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Grain-size distribution and chemical composition of water-insoluble components in aeolian dust collected in Japan in spring 2002

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Abstract: We have studied chemical characteristics of aeolian dust (Kosa) during the transportation from China to Japan. Dust samples were collected at Naha, Fukuoka, Nagoya and Tsukuba in Japan from March to May 2002 and analyzed for the chemical composition of water-insoluble components. The chemical concentrations of most elements in the air were the highest during a dust event, and their particle size distribution patterns are characterized by having one large peak at 2.1-7.0 μm. Most elements are considered to originate in mineral aerosol because mass concentrations have the same distribution patterns. However, some elements, Cd, Sn, Sb, Pb and Bi, were highly enriched in small particles (under 1 μm), which consist of mainly carbon aerosol released by a vehicle, plant and heating system. These elements are considered to originate in anthropogenic materials.

The elemental concentrations divided by Al_2O_3 contents are useful to examine the change of mineralogical composition or estimate the contribution of anthropogenic materials. The concentration ratios of most elements were almost constant through the particle size, but often high in the smaller particle (0.65-1.01 μm). The particle's mineral composition probably changes at 1 μm. On the contrary, distribution patterns of some heavy metals having anthropogenic origins revealed that the fraction of anthropogenic materials in aeolian dust gradually increased with the decrease of particle size and suddenly rose at 2 μm of particle size.

Finally, we examined geochemical differences between two periods (dust event and non-dust event) and among observation stations. Some elements (P_2O_5, K_2O, T-Fe_2O_3, Zr, Nb and Mo) have systematic differences among sampling locations during non-dust event and showed that aeolian dust contains some local materials. However, these systematic differences disappeared when a dust event was observed and revealed that a dust storm carried a large amount of aerosol from East Asia to Japan and made a nearly homogeneous chemical composition of dust at each station.

Keywords: Kosa, aeolian dust, water-insoluble components, grain-size distribution, mass concentration, chemical composition, Naha, Fukuoka, Nagoya, Tsukuba

1. Introduction

"Kosa" is aeolian dust transported from the arid or semi-arid region in East Asia to Japan during spring. Aeolian dust that has been transported over a long distance has large influences on life, agriculture and traffic, and more serious effects on the global climate. Recently, the National Institute for Environmental Studies (2001) reported some observations on atmospheric aerosols in China from the point of view of atmospheric pollution. On the contrary, to investigate the impact of aeolian dust on the climate, the "Aeolian

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Dust Experiment on Climate Impact (ADEC)" started in April 2000 with a five-year plan. We have joined the ADEC project and mainly studied chemical characteristics of aeolian dust during the transportation from China to Japan.

The chemical studies of aeolian dust in China and Japan have been carried out by many researchers (Nishikawa *et al.*, 1987, Inst. Hydrospheric Sci. Nagoya Univ., 1991, Zhang and Iwasaka, 1998, Mori *et al.*, 2003, and references there in). The larger particles (>1 μm) mainly consist of mineral aerosol, whose mineralogical composition is quartz, K-feldspar, plagioclase, mica group (e.g. chlorite), clay minerals (e.g. kaolinite and illite) and amorphous materials (Inst. Hydrospheric Sci. Nagoya Univ., 1991, Kanai *et al.*, 2003). The Al_2O_3 is the most abundant mineral in mineral aerosol and its concentration in dust is used as a good indicator of the contribution of minerals. Most elements are considered to originate in mineral aerosol because they have good linear relationships to Al_2O_3 contents. However, some elements such as Cu, Zn and Pb sometimes have poor relationships to Al_2O_3, and they are enriched in small particles under 1 μm. The smaller particles (< 1 μm) consist mainly of carbon aerosol, which are released by a vehicle, plant and heating system. However, most studies were carried out in China and Japan independently. There are few studies that examine the chemical compositions of aeolian dust (Mori *et al.*, 2003).

In the ADEC project, we have a valuable opportunity to simultaneously collect aeolian dust in China and Japan through the year and examine their chemical composition variations during their transportation. The dust samples were collected at three Chinese stations (Beijing, Qingdao and Hefei) and four Japanese stations (Naha, Fukuoka, Nagoya and Tsukuba) by using high and low volume air samplers (Fig. 1). We have tried to determine many elemental concentrations of aeolian dust for as long as possible and examine the grain-size distribution of chemical composition from three points of view; (1) the similarity and difference among sampling locations in Japan and China and influence of the dust event, (2) the chemical compositional change during the transportation of aeolian dust from China to Japan and (3) the seasonal variation of the chemical composition of aeolian dust at the Tsukuba station, Japan. In this paper, we focus on the data collected at four Japanese stations during the first intensive observation period (IOP) (IOP 1: 8-21 April 2002) (Kanai *et al.*, 2003). The other points will be reported in future papers.

2. Sampling locations and period

The high volume air sampler (HV-1000F, Shibata Co. Ltd.) and Andersen-type low volume air sampler (AN-200, Shibata Co. Ltd.) were used at four Japanese stations shown in Fig. 1. The air samplers were fixed on the roof of a building to prevent from collecting local surface materials raised by the wind. The HV-1000F was used to collect bulk dust samples for isotopic studies, and AN-200 was used to examine the physical and chemical properties of aeolian dust. The AN-200 obtains the grain size distribution data of aeolian dust: the particle size classification is >11 μm, 11-7.0 μm, 7.0-4.7 μm, 4.7-3.3 μm, 3.3-2.1 μm, 2.1-1.1 μm, 1.1-0.65 μm, 0.65-0.43 μm, and <0.43 μm. The AN-200 was operated for 14-20 days during the usual observation period and 4-8 days during the IOP-1. The quartz filter (Tokyo Dylec, 2500QAT-UP) was used to collect small particles (0.65-0.43 μm and <0.43 μm) and other particles were trapped by the PF-050 polyflon filter (Advantec Co.Ltd.). Each filter was dried in desiccators before and after sampling to be weighed.

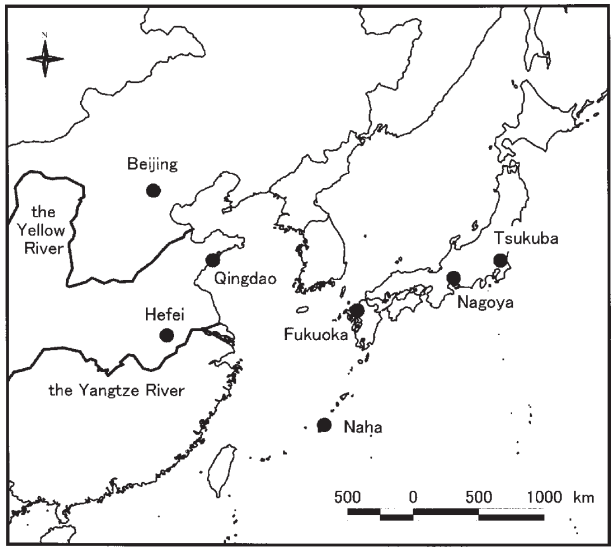

Fig. 1. Sampling locations in China and Japan.

3. Analytical Methods

The dust samples were analyzed to determine the water-soluble and water-insoluble components. A quarter of the filter obtained by AN-200 was soaked in a Teflon beaker with 1 ml of ethanol and 25 ml of Milli Q water (MQ). An ultrasonic cleaner was operated for 30 minutes, and the soluble component was leached. The eluent and insoluble fractions were separated by a 0.22 μm cellulose acetate-type membrane filter. The eluent was weighed and its composition was determined by an ion chromatograph. The principal ions determined for the soluble fraction are Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, Na^+, K^+, Ca^{2+} and Mg^{2+} (Kamioka and Kanai, 2002).

The insoluble fraction trapped on the quarter of the polyflon filter and membrane filter was decomposed

with 3 ml of HNO_3, 1 ml of $HClO_4$ and 3 ml of HF in a Teflon beaker at 120 ℃ for 2 hours. After the polyflon filter was removed, the degradation product was evaporated to dryness under 200 ℃. The residue was dissolved in 0.5 ml of 7N HNO_3 and diluted to 10 ml by MQ (Imai, 1990). Fifty-one elements were determined by ICP- AES (Na_2O, MgO, Al_2O_3, P_2O_5, K_2O, CaO, TiO_2, MnO, Total Fe_2O_3, V, Sr, Ba) and ICP-MS (Li, Be, Sc, Cr, Co, Ni, Cu, Zn, Ga, Rb, Y, Zr, Nb, Mo, Cd, Sn, Sb, Cs, REE, Hf, Ta, Tl, Pb, Bi, Th and U).

4. Result

4.1 Aerosol concentrations at Naha, Fukuoka, Nagoya and Tsukuba in spring 2002

Figure 2 shows the aerosol concentrations against particle size at four stations in spring 2002. Every station has the highest dust concentrations during April 6-12 because a large-scale dust event was observed on April 7-10. The total dust concentrations decreased in the order of Fukuoka, Nagoya, Tsukuba and Naha (Kanai *et al.*, 2003). The distribution of particle size showed two peaks at 2.1-7.0 μm and 0.43-0.65 μm. When a large-scale dust event was observed, the concentrations of dust samples with 2.1-7.0 μm particle size especially increased. However, the peak particle size at 0.43-0.65 μm was not noticeable during the dust event. The distribution patterns of dust concentrations in Fig. 2 are well consistent with the previous data (e.g. Institute of Hydrospheric Sciences, Nagoya University, 1991) and indicate that a large- scale dust storm conveyed a large amount of mineral aerosol from East Asia to Japan.

4.2 Blank test and repeated measurements of JB-1

The weight of dust samples collected on a piece of filter in this study ranged from 0.1 mg to 12 mg, and the mean was 2.4 mg. The analytical method according to Imai (1990) usually requires about 200-250 mg of geological sample. The weight of a dust sample is only one-hundredth of that amount. Therefore, it is necessary to confirm whether or not the analytical

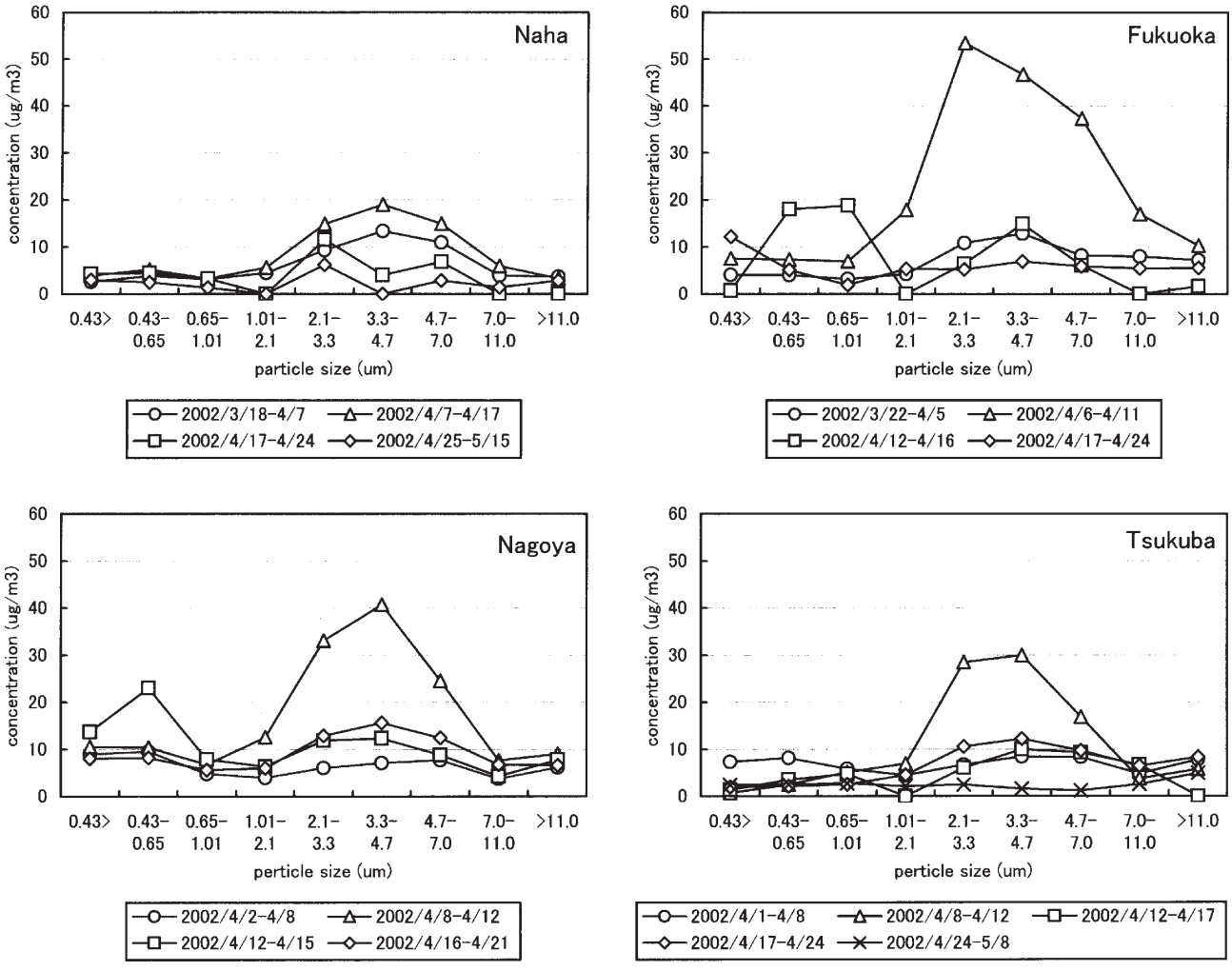

Fig. 2. Distribution of aerosol concentrations among particle size at Naha, Fukuoka, Nagoya and Tsukuba stations before, during and after the first intensive observation period (IOP) (IOP 1: 8-21, April in 2002).

method is applicable to a few mg of aeolian dust sample. We examined the blank of quartz, polyflon and membrane filters (Kanai *et al.*, 2002) and the accuracy of the analytical method by the repeated measurements of geological samples.

The blank test showed that blank values of polyflon and membrane filters are quite low, but quartz filters have high blank values for analyzed elements (Kanai *et al.*, 2003). Unfortunately, the concentrations of almost all elements in the two smallest fractions (0.65-0.43 μm and <0.43 μm), which were trapped by quartz filters, were near the blank values so their chemical composition data were eliminated.

The accuracy of the analytical method was examined by repeated measurements of JB-1, which is a basaltic rock and one of the Geochemical Reference Samples (Imai *et al.*, 1995). The 2 mg of JB-1 with a quarter of the PF-050 polyflon filter and membrane filter was digested with a mixed acid. The degradation product was finally diluted to 10 ml with MQ and 7N HNO_3. Table 1 shows the results of five repetitions. The recovery of most elements ranged from 80 to 120 % with an average of about 100 %. However, Cu, Cd, Sn, Sb and Ta have much larger values than the recommended or preferable values. Their excess recovery may be caused by some contamination, their poor sensitivity to the ICP-MS method and their very low concentrations. The standard deviations of alkali metals, V, Cu, Nb, Mo, Cd, Sb, Ta, Tl, Pb and Bi were about 20-50 % (90 % for Cd) and those of the other elements were about 5-10 %. Consequently, the analysis of a few mg of aeolian dust is semi-quantitative.

4.3 Analytical results of water-insoluble component

Table 2 shows the chemical composition of water-insoluble components of aeolian dust collected in Japan. Figure 3 shows the variation of chemical compositions in the air with grain size for some elements (Na_2O, Al_2O_3, CaO, Mo, Sb and Pb). Most elements have the same distribution patterns as Al_2O_3, which had two peaks at > 11 μm and 2.1- 4.7 μm (bimodal distribution). When a dust event was observed (April 6-12, 2002), elemental concentrations sharply increased at 2.1-7.0 μm of particle size. The distribution patterns of chemical composition are consistent with those of size- segregated measurements of aeolian dust (Fig. 2). It is obvious that most elements originate in mineral aerosol, as mentioned above.

However, some elements, Na_2O, CaO, Mo, Cd, Sb, Pb and Bi, have different distribution patterns from Al_2O_3. The Na_2O concentrations systematically decreased with decreasing particle size and have no clear peak, although its concentration for many samples was not detectable. The CaO had similar distribution patterns to Al_2O_3 during a dust event and to Na_2O during a non-dust event. The distribution pattern of Mo had two peaks at > 11 μm and 2.1-3.3 μm (Fig. 3) and was characterized by having a very high concentration in the largest particles (> 11 μm). The Cd, Sn, Pb and Bi concentrations gradually increased with the decrease of particle size (see Pb in Fig. 3). The Sb had one peak at 2.1-4.7 μm, but still had a high concentration in small particles (0.65-2.1 μm). The Na_2O, CaO, Mo, Cd, Sn, Sb, Pb and Bi do not always have high concentrations during a dust event, differing from Al_2O_3. They probably have different origins from mineral aerosol transported from Asia to Japan.

5. Discussion

5.1 Distribution patterns of elemental concentration normalized by Al_2O_3 content

Figure 3 shows that most elements have similar patterns to Al_2O_3 and originate in mineral aerosol. Except for SiO_2, Al_2O_3 is the most abundant in mineral aerosol, especially in clay minerals. The elemental concentrations divided by Al_2O_3 contents can be used to examine the change of mineralogical composition or estimate the influence of non-silicate minerals (calcite, gypsum and sea salt) and anthropogenic materials (Inst. Hydrosphere Sci. Nagoya Univ., 1991). Figure 4 shows the chemical compositions normalized by Al_2O_3 content at the Fukuoka and Nagoya stations. The concentration ratios of most elements to Al_2O_3 were constant from >11 μm to 1.01 μm, but suddenly increased at 0.65-1.01 μm. These features did not change whether or not a dust event was observed. It is concluded that the mineral composition is constant from >11 μm to 1.01 μm, but changes under 1 μm.

The concentration ratios of Na_2O, K_2O, CaO and Mo to Al_2O_3 for some samples gradually decrease with the decrease of particle size (Fig. 4). As mentioned above, the largest particle (>11 μm) contains a fairly high Mo concentration, but the other particles are poor in Mo (Fig. 3). The decreasing trend suggests that the mineralogical composition may change at a particle size >11 μm. On the other hand, the decreasing trend of Na_2O, K_2O and CaO might indicate that the compositions of K-feldspar, plagioclase, and mica and clay minerals gradually change. If it is true, the other elements will show compositional change with the decrease of the particle size, but this did not happen. Accordingly, it is unlikely that the change of mineral composition causes the decrease of concentration ratios of Na_2O, K_2O and CaO to Al_2O_3. We suppose that Na^+, K^+ and Ca^{2+} are trapped in a layer of clay mineral and easily dissolved in water when the size of clay mineral is small or non-silicate minerals (e.g. calcite, gypsum and sea salts) were not dissolved enough in MQ for the larger particles.

The concentration ratios of Cd, Sn, Sb, Pb and Bi to Al_2O_3 for dust samples with >11-2.1 μm of particle

Table 1 Analytical result of repeated test by using 2 mg of JB-1.

	n	mean	standard deviation (%)	recovery (%)	Recommended or preferable values*
(%)					
Na_2O	3	3.26	26	118	2.77
MgO	5	7.91	7	103	7.71
Al_2O_3	5	13.84	6	95	14.53
P_2O_5	5	0.254	5	100	0.255
K_2O	5	1.76	27	123	1.43
CaO	5	8.87	10	96	9.25
TiO_2	5	1.29	4	98	1.32
MnO	5	0.152	8	99	0.153
T-Fe_2O_3	5	8.19	5	91	8.99
(ppm)					
Li	5	10.5	19	92	11.5
Be	5	1.27	5	96	1.33
Sc	5	27.4	6	100	27.5
V	5	228	26	108	211
Cr	5	364	13	86	425
Co	5	37.6	6	99	38.2
Ni	5	136	11	102	133
Cu	3	250	50	454	55.1
Zn	2	77.7	–	91	85.2
Ga	5	17.4	7	97	17.9
Rb	5	42.9	9	104	41.3
Sr	4	443	4	100	444
Y	5	24.2	6	100	24.3
Zr	5	138	8	98	141
Nb	5	33.5	34	100	33.3
Mo	5	30.5	34	111	27.4
Cd	3	0.20	90	182	0.11
Sn	1	5.87	–	306	1.92
Sb	5	0.42	50	151	0.28
Cs	4	1.12	26	91	1.23
Ba	5	485	4	98	493
La	5	38.7	7	100	38.6
Ce	5	67.5	6	100	67.8
Pr	5	6.99	6	100	7.01
Nd	5	26.6	6	99	26.8
Sm	5	5.14	7	100	5.13
Eu	5	1.50	8	101	1.49
Gd	5	4.95	8	101	4.9
Tb	5	0.82	8	100	0.82
Dy	5	4.18	7	101	4.14
Ho	5	0.78	6	99	0.79
Er	5	2.27	10	100	2.27
Tm	5	0.34	5	97	0.35
Yb	5	2.09	7	98	2.13
Lu	5	0.31	9	102	0.31
Hf	5	3.27	11	99	3.31
Ta	5	6.23	33	213	2.93
Tl	5	0.08	35	77	0.1
Pb	4	12.2	44	122	10
Bi	4	0.035	49	105	0.033
Th	5	9.33	7	100	9.3
U	5	1.63	9	97	1.67

* Imai et al. (1995)

Table 2 Analytical results of aeolian dust in spring of 2002.

Stage	Size (um)	weight (mg)	Na2O (%)	MgO (%)	Al2O3 (%)	P2O5 (%)	K2O (%)	CaO (%)	TiO2 (%)	MnO (%)	Fe2O3 (%)	Li (ppm)	Be (ppm)	Sc (ppm)	V (ppm)	Cr (ppm)	Co (ppm)	Ni (ppm)
Nagoya no. 17: 2002/4/2-4/8, total flow rate = 229 m^3																		
0	>11.0	1.41	0.57	0.90	6.15	0.21	1.00	2.04	0.25	0.05	1.03	13	1.0	4.4		107	12	123
1	7.0-11.0	0.86		1.19	7.18	0.14		2.57	0.28	0.05	1.09	16	1.2	5.6			5.0	
2	4.7-7.0	1.77		0.73	5.54	0.11	0.03	0.96	0.21	0.03	1.16	14	1.1	4.2		50	6.3	
3	3.3-4.7	1.63		0.87	5.87	0.11		0.75	0.22	0.04	1.93	15	1.1	4.5		16	8.4	49
4	2.1-3.3	1.39		1.08	6.04	0.11		1.05	0.24	0.03	1.24	15	1.2	4.8			6.2	
5	1.01-2.1	0.91		0.57	3.39	0.07		1.19	0.15	0.02	0.50	2	0.6	2.4		106	2.7	100
6	0.65-1.01	1.10		0.45	0.63	0.05		4.07	0.07	0.02			0.01	1.4				
Nagoya no. 18: 2002/4/8-4/12, total flow rate = 166 m^3																		
0	>11.0	1.50		0.66	4.41	0.19	0.03	1.14	0.23	0.04	1.55	14	1.1	2.8			4.2	
1	7.0-11.0	1.27		1.46	7.81	0.17	0.19	1.03	0.43	0.05	1.71	31	2.2	8.0			10	
2	4.7-7.0	4.09		1.04	7.00	0.12	0.78	0.69	0.32	0.05	2.60	34	2.2	8.8			12	0.2
3	3.3-4.7	6.77		1.47	8.37	0.14	1.02	1.00	0.34	0.05	3.29	40	2.1	11	38		13	17
4	2.1-3.3	5.51		1.13	6.95	0.13	0.95	0.61	0.30	0.04	2.81	32	2.0	8.8			10	3
5	1.01-2.1	2.09		1.01	6.07	0.12	0.70	0.47	0.37	0.03	2.16	11	1.2	5.7			2.2	
6	0.65-1.01	1.13		0.55	1.84	0.09		1.60	0.35	0.03	0.25							
Nagoya no. 19: 2002/4/12-4/15, total flow rate = 123 m^3																		
0	>11.0	0.96	1.08	0.93	4.77	0.17	0.84	1.96	0.33	0.04	0.97							
1	7.0-11.0	0.53		1.47	5.42	0.18	0.27	3.24	0.48	0.04								
2	4.7-7.0	1.09		1.27	6.94	0.15	0.18	1.76	0.25	0.04	1.55							
3	3.3-4.7	1.52		0.85	5.41	0.11	0.04	0.78	0.20	0.03	1.10							
4	2.1-3.3	1.47		0.73	5.82	0.15		0.12	0.39	0.03	1.75							
5	1.01-2.1	0.78		0.51	3.82	0.11		0.06	0.19	0.02	0.36							
6	0.65-1.01	0.97			0.20	0.07				0.02								
Nagoya no. 20: 2002/4/16-4/21, total flow rate = 195 m^3																		
0	>11.0	1.29		0.55	3.28	0.15		0.56	0.15	0.02	0.45							
1	7.0-11.0	1.32		1.08	6.68	0.13		0.57	0.33	0.04	1.93		0.5	2.1				
2	4.7-7.0	2.43		1.05	5.91	0.12	0.41	0.42	0.29	0.03	1.87	10	0.8	4.8			5.1	
3	3.3-4.7	3.06		0.79	4.32	0.09	0.41	0.57	0.20	0.02	1.23	5	0.5	2.9			3.0	
4	2.1-3.3	2.52		1.20	7.00	0.13	0.48	0.44	0.33	0.04	2.21	29	1.7	8.6			8.9	3
5	1.01-2.1	1.17		0.92	5.08	0.12		0.38	0.25	0.03	1.65	13	1.0	6.1			3.4	
6	0.65-1.01	1.10		0.21	0.44	0.05		0.41	0.03	0.02								
Tsukuba no. 19: 2002/4/1-4/8, total flow rate = 283 m^3																		
0	>11.0	2.20		1.18	5.40	0.25	0.48	1.17	0.26	0.04	1.09	17	0.9	7.9			6.9	0.5
1	7.0-11.0	1.36	1.47	1.20	5.59	0.24	1.19	2.20	0.27	0.04	1.06	22	1.0	8.6			8.0	99
2	4.7-7.0	2.36		0.79	4.04	0.12	0.65	0.78	0.19	0.02	1.01	16	0.8	6.0			5.8	9
3	3.3-4.7	2.39		1.00	4.88	0.12	0.46	1.39	0.20	0.02	1.40	19	0.9	6.8			6.6	
4	2.1-3.3	1.90		0.77	4.22	0.13	0.40	1.07	0.19	0.02	1.25	17	1.0	6.3			4.7	
5	1.01-2.1	1.27		0.61	3.09	0.12	0.11	0.57	0.17	0.02		10	0.7	6.4			2.3	
6	0.65-1.01	1.65		0.21	0.15	0.07		0.34	0.004	0.01			0.1	2.0				
Tsukuba no. 20: 2002/4/8-4/12, total flow rate = 162 m^3																		
0	>11.0	0.93		0.74	4.38	0.28		0.87	0.18	0.03		12	1.0	8.0			2.4	
1	7.0-11.0	0.61		0.99	5.82	0.26		0.86	0.24	0.03		19	1.4	14			6.2	
2	4.7-7.0	2.73		1.47	8.75	0.20	1.12	0.95	0.37	0.05	3.04	34	1.8	11			12	2
3	3.3-4.7	4.85	0.66	1.17	6.81	0.14	1.30	0.69	0.33	0.04	2.57	28	1.5	8.4			9.3	16
4	2.1-3.3	4.61	0.10	1.30	7.73	0.15	1.29	0.57	0.36	0.04	2.76	28	1.6	10			10	17
5	1.01-2.1	1.12	0.34	1.23	8.64	0.21	1.62	0.42	0.47	0.04	1.96	29	2.0	13			7.7	
6	0.65-1.01	0.83	0.68	0.25	1.44	0.18	0.67	0.33	0.17	0.01		2	0.3	6.8				
Tsukuba no. 21: 2002/4/12-4/17, total flow rate = 197 m^3																		
0	>11.0	-																
1	7.0-11.0	1.32	4.81	0.69	4.51	0.24	1.71	0.98	0.21	0.04	0.71	11	0.6	8.2			5.0	
2	4.7-7.0	1.84	3.73	1.02	7.13	0.23	1.75	0.82	0.34	0.05	2.03	25	1.2	10			8.8	
3	3.3-4.7	1.97	0.32	0.92	5.94	0.18	0.82	0.46	0.28	0.04	1.41	22	1.1	8.5			6.8	
4	2.1-3.3	1.20		1.19	8.69	0.22	0.80	0.82	0.39	0.04	2.17	32	1.7	11			12	
5	1.01-2.1	0.00																
6	0.65-1.01	0.94	4.56	0.23	0.33	0.16	1.39	0.56	0.01	0.02			0.1	3.5				
Tsukuba no. 22: 2002/4/17-4/24, total flow rate = 285 m^3																		
0	>11.0	2.36	2.11	0.86	11.04	0.45	1.07	1.76	0.60	0.10	3.95	21	1.1	17			15	44
1	7.0-11.0	1.83	2.31	0.24	1.95	0.12	0.78	0.34	0.11	0.01		4	0.3	3.5			1.2	
2	4.7-7.0	2.76	0.71	0.58	4.08	0.12	0.63	0.47	0.19	0.03	1.16	14	0.7	5.9			4.7	
3	3.3-4.7	3.48	0.68	0.95	5.98	0.16	0.96	0.66	0.37	0.04	1.70	23	1.2	7.5			7.7	
4	2.1-3.3	3.01	0.23	0.45	2.48	0.09	0.49	0.30	0.14	0.01	0.76	9	0.7	3.3	27		3.4	
5	1.01-2.1	1.29		0.74	3.19	0.12	0.38	1.12	0.24	0.02	0.71	10	0.7	4.7	62		1.9	
6	0.65-1.01	0.72	3.55	0.63	0.39	0.13	0.89	1.41		0.01			0.2	5.3				
Tsukuba no. 23: 2002/4/24-4/, total flow rate = 563 m^3																		
0	>11.0	2.77		0.31	4.13	0.20	0.10	2.37	0.19	0.04	1.19	43	0.3	5.1			4.4	27
1	7.0-11.0	1.44		0.33	3.89	0.15		0.62	0.21	0.04	1.41	82	0.6	6.4			5.5	
2	4.7-7.0	0.69		0.89	10.53	0.33		0.31	0.57	0.09	4.52	218	1.6	15		166	12	
3	3.3-4.7	0.89		0.47	5.54	0.22			0.33	0.04	2.45	155	1.2	9.1			7.3	
4	2.1-3.3	1.38		0.38	3.07	0.14		0.16	0.17	0.02	1.93	90	0.6	4.5			3.2	
5	1.01-2.1	1.21		0.18	2.20	0.04		1.89	0.10	0.02	0.90	116	0.3	2.9			0.4	
6	0.65-1.01	1.56				0.03			0.01	0.01	0.10	88	0.1	0.7				

Table 2 Analytical results of aeolian dust in spring of 2002 (continue).

Stage	Size (um)	weight (mg)	Na2O (%)	MgO (%)	Al2O3 (%)	P2O5 (%)	K2O (%)	CaO (%)	TiO2 (%)	MnO (%)	Fe2O3 (%)	Li (ppm)	Be (ppm)	Sc (ppm)	V (ppm)	Cr (ppm)	Co (ppm)	Ni (ppm)
Naha no. 19: 2002/3/18-4/7, total flow rate = 799 m^3																		
0	>11.0	2.93		1.06	9.64	0.13		3.21	0.24	0.04	1.87	58	1.1	6.4			8.7	51
1	7.0-11.0	3.14		1.83	8.94	0.21	0.79	3.24	0.37	0.05	3.14	61	1.8	9.4			11	28
2	4.7-7.0	8.75		1.17	6.51	0.14	0.74	3.54	0.23	0.04	2.24	30	1.2	6.6			8.1	16
3	3.3-4.7	10.70		1.06	5.55	0.15	0.94	1.26	0.27	0.04	2.76	26	1.4	6.4			10	21
4	2.1-3.3	7.46		1.21	6.98	0.17	1.04	0.72	0.33	0.05	3.37	39	1.7	8.4			12	43
5	1.01-2.1	3.55		1.17	6.58	0.12	0.58	0.17	0.32	0.03	2.56	60	1.5	8.2			7.5	10
6	0.65-1.01	2.51		0.10	1.00				0.05	0.02	0.12	67	0.3	1.6			0.6	
Naha no. 20: 2002/4/7-4/17, total flow rate = 403 m^3																		
0	>11.0	1.34		0.57	3.82	0.04		1.49	0.17	0.02	0.81	106	0.6	4.3	65		6.5	1
1	7.0-11.0	2.40		0.89	5.50	0.09		1.60	0.22	0.03	2.09	54	0.9	6.1	138		6.6	109
2	4.7-7.0	6.02		1.19	6.63	0.12	0.78	0.99	0.28	0.04	2.66	31	1.3	8.2	110		9.4	18
3	3.3-4.7	7.66		1.17	7.19	0.12	1.04	0.90	0.33	0.04	2.93	28	1.4	8.4	125		11	24
4	2.1-3.3	6.00		1.34	8.01	0.14	1.15	0.57	0.40	0.04	3.30	33	1.6	9.3	123		11	33
5	1.01-2.1	2.27		0.95	7.35	0.09	0.50	1.42	0.34	0.03	2.49	52	1.4	8.2	134		8.2	5
6	0.65-1.01	1.31		0.08	1.59				0.06	0.02	1.06	64	0.3	1.7	248		1.0	63
Naha no. 21: 2002/4/17-4/24, total flow rate = 299 m^3																		
0	>11.0	-																
1	7.0-11.0	-																
2	4.7-7.0	2.05		0.35			0.50		0.04	0.01	0.40	0.6	0.4	1.6			3.7	18
3	3.3-4.7	1.21		1.72	5.35		1.20		0.30	0.04	3.35	27	1.7	8.7			16	45
4	2.1-3.3	3.47		0.25			0.05		0.04	0.01	0.84	1	0.3	1.2			2.8	3
5	1.01-2.1	-																
6	0.65-1.01	0.97															1.7	23
Naha no. 22: 2002/4/25-5/15, total flow rate = 816 m^3																		
0	>11.0	2.33								0.003							1.1	63
1	7.0-11.0	1.18									0.27						2.2	19
2	4.7-7.0	2.35							0.01	0.01	0.54		0.1				2.4	17
3	3.3-4.7	-																
4	2.1-3.3	5.06		0.01			0.11		0.01		0.19		0.07		3		1.1	4
5	1.01-2.1	-																
6	0.65-1.01	1.07									0.15						0.5	117
Fukuoka no. 18: 2002/3/22-4/5, total flow rate = 575 m^3																		
0	>11.0	4.16	3.94	1.40	6.32	0.13	2.30		0.34	0.05	2.65	25	1.5	6.9	127		11	46
1	7.0-11.0	4.60	1.11	0.65	2.69	0.04	0.97		0.16	0.02	1.25	11	0.8	3.5			6	35
2	4.7-7.0	4.71	1.20	0.83	3.36	0.08	1.03		0.19	0.02	1.49	17	0.9	4.8			9	850
3	3.3-4.7	7.40	0.25	1.11	5.60	0.12	1.05		0.28	0.04	2.65	26	1.3	6.5	21		9	26
4	2.1-3.3	6.26		1.27	7.13	0.15	1.28		0.39	0.04	3.42	31	1.7	8.4	61		11	46
5	1.01-2.1	2.43		0.53	1.62	0.03	0.33		0.19	0.02	1.69	8	0.9	3.5	5		5	21
6	0.65-1.01	1.85								0.03	0.68		0.1		2		3	140
Fukuoka no. 19: 2002/4/6-4/11, total flow rate = 215 m^3																		
0	>11.0	2.21	3.80	1.67	8.91	0.15	2.35	1.10	0.35	0.06	2.46	36	1.7	10	21	67	13	114
1	7.0-11.0	3.64	0.67	1.07	6.35	0.10	1.11	0.55	0.24	0.04	1.98	24	1.2	7.0	28	61	8	39
2	4.7-7.0	8.00	0.36	1.18	7.39	0.12	1.21	0.74	0.30	0.06	2.67	28	1.5	8.0	35	62	10	89
3	3.3-4.7	10.02	0.11	1.36	8.95	0.15	1.46	0.70	0.35	0.07	3.69	35	1.8	9.8	59	65	12	27
4	2.1-3.3	11.46	0.12	1.34	8.76	0.14	1.41	0.75	0.35	0.07	3.69	32	1.7	9.7	44	52	12	49
5	1.01-2.1	3.83		1.09	6.94	0.09	0.86	0.91	0.30	0.04	2.19	25	1.3	7.8	3	20	7	16
6	0.65-1.01	1.49		0.21	1.40				0.07	0.02	0.74	7	0.4	1.9	27	26	1	
Fukuoka no. 20: 2002/4/12-4/16, total flow rate = 165 m^3																		
0	>11.0	0.27		3.39		0.15	2.36		0.89	0.12	3.53	72	3.4	17			25	60
1	7.0-11.0	-																
2	4.7-7.0	1.01		2.59		0.15	1.69	1.96	0.57	0.08	4.22	65	2.8	17	122	104	20	17
3	3.3-4.7	2.45		1.44	7.64	0.10	0.93	0.44	0.30	0.04	2.25	34	1.5	8	53	49	11	27
4	2.1-3.3	1.06		3.16		0.25	2.52	2.28	0.77	0.10	6.54	83	4.0	21	246	80	25	67
5	1.01-2.1	-																
6	0.65-1.01	3.10		0.04					0.01	0.01	0.05	2	0.1	0.1			0.1	
Fukuoka no. 20: 2002/4/17-4/24, total flow rate = 323 m^3																		
0	>11.0	1.80	12.0	1.31	6.31	0.13	4.30	1.12	0.30	0.04	1.76	26	1.1	6.5	299	148	9	90
1	7.0-11.0	1.76	4.23	0.96	4.84	0.09	2.04	0.15	0.22	0.03	1.44	21	1.1	5.1	88		7	42
2	4.7-7.0	1.91		1.32	6.96	0.12	0.99	0.39	0.31	0.04	2.55	29	1.4	8.1			10	17
3	3.3-4.7	2.25		1.04	6.40	0.13	0.85	0.71	0.31	0.04	2.37	27	1.5	6.7			9	29
4	2.1-3.3	1.69		1.67	10.24	0.21	1.07	2.52	0.46	0.05	3.24	44	1.9	11	30	55	13	18
5	1.01-2.1	1.72		0.39	2.35	0.02			0.10	0.01	1.27	9	0.5	2.5			2	
6	0.65-1.01	0.63								0.02	1.48		0.2					

Table 2 Analytical results of aeolian dust in spring of 2002 (continue).

Stage	Cu (ppm)	Zn (ppm)	Ga (ppm)	Rb (ppm)	Sr (ppm)	Y (ppm)	Zr (ppm)	Nb (ppm)	Mo (ppm)	Cd (ppm)	Sn (ppm)	Sb (ppm)	Cs (ppm)	Ba (ppm)	La (ppm)	Ce (ppm)	Pr (ppm)	Nd (ppm)	Sm (ppm)	Eu (ppm)
Nagoya no. 17: 2002/4/2-4/8, total flow rate = 229 m^3																				
0		388	8	59	81	12	52	8	14	0.1		11	3.5	324	17	26	2.8	11	1.9	0.5
1		461	11	51	95	14	103	7	15			34	4.8	392	28	35	3.9	14	3.0	0.5
2		270	9	41	58	9	93	6	10	0.3		36	3.7	360	14	28	2.7	11	2.3	0.4
3		126	10	43	54	9	87	5	15	2.0		52	3.6	444	15	31	3.0	12	2.4	0.4
4		579	11	44	82	11	121	4	23	0.5		71	4.3	580	18	35	3.6	13	2.4	0.5
5			9	13	52	6	51	4	15	1.0		61	3.1	295	18	20	2.3	8	1.3	0.1
6			5		102	6	57	6		1.8		43	0.6	28	14	7	0.7	2	0.5	0.02
Nagoya no. 18: 2002/4/8-4/12, total flow rate = 166 m^3																				
0			7	30	48	7	29	8	45			6	2.5	195	6	20	2.2	8	1.9	0.3
1			16	68	75	21	87	10	17			12	5.6	347	30	46	5.0	20	3.6	0.6
2			14	67	61	15	66	7	6	0.01		8	4.7	293	17	40	4.3	17	3.0	0.7
3			15	83	81	18	63	7	5	0.3		9	6.1	354	24	50	5.4	22	4.4	0.9
4			14	67	58	16	70	6	8	1.8		10	4.8	293	18	40	4.3	17	3.1	0.6
5			13	51	52	15	60	6	5	0.7		22	5.0	256	16	41	4.3	16	3.1	0.5
6			5		71	9	143	6	3	2.3		41	1.9	74	18	17	1.9	5	0.8	0.1
Nagoya no. 19: 2002/4/12-4/15, total flow rate = 123 m^3																				
0	615		3	31	74	14	18	2	97			9	3.3	225	7	27	2.8	9	2.4	0.3
1	749		1		121	25	35	5	33			20	2.4	238	25	22	2.5	8	1.3	0.4
2	463		6	15	92	6			10			35	3.9	337	9	30	3.0	11	1.7	0.4
3	532		6	16	57	5			8			35	3.2	272	5	26	2.9	9	1.7	0.4
4	685		8	14	39	7	93	12	23	0.1		43	3.5	308	8	37	3.7	14	2.0	0.4
5	1250		8		24	0.2	16	2	10	1.0		59	3.4	159		18	1.9	5	0.7	0.1
6	1030		6						5	1.3		40	0.7							
Nagoya no. 20: 2002/4/16-4/21, total flow rate = 195 m^3																				
0	1000		2		30	3			80			7	2.0	160		13	1.3	5	0.7	0.3
1	950		7	31	60	8	16	0.4	29			18	4.3	313	8	35	3.7	14	2.3	0.5
2	399		9	44	54	11	37	4	16			17	4.1	300	12	35	3.7	14	2.5	0.6
3	440		6	28	37	7	28	2	11			16	2.8	249	9	25	2.4	9	1.7	0.3
4	509		14	63	61	14	61	5	17	0.1		23	5.5	350	16	44	4.8	19	3.8	0.6
5			13	40	55	8	56	1	19	1.1		39	5.0	345	7	32	3.4	12	2.3	0.5
6	374		3		13				7	0.8		35	0.9	17		2	0.1			
Tsukuba no. 19: 2002/4/1-4/8, total flow rate = 283 m^3																				
0	664	1046	9	29	60	10	59	6	0.4		10	10	2.8	196	11	25	2.7	11	2.4	0.5
1	1540	1747	10	36	94	10	44	6		17	12	16	3.8	253	20	28	2.9	12	2.5	0.5
2	1270	208	7	28	49	8	38	3		1	11	14	2.8	181	9	22	2.5	10	2.1	0.4
3	1610		9	32	78	9	46	4	0.2		102	24	3.6	250	11	29	3.1	12	2.2	0.5
4	1580	106	9	21	61	7	62	4		1	178	33	3.2	250	9	28	2.8	11	2.0	0.5
5	1150	1099	9	4	26	6	69	10		10	2566	39	2.9	168	4	23	2.3	9	1.5	0.3
6	951	101	4		3	1	6	5		10	1209	25	0.3	2		4	0.5	1	0.2	0.02
Tsukuba no. 20: 2002/4/8-4/12, total flow rate = 162 m^3																				
0	1170		7	2	43	8	235	5	53			7	2.9	156		24	2.4	10	1.7	0.4
1	1240		11		35	19	400	10	15			8	3.6	201		45	4.9	20	3.7	0.7
2	365	265	15	72	73	16	151	8	0.1			8	6.0	348	19	51	5.4	21	3.8	0.8
3	528	454	12	65	61	13	77	7	0.9	2	9	8	5.1	279	18	40	4.3	16	3.2	0.7
4	239	650	14	69	64	15	77	8	1.1	2	21	9	5.6	301	20	48	5.0	19	3.6	0.7
5	1160	587	18	55	59	17	252	10	1.1	4	160	29	5.9	336	16	53	5.3	21	3.9	0.8
6	1850	560	5		14	5	358	14		12		38	1.5	71		12	1.3	5	0.9	0.1
Tsukuba no. 21: 2002/4/12-4/17, total flow rate = 197 m^3																				
0																				
1	659		6	20	34	10	75	2	5			9	2.3	151	7	18	2.0	8	1.6	0.3
2	429	263	10	47	57	13	46	7	2	0.6		16	4.0	258	12	32	3.5	15	2.7	0.6
3	55		10	36	43	12	62	4	1			15	3.7	244	10	33	3.5	13	2.8	0.5
4	2130	190	16	43	71	14	80	7	5	0.9		35	5.3	362	12	48	4.8	18	3.4	0.8
5																				
6	869		6		8	0	27	8		12		30	0.6	26		6	0.6	2	0.6	0.04
Tsukuba no. 22: 2002/4/17-4/24, total flow rate = 285 m^3																				
0	1020	25	14	31	69	19	72	12	24			4	3.1	220	15	39	4.4	18	3.9	0.8
1	488		3	2	12	3	14	19	4			2	1.1	69	1	8	1.0	4	0.7	0.2
2	246		7	23	33	7	30	3	1			6	2.2	157	6	21	2.2	9	1.8	0.4
3	492	127	10	44	54	11	52	8	2	1.9		16	3.6	254	12	32	3.5	14	2.6	0.6
4	241		5	14	22	5	25	3	0.01	1.5		12	1.7	128	3	16	1.5	6	1.3	0.2
5	1140	23	6	5	53	5	21	3		7.1		25	2.2	193	1	20	2.0	7	1.5	0.3
6	1120		4		35	0	74	9		17		36	0.7	23		7	0.7	3	0.3	0.02
Tsukuba no. 23: 2002/4/24-4/, total flow rate = 563 m^3																				
0	929		5	13	139	5	40	2	3			7	2.6	114	7	13	1.5	5	1.2	0.3
1	1150		7	12	45	7	26	0.4	1			18	1.3	155	8	15	1.8	7	1.9	0.5
2			15	53	64	16	80	1	5			80	3.9	520	15	44	5.2	18	3.7	1.2
3			10	17	32	10	67					96	1.7	401	7	24	2.7	10	2.3	0.7
4			7	19	34	4	44					102	1.5	307	6	15	1.9	7	1.5	0.3
5			5		133	2	38	1				77	1.3	110	4	10	1.3	3	0.6	0.1
6			1									58		12		0.3	0.2	0.04		0.1

Table 2 Analytical results of aeolian dust in spring of 2002 (continue).

Stage	Cu (ppm)	Zn (ppm)	Ga (ppm)	Rb (ppm)	Sr (ppm)	Y (ppm)	Zr (ppm)	Nb (ppm)	Mo (ppm)	Cd (ppm)	Sn (ppm)	Sb (ppm)	Cs (ppm)	Ba (ppm)	La (ppm)	Ce (ppm)	Pr (ppm)	Nd (ppm)	Sm (ppm)	Eu (ppm)
Naha no. 19: 2002/3/18-4/7, total flow rate = 799 m^3																				
0	1520		10	48	92	9	29	1	17			20	3.5	230	11	25	2.9	11	2.0	0.4
1	1220		14	79	121	15	46	2	2			22	5.7	364	25	45	5.2	18	3.8	1.0
2	141		10	54	167	9	49	2	1			13	4.9	280	14	32	3.4	13	2.3	0.5
3	139		11	52	64	9	41	2	2			16	3.4	248	13	30	3.1	12	2.2	0.5
4			14	71	70	10	54	3	3			22	4.4	354	16	43	3.8	15	3.3	0.6
5			18	72	49	10	41	2	2			37	7.0	293	17	40	4.1	15	3.4	0.7
6			11	14	14	1	14					23	1.5	30		5	0.5	2	0.2	0.1
Naha no. 20: 2002/4/7-4/17, total flow rate = 403 m^3																				
0			5	27	41	5	7		22			7	1.3	124	2	12	1.6	5	1.0	0.5
1			8	37	98	8	36	0.3	1			8	3.2	206	10	24	2.7	10	1.7	0.7
2			10	60	68	11	44	3	1			7	4.3	264	15	34	3.7	14	2.7	0.7
3			12	61	71	12	53	3	1			7	4.4	280	17	38	4.0	15	3.0	0.6
4			14	73	66	14	64	4	1			9	5.1	316	20	44	4.6	19	3.5	0.8
5			15	53	125	11	63	2	2			17	6.1	293	15	41	3.8	16	2.5	0.7
6			8		18	1						15	1.7	66		7	1.0	2	0.4	0.3
Naha no. 21: 2002/4/17-4/24, total flow rate = 299 m^3																				
0																				
1																				
2	787	238		28		3			0.31	0.3	53		262	93	3	8	0.9	4	0.8	0.3
3	232	536	7	83		15		0.34	3.49	0.4	70		319	390	21	49	5.4	21	4.3	0.9
4	159	67		14		2			11	0.02	28		149	54	2	6	0.6	3	0.6	0.1
5																				
6	997								1.92	1.2	65	1.8	547							
Naha no. 22: 2002/4/25-5/15, total flow rate = 816 m^3																				
0		28		0.08					9.2	0.08	38		174	9						0.01
1	670	202							7.7		73		327							
2	603	141		6.4					4.8	0.2	30		146	59						0.04
3																				
4	281			4.4					3.2	0.3	14		65	34						0.05
5																				
6	430								3.0	3.8	58		300							
Fukuoka no. 18: 2002/3/22-4/5, total flow rate = 575 m^3																				
0	1520	412	8	70		12	12	5	22	1.3	10		13	319	17	36	4.0	15	3.1	0.6
1	421	120	4	37		6		2	7.7	0.3	13		30	175	8	17	2.0	7	1.6	0.4
2	492	142	6	45		8	10	4	6.4	1.2	3		20	212	12	24	2.7	10	1.9	0.4
3	168	174	10	55		11	37	5	5.8	1.5				315	16	35	3.9	15	2.7	0.5
4	493	306	14	65		13	55	7	8.8	3.4				385	21	47	4.6	18	3.3	0.7
5		259	8	37		7		2	6.7	5.1			53	155	11	23	2.3	9	2.0	0.3
6	267	321	6	4					21	6.7			112	13						0.1
Fukuoka no. 19: 2002/4/6-4/11, total flow rate = 215 m^3																				
0	3270	318	13	91	75	16	30	8.5	6.3	2.0		4	5.2	361	23	46	5.2	21	3.8	0.8
1	958	119	10	60	50	11	24	4.7	1.8	0.7		2	4.3	258	15	32	3.6	14	2.8	0.6
2	335	92	12	66	67	13	47	5.6	1.2	0.7	0.3	2	5.1	295	18	40	4.1	16	3.1	0.7
3	133	114	14	82	74	14	58	7.1	1.2	0.7	0.2	3	6.2	361	20	46	4.8	19	3.8	0.8
4	778	195	14	76	78	15	59	6.7	1.5	1.4	0.7	3	6.5	346	21	49	5.0	20	3.9	0.8
5	512	264	12	62	82	14	45	7.0	2.5	3.9	1.5	6	5.6	257	19	42	4.5	16	3.1	0.6
6	302	393	7	16		3		0.6		5.5	101	10	0.2	61	3	10	1.0	4	0.8	0.1
Fukuoka no. 20: 2002/4/12-4/16, total flow rate = 165 m^3																				
0	3400	953	26	186		25		12	203	17		5		806	33	79	9	35	8	1.9
1																				
2	815	315	24	131	179	28	39	12	6	0.4		9	8	592	38	80	9	32	6	1.3
3	7700	172	13	68	54	14	24	6	3	0.8		5	4	323	24	43	5	18	3	0.7
4	1980	729	36	158	220	35	91	16	12	6.2	9.4	18	12	765	52	111	11	42	8	1.5
5																				
6	624	139	3.7	4.7		0.1			0.4	2.2	45.3	6.1		14.6	0.5	2.0	0.2	0.5	0.2	0.0
Fukuoka no. 20: 2002/4/17-4/24, total flow rate = 323 m^3																				
0	5240	346	8.2	80	52	11	2.5	5.2	37	1.8		8	3.5	295	15	32	3.6	14	2.6	0.6
1	1920	238	7.1	57	11	8.5		2.8	14	0.8		7	1.9	239	11	25	2.8	10	2.1	0.5
2	738	336	11	67	48	14	29	8.1	12	0.8	853	25	3.8	347	18	39	4.3	17	2.9	0.7
3	539	228	12	53	63	11	39	6.3	8	1.9		18	3.2	297	16	37	3.6	14	2.8	0.6
4	353	367	19	80	202	18	93	10	11	4.0		26	9.6	471	26	59	6.0	24	4.0	0.8
5		185	4.1	20	12	4.1		1.6	5	4.2		10	1.0	133	8	15	1.6	6	1.2	0.2
6	440	305	0.9	0.6					7	13		21		45		3	0.1		0.0	

Table 2 Analytical results of aeolian dust in spring of 2002 (continue).

Stage	Gd (ppm)	Tb (ppm)	Dy (ppm)	Ho (ppm)	Er (ppm)	Tm (ppm)	Yb (ppm)	Lu (ppm)	Hf (ppm)	Ta (ppm)	Tl (ppm)	Pb (ppm)	Bi (ppm)	Th (ppm)	U (ppm)
Nagoya no. 17: 2002/4/2-4/8, total flow rate = 229 m^3															
0	2.3	0.7	1.7	0.4	1.4	0.2	1.2	0.2	3	0.2	0.3	109	0.5	5.2	1.7
1	2.5	0.4	2.4	0.4	1.2	0.2	1.4	0.2	3	35	0.4	137	1.2	7.0	2.2
2	1.9	0.3	1.5	0.3	1.0	0.1	0.6	0.1	3		0.4	97	1.1	4.9	1.5
3	2.5	0.4	1.8	0.3	0.9	0.1	0.7	0.1	2	1	0.6	118	1.2	5.3	1.5
4	2.4	0.3	2.0	0.4	1.1	0.1	0.9	0.1	3	10	0.7	256	3.2	5.7	2.7
5	2.1	0.2	1.2	0.2	0.4	0.1	0.4	0.1	1		0.6	783	8.4	3.4	2.2
6	0.5	0.1	0.9	0.2	0.4	0.1	0.9	0.1	2	23	0.3	930	6.7	2.1	1.7
Nagoya no. 18: 2002/4/8-4/12, total flow rate = 166 m^3															
0	1.5	0.2	1.3	0.2	0.6	0.1	0.7	0.1	1	21	0.1	68	0.7	4.5	2.7
1	4.1	0.6	3.1	0.6	2.4	0.4	3.2	0.3	3	16	0.4	87	1.1	8.9	2.5
2	3.3	0.5	2.5	0.5	1.3	0.2	1.5	0.2	2	4	0.5	54	0.6	6.6	1.8
3	3.8	0.6	3.1	0.6	1.6	0.3	1.6	0.2	2	3	0.6	73	0.7	8.1	1.9
4	2.9	0.5	2.3	0.5	1.5	0.2	1.3	0.2	2		0.5	84	1.3	6.9	1.8
5	2.7	0.4	2.3	0.5	1.3	0.2	1.6	0.2	2		0.5	361	5.2	7.4	1.7
6	1.0	0.2	1.3	0.3	1.1	0.1	1.4	0.2	6	16	0.3	956	9.6	4.5	2.3
Nagoya no. 19: 2002/4/12-4/15, total flow rate = 123 m^3															
0	2.1	0.3	2.6	0.5	1.6	0.3	1.4	0.3	2	2		178	1.0	9.3	2.5
1	2.2	0.5	3.4	0.9	3.0	0.4	3.0	0.6	6			233	1.1	10	4.3
2	1.7	0.3	1.4	0.3	0.6	0.1	0.7	0.04	0	12	0.2	194	1.2	5.5	2.0
3	1.8	0.2	1.3	0.3	0.6	0.1	0.6	0.1	1	13	0.2	127	1.3	4.8	1.5
4	2.0	0.3	1.7	0.3	0.6	0.1	0.6	0.1	3	5	0.4	268	3.5	5.7	1.5
5	1.0	0.0	0.8	0.1	0.3		0.03	0.01	1	24	0.6	1080	11	3.7	1.1
6										9	0.5	1240	9.6	0.4	1.0
Nagoya no. 20: 2002/4/16-4/21, total flow rate = 195 m^3															
0	1.0	0.1	1.0	0.2	0.4	0.0	0.0	0.04	0.4	17		29	0.2	3.3	1.0
1	2.3	0.3	2.0	0.3	0.8	0.1	1.0	0.1	0.7	0.3	0.2	81	0.9	6.0	1.7
2	2.5	0.3	1.8	0.4	0.9	0.1	0.9	0.2	1	9	0.4	73	0.7	5.6	1.7
3	1.9	0.3	1.2	0.3	0.6	0.1	0.7	0.1	1	2	0.3	105	1.2	4.4	1.6
4	3.2	0.5	2.2	0.5	1.2	0.2	1.3	0.2	1	10	0.5	99	1.1	6.7	1.8
5	2.5	0.3	1.7	0.3	0.7	0.1	0.8	0.1	3	17	0.7	679	7.6	4.5	1.7
6	0.1									16	0.1	724	6.7	0.6	0.6
Tsukuba no. 19: 2002/4/1-4/8, total flow rate = 283 m^3															
0	2.1	0.4	1.8	0.3	1.0	0.2	1.0	0.2	2	8	0.4	55	1.2	4.9	1.4
1	2.0	0.3	1.7	0.3	0.9	0.2	0.8	0.2	1	7	0.5	658	2.6	5.1	1.7
2	1.6	0.3	1.4	0.2	0.7	0.1	0.8	0.1	1	5	0.3	243	2.1	4.0	1.1
3	1.9	0.3	1.6	0.3	0.8	0.2	0.8	0.1	1	0.4	0.4	314	2.8	4.6	1.0
4	1.9	0.4	1.5	0.3	0.6	0.1	0.8	0.1	1	2	0.5	1130	7.4	5.1	1.1
5	1.2	0.2	1.1	0.2	0.5	0.1	0.5	0.2	2	12	0.6	5370	17	3.8	0.9
6	0.3	0.02	0.3	0.007	0.2	0.04	0.1	0.04	0.2	6	0.3	4880	12	1.0	0.3
Tsukuba no. 20: 2002/4/8-4/12, total flow rate = 162 m^3															
0	2.0	0.3	1.5	0.2	0.9	0.2	0.7	0.1	13	16	0.4		1.3	5.5	1.1
1	3.0	0.6	3.3	0.5	1.5	0.3	2.2	0.4	22	42	1.7		2.7	12	2.3
2	3.9	0.6	3.0	0.5	1.5	0.2	1.4	0.2	6	6	0.7	92	1.9	8.5	1.9
3	2.9	0.4	2.2	0.4	1.2	0.2	1.2	0.2	3	12	0.6	221	1.9	6.8	1.4
4	3.2	0.5	2.7	0.5	1.4	0.2	1.5	0.2	3	4	0.6	431	3.9	8.1	1.6
5	3.7	0.5	3.0	0.5	1.7	0.3	1.7	0.2	11	5	1.0	1930	18	10	2.1
6	1.0	0.1	0.9	0.1	0.7	0.1	0.5	0.1	18	23	0.8	3860	26	6	0.9
Tsukuba no. 21: 2002/4/12-4/17, total flow rate = 197 m^3															
0															
1	1.9	0.3	1.5	0.3	0.8	0.2	0.9	0.1	3	13	0.2	21	1.2	3.5	1.0
2	2.6	0.5	2.2	0.4	0.9	0.2	1.1	0.1	1	11	0.5	293	2.4	5.3	1.7
3	2.4	0.4	2.0	0.4	1.1	0.2	1.1	0.2	2	6	0.4	238	2.8	5.6	1.3
4	3.2	0.5	2.3	0.4	1.2	0.2	1.2	0.2	2	14	0.8	1170	10	7.8	2.9
5															
6	0.4	0.06	0.2	0.03	0.1	0.1	0.2	0.01	0.3	27	0.6	4280	20	1.8	1.2
Tsukuba no. 22: 2002/4/17-4/24, total flow rate = 285 m^3															
0	3.6	0.6	3.2	0.6	1.5	0.3	1.4	0.2	1.9	7	0.3	69	1.1	6.2	2.4
1	0.8	0.1	0.6	0.1	0.3	0.1	0.4	0.1	0.3	23	0.1		0.6	1.9	0.8
2	1.7	0.3	1.3	0.3	0.6	0.1	0.6	0.1	0.7	1	0.3		1.0	3.5	1.4
3	2.4	0.4	2.0	0.3	1.0	0.2	1.0	0.1	1.4	3	0.4	146	1.6	5.4	1.6
4	1.1	0.1	0.8	0.2	0.5	0.1	0.5	0.1	0.6	2	0.3	93	1.6	2.6	1.4
5	1.1	0.1	1.1	0.2	0.5	0.1	0.5	0.1	0.5	7	0.4	1080	7.3	3.4	1.6
6	0.5	0.1	0.3		0.0	0.1	0.1	0.03	3.2	27	0.5	3800	15	2.5	0.9
Tsukuba no. 23: 2002/4/24-4/, total flow rate = 563 m^3															
0	0.8	0.2	1.0	0.1	0.6		0.3	0.1	0.9	2.9	0.01	72	0.3	2.1	1.0
1	1.7	0.3	1.0	0.2	1.0		0.5	0.08	1.1	4.8	0.1	166	0.7	3.0	1.3
2	3.6	0.5	2.7	0.5	2.9		0.8	0.3	1.8	5.7	0.3	427	2.3	5.5	2.4
3	1.8	0.1	1.7	0.1	1.3		0.9	0.1	1.5	2.5	0.6	427	2.5	3.9	2.3
4	1.5	0.2	0.7		0.6		0.4		0.1		0.2	1070	4.1	2.1	1.9
5	1.0	0.1	0.4		0.3		0.1	0.05	0.6			2860	8.0	1.6	0.6
6	0.1	0.1			0.1			0.1				3150	6.3	0.2	0.5

Table 2 Analytical results of aeolian dust in spring of 2002 (continue).

Stage	Gd (ppm)	Tb (ppm)	Dy (ppm)	Ho (ppm)	Er (ppm)	Tm (ppm)	Yb (ppm)	Lu (ppm)	Hf (ppm)	Ta (ppm)	Tl (ppm)	Pb (ppm)	Bi (ppm)	Th (ppm)	U (ppm)
Naha no. 19: 2002/3/18-4/7, total flow rate = 799 m^3															
0	2.0	0.3	1.3	0.2	0.7	0.1	0.6	0.2	0.8	1.5	0.2	252	0.9	4.6	1.3
1	3.4	0.6	2.6	0.4	1.3	0.1	1.0	0.1	1.6	2.0	0.4	446	1.3	7.8	2.0
2	2.2	0.3	1.7	0.3	0.9	0.1	1.0	0.1	1.2		0.4	336	1.0	5.2	1.6
3	2.0	0.3	1.7	0.2	0.8	0.1	0.8	0.1	1.0		0.5	470	1.6	5.0	1.7
4	2.5	0.4	1.8	0.3	1.0	0.2	0.7	0.1	1.4		0.7	1340	4.7	6.7	2.0
5	2.3	0.3	1.9	0.3	1.1	0.1	0.7	0.1	1.1		1.0	6330	19	6.6	1.8
6	0.6	0.04	0.6	0.1	0.2			0.03	0.1		0.5	7100	22	1.4	0.2
Naha no. 20: 2002/4/7-4/17, total flow rate = 403 m^3															
0	1.6	0.2	0.7		0.7		0.1	0.1					1.0	2.4	0.8
1	1.9	0.3	1.5	0.2	0.8	0.1	0.4	0.1	1.0			71	0.9	4.8	1.1
2	2.0	0.4	2.1	0.3	1.3	0.1	0.9	0.2	1.1	0.7	0.5	264	1.0	5.6	1.4
3	2.7	0.4	2.3	0.3	1.1	0.2	1.1	0.1	1.4	0.9	0.5	296	1.3	6.7	1.7
4	2.9	0.4	2.5	0.4	1.3	0.2	1.2	0.2	1.7		0.7	575	2.9	7.6	1.7
5	2.1	0.3	2.3	0.2	1.3	0.1	0.6	0.2	1.5	2.0	1.2	2390	12	7.2	2.0
6	0.7		0.6		0.3			0.1		4.2	1.2	4280	16	1.3	0.7
Naha no. 21: 2002/4/17-4/24, total flow rate = 299 m^3															
0															
1															
2	0.7	0.1	0.3	0.1		0.0	0.2				0.1	110	1.1	2.3	
3	3.6	0.5	2.3	0.5	1.0	0.2	1.2	0.2			0.7	140	4.8	9.7	1.4
4	0.4	0.1	0.3	0.04	0.1	0.01	0.02	0.01			0.1	36	1.9	1.5	
5															
6											0.6	689	21		
Naha no. 22: 2002/4/25-5/15, total flow rate = 816 m^3															
0											0.03	22	0.8		
1												55	1.5		
2											0.14	58	1.6		
3															
4											0.06	160	1.8	0.2	
5															
6											0.04	736	16		
Fukuoka no. 18: 2002/3/22-4/5, total flow rate = 575 m^3															
0	2.4	0.4	2.0	0.4	1.0	0.2	0.9	0.1	0.3		0.5	125	2.1	6.2	1.1
1	1.2	0.3	1.1	0.2	0.4	0.1	0.3	0.05	0.01		0.3	69	1.7	3.1	0.5
2	1.8	0.3	1.3	0.2	0.6	0.1	0.4	0.1	0.2		0.4	109	2.4	4.0	0.7
3	2.5	0.4	1.8	0.3	1.0	0.2	1.0	0.1	0.9		0.6	141	4.1	5.8	1.4
4	3.1	0.5	2.2	0.4	1.2	0.2	1.1	0.2	1.4		0.9	410	14	7.5	2.0
5	1.5	0.2	1.0	0.2	0.2	0.1	0.6	0.03			1.1	1490	50	3.6	0.5
6											1.1	3420	72	0.1	
Fukuoka no. 19: 2002/4/6-4/11, total flow rate = 215 m^3															
0	3.1	0.5	3.0	0.5	1.6	0.2	1.4	0.2	0.9		0.6	99	0.8	7.5	1.7
1	2.4	0.4	1.8	0.4	1.1	0.2	0.9	0.1	0.6		0.4	51	0.4	5.3	1.2
2	2.8	0.4	2.2	0.4	1.1	0.2	1.1	0.2	1.2		0.5	43	0.4	6.3	1.4
3	3.2	0.6	2.6	0.5	1.3	0.2	1.2	0.2	1.4		0.6	52	0.6	7.7	1.6
4	3.2	0.6	2.5	0.5	1.4	0.2	1.2	0.2	1.5		0.6	76	0.9	8.3	1.6
5	3.0	0.5	2.4	0.4	1.2	0.2	1.1	0.2	1.0		0.7	413	4.2	6.8	1.4
6	0.5	0.1	0.5	0.1	0.4	0.04	0.5	0.04			0.8	1270	8.2	1.4	0.6
Fukuoka no. 20: 2002/4/12-4/16, total flow rate = 165 m^3															
0	7	1.0	5.3	0.7	3.3	0.4	3.0	0.3			1.6	423	3.1	12	4.9
1															
2	6	0.9	4.6	0.8	2.7	0.4	2.6	0.3	1.3		1.2	233	2.3	12	3.5
3	3	0.5	2.3	0.4	1.5	0.2	1.2	0.2	0.6		0.6	146	1.4	7.0	1.8
4	7	1.2	5.6	1.0	3.1	0.5	3.1	0.4	2.4		1.6	591	7.0	18	5.2
5															
6	0.2	0.02	0.1		0.1	0.01	0.1	0.01			0.3	711	6.3	0.3	0.2
Fukuoka no. 20: 2002/4/17-4/24, total flow rate = 323 m^3															
0	2.6	0.5	1.9	0.3	1.0	0.1	0.9	0.1	0.04		0.4	107	0.6	5.0	1.4
1	1.7	0.3	1.4	0.3	1.0	0.1	0.8	0.1			0.4	92	0.5	3.8	1.1
2	2.8	0.4	2.2	0.4	1.1	0.2	1.2	0.2	0.7		0.5	550	0.9	6.0	1.7
3	2.4	0.4	2.1	0.3	1.1	0.1	0.9	0.1	0.9		0.6	123	1.1	5.9	1.8
4	4.1	0.6	3.2	0.5	1.9	0.2	1.8	0.3	1.9		0.8	267	2.9	9.1	2.6
5	1.0	0.1	0.8	0.1	0.6	0.1	0.5	0.05			0.4	354	3.8	2.3	0.9
6	0.2		0.1		0.3						0.7	1360	11		0.9

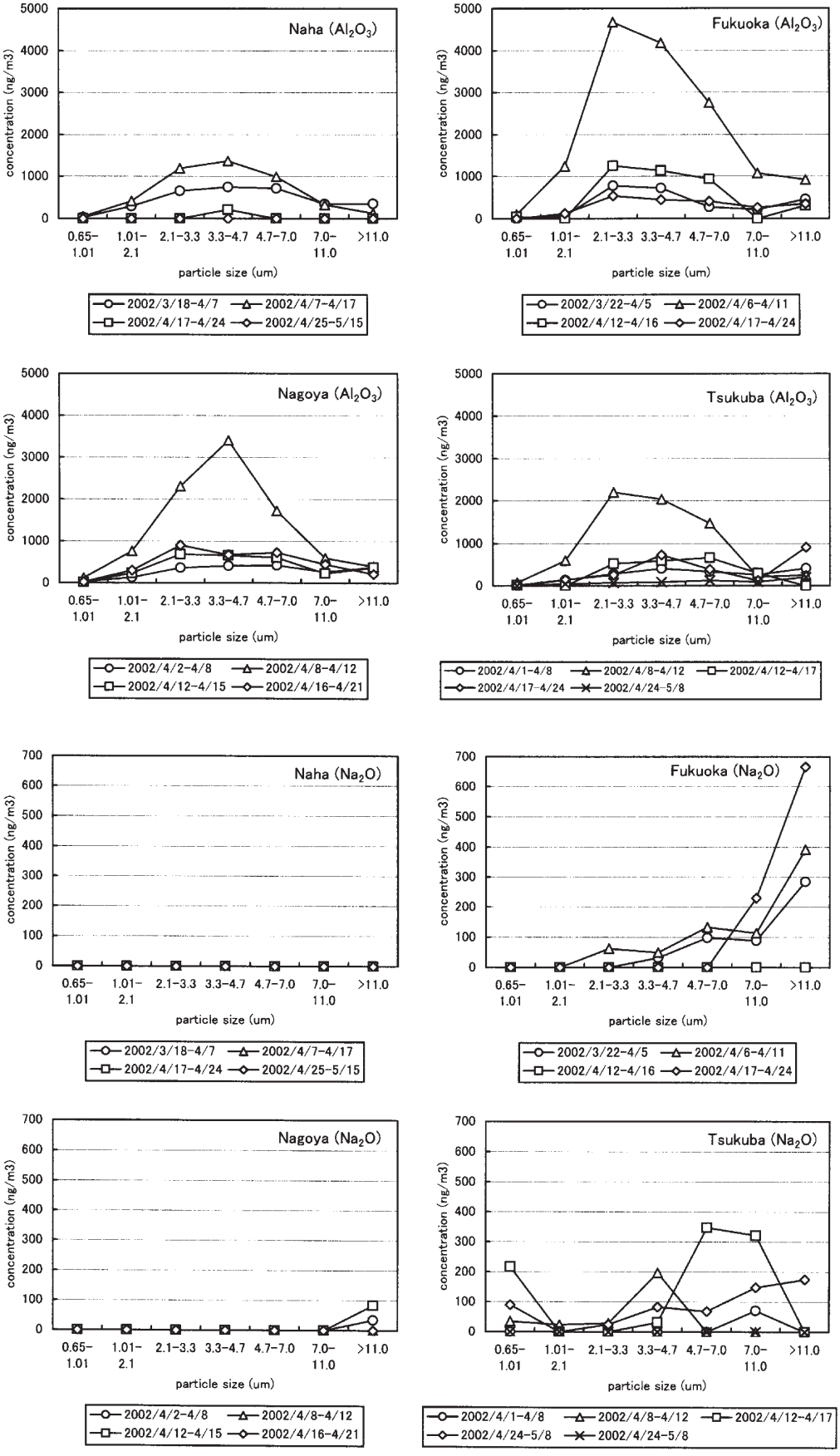

Fig. 3. The distribution of Na_2O, Al_2O_3, CaO, Mo, Sb and Pb concentrations in dust samples at four Japanese stations.

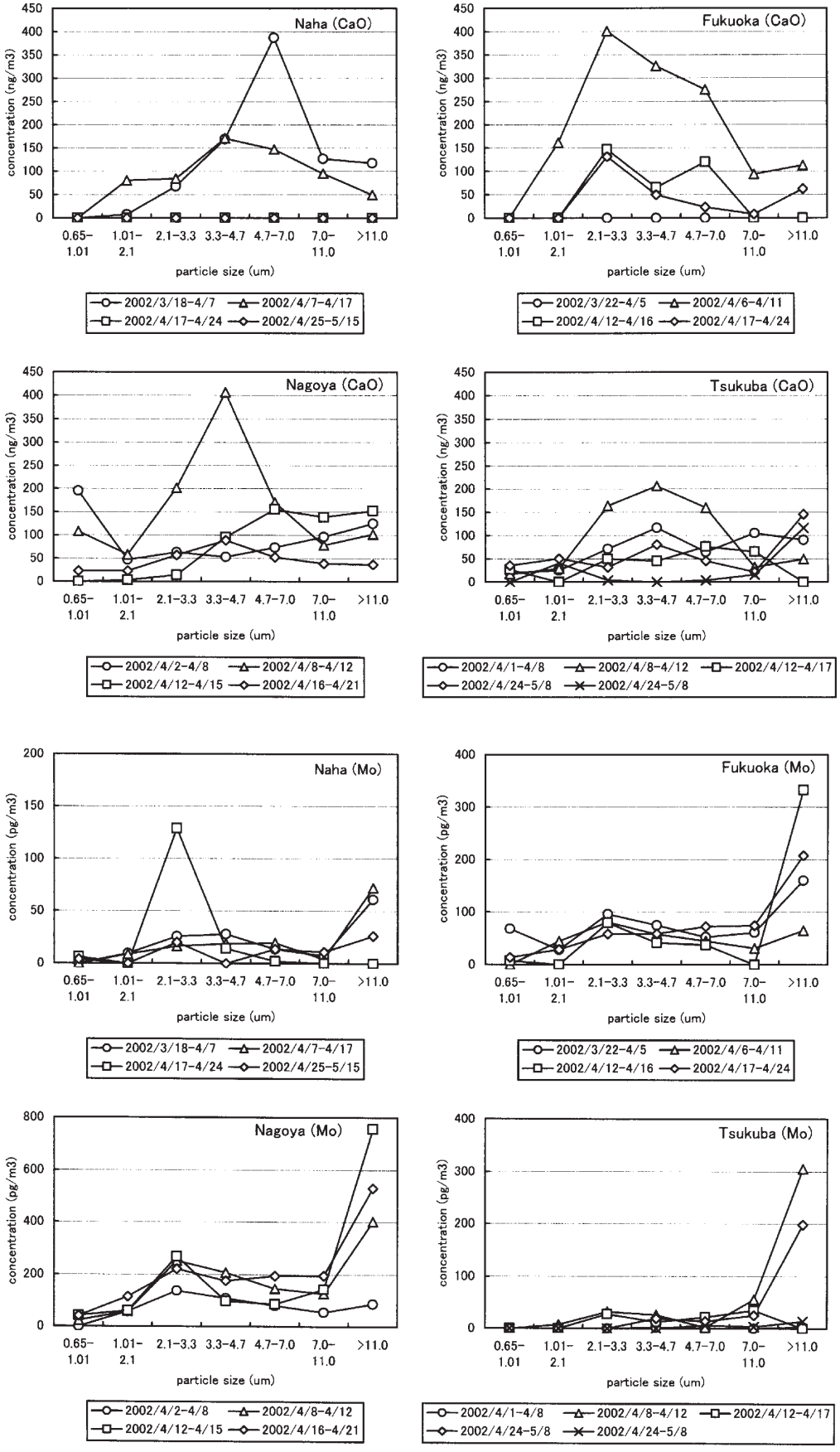

Fig. 3 Continued.

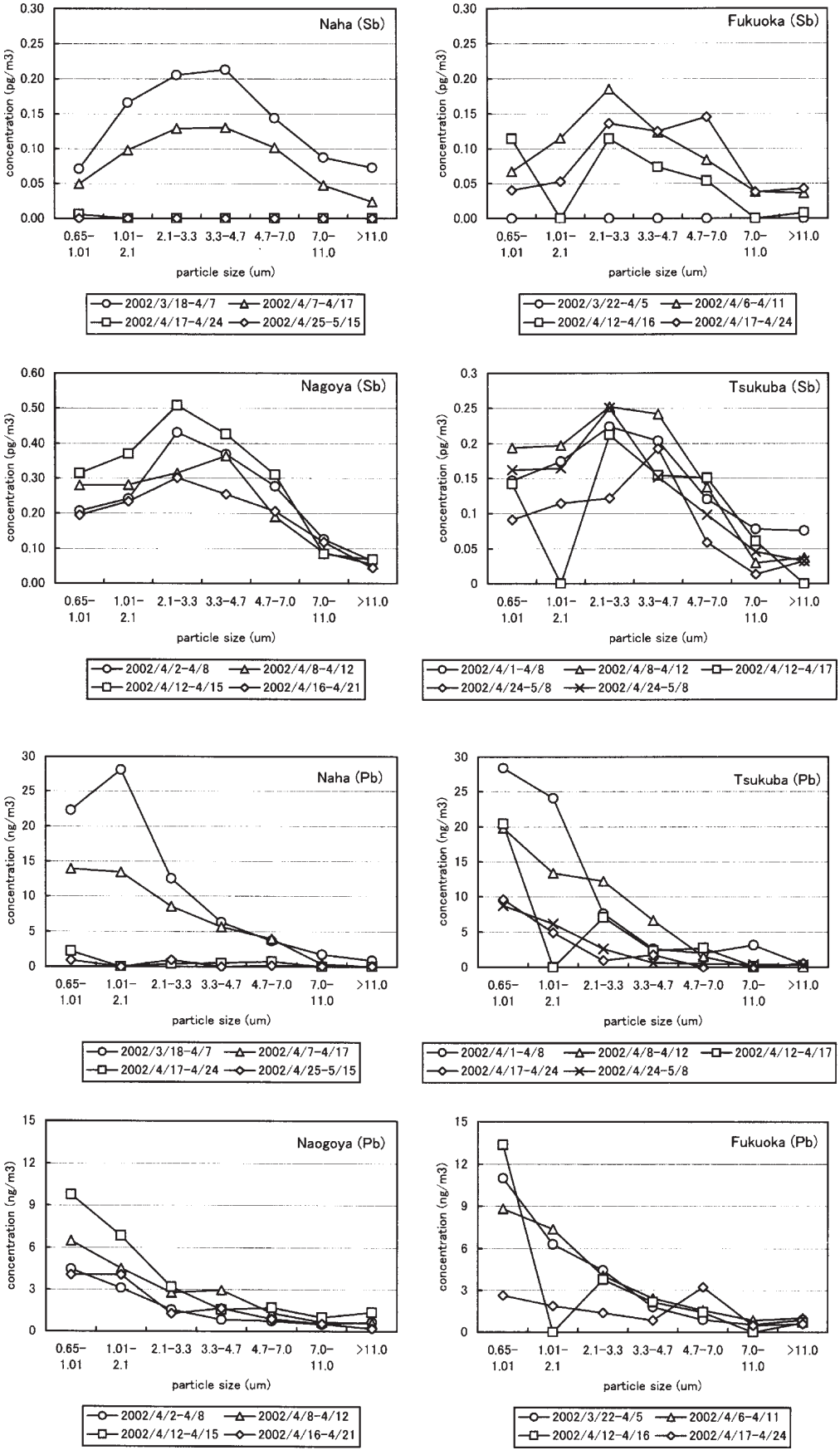

Fig. 3 Continued.

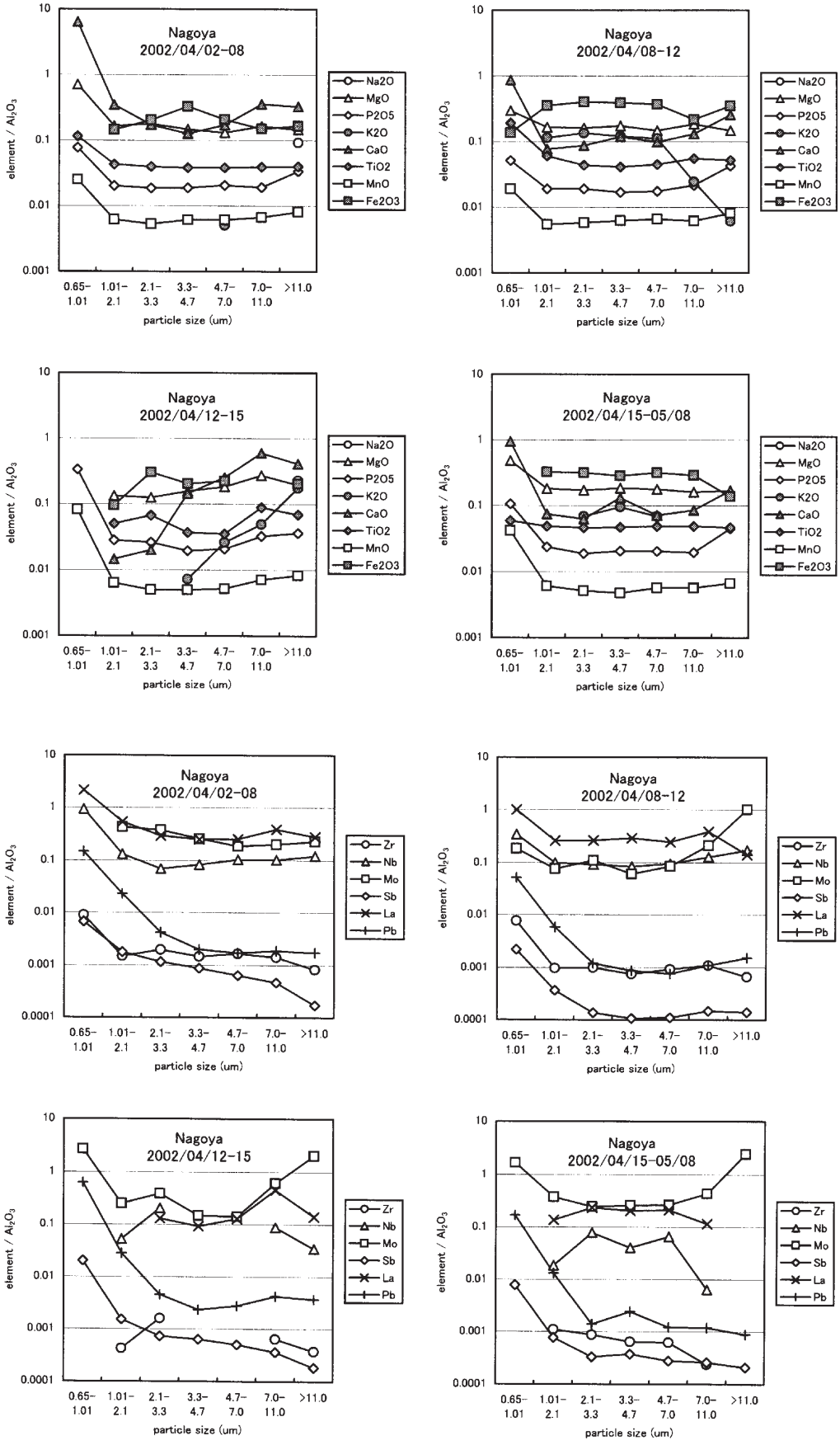

Fig. 4. The concentration ratios of major and some minor elements to Al_2O_3 at the Nagoya and Fukuoka stations.

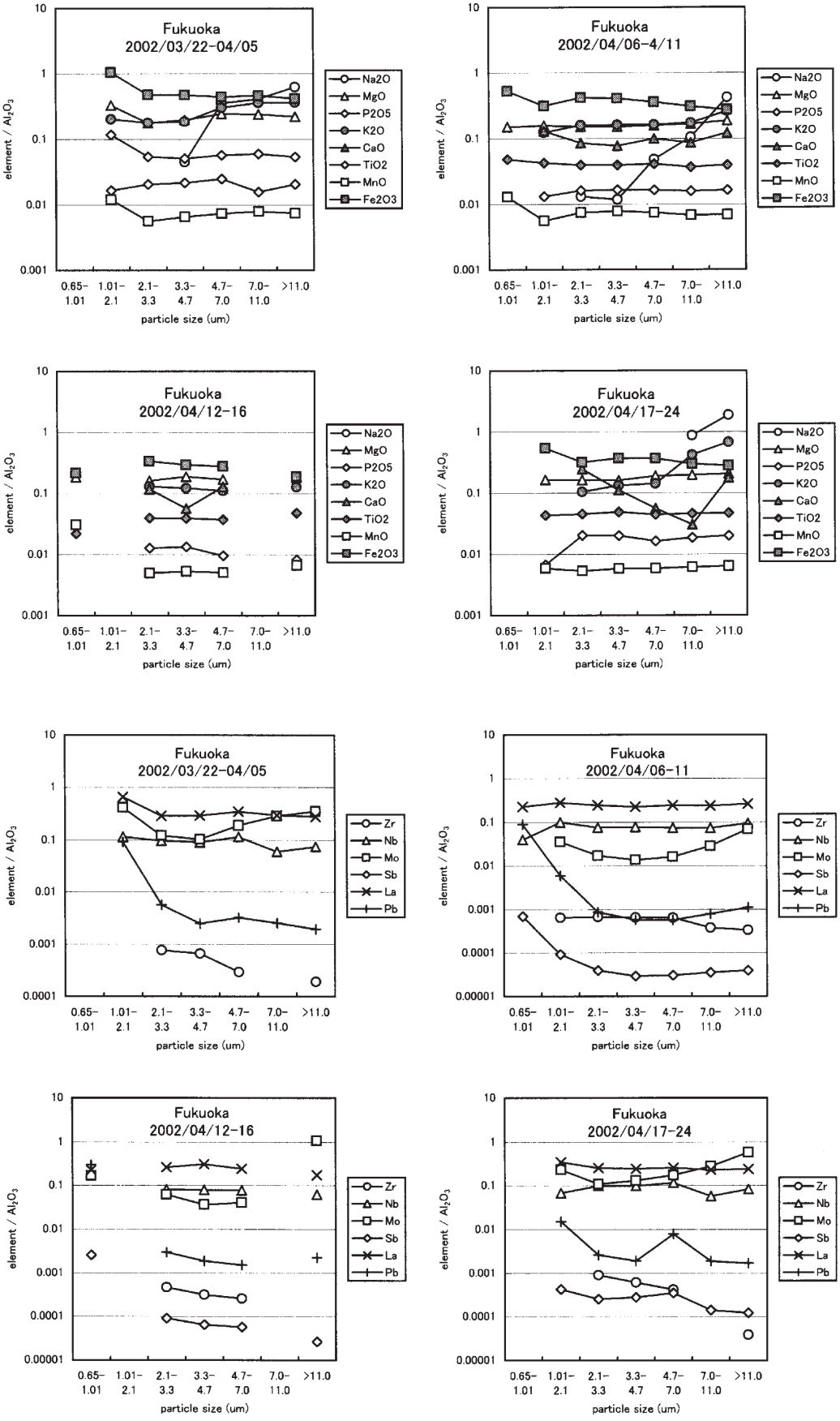

Fig. 4 Continued.

size increase with the decrease of the particle size during a non-dust event, but were constant during a dust event (Fig. 4). The small particles (0.65-2.1 μm) always have very high concentration ratios for these elements, which show that small particles contain much of the anthropogenic materials. The contribution of anthropogenic materials to aeolian dust is supposed to be slight in the range of 2.1-7.0 μm. However, the gentle increasing trend during a non-dust event indicates that the fraction of anthropogenic materials gradually increased with decreasing particle size. A large amount of mineral aerosol, whose Cd, Sn, Sb, Pb and Bi concentrations are low, probably hid the influence of anthropogenic materials on the large particles when a large-scale dust event was observed.

5.2 Geochemical differences between two periods (dust event and non-dust event) and among observation stations

When a dust event was observed, the dust samples with 2.1-7.0 μm of particle size are characterized by having extremely high dust concentrations and chemical concentrations (Figs. 2 and 3). The Al_2O_3 normalized values for most elements were almost constant whether or not a dust event was observed (Fig. 4). Dust samples with 2.1-7.0 μm of particle size have constant mineral compositions and a slight influence of anthropogenic materials. Consequently, the mean of Al_2O_3 normalized values of dust samples with 2.1-7.0 μm is most suitable to examine the change of chemical compositions between the two periods (dust event and non-dust event) and among observation stations.

Figure 5 shows that the mean of Al_2O_3 normalized values at four observation stations in spring 2002. The dispersion of the means was too large to find a significant difference between dust event and non-dust event. However, the means for P_2O_5, Mo, Sb and Pb seem to be relatively low during a dust event. Because Sb and Pb are enriched in anthropogenic materials and low in minerals, their low concentration ratios indicate the contribution of mineral aerosol transported from East Asia. Simultaneously the low concentration ratios of P_2O_5 and Mo to Al_2O_3 indicate that aeolian dust transported from Asia may be originally poor in these elements compared with Japan's surface materials.

When we focused on geochemical differences among observation stations, some elements have systematic differences. For example, samples at the Naha station were relatively rich in Pb and poor in Nb. Those at Nagoya station were enriched in Zr, Mo and Sb, but were poor in K_2O. Those at Tsukuba station were enriched in P_2O_5 and Pb. These differences may indicate the contamination of local surface materials to the collected dust samples. When a dust storm was observed, however, four observation stations had almost the same values to each other. Their constant values over the sampling locations indicate that a large amount of mineral aerosol transported from Asia is supplied to Japan.

6. Summary

We have studied chemical characteristics of aeolian dust (Kosa) during the transportation from China to Japan. We collected aeolian dust by air samplers at four Japanese stations (Naha, Fukuoka, Nagoya and Tsukuba) in spring 2002 and analyzed chemical compositions of water-insoluble components.

Every station observed the highest aerosol concentrations and aerosols were characterized by having one peak at 2.1-7.0 μm of the particle size during April 6-12 when a large-scale dust event was observed. The patterns of chemical concentrations of most elements in the air have similar trends to dust concentrations. The similarity among their distribution patterns shows that most elements originate in mineral aerosol. However, some elements, Cd, Sn, Sb and Pb, have high concentrations in small particles, which indicates that they have anthropogenic origins.

The Al_2O_3 normalized values of elemental concentrations are helpful to examine the change of mineralogical compositions and influence of non-silicate minerals and anthropogenic materials. The Al_2O_3 normalized values for most elements have constant ratios from >11 μm to 1.01 μm, but suddenly increased at 0.65-1.01 μm. The mineral composition is constant from >11 μm to 1.01 μm and change at 1 μm. However, the concentration ratios for Cd, Sn, Sb and Pb gradually increased with the decrease of particles size and suddenly rose at 2 μm. Their increasing trends correspond to the increase of the fraction of anthropogenic materials in aeolian dust.

Dust samples with 2.1-7.0 μm of particle size mainly consist of mineral aerosol and have little anthropogenic materials. The means of the Al_2O_3 normalized values in 2.1- 7.0 μm of particle size are useful to examine the contribution of local surface materials to the collected dust samples. Although the means of the Al_2O_3 normalized values varied widely, we found the systematic difference of P_2O_5, K_2O, T-Fe_2O_3, Zr, Nb and Mo among the sampling locations. However, when a dust event was observed, the means of Al_2O_3 normalized values became almost constant among the sampling locations. We recognized that a dust storm carried a large amount of mineral aerosol from East Asia to Japan.

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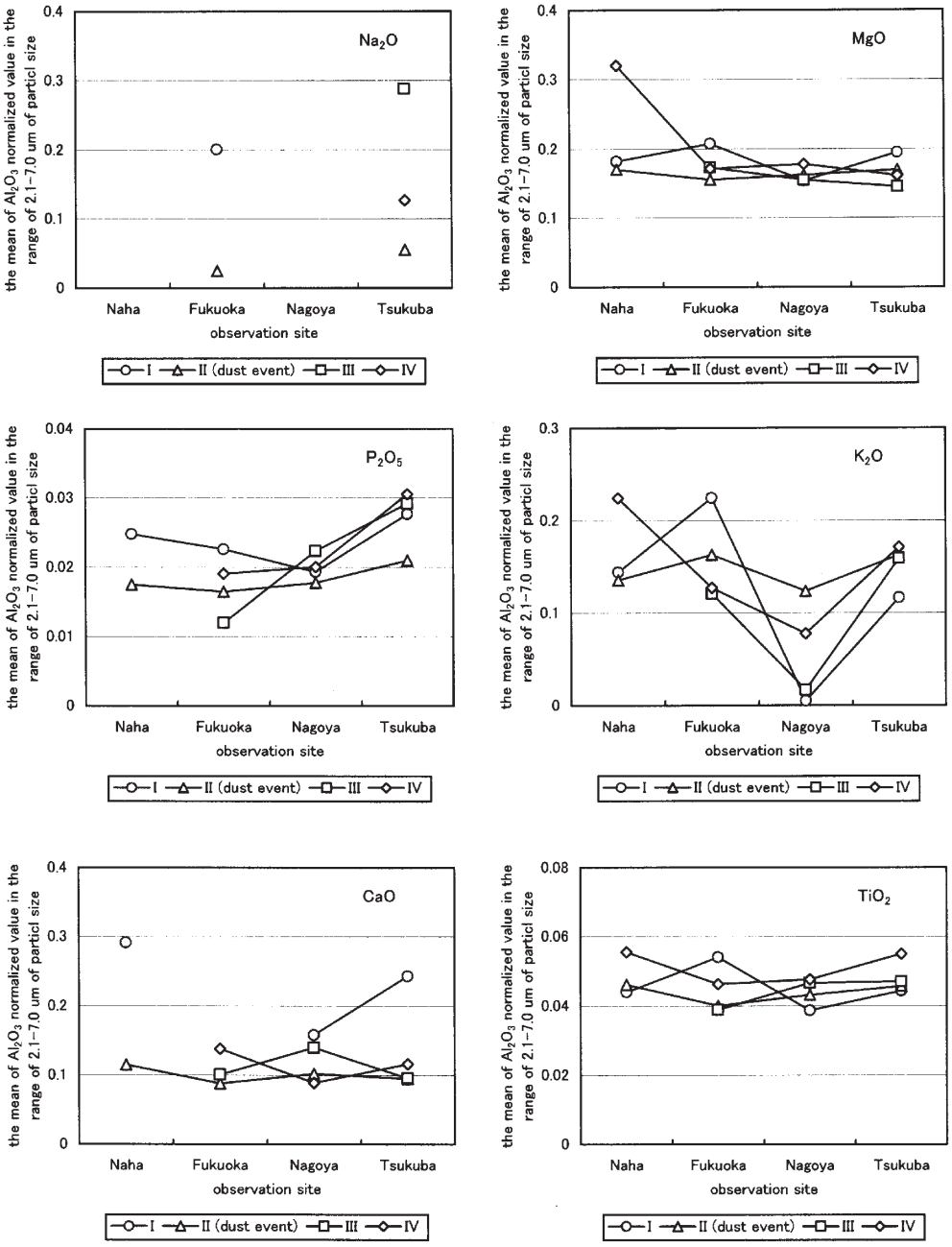

Fig. 5. The mean of concentration ratios of dust samples among 2.1-7.0 μm of particle size. I － IV indicates the sampling period. The I means the observation period before IOP 1: 3/18-4/7 for Naha, 3/22-4/5 for Fukuoka, 4/2-4/8 for Nagoya and 4/1-4/8 for Tsukuba. The II means the first half of IOP 1: 4/7-4/17 for Naha, 4/6-4/11 for Fukuoka, and 4/8-4/12 for Nagoya and Tsukuba. The III means the middle of IOP-1: 4/12-4/16 for Fukuoka, 4/12-4/15 for Nagoya and 4/12-4/17 for Tsukuba. The IV means the latter half of IOP 1: 4/17-4/24 for Naha, Fukuoka and Tsukuba, and 4/16-4/21 for Nagoya. A large-scale dust event was observed in the second observation period (II).

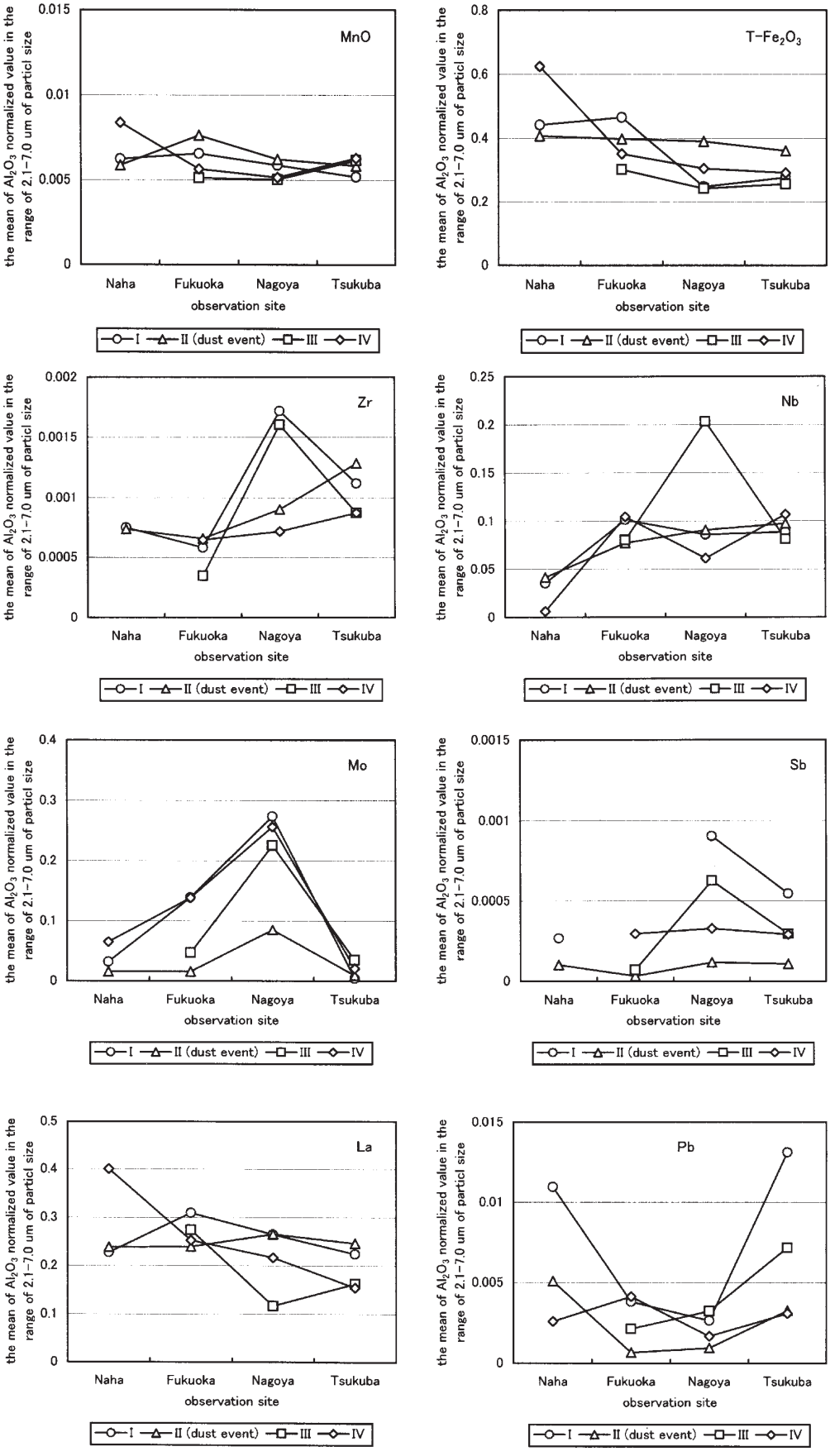

Fig. 5 Continued.

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日本で2002年春に採取した風送ダストについて その非水溶性成分の化学組成を粒径分布から見た特徴

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要　旨

2002年3月から5月にかけて、那覇、福岡、名古屋、つくばの4地点で風送ダスト試料を採取し、その非水溶性成分中の化学組成を測定した。ほとんどの元素は、その粒径分布の特徴として2.1～7.0 μmの粒径に一つのピークを示すことから、主に鉱物エアロゾルに含まれていると考えられる。しかし、Cd, Sn, Sb, Pb, Biなどの元素は1 μmよりも細かいダスト粒子に多く含まれ、人為起源の炭素エアロゾルに由来すると考えられた。

Al_2O_3濃度で各元素濃度を規格化した値の粒径分布の特徴から、風送ダスト中の鉱物組成は1 μmを境に変化し、2 μmより細かい粒子では人為起源物質の混入率が高くなることなどが明らかになった。次に、Al_2O_3規格値の空間的及び時系列的な変化に着目すると、一部の元素に系統的な地域差、すなわち観測点周辺からの物質の混入が認められた。しかし、一度ダストイベントが発生すると、ほぼすべての元素の濃度比は試料採取地点に関係なくほぼ一定の値を示し、非常に大量の風送ダストが東アジア地域から日本へ運ばれていることが明らかになった。