E+02 1.5E+01 4.4E+00 2.8E+00 1.4E+01 2.6E+01 4.5E+00 2.7E+00 2.3E+00 1.2E+01 4.8E+00 3.0E+00	Mn 2.1E+00 1.1E+01 3.2E+00 2.3E+00 2.5E+00 1.5E+01 3.6E+00 2.4E+00 1.5E+00 1.5E+01 4.4E+00 2.8E+00 2.2E+00 1.5E+00 9.2E+00 4.7E+00 2.1E+00	Fe 1.6E+00 2.4E+00 1.6E+00 1.8E+00 2.9E+00 1.7E+00 2.2E+00 1.7E+00 1.6E+00 1.6E+00 1.6E+00 1.2E+00 2.0E+00 2.0E+00 1.9E+00 1.7E+00 1.9E+00 1.7E+00 1.4E+00	Cu 9.8E+01 2.6E+01 1.5E+01 2.7E+01 8.8E+01 6.5E+02 6.3E+01 3.2E+01 2.6E+01 2.6E+01 2.7E+01 2.5E+01 4.2E+00 5.0E+01 9.0E+01 3.2E+01 3.2E+01 3.2E+01	As 1.5E+02 7.6E+02 1.0E+02 2.2E+01 1.9E+01 1.6E+03	Se 5.7E+00 2.5E+01 1.5E+00 2.8E-01 - 5.1E+01 4.8E+00 1.5E+00 1.5E+00 1.9E+01 1.9E+01 1.6E-01 6.6E+00 1.8E+01 4.5E+00 7.1E-01	Sr 9.6E-01 1.4E+00 1.3E+00 1.6E+00 1.7E+00 1.2E+01 3.6E+00 2.1E+00 8.5E-01 1.5E+00 1.5E+00 1.4E+00 1.4E+00 1.4E+00 1.4E+00 1.4E+00 1.4E+00 1.4E+00 1.4E+00 1.3E+00	Cd 5.4E+02 2.1E+03 3.4E+02 6.8E+01 3.2E+01 4.5E+03 1.1E+03 2.9E+03 4.9E+02 7.0E+01 1.3E+02 1.7E+03 2.1E+03 8.2E+02 1.3E+02 1.3E+02	Sb 8.7E+02 3.3E+03 4.6E+02 1.7E+02 1.3E+003 9.3E+02 - 1.6E+03 5.4E+03 9.2E+02 3.2E+02 2.0E+02 2.2E+03 3.6E+03 1.1E+03 4.0E+02	Ba 1.8E+00 3.3E+00 2.3E+00 1.8E+00 2.2E+00 2.0E+00 1.9E+01 4.6E+00 2.5E+00 1.4E+00 3.3E+00 3.4E+00 2.4E+00 1.4E+00 3.4E+00 3.4E+00 3.4E+00	Pb 8.0E+01 4.8E+02 6.9E+01 1.2E+01 9.4E+00 9.2E+02 1.3E+02 2.1E+01 5.5E+02 7.4E+01 1.2E+01 1.9E+01 3.0E+01 5.5E+02 1.8E+02 1.6E+01	Bi 3.4E+02 2.7E+03 3.1E+02 3.8E+01 3.3E+01 5.9E+03 5.8E+02 9.4E+01 5.7E+02 3.4E+03 4.6E+02 5.4E+01 4.1E+01 5.4E+02 2.1E+03 6.7E+02 1.0E+02

Table S1 Summary of EF values of 16 metallic elements by particle size in each season at Kumamoto

EF (sea salt)	Size	Na	Mg	Al	K	V	Mn	Fe	Cu	As	Se	\mathbf{Sr}	Cd	Sb	Ba	Pb	Bi
Spring	< 0.1	1.0E+00	5.5E-01	5.2E+06	-	1.4E+04	-	-	2.1E+06	1.8E+04	-	3.6E+00	-	2.1E+04	8.8E+03	4.5E+06	2.4E+06
	0.5 - 1	1.0E+00	8.1E-01	2.0E+06	1.4E+01	3.1E+04	4.0E+05	2.7E+06	1.0E+06	2.1E+04	4.6E+05	2.9E+00	4.6E+05	3.2E+04	3.8E+03	7.9E+07	2.1E+08
	1 - 2.5	1.0E+00	9.8E-01	2.7E+06	3.1E+00	2.4E+03	1.2E+05	4.6E+05	7.7E+04	2.6E+03	1.5E+04	1.8E+00	4.7E+04	3.9E+03	9.2E+02	8.5E+06	2.5E+07
	2.5 - 10	1.0E+00	1.0E+00	1.4E+06	1.9E+00	6.0E+02	5.2E+04	2.3E+05	4.8E+04	3.7E+02	-	1.4E+00	1.6E+03	5.6E+02	3.4E+02	3.3E+05	1.5E+06
	10<	1.0E+00	1.1E+00	1.1E+06	1.6E+00	4.1E+02	3.9E+04	4.3E+05	3.7E+04	2.1E+02	-	1.2E+00	-	1.1E+03	2.0E+02	1.0E+05	1.6E+06
Autumn	< 0.1	1.0E+00	2.8E+00	2.0E+07	-	2.3E+04	9.7E+05	-	-	-	-	1.3E+01	5.8E+05	-	3.2E+03	5.6E+07	1.2E+08
	0.5 - 1	1.0E+00	1.5E+00	8.0E+06	3.5E+01	4.1E+04	1.8E+06	2.6E+06	3.8E+06	6.8E+04	6.6E + 05	3.7E+00	1.8E+06	1.2E+05	2.9E+03	2.6E+08	6.8E+08
	1-2.5	1.0E+00	1.2E+00	3.8E+06	3.3E+00	2.3E+03	2.3E+05	1.2E+06	9.1E+04	3.6E+03	3.9E+04	2.2E+00	1.1E+05	7.8E+03	1.0E+03	1.2E+07	3.7E+07
	2.5 - 10	1.0E+00	1.0E+00	2.3E+06	2.1E+00	9.1E+02	9.9E+04	4.5E+05	2.3E+04	7.9E+02	6.3E+03	1.6E+00	8.6E+03	1.3E+03	5.9E+02	1.2E+06	4.2E+06
	10<	1.0E+00	1.1E+00	1.1E+06	1.3E+00	3.8E+02	4.8E+04	2.4E+05	1.6E+04	5.9E+01	7.9E+03	1.5E+00	2.1E+04	-	2.5E+02	4.8E+05	-
Winter	< 0.1	-	-	-	-	-	-	-	-	-	-	-	-		-	-	-
	0.5 - 1	1.0E+00	7.0E-01	5.4E+06	8.1E+01	7.0E+04	4.1E+06	-	-	1.7E+05	2.2E+06	2.6E+00	4.8E+06	4.8E+05	1.4E+03	9.7E+08	3.5E+09
	1-2.5	1.0E+00	8.7E-01	1.8E+06	3.0E+00	1.7E+03	2.4E+05	5.5E+05	-	3.9E+03	3.6E+04	1.5E+00	1.9E+05	9.8E+03	6.7E+02	2.3E+07	1.1E+08
	2.5 - 10	1.0E+00	8.3E-01	9.2E+05	1.6E+00	4.9E+02	4.3E+04	2.0E+05	-	-	1.4E+03	1.1E+00	1.0E+04	7.0E+02	3.1E+02	1.5E+06	5.1E+06
	10<	1.0E+00	1.0E+00	6.2E+05	1.3E+00	1.6E+02	1.8E+04	6.2E+04	-	-	-	1.0E+00	-	2.0E+02	1.1E+02	5.0E+05	1.1E+06
EF (soil)	Size	Na	Mg	Al	K	V	Mn	Fe	Cu	As	Se	\mathbf{Sr}	Cd	Sb	Ba	Pb	Bi
Spring	< 0.1	1.1E+01	1.8E+00	1.0E+00	-	1.2E+01	-	-	2.6E+02	3.1E+02	-	1.9E+00	-	6.8E+02	5.2E+00	2.1E+01	1.1E+01
	0.5 - 1	3.4E+01	6.4E+00	1.0E+00	1.4E+01	1.3E+02	1.0E+01	9.3E+00	2.5E+02	1.8E+03	4.6E+01	5.1E+00	3.7E+03	3.2E+03	1.2E+01	8.7E+02	2.7E+03
	1 - 2.5	3.2E+01	7.3E+00	1.0E+00	3.1E+00	9.2E+00	2.8E+00	1.4E+00	1.6E+01	1.9E+02	3.4E+00	2.9E+00	3.6E+02	4.6E+02	1.9E+00	8.6E+01	3.2E+02
	2.5 - 10	7.6E+01	1.9E+01	1.0E+00	4.2E+00	3.1E+00	2.0E+00	1.5E+00	2.0E+01	4.8E+01	-	5.4E+00	5.7E+01	1.3E+02	1.4E+00	1.7E+01	3.3E+01
	10<	1.4E+02	3.5E+01	1.0E+00	6.3E+00	3.1E+00	1.5E+00	1.9E+00	2.5E+01	4.3E+01	-	8.4E+00	-	3.2E+02	9.6E-01	1.0E+01	2.0E+01
Autumn	<0.1	3.0E+00	2.3E+00	1.0E+00	-	2.8E+01	5.5E+00	- 0.0E+00	-	-	- 0.0E+01	1.6E+00	4.8E+02	-	8.3E-01	1.1E+02	1.6E+02
	0.5-1	7.5E+00	2.9E+00	1.0E+00	9.7E+00	3.4E+01	1.2E+01	3.0E+00	3.1E+02	1.4E+03	2.2E+01	1.7E+00	3.6E+03	2.8E+03	2.1E+00	7.8E+02	2.2E+03
	1-2.5 9 5-10	1.6E+01	4.9E+00	1.0E+00 1.0E+00	1.9E+00	4.1E+00 2.7E+00	3.3E+00 2.2E+00	2.8E+00	1.5E+01	1.5E+02 5 4E+01	2.8E+00 7.4E-01	2.2E+00	4.6E+02 6.1E+01	4.0E+02 1.0E+02	1.6E+00	7.4E+01	2.5E+02 4.9F±01
	2.5 IU 10<	5.5E+01	1.0E+00 1.5E+01	1.0E+00 1.0E+00	2.0E+00 2.6E+00	2.7E+00 2.3E+00	2.3E+00 2.3E+00	1.0E+00 1.9E+00	9.9E+00	9.3E+00	1.4E 01 1.8E+00	2.0E+00 5.0E+00	0.1E+01 2.9E+02	1.0E+02	1.3E+00 1.3E+00	1.2E+01 1.0E+01	4.514+01
Winter	<0.1	-	-	1.0E+00	-	1.7E+01	-	-	-	-	-	-		1 7E+02	-	5.9E+01	1.6E+02
,, 11001	0.5-1	1.1E+01	2.0E+00	1.0E+00	3.3E+01	8.7E+01	4.1E+01	-	-	5.1E+03	1.1E+02	1.8E+00	1.5E+04	1.7E+04	1.5E+00	4.3E+03	1.6E+04
	1-2.5	3.3E+01	7.5E+00	1.0E+00	3.6E+00	6.2E+00	7.0E+00	2.8E+00	-	3.5E+02	5.3E+00	3.0E+00	1.7E+03	1.0E+03	2.2E+00	3.0E+02	1.5E+03
	2.5 - 10	6.5E+01	1.4E+01	1.0E+00	3.7E+00	3.5E+00	2.5E+00	2.0E+00	-	-	4.1E-01	4.4E+00	1.8E+02	1.5E+02	2.0E+00	3.8E+01	1.4E+02
	10<	9.7E+01	2.5E+01	1.0E+00	4.6E+00	1.7E+00	1.5E+00	9.2E-01	-	-	-	6.2E+00	-	6.3E+01	1.0E+00	2.0E+01	4.7E+01

Table S2 Summary of EF values of 16 metallic elements by particle size in each season at Cape Hedo

Fig. S1 ^N 50 40 30 ° Kumamoto 30 (a) °Cape Hedo 20 (a) (b) 110 120 130 140 150 E

(C)

(d)



Fig. S2 (a) (b)

Fig. S3 (a)(b)(c)(d)







Fig. S4

