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 Taniguchi[1], Kojiro Shimada[2], Akinori Takami[3], Neng-Huei Lin[2,4], Chak K. Chan[5], Yong Pyo Kim[2,6], Shiro Hatakeyama[1,2,7]*

[1]*Graduate School of Agriculture, Tokyo University of Agriculture and Technology, Fuchu, Tokyo 183-8509, Japan*

[2]*Global Innovation Research Organization, Tokyo University of Agriculture and Technology, Fuchu, Tokyo 183-8538, Japan*

[3]*National 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*

[5]*School of Energy and Environment, City University of Hong Kong, Hong Kong, China*

[6]*Department of Chemical Engineering & Materials Science and Department of Environmental Science & Engineering, Ewha Womans University, Seoul 03760, Republic of Korea*

[7]*Center 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 μm size fraction, and Cu concentration was measured in the 2.5–10 μm 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.

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	Sr	Cd	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
	2.5-10	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

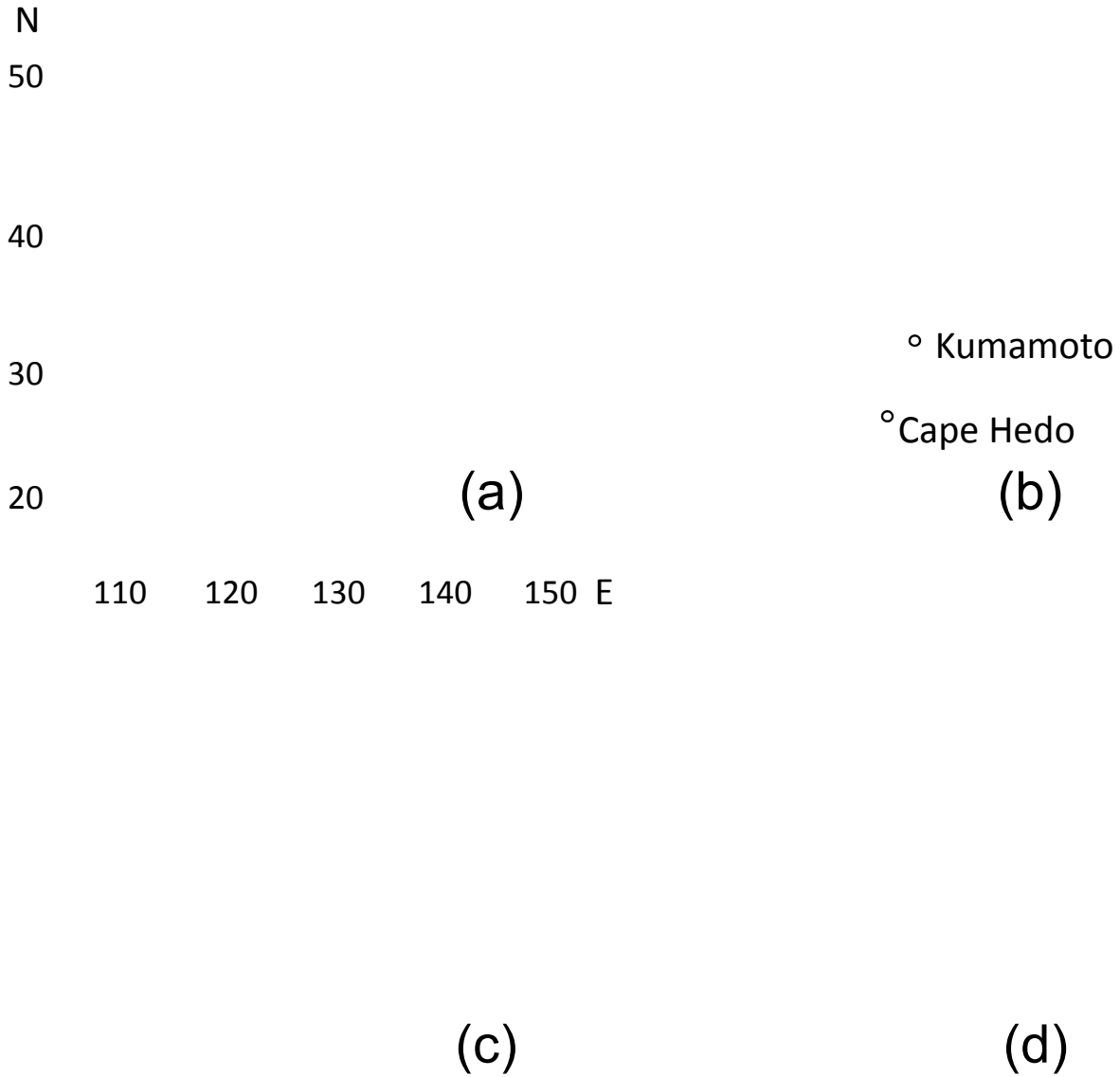
EF (soil)	Size	Na	Mg	Al	K	V	Mn	Fe	Cu	As	Se	Sr	Cd	Sb	Ba	Pb	Bi
Spring	<0.1	2.6E+00	1.6E+00	1.0E+00	3.8E+00	9.4E+00	2.1E+00	1.6E+00	4.3E+02	1.5E+02	5.7E+00	9.6E-01	5.4E+02	8.7E+02	1.8E+00	8.0E+01	3.4E+02
	0.5-1	2.9E+00	1.8E+00	1.0E+00	7.8E+00	1.9E+01	1.1E+01	2.4E+00	9.8E+01	7.6E+02	2.5E+01	1.4E+00	2.1E+03	3.3E+03	3.3E+00	4.8E+02	2.7E+03
	1-2.5	4.2E+00	2.6E+00	1.0E+00	1.9E+00	4.1E+00	3.2E+00	1.6E+00	2.6E+01	1.0E+02	1.5E+00	1.3E+00	3.4E+02	4.6E+02	2.3E+00	6.9E+01	3.1E+02
	2.5-10	9.9E+00	4.1E+00	1.0E+00	1.9E+00	2.6E+00	2.3E+00	1.8E+00	1.5E+01	2.2E+01	2.8E-01	1.6E+00	6.8E+01	1.7E+02	1.8E+00	1.2E+01	3.8E+01
	10<	1.7E+01	5.7E+00	1.0E+00	3.0E+00	2.3E+00	2.5E+00	2.9E+00	2.7E+01	1.9E+01	-	1.7E+00	3.2E+01	1.3E+02	2.2E+00	9.4E+00	3.3E+01
Summer	<0.1	6.6E-01	5.5E-01	1.0E+00	2.9E+00	5.3E+01	3.1E+00	1.7E+00	8.8E+01	-	-	1.0E+00	-	-	2.0E+00	-	9.7E+02
	0.5-1	6.6E+00	3.0E+00	1.0E+00	2.1E+01	3.4E+02	1.5E+01	2.2E+00	6.5E+02	1.6E+03	5.1E+01	1.2E+01	4.5E+03	7.8E+03	1.9E+01	9.2E+02	5.9E+03
	1-2.5	2.4E+01	7.0E+00	1.0E+00	2.8E+00	1.5E+01	3.6E+00	1.7E+00	6.3E+01	-	4.8E+00	3.6E+00	1.1E+03	9.3E+02	4.6E+00	1.3E+02	5.8E+02
	2.5-10	2.3E+01	6.5E+00	1.0E+00	1.8E+00	4.4E+00	2.4E+00	1.6E+00	3.2E+01	-	1.5E+00	2.1E+00	-	-	2.5E+00	2.1E+01	9.4E+01
	10<	3.4E+00	1.7E+00	1.0E+00	6.8E-01	2.8E+00	1.5E+00	1.2E+00	2.6E+01	-	-	8.5E-01	-	-	1.4E+00	-	-
Autumn	<0.1	7.3E-01	9.0E-01	1.0E+00	2.2E+00	1.4E+01	4.5E+00	6.5E+00	1.4E+00	1.9E+02	4.4E+00	8.5E-01	1.0E+03	1.6E+03	1.5E+00	8.4E+01	5.7E+02
	0.5-1	4.0E+00	1.7E+00	1.0E+00	7.7E+00	2.6E+01	1.5E+01	2.6E+00	2.6E+01	1.0E+03	1.9E+01	1.5E+00	2.9E+03	5.4E+03	3.3E+00	5.5E+02	3.4E+03
	1-2.5	4.6E+00	2.4E+00	1.0E+00	1.6E+00	4.5E+00	4.4E+00	2.0E+00	2.7E+01	1.7E+02	1.9E+00	1.5E+00	4.9E+02	9.2E+02	3.4E+00	7.4E+01	4.6E+02
	2.5-10	5.0E+00	2.5E+00	1.0E+00	1.1E+00	2.7E+00	2.8E+00	1.9E+00	2.5E+01	3.2E+01	2.6E-01	1.3E+00	7.0E+01	3.2E+02	2.4E+00	1.2E+01	5.4E+01
	10<	4.6E+00	2.4E+00	1.0E+00	7.4E-01	2.3E+00	2.2E+00	1.7E+00	4.2E+00	1.8E+01	1.6E-01	1.1E+00	1.3E+02	2.0E+02	1.4E+00	1.9E+01	4.1E+01
Winter	<0.1	2.7E+00	9.4E-01	1.0E+00	6.3E+00	4.8E+00	1.5E+00	2.6E+00	5.0E+01	2.7E+02	6.6E+00	1.4E+00	1.7E+03	2.2E+03	-	3.0E+01	5.4E+02
	0.5-1	3.3E+00	1.4E+00	1.0E+00	6.3E+00	1.2E+01	9.2E+00	1.9E+00	9.0E+01	7.9E+02	1.8E+01	1.2E+00	2.1E+03	3.6E+03	-	5.5E+02	2.1E+03
	1-2.5	5.5E+00	2.5E+00	1.0E+00	1.9E+00	5.2E+00	4.7E+00	1.6E+00	6.0E+01	2.7E+02	4.5E+00	1.4E+00	8.2E+02	1.1E+03	5.8E+00	1.8E+02	6.7E+02
	2.5-10	6.9E+00	2.8E+00	1.0E+00	8.9E-01	3.0E+00	2.1E+00	1.4E+00	3.2E+01	3.8E+01	7.1E-01	1.3E+00	1.3E+02	4.0E+02	3.4E+00	1.6E+01	1.0E+02
	10<	1.1E+01	4.1E+00	1.0E+00	1.0E+00	2.8E+00	2.0E+00	1.3E+00	2.1E+01	-	-	1.3E+00	-	2.9E+02	-	7.9E+00	1.2E+02

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

EF (sea salt)	Size	Na	Mg	Al	K	V	Mn	Fe	Cu	As	Se	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	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	-	-	-	-	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	1.6E+01	4.9E+00	1.0E+00	1.9E+00	4.1E+00	3.3E+00	2.8E+00	1.5E+01	1.5E+02	2.8E+00	2.2E+00	4.6E+02	4.0E+02	1.6E+00	7.4E+01	2.5E+02
	2.5-10	2.7E+01	7.0E+00	1.0E+00	2.0E+00	2.7E+00	2.3E+00	1.8E+00	7.1E+00	5.4E+01	7.4E-01	2.6E+00	6.1E+01	1.0E+02	1.5E+00	1.2E+01	4.3E+01
	10<	5.5E+01	1.5E+01	1.0E+00	2.6E+00	2.3E+00	2.3E+00	1.9E+00	9.9E+00	9.3E+00	1.8E+00	5.0E+00	2.9E+02	-	1.3E+00	1.0E+01	-
Winter	<0.1	-	-	1.0E+00	-	1.7E+01	-	-	-	-	-	-	-	1.7E+02	-	5.9E+01	1.6E+02
	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

Fig. S1



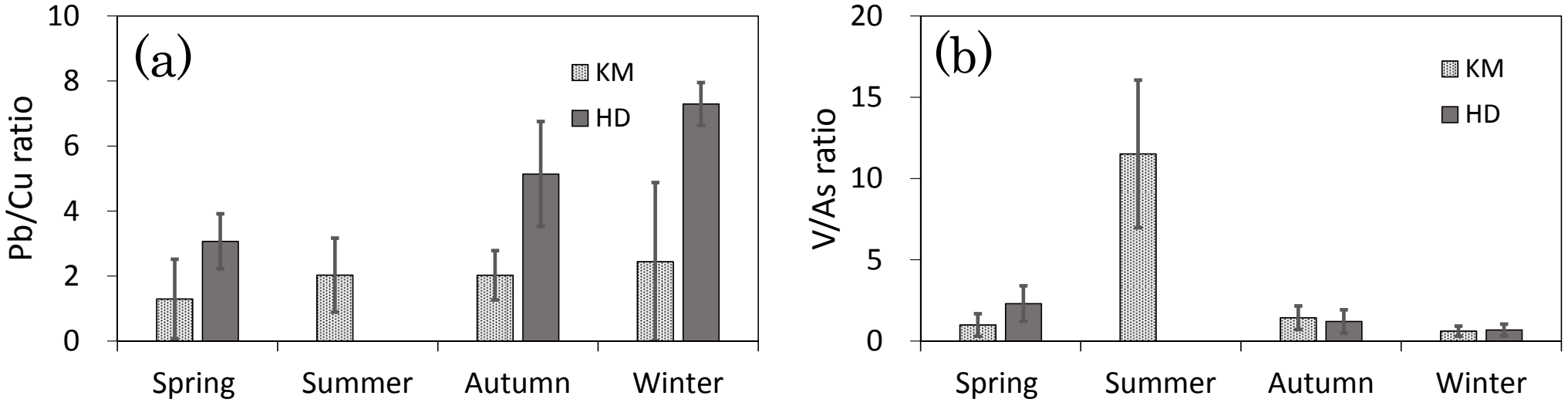
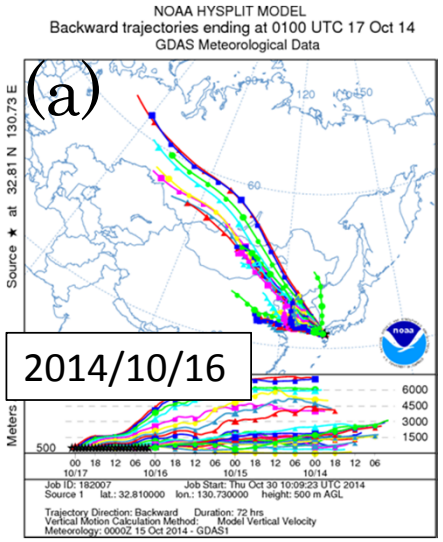

(a)
Pb/Cu ratio
10
8
6
4
2
0
KM
HD
Spring
Summer
Autumn
Winter
(b)
V/As ratio
20
15
10
5
0
KM
HD
Spring
Summer
Autumn
Winter

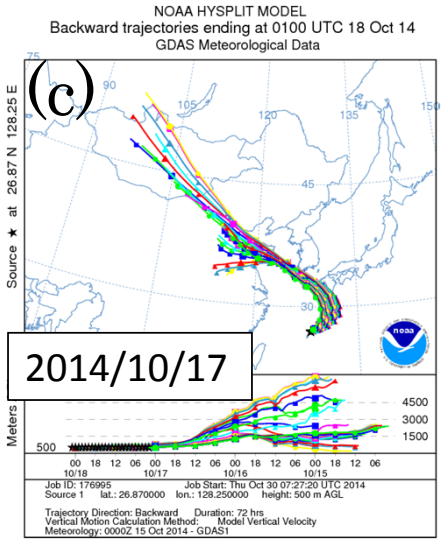
Fig. S2 (a) (b)

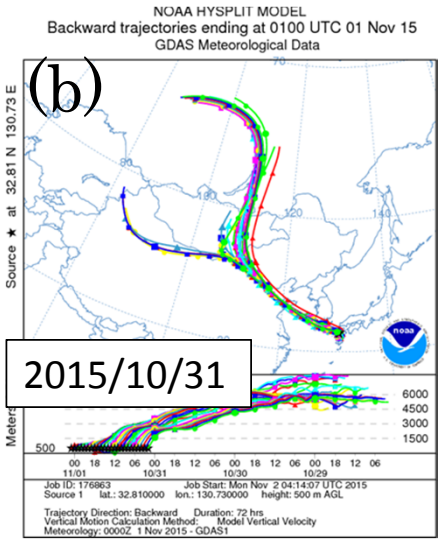
Fig. S3 (a)(b)(c)(d)

In KM

In HD







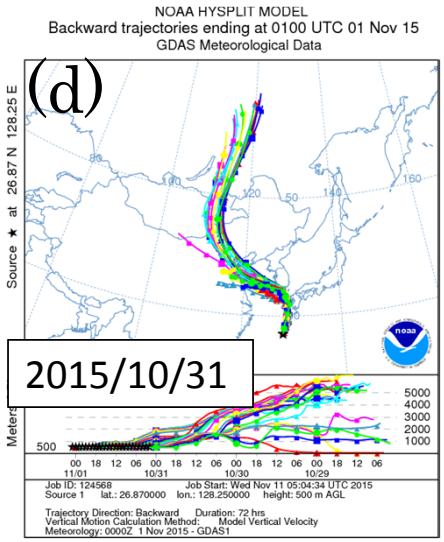
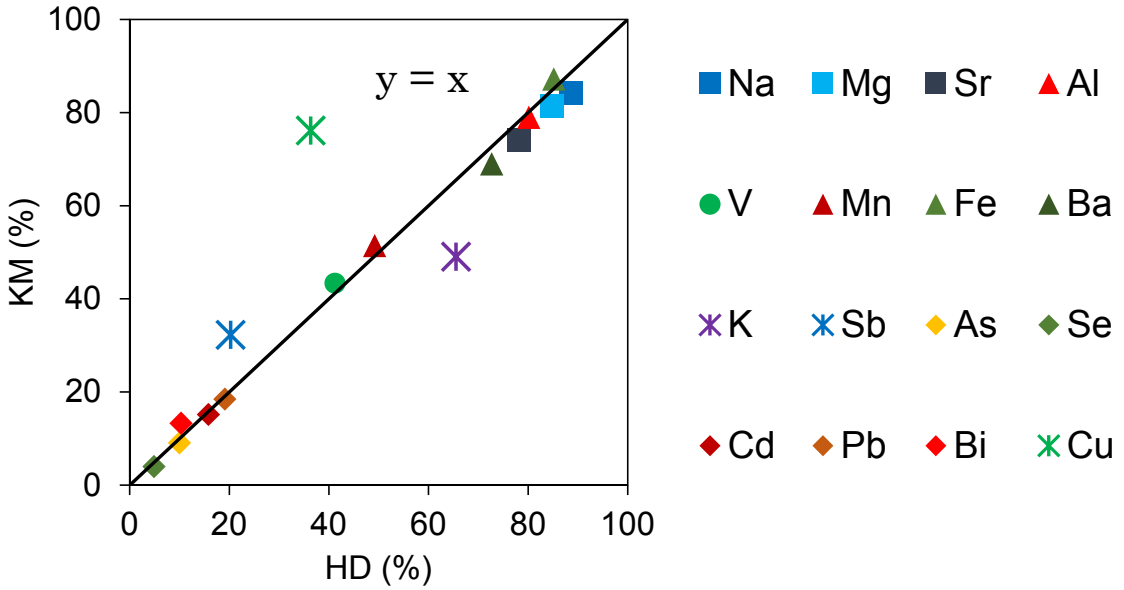


Fig. S4


100
80
60
40
20
0
KM (%)
y = x
0
20
40
60
80
100
HD (%)
Na
Mg
Sr
Al
V
Mn
Fe
Ba
K
Sb
As
Se
Cd
Pb
Bi
Cu