

Chemical characteristics of water-insoluble components in aeolian dust collected in China in spring 2002

Atsuyuki Ohta¹, Renjian Zhang², Shigeru Terashima¹, Yutaka Kanai³, Hikari Kamioka³, Noboru Imai¹, Yukihiro Matsuhisa¹, Hiroshi Shimizu⁴, Yoshio Takahashi⁴, Kenji Kai⁵, and Masahiko Hayashi⁶

Atsuyuki Ohta, Renjian Zhang, Shigeru Terashima, Yutaka Kanai, Hikari Kamioka, Noboru Imai, Yukihiro Matsuhisa, Hiroshi Shimizu, Yoshio Takahashi, Kenji Kai and Masahiko Hayashi (2005) Chemical characteristics of water-insoluble components in aeolian dust collected in China in spring 2002. *Bull. Geol. Surv. Japan*, vol. 56 (7/8), 259-272, 5 figs, 1 appendix.

Abstract: Aeolian dust collected at three stations in China (Beijing, Qingdao, and Hefei) in spring 2002 has been analyzed and their chemical features have been thoroughly discussed. The mass concentrations of aeolian dust collected were high in coarse grains, and the distribution patterns against particle size were different among the sampling stations. When large-scale dust events were observed, the concentrations of suspended particle with a particle size over 2 μm especially increased. The chemical compositions (Al_2O_3 , Na_2O , P_2O_5 , Total Fe_2O_3 , Rb, Zr, and La) of all but one aeolian dust sample were almost constant in coarse grains and quickly decreased below 1.1-2.1 μm . This result suggests that the contribution of mineral aerosol to aeolian dust sharply decreased in fine grains. In the dust event of March at Beijing, however, these elemental concentrations were almost constant over the variations of particle size. This fact indicates that the large-scale dust event supplied a large amount of mineral aerosol even in fine grains. The elemental concentration ratios to Al_2O_3 were almost constant in coarse-medium grains, but suddenly increased below 1-2 μm : the mineralogical composition was homogenous in coarse-middle grains, but changed in fine grains. The grain-size distribution pattern of the elemental concentration ratio has no systematic variations with or without a dust event or among three sampling stations. Therefore, chemical features of aeolian dust coming from inland China and suspended particle accumulated around sampling locations are very similar. On the contrary, some heavy elements (Cr, Ni, Cu, Zn, Mo, Cd, Sb, Sn, Pb, and Bi) had different features from elements that originated from mineral aerosol such as Al_2O_3 . The concentrations and metal/ Al_2O_3 ratios for these heavy metals increased with decreasing particle size. For example, the Cu/ Al_2O_3 and Pb/ Al_2O_3 ratios dramatically increased tenfold to hundredfold with decreasing particle size. These distribution patterns against the particle size suggest that anthropogenic materials were contaminated to finer grains.

Keywords: aeolian dust, water-insoluble components, grain-size distribution, mass concentration, chemical composition, Beijing, Hefei, Qingdao

1. Introduction

Atmospheric aerosols have serious effects to life, agriculture, and traffic, and heavily impact human health and the global climate. From the viewpoint of atmospheric pollution, the National Institute for Environmental Studies (2001) reported some observations on atmospheric aerosols in China. Zhang *et al.* (2002,

2003a, b) have studied the mass concentrations of total suspended particles and its chemical composition in Beijing for this purpose. These studies mainly targeted fine grains (below 2.5 μm), which have the most important effect on human health. Therefore, the investigation on the impact of aeolian dust on the climate is another important problem. The “Aeolian Dust Experiment on Climate Impact (ADEC)” project has

¹ Institute of Geology and Geoinformation, GSJ, AIST, Central 7 1-1-1 Higashi, Tsukuba, Ibaraki, 305-8567, Japan.

² Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, 100029, P. R. China.

³ Research Center for Deep Geological Environments, GSJ, AIST, Central 7 1-1-1 Higashi, Tsukuba, Ibaraki, 305-8567, Japan.

⁴ Hiroshima University, 1-3-1 Kagamiyama, Higashi-Hiroshima, 739-8526, Japan.

⁵ Nagoya University, Furo-cho, Chikusa-ku, Nagoya, 464-8601, Japan.

⁶ Fukuoka University, 8-19-1 Nanakuma, Jonan-ku, Fukuoka, 814-0180, Japan.

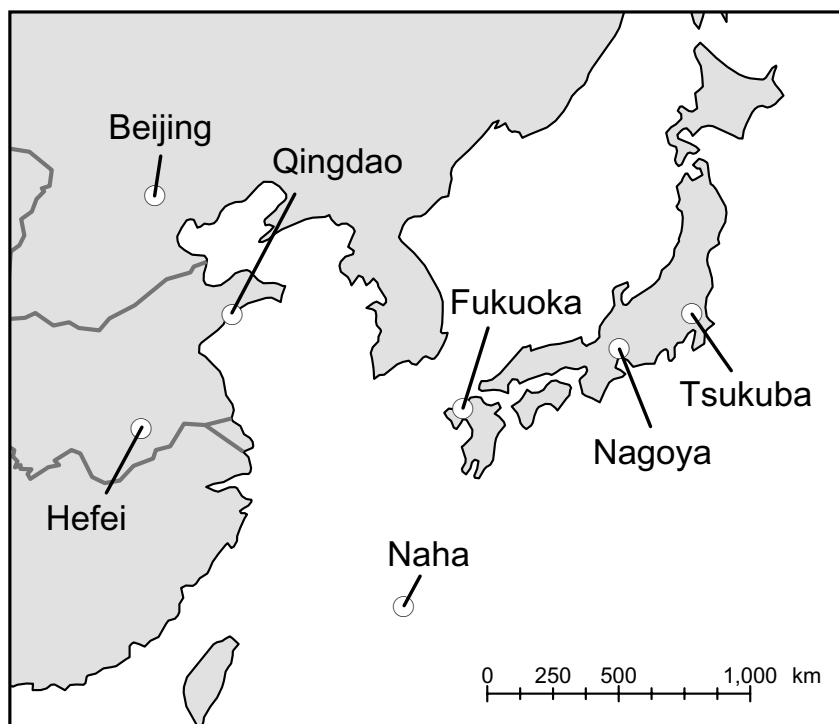


Fig. 1 Sampling locations in China and Japan for the ADEC project.

the above-mentioned goal and observed aeolian dust at several stations in China and Japan from April 2000 to March 2005. In ADEC project, the authors have tried to explain the physical and chemical characteristics and grain-size distribution of aeolian dust, which is transported from China to Japan. 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).

Our purpose is to explain the following three points; 1) the similarity and difference among sampling locations in Japan or China and influence of a dust event (Ohta *et al.*, 2003), 2) the chemical compositional change during the transportation of aeolian dust from China (Beijing) to Japan (Ohta *et al.*, 2004; 2005a), and 3) the seasonal variation of the chemical composition of aeolian dust at the Tsukuba station, Japan (Ohta *et al.*, 2005b). In this paper, the authors focus on the data at three Chinese stations in spring 2002, which have not been reported yet. Beijing and Qingdao were hit by aeolian dust coming from southern Mongolia, western Inner Mongolia, and inland China (Takla Makan Desert and Talim Basin), but the dust event was scarcely observed in Hefei. The samples collected at Hefei are useful to examine chemical and physical property of aeolian dust and those at Beijing and Qingdao are used to determine the influence of dust event.

2. Sampling locations and period

The Andersen-type low volume air sampler (AN-200, Shibata Co. Ltd.) was used at three Chinese stations shown in Fig. 1. The air sampler was fixed on the roof of a building to prevent as much as possible from collecting local surface material carried by the wind. The AN-200 obtains the grain size distribution data of aeolian dust: the particle size classification is $>11\ \mu\text{m}$, $11-7.0\ \mu\text{m}$, $7.0-4.7\ \mu\text{m}$, $4.7-3.3\ \mu\text{m}$, $3.3-2.1\ \mu\text{m}$, $2.1-1.1\ \mu\text{m}$, $1.1-0.65\ \mu\text{m}$, $0.65-0.43\ \mu\text{m}$, and $<0.43\ \mu\text{m}$. The quartz filter (Tokyo Dylec, 2500QAT-UP) was used to collect fine particles ($0.65-0.43\ \mu\text{m}$ and $<0.43\ \mu\text{m}$), and other particles were trapped by the PF-050 polyflon filter (Advantec Co. Ltd.). The AN-200 was operated for 7-20 days during the usual observation period and 1-8 days during the first intensive observation period (IOP) (IOP 1: 8-21 April 2002). According to Zhang *et al.* (2002) and Kanai *et al.* (2003), large-scale dust events were observed on March 19-22 and April 8-12, 2002 in China. Eight aeolian dust samples (four for Beijing and two for Hefei and Qingdao) were analyzed here. The three samples of Beijing no. 002 and no. 003 and Qingdao no. 022 were collected during a dust event. No sample was collected at Hefei in a dust event.

3. Analytical Methods

The water-soluble and water-insoluble components in dust samples were analyzed based on the reports of

Kanai *et al.* (2002) and Ohta *et al.* (2003). The soluble component of a quarter of the filter obtained by AN-200 was leached with a mixture of ethanol and Milli Q water (MQ). The insoluble fractions were separated by a cellulose acetate-type membrane filter. The composition of the eluent determined by an ion chromatograph will be discussed in another paper.

The insoluble fraction trapped on a quarter of the polyflon filter and membrane filter was decomposed with HNO₃, HClO₄, and HF. Fifty-one elements were determined by a Thermo Jarrell Ash IRIS ICP-AES (Na₂O, MgO, Al₂O₃, P₂O₅, K₂O, CaO, TiO₂, MnO, Total Fe₂O₃ (T-Fe₂O₃), V, Sr, and Ba) and an Agilent Technologies HP4500 ICP-MS (Li, Be, Sc, Cr, Co, Ni, Cu, Zn, Ga, Rb, Zr, Nb, Mo, Cd, Sn, Sb, Cs, REE (Y and lanthanide), Hf, Ta, Tl, Pb, Bi, Th, and U). The analytical results are shown in the Appendix. The analytical values in two fine fractions (0.65-0.45 μm and <0.45 μm) were semiquantitative because of the high blank values in the quartz filter (Ohta *et al.*, 2003, 2005b).

4. Results and Discussion

4.1 Aerosol concentrations at Beijing, Hefei, and Qingdao in spring 2002

Figure 2 shows the mass concentrations of aeolian dust samples at three Chinese stations in spring 2002, and the data are quoted from Kanai *et al.* (2003). When dust events were observed, total dust concentrations were extremely high at the Beijing and Qingdao stations, and the mass concentrations in coarse grains of over 2.1 μm markedly increased (see BJ002, BJ003, and QD022 in Fig. 2).

The mass concentrations of four dust samples collected at Beijing were all high in coarse grains, but their distribution patterns were not very similar to one another. When dust events were observed, the mass concentrations of BJ002 increased in coarse grains (over 2.1 μm), but those of BJ003 increased throughout all particle sizes. At the Qingdao and Hefei stations, the mass concentrations of aeolian dust were characterized by a broad peak from 7 μm to 2 μm. In the dust event, the concentrations of dust samples (QD022) with 7.0-2.1 μm particle size remarkably increased. Except for QD022, the mass concentrations of aeolian dust collected at Qingdao and Hefei had similar patterns against particle size to one another. The mass concentration of BJ002 in coarse grains (over 7 μm) was much higher than those of QD022, although they were collected in the dust event of March. Their difference will indicate that the coarse grains effectively fell out during transportation.

4.2 Analytical result of a water-insoluble component

The insoluble fraction of a coarse-medium grain of

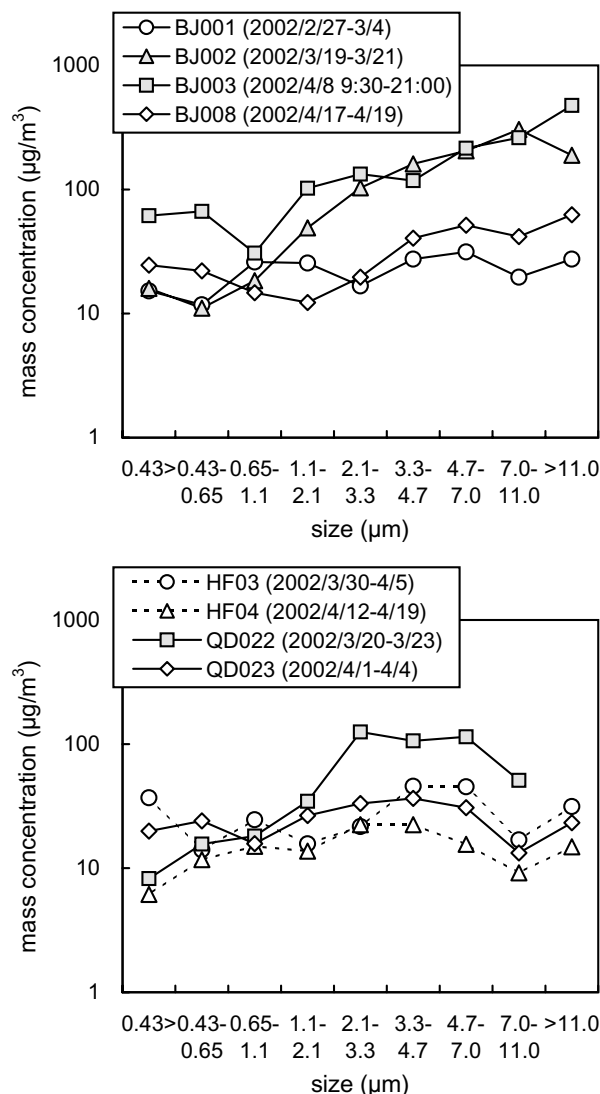


Fig. 2 Distribution of aerosol concentrations among particle size at the Beijing, Hefei, and Qingdao stations. The data are quoted from Kanai *et al.* (2003). The solid symbols indicate that the sample was collected in a dust event.

aeolian dust (>2-1 μm) consists mainly of mineral aerosol and that of a fine fraction (<2-1 μm) is composed mainly of carbon aerosol, which is released by a vehicle, plant and heating system and contains large amounts of anthropogenic materials. Next to Si, aluminum is the second most abundant element in minerals such as feldspar and clay minerals (illite and chlorite), and its concentration is used as a good indicator of the contribution of mineral aerosol (Inst. Hydro-spheric Sci. Nagoya Univ., 1991; Yabuki *et al.*, 2002).

Figure 3 shows the Al₂O₃ concentration of an insoluble fraction. The Al₂O₃ concentrations for Beijing samples were constant from >11 μm to 2.1 (or 3.3) μm, but they steeply decreased below 2.1 (or 3.3) μm. At the Qingdao and Hefei stations, the Al₂O₃ concen-

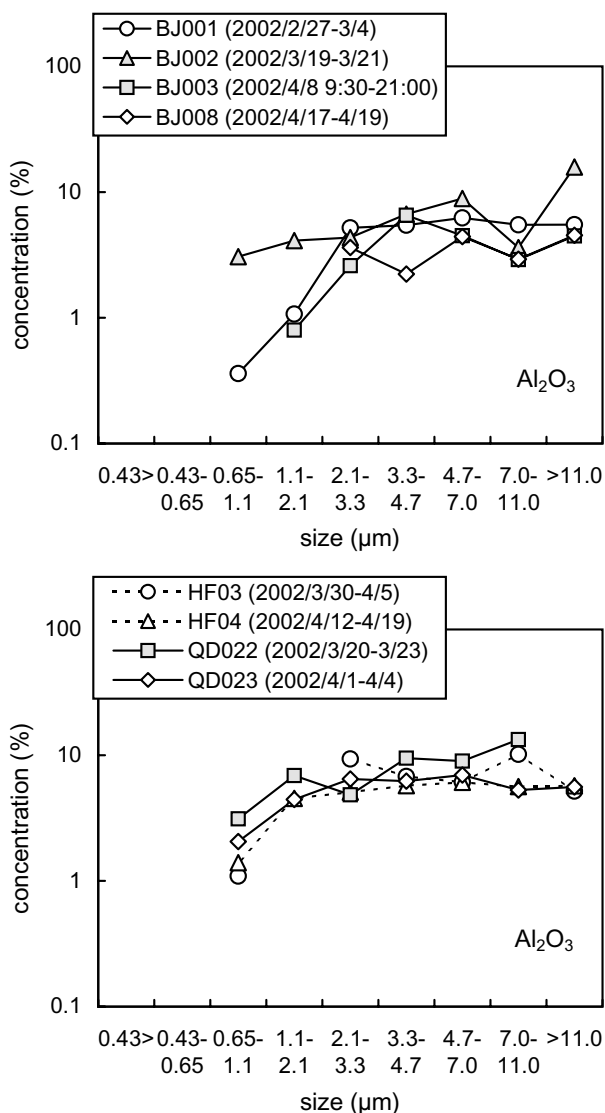


Fig. 3 The distribution of Al₂O₃ concentration in dust samples against particle size at three Chinese stations. The symbols are the same as those in Fig. 2.

trations for aeolian dust samples were constant from >11 μm to 1.1 μm and suddenly decreased below 1.1 μm. These results suggest that the contribution of mineral aerosol sharply decreased below 1.1-3.3 μm. Nevertheless, the BJ002, which was collected on March 19-22 in a large-scale dust event, is only sample without a sudden decrease of Al₂O₃ concentration. This fact indicates that the dust event of March provided a large amount of mineral aerosol even in the fine grains at Beijing. Although the QD022 was collected in the same dust event in March, it had a steep fall of Al₂O₃ concentration in the fine grains. It is assumed that the aeolian dust fell out during its transportation from Beijing to Qingdao so it made a small contribution to the local materials accumulated around Qingdao station. The BJ003 collected in another dust event of April

had comparable mass concentrations of aeolian dust to BJ002. However, the Al₂O₃ concentration of BJ003 was almost the same as those of BJ001 and BJ008. The high mass concentrations of medium-fine grains (below 2.1-3.3 μm) for BJ003 were probably caused not only by mineral aerosol conveyed from inland China in the dust event but also by local minerals, carbon aerosol, and soluble fractions ((NH₄)₂SO₄, NaNO₃, CaCO₃ and other fractions).

Figure 4 shows the variation of chemical compositions with grain size for eight elements (Na₂O, P₂O₅, T-Fe₂O₃, Cu, Rb, Zr, La, and Pb). The Na₂O, P₂O₅, T-Fe₂O₃, Rb, Zr, and La have almost the same distribution patterns as Al₂O₃. Accordingly, they are also originated from mineral aerosol. Some elements, Cr, Ni, Cu, Zn, Mo, Cd, Sn, Sb, Pb, and Bi, have partly or totally different distribution patterns from Al₂O₃. At the Beijing station, there was a dramatic rise of Cu and Pb concentrations below 3.3 μm in a normal period (BJ001 and BJ008) and below 2.1 μm in a dust event (BJ002 and BJ003). At Qingdao and Hefei stations, there were also sudden increases of Cu and Pb concentrations below 2.1 μm or 3.3 μm. The high concentrations for Cu and Pb in the finer grains suggest that these elements originated mainly from anthropogenic materials. However, the Cu and Pb concentrations below 2.1 μm in a dust event were relatively lower than those absent from a dust event both at the Beijing and Qingdao stations with some exceptions. This fact indicates that a large amount of mineral aerosol, whose Cd, Sn, Sb, Pb and Bi concentrations are low, diminished the influence of anthropogenic materials on large particles when a large-scale dust event was observed.

4.3 Elemental concentrations normalized by Al₂O₃ content

As mentioned above, the Al₂O₃ concentration is used as an indicator of the contribution of mineral aerosol. Therefore, dividing the elemental concentrations by the Al₂O₃ contents is useful for examining the change of mineralogical composition or estimating the influence of non-silicate minerals and anthropogenic materials (Inst. Hydrosphere Sci. Nagoya Univ., 1991). Figure 5 shows the chemical compositions normalized by the Al₂O₃ content. The P₂O₅, T-Fe₂O₃, Rb, Zr, and La had a flat or slightly increasing trend with the decrease of particle size, but the fine particles below 1-2 μm had a sudden increase of these ratios. The element concentration ratio showed no systematic differences among sampling location and between the dust events and non-dust event. Therefore, it is concluded that the mineral composition is constant from >11 μm to 1-2 μm, but changes under 1-2 μm; the mineralogical composition of aeolian dust coming from inland China is not much different from those of local materials.

The concentration ratios of Na₂O and CaO to Al₂O₃

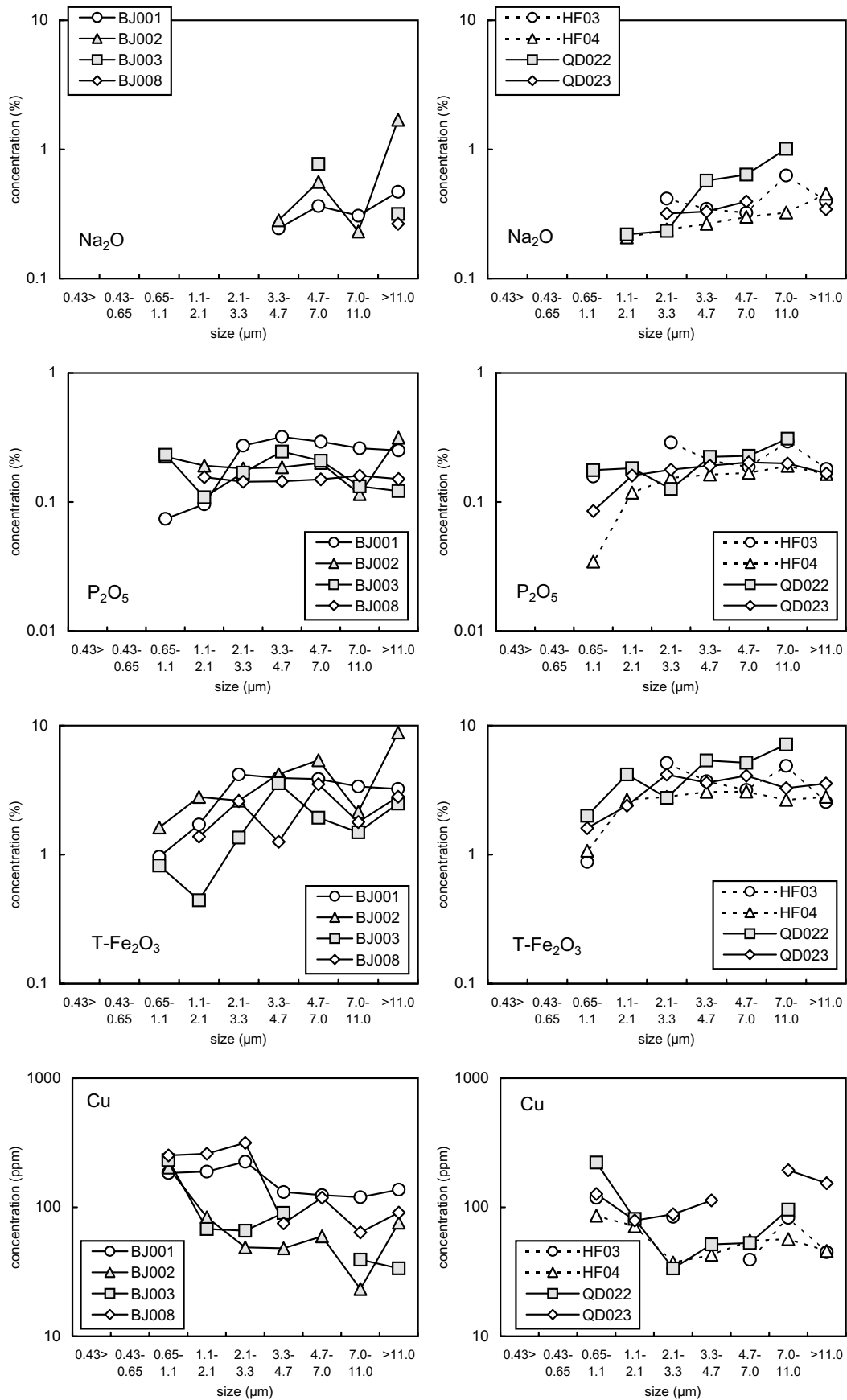


Fig. 4 The distribution of Na₂O, P₂O₅, T-Fe₂O₃, Cu, Rb, Zr, La and Pb concentrations in dust samples against particle size at three Chinese stations. The symbols are the same as those in Fig. 2.

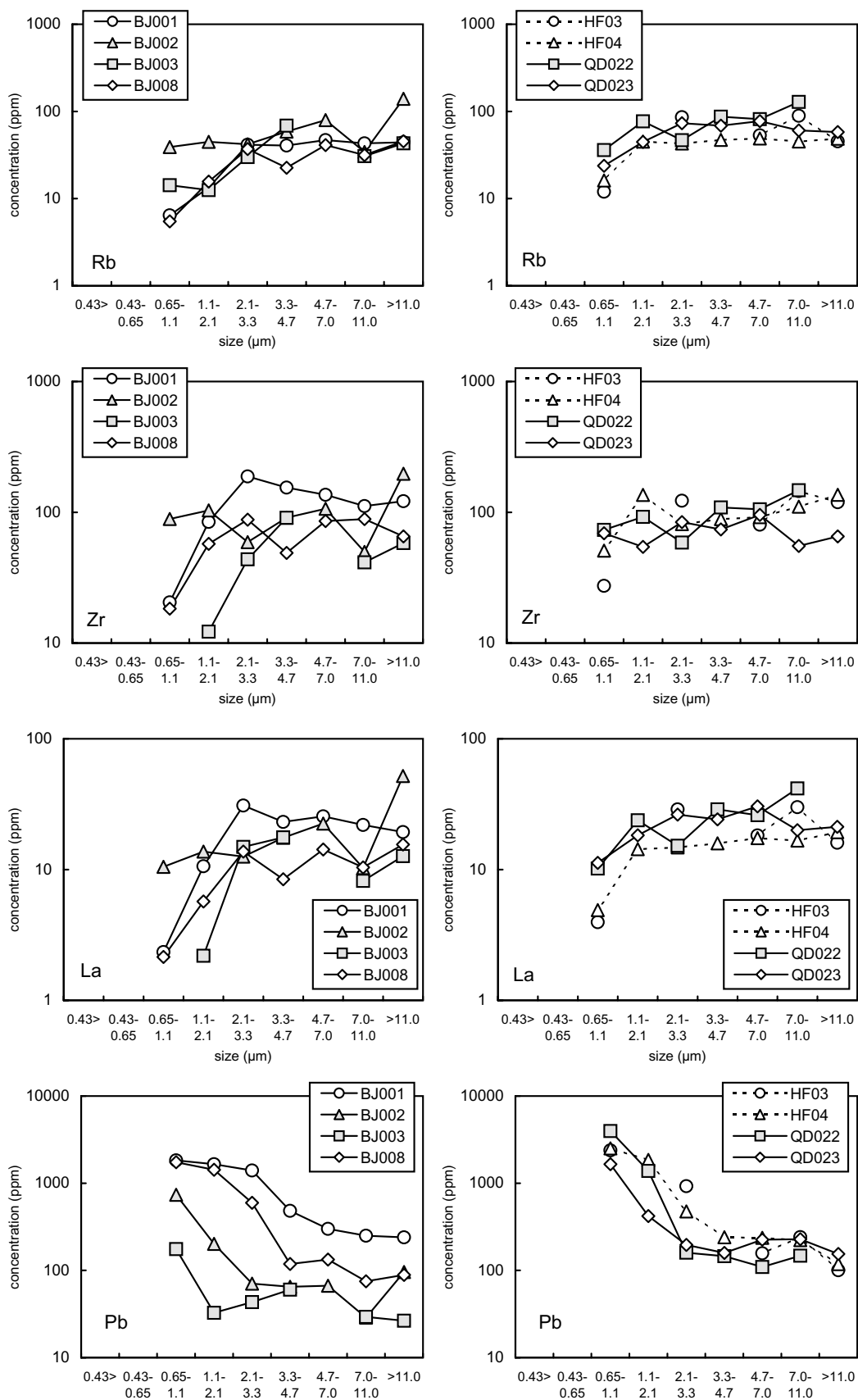


Fig. 4 (continue)

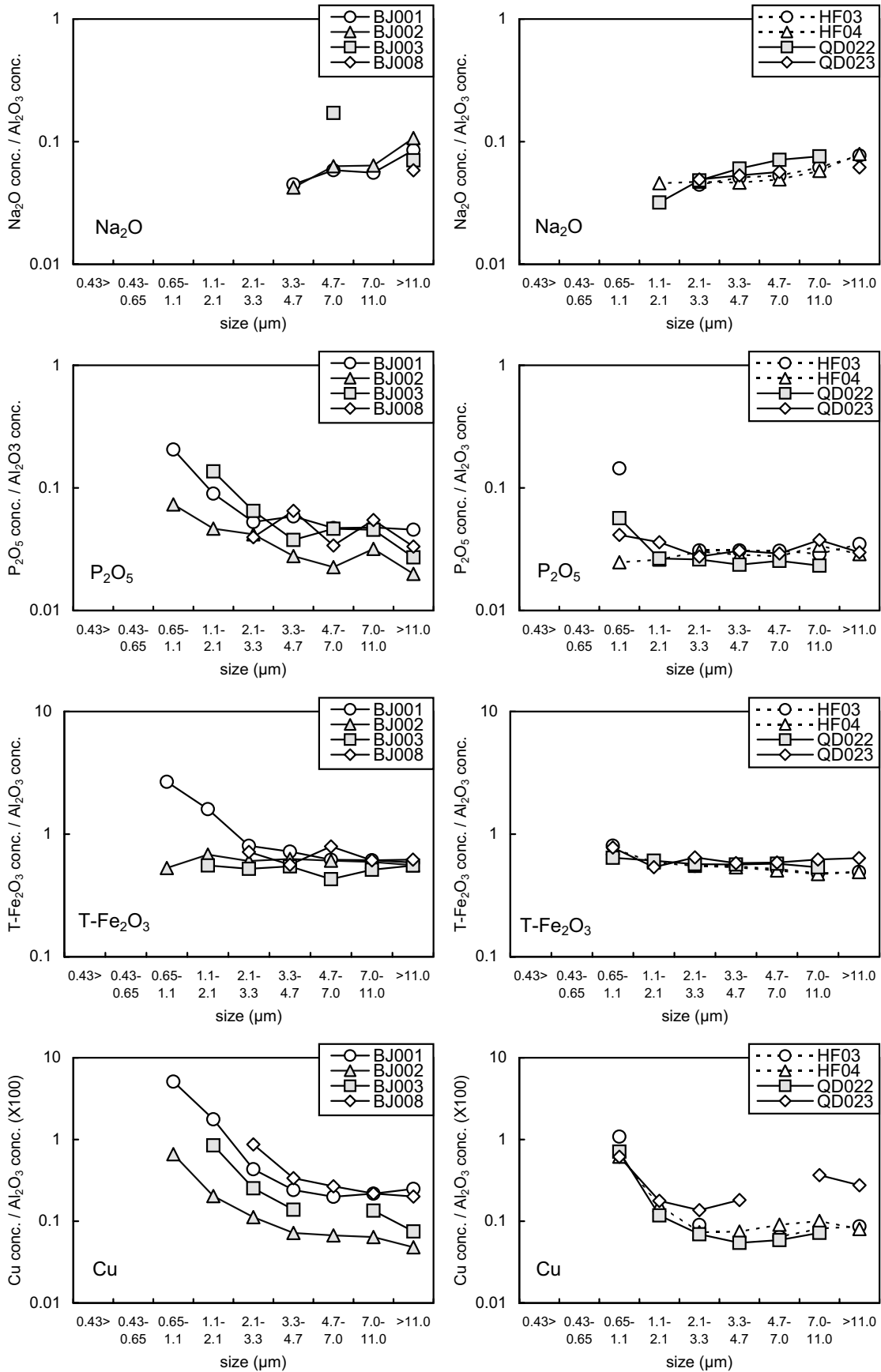


Fig. 5 The concentration ratios of major and some minor elements to Al_2O_3 . The symbols are the same as those in Fig. 2.

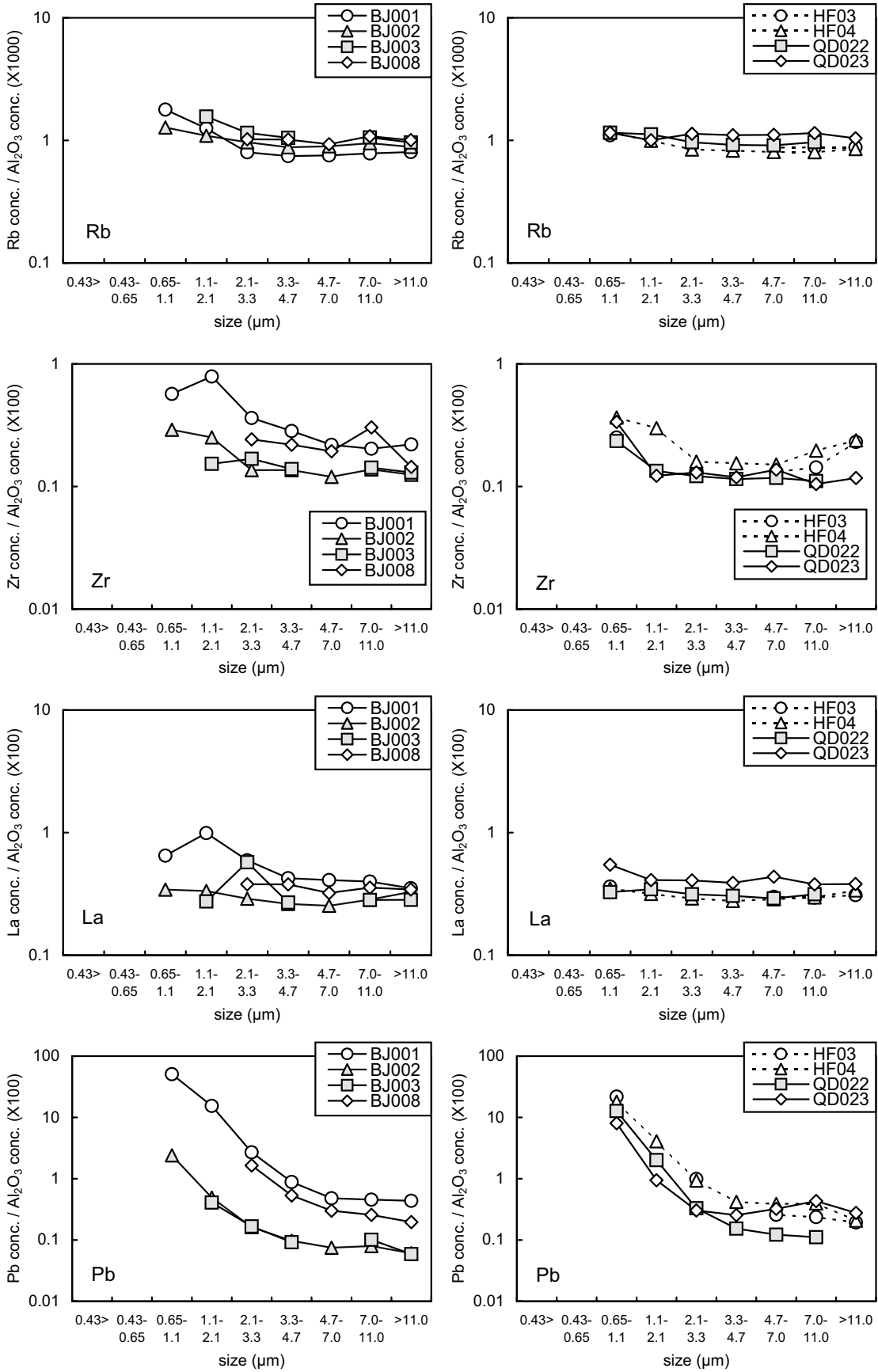


Fig. 5 (continue)

for all samples moderately decreased with the decrease of particle size (see Na₂O in Fig. 5). The decreasing trend suggests that the mineralogical composition may gradually change. Ohta *et al.* (2003) presumed that Na⁺, K⁺ and Ca²⁺ trapped in a clay mineral and non-silicate mineral (e.g. calcite, gypsum and sea salts) agglutinating with coarser grains were not dissolved enough in MQ.

The concentration ratios of Cr, Ni, Cu, Zn, Mo, Cd, Sn, Sb, Pb, and Bi to Al₂O₃ for dust samples with particle size over 3.3-2.1 μm were constant, but increased with the decrease of the particle size below 3.3-2.1 μm (see Cu and Pb in Fig. 5). The gentle increasing trend is an inherent nature of aeolian dust because a similar trend has also been found in the concentration ratios for other elements except for Na₂O and CaO. The concentration ratios of fine grains for most elements increased several fold than those of coarse grains, and those for heavy metals increased tenfold to hundred-fold with decreasing particle size. Therefore, the increasing trend for these elements indicates that the fraction of anthropogenic materials increased with decreasing particle size.

Interestingly, the elemental concentration ratios in Fig. 5 had basically the same trend against particle size irrespective of sampling stations or the occurrence of a dust storm. This result indicates that chemical features of aeolian dust are similar. In other words, it is difficult to specify the origin of aeolian dust by using its chemical composition. However, some elements had systematic differences. The Cu, Zr, La, and Pb to Al₂O₃ concentration ratios for samples collected at the Beijing station were smaller when a dust event was observed, although there were little systematic changes in the distribution pattern. The concentration ratios of Qingdao and Hefei had similar values to those of Beijing in a dust event, but were lower than those of Beijing in a normal period. However, there was no systematic change of concentration ratios at the Qingdao station between a dust event and non-dust event. Accordingly, local materials in Beijing have a relatively high concentration for these elements.

5. Conclusion

The authors have examined the chemical differences of aeolian dust among sampling locations (Beijing, Qingdao, and Hefei) and influence of aeolian dust coming from inland China on local suspended materials. The chemical compositions of aeolian dust conveyed from inland China are not much different from those of local materials, except for some heavy metals such as Cu and Pb, which are released by an anthropogenic activity. The elements/Al₂O₃ values, which exhibit a change of mineralogical composition or contribution of non-silicate minerals and anthropogenic

materials, did not systematically change among sampling locations or between a dust event and non-dust event. In conclusion, the chemical properties of aeolian dust are very constant.

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中国で2002年春に採取した風送ダストの非水溶性成分の化学的特徴

太田充恒¹・張 仁健²・寺島 滋¹・金井 豊³・上网 晃³・今井 登¹・
松久幸敬¹・清水 洋⁴・高橋嘉夫⁴・甲斐憲次⁵・林 政彦⁶

要 旨

2002年春に中国の北京、青島、合肥の3地点で風送ダスト試料を採取し、化学分析を行い、その特徴について検討を行った。大気中ダストは粗い粒径側で濃度が高く、粒径に対する濃度変化は試料採取地点によって異なっていた。ダストイベントが発生した時は、 $2\mu\text{m}$ 以上の粒子に著しい増加が認められた。一例外を除く全ての試料の化学組成 (Al_2O_3 , Na_2O , P_2O_5 , Total Fe_2O_3 , Rb, Zr, and La) は、粗大粒子側ではほぼ濃度が一定であるが $1.1\sim 2.1\mu\text{m}$ よりも細かい粒子において急激に減少した。この結果は、鉱物質エアロゾルの寄与が細粒粒子で著しく減少することを表している。ただし、3月に北京で観測されたダストイベントでは、これらの元素は粒径変化に関係なくほぼ一定の濃度を示した。この結果は、大規模なダストイベントは細粒粒子においても大量の鉱物質エアロゾルを供給していることを表している。元素濃度と Al_2O_3 濃度の比を見たところ、粗粒-中粒にかけてほぼ一定の値を示すものの $1\sim 2\mu\text{m}$ を境に急激に減少する。すなわち、鉱物組成は粗粒-中粒ダストではほぼ均質であるが細粒粒子側で変化することを示している。元素濃度比の粒径に対する変化に着目すると、ダストイベントの有無や3試料採取地点間に系統的な違いは認められなかった。したがって、中国内陸部から運ばれる風送ダストと試料採取地点周辺から巻き上げられた物質の化学組成には共通点が多いことを示している。一方、いくつかの重金属元素 (Cr, Ni, Cu, Zn, Mo, Cd, Sb, Sn, Pb, and Bi) は、鉱物質エアロゾル起源の元素 (Al_2O_3 など) とは異なる特徴を示した。これらの元素の濃度及び Al_2O_3 濃度比は粒径が細くなるに従って著しく増加する。例えば、 $\text{Cu}/\text{Al}_2\text{O}_3$ 比と $\text{Pb}/\text{Al}_2\text{O}_3$ 比は粒径が細くなるに従って、 $10\sim 100$ 倍も劇的な増加を示した。これらの粒径に対する変化は細かい粒子ほど人為起源物質の寄与が多いことを示している。

Appendix. Analytical results of aeolian dust collected in Beijing, Hefei and Qingdao from February to spring in 2002.

Stage	size (um) (μm)	weight (mg)	Na ₂ O (%)	MgO (%)	Al ₂ O ₃ (%)	P ₂ O ₅ (%)	K ₂ O (%)	CaO (%)	TiO ₂ (%)	MnO (%)	T-Fe ₂ O ₃ (%)	Li ppm	Be ppm	Sc ppm
Beijing no.001: 2002/2/27-3/4, total flow = 204 m³														
0	>11.0	5.6	0.5	1.4	5.5	0.3	0.9	2.46	0.3	0.04	3.2	32	3	9
1	7.0-11.0	4.0	0.3	1.5	5.5	0.3	0.8	1.88	0.2	0.05	3.4	34	4	8
2	4.7-7.0	6.4	0.4	1.5	6.2	0.3	0.9	2.60	0.3	0.05	3.9	42	5	10
3	3.3-4.7	5.6	0.2	1.1	5.5	0.3	0.8	1.61	0.3	0.04	3.9	36	5	8
4	2.1-3.3	3.4		0.8	5.2	0.3	0.6		0.3	0.04	4.2	37	5	10
5	1.1-2.1	5.2			1.1	0.1			0.07	0.03	1.7	11	2	4
6	0.65-1.1	5.3			0.4	0.07			0.02	0.03	1.0	5	1	0.6
7	0.43-0.65	2.4								0.03				
BU	0.43>	3.1								0.02				
Beijing no.002: 2002/3/19-3/21, total flow = 81.5 m³														
0	>11.0	15.4	1.7	3.1	15.8	0.3	3.1	4.26	1.0	0.14	8.8	67	4	21
1	7.0-11.0	24.8	0.2	0.8	3.6	0.1	0.7	0.85	0.2	0.03	2.1	16	1	5
2	4.7-7.0	16.7	0.6	1.6	8.9	0.2	1.7	1.80	0.5	0.09	5.4	44	3	12
3	3.3-4.7	13.1	0.3	1.3	6.7	0.2	1.3	1.24	0.4	0.06	4.2	32	2	9
4	2.1-3.3	8.4		1.0	4.3	0.2	0.7	0.65	0.2	0.04	2.6	21	1	6
5	1.1-2.1	4.0		0.9	4.1	0.2	0.7		0.2	0.04	2.8	23	1	10
6	0.65-1.1	1.5			3.1	0.2			0.10	0.03	1.6	18	1	10
7	0.43-0.65	0.9												
BU	0.43>	1.3												
Beijing no.003: 2002/4/8 9:30 - 21:00, total flow = 1.95 m³														
0	>11.0	9.3	0.3	0.9	4.5	0.1	0.7	0.84	0.3	0.04	2.5	19	1	6
1	7.0-11.0	5.1		0.5	2.9	0.1	0.4		0.14	0.02	1.5	15	0.7	4
2	4.7-7.0	4.2		1.1	4.5	0.2	0.8	1.60	0.2	0.04	1.9			6
3	3.3-4.7	2.3		1.1	6.5	0.2	1.0		0.3	0.06	3.6	32	2	9
4	2.1-3.3	2.6			2.6	0.2	0.4		0.10	0.02	1.4	15	0.7	4
5	1.1-2.1	2.0			0.8	0.1					0.4	6	0.3	1
6	0.65-1.1	0.6									0.8		0.3	
7	0.43-0.65													
BU	0.43>					0.1								
Beijing no.008: 2002/4/17-4/19, total flow = 81.5 m³														
0	>11.0	5.1	0.3	1.3	4.5	0.2	0.8	3.43	0.3	0.05	2.8	26	2	5
1	7.0-11.0	3.4		0.8	2.9	0.2	0.4	1.63	0.14	0.03	1.8	17	1	7
2	4.7-7.0	4.2		1.2	4.4	0.1	0.7	2.47	0.2	0.05	3.5	28	2	7
3	3.3-4.7	3.3		0.5	2.2	0.1	0.3	1.05	0.11	0.02	1.3	16	1	3
4	2.1-3.3	1.6			3.6	0.1			0.12	0.04	2.6	26	2	6
5	1.1-2.1	1.0				0.2				0.04	1.4	9	0.5	2
6	0.65-1.1	1.2								0.04			0.2	
7	0.43-0.65									0.03				
BU	0.43>									0.01				
Hefei no.03: 2002/3/30- 4/5, total flow = 214 m³														
0	>11.0	6.7	0.4	0.9	5.2	0.2	0.8	2.57	0.3	0.03	2.5	26	1	9
1	7.0-11.0	3.6	0.6	1.9	10.2	0.3	1.6	4.10	0.5	0.07	4.9	47	2	11
2	4.7-7.0	9.7	0.3	1.1	6.1	0.2	1.1	2.48	0.3	0.04	3.2	28	1	7
3	3.3-4.7	9.8	0.3	1.3	6.8	0.2	1.3	2.31	0.4	0.05	3.7			10
4	2.1-3.3	4.6	0.4	1.8	9.3	0.3	1.6	1.85	0.5	0.06	5.1	39	2	10
5	1.1-2.1	3.4												
6	0.65-1.1	5.3		0.1	1.1	0.2	0.2		0.06	0.02	0.9	5	0.3	0.7
7	0.43-0.65	3.0			1.0	0.1			0.04	0.02	0.6			
BU	0.43>			0.3	0.04				0.02	0.01	0.1			
Hefei no.04: 2002/4/12- 4/19, total flow = 264 m³														
0	>11.0	3.9	0.5	0.9	5.7	0.2	0.9	2.16	0.3	0.04	2.8	24	1	6
1	7.0-11.0	2.4	0.3	0.9	5.6	0.2	0.8	1.23	0.3	0.03	2.7	24	1	5
2	4.7-7.0	4.1	0.3	1.0	6.1	0.2	1.0	1.46	0.3	0.04	3.1	27	1	6
3	3.3-4.7	5.9	0.3	0.9	5.7	0.2	1.0	0.76	0.3	0.03	3.1	25	1	6
4	2.1-3.3	5.9	0.2	0.8	5.1	0.2	0.8	0.46	0.3	0.03	2.8	22	1	6
5	1.1-2.1	3.6	0.2	0.7	4.5	0.1	0.8		0.2	0.03	2.7	20	1	5
6	0.65-1.1	4.0		0.2	1.4	0.03	0.2		0.06	0.02	1.1	7	0.4	2
7	0.43-0.65	3.1			1.0				0.04	0.03	0.6	7		
BU	0.43>	1.6			1.6				0.06	0.03	0.6			
Qingdao no.022: 2002/3/20-3/23, total flow = 121 m³														
0	>11.0	0.0												
1	7.0-11.0	6.2	1.0	3.7	13.3	0.3	2.7	1.91	0.7	0.10	7.1	61	3	16
2	4.7-7.0	13.9	0.6	2.2	9.0	0.2	1.9	1.68	0.5	0.07	5.2	43	2	11
3	3.3-4.7	12.9	0.6	2.6	9.5	0.2	2.0	1.28	0.5	0.08	5.4	50	2	13
4	2.1-3.3	15.2	0.2	1.5	4.8	0.1	0.9	0.49	0.3	0.04	2.8	24	1	7
5	1.1-2.1	4.2	0.2	2.1	6.9	0.2	1.3		0.3	0.05	4.2	36	2	10
6	0.65-1.1	2.2		0.5	3.1	0.2	0.5		0.14	0.05	2.0	17	1	3
7	0.43-0.65	1.9												
BU	0.43>	1.0												
Qingdao no.023: 2002/4/1-4/4, total flow = 121 m³														
0	>11.0	2.8	0.3	1.0	5.6	0.2	1.0		0.2	0.04	3.6	26	1	5
1	7.0-11.0	1.6		0.9	5.3	0.2	0.9		0.2	0.04	3.3	27	2	5
2	4.7-7.0	3.7	0.4	1.5	7.0	0.2	1.3		0.3	0.06	4.1	35	2	8
3	3.3-4.7	4.4	0.3	1.4	6.2	0.2	1.2	0.79	0.3	0.05	3.6	32	2	8
4	2.1-3.3	4.0	0.3	1.4	6.5	0.2	1.1	0.83	0.3	0.05	4.2	34	2	8
5	1.1-2.1	3.2		0.8	4.4	0.2	0.7		0.2	0.02	2.4	21	1	5
6	0.65-1.1	1.9			2.1	0.1			0.13	0.03	1.6	12	0.7	1
7	0.43-0.65				0.9	0.06			0.06	0.03	0.8	6	0.4	
BU	0.43>								0.04	0.04	1.5	7	0.7	

Italic type indicates that the value is semiquantitative.

Appendix. (continue)

Stage	size (um) (μm)	weight (mg)	V ppm	Cr ppm	Co ppm	Ni ppm	Cu ppm	Zn ppm	Ga ppm	Rb ppm	Sr ppm	Y ppm	Zr ppm	Nb ppm	Mo ppm
Beijing no.001: 2002/2/27-3/4, total flow = 204 m³															
0	>11.0	5.6	240	59	14		140	408	14	44	240	15	120	11	8
1	7.0-11.0	4.0	360		15		120	396	17	43	260	17	110	9	
2	4.7-7.0	6.4	230	59	20	33	120	854	22	47	330	20	140	11	
3	3.3-4.7	5.6	280		19	13	130	1130	26	41	290	20	150	10	
4	2.1-3.3	3.4	850		20	16	230	571	47	42	250	25	190	11	23
5	1.1-2.1	5.2	250		8		190	571	28	13	49	8	84	5	21
6	0.65-1.1	5.3	420		3		180	527	33	6	20	2	20	2	
7	0.43-0.65	2.4													
BU	0.43>	3.1													
Beijing no.002: 2002/3/19-3/21, total flow = 81.5 m³															
0	>11.0	15.4	120	100	25	69	76	180	29	140	250	31	200	22	8
1	7.0-11.0	24.8	37	24	6	17	23	46	7	34	53	8	50	6	1
2	4.7-7.0	16.7	77	130	17	110	60	130	17	80	120	16	110	11	2
3	3.3-4.7	13.1	58	43	12	33	48	100	14	59	83	13	91	10	
4	2.1-3.3	8.4	67		7	21	49	130	9	42	53	10	59	6	
5	1.1-2.1	4.0	58		15		84	280	10	45	46	10	100	7	33
6	0.65-1.1	1.5			3		200	290	13	39		8	89	4	18
7	0.43-0.65	0.9													
BU	0.43>	1.3													
Beijing no.003: 2002/4/8 9:30 - 21:00, total flow = 1.95 m³															
0	>11.0	9.3	42	62	7	50	34	62	8	43	65	9	58	6	
1	7.0-11.0	5.1	69		4	20	39	54	6	31	34	7	41	4	
2	4.7-7.0	4.2													
3	3.3-4.7	2.3			10	71	90	113	13	69	56	14	91	7	71
4	2.1-3.3	2.6			3		66	47	6	30	17	6	44	4	16
5	1.1-2.1	2.0	120		3		68	89	2	13		1	12		
6	0.65-1.1	0.6	390		21					14					
7	0.43-0.65	1.3													
BU	0.43>	1.2													
Beijing no.008: 2002/4/17-4/19, total flow = 81.5 m³															
0	>11.0	5.1	67		9	120	91	160	10	45	130	10	65	7	
1	7.0-11.0	3.4	69	150	7	120	64	66	7	32	67	8	89	5	33
2	4.7-7.0	4.2	68	810	21	560	120	240	10	41	120	10	86	7	12
3	3.3-4.7	3.3	71			22	75	150	5	23	51	5	49	3	
4	2.1-3.3	1.6	210	690	20	580	320	750	12	37	78	10	88	5	
5	1.1-2.1	1.0	650	1230	19	920	260	560	7	16		2	57		
6	0.65-1.1	1.2	320			43	250	520	8	5			18		
7	0.43-0.65	1.8													
BU	0.43>	2.0													
Hefei no.03: 2002/3/30- 4/5, total flow = 214 m³															
0	>11.0	6.7	54	55	9	24	45	140	11	45	110	12	120	10	5
1	7.0-11.0	3.6	73	92	15	39	83	280	19	89	180	21	150	14	
2	4.7-7.0	9.7	53	42	10	23	39	190	12	53	100	13	80	8	
3	3.3-4.7	9.8	55					310			110				
4	2.1-3.3	4.6	110	60	14	36	84	810	21	86	120	19	120	11	
5	1.1-2.1	3.4													
6	0.65-1.1	5.3	45		1	4	120	380	18	12	13	2	27	2	
7	0.43-0.65	3.0									11				
BU	0.43>	7.9									7				
Hefei no.04: 2002/4/12- 4/19, total flow = 264 m³															
0	>11.0	3.9	67		8	17	46	120	10	49	110	12	140	10	6
1	7.0-11.0	2.4	59		7	9	57	210	11	45	83	11	110	8	
2	4.7-7.0	4.1	80		9	19	55	220	13	49	88	12	92	8	
3	3.3-4.7	5.9	62		8	33	43	210	12	47	66	11	89	8	
4	2.1-3.3	5.9	60		6	14	37	240	12	43	54	10	81	6	
5	1.1-2.1	3.6	81		5	8	71	290	17	45	47	9	140	8	8
6	0.65-1.1	4.0	51		2		86	250	14	16	16	3	51	3	
7	0.43-0.65	3.1	57								10				
BU	0.43>	1.6									18				
Qingdao no.022: 2002/3/20-3/23, total flow = 121 m³															
0	>11.0	0.0													
1	7.0-11.0	6.2	700	95	21	9	96		27	130	200	29	150	17	
2	4.7-7.0	13.9	300	71	15	21	53		19	82	140	17	110	11	
3	3.3-4.7	12.9	390	80	16	18	52		21	87	140	20	110	12	
4	2.1-3.3	15.2	250	37	8	4	34		11	47	71	11	59	6	
5	1.1-2.1	4.2	530		11		81		27	77	87	18	92	9	
6	0.65-1.1	2.2	1040		4		220		60	36	40	7	73	8	
7	0.43-0.65	1.9													
BU	0.43>	1.0													
Qingdao no.023: 2002/4/1-4/4, total flow = 121 m³															
0	>11.0	2.8	830		13		150		12	58	110	14	65	8	
1	7.0-11.0	1.6	1980		12		190		12	61	92	14	55	8	
2	4.7-7.0	3.7	940		15	160			16	77	120	19	95	9	
3	3.3-4.7	4.4	760		13		110		14	69	99	17	74	8	
4	2.1-3.3	4.0	460		12		88		16	73	100	19	84	8	
5	1.1-2.1	3.2	720		6		79		13	45	63	13	54	6	
6	0.65-1.1	1.9			3		130		14	24	37	7	69	11	
7	0.43-0.65	2.9									21			9	
BU	0.43>	2.4									34			5	

Italic type indicates that the value is semiquantitative.

Appendix. (continue)

Stage	size (um) (µm)	weight (mg)	Cd ppm	Sn ppm	Sb ppm	Cs ppm	Ba ppm	La ppm	Ce ppm	Pr ppm	Nd ppm	Sm ppm	Eu ppm	Gd ppm	Tb ppm
Beijing no.001: 2002/2/27-3/4, total flow = 204 m³															
0	>11.0	5.6	2	30	6	3.1	550	19	41	4	15	3	0.7	3	0.4
1	7.0-11.0	4.0	3	20	7	3.3	570	22	45	5	16	3	0.7	3	0.6
2	4.7-7.0	6.4	5	20	10	3.6	670	26	56	5	19	4	0.8	3	0.6
3	3.3-4.7	5.6	9	110	20	3.5	630	23	52	5	17	3	0.6	9	0.6
4	2.1-3.3	3.4	10	60	50	4.9	690	31	62	6	22	4	0.8	4	0.7
5	1.1-2.1	5.2	8	120	50	1.7	270	11	19	2	8	1	0.3	1	0.2
6	0.65-1.1	5.3	7	90	60	0.8	200	2	5	0.4	2	0.3	0.1	0.2	0.0
7	0.43-0.65	2.4	6												
BU	0.43>	3.1	6												
Beijing no.002: 2002/3/19-3/21, total flow = 81.5 m³															
0	>11.0	15.4	1	10		9.2	770	52	110	12	45	8	1.7	8	1.1
1	7.0-11.0	24.8	0.3	3	0.5	2.5	180	10	22	2	9	2	0.3	2	0.3
2	4.7-7.0	16.7	1	6		5.7	420	22	53	5	20	4	0.8	4	0.6
3	3.3-4.7	13.1	2	6		4.3	300	18	41	4	15	3	0.6	3	0.4
4	2.1-3.3	8.4	3	5	1	3.3	200	13	28	3	11	2	0.4	2	0.3
5	1.1-2.1	4.0	8	10	2	3.8	180	14	31	3	12	2	0.5	2	0.3
6	0.65-1.1	1.5	9	30		3.8	100	10	21	2	9	2	0.3	2	0.2
7	0.43-0.65	0.9	11												
BU	0.43>	1.3													
Beijing no.003: 2002/4/8 9:30 - 21:00, total flow = 1.95 m³															
0	>11.0	9.3	1	3		2.8	210	13	26	3	11	2	0.5	2	0.3
1	7.0-11.0	5.1	0.4	6		2.2	120	8	16	2	7	1	0.3	1	0.2
2	4.7-7.0	4.2				1.3									
3	3.3-4.7	2.3	0.7	10		5.2	270	18	38	5	15	3	0.6	3	0.5
4	2.1-3.3	2.6	0.5	20	10	2.4	92	15	22	3	7	1	0.3	1	0.2
5	1.1-2.1	2.0	0.5	20	10	1.0		2	5	1	2	0.5	0.1		0.1
6	0.65-1.1	0.6	2	30		1.4			5	1		0.7			
7	0.43-0.65	1.3													
BU	0.43>	1.2													
Beijing no.008: 2002/4/17-4/19, total flow = 81.5 m³															
0	>11.0	5.1	1	30	5	2.7	340	16	32	4	13	2	0.5	2	0.4
1	7.0-11.0	3.4	1	8		2.3	200	10	22	2	9	2	0.4	2	0.3
2	4.7-7.0	4.2	2	30		2.9	360	14	30	3	12	2	0.4	2	0.3
3	3.3-4.7	3.3	1	10	4	1.7	200	8	16	2	7	1	0.2	1	0.2
4	2.1-3.3	1.6	7	40		3.3	340	14	29	4	12	2	0.4	2	0.3
5	1.1-2.1	1.0	6	90	9	2.5		6	12	2	5	1			
6	0.65-1.1	1.2	5	150		1.4		2	4	0.5	2				
7	0.43-0.65	1.8	7												
BU	0.43>	2.0	5												
Hefei no.03: 2002/3/30- 4/5, total flow = 214 m³															
0	>11.0	6.7	1	9		3.1	270	16	32	3	14	2	0.5	2	0.3
1	7.0-11.0	3.6	2	20		7.2	470	30	62	7	25	5	1.0	4	0.7
2	4.7-7.0	9.7	2	10		4.3	280	18	38	4	15	3	0.6	3	0.4
3	3.3-4.7	9.8					320								
4	2.1-3.3	4.6	9	30		9.1	460	29	65	6	24	4	0.9	4	0.6
5	1.1-2.1	3.4													
6	0.65-1.1	5.3	6	80		2.6	79	4	8	1	3	0.5	0.1	0.4	0.1
7	0.43-0.65	3.0	9												
BU	0.43>	7.9	4												
Hefei no.04: 2002/4/12- 4/19, total flow = 264 m³															
0	>11.0	3.9	1	10		3.3	290	19	38	4	15	3	0.5	2	0.4
1	7.0-11.0	2.4	5	20		3.8	280	17	35	3	14	2	0.5	2	0.4
2	4.7-7.0	4.1	4	20		4.1	290	17	37	4	14	3	0.4	2	0.4
3	3.3-4.7	5.9	4	10		4.1	260	16	36	4	13	2	0.5	2	0.4
4	2.1-3.3	5.9	6	20		4.6	230	15	34	3	12	2	0.5	2	0.3
5	1.1-2.1	3.6	8	50		6.9	200	14	31	3	10	2	0.3	2	0.3
6	0.65-1.1	4.0	10	60		2.7	78	5	10	1	3	0.6	0.1	0.5	0.1
7	0.43-0.65	3.1	9												
BU	0.43>	1.6	8												
Qingdao no.022: 2002/3/20-3/23, total flow = 121 m³															
0	>11.0	0.0													
1	7.0-11.0	6.2	1	10	1	9.3	740	42	90	9	36	7	1.3	6	1.0
2	4.7-7.0	13.9	0.7	8	3	5.9	500	26	60	6	23	4	0.9	4	0.6
3	3.3-4.7	12.9	1	20	3	6.7	490	29	67	7	25	5	1.0	4	0.7
4	2.1-3.3	15.2	1	7	3	3.9	260	15	34	3	13	3	0.5	2	0.4
5	1.1-2.1	4.2	4	40	20	10.3	380	24	53	5	20	4	0.7	3	0.6
6	0.65-1.1	2.2	4	90	40	7.5	200	10	21	2	8	2	0.3	1	0.3
7	0.43-0.65	1.9													
BU	0.43>	1.0													
Qingdao no.023: 2002/4/1-4/4, total flow = 121 m³															
0	>11.0	2.8	1	40		3.6	650	21	42	5	17	3	0.7	3	0.5
1	7.0-11.0	1.6	0.6			4.1	520	20	41	5	17	3	0.7	3	0.4
2	4.7-7.0	3.7	1	20	0.6	5.2	550	30	64	7	26	5	0.8	4	0.7
3	3.3-4.7	4.4	1	20	30	4.7	430	24	50	5	20	3	0.7	3	0.5
4	2.1-3.3	4.0	2	20	20	5.4	440	26	55	6	21	4	0.9	4	0.6
5	1.1-2.1	3.2	2	20	4	3.9	310	18	37	4	14	3	0.6	3	0.4
6	0.65-1.1	1.9	4	40	20	3.0	250	11	21	2	7	1	0.3	1	0.2
7	0.43-0.65	2.9	6												
BU	0.43>	2.4	4												

Italic type indicates that the value is semiquantitative.

Appendix. (continue)

Stage	size (um) (µm)	weight (mg)	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
Beijing no.001: 2002/2/27-3/4, total flow = 204 m³															
0	>11.0	5.6	2	0.4	1	0.2	1	0.2	2	3	2	240	2	7	2
1	7.0-11.0	4.0	2	0.4	1	0.2	1	0.2	2	3	3	250	4	7	3
2	4.7-7.0	6.4	3	0.5	2	0.3	2	0.3	3	2	4	300	4	8	3
3	3.3-4.7	5.6	3	0.5	2	0.3	2	0.2	3	4	5	480	7	8	4
4	2.1-3.3	3.4	3	0.7	2	0.4	2	0.3	4	4	6	1400	20	10	4
5	1.1-2.1	5.2	1	0.2	0.7	0.1	1	0.1	2	4	2	1650	20	4	1
6	0.65-1.1	5.3	0.2	0.1	0.2	0.0	0.2	0.0	0.3	5	2	1830	20	1	1
7	0.43-0.65	2.4										2810			
BU	0.43>	3.1										3560			
Beijing no.002: 2002/3/19-3/21, total flow = 81.5 m³															
0	>11.0	15.4	5	1.0	3	0.4	3	0.4	5	2	1	100	1	20	4
1	7.0-11.0	24.8	1	0.2	0.7	0.1	0.7	0.1	1	1	0.3	30	0.4	4	1
2	4.7-7.0	16.7	3	0.5	2	0.2	1	0.2	3	2	1	70	1	8	2
3	3.3-4.7	13.1	2	0.4	1	0.2	1	0.2	2	2	0.7	60	1	6	2
4	2.1-3.3	8.4	2	0.3	1	0.1	1	0.1	2	2	0.5	70	1	5	1
5	1.1-2.1	4.0	2	0.3	1	0.1	1	0.1	3	3	1	200	3	8	1
6	0.65-1.1	1.5	1	0.2	0.6	0.1	0.7	0.1		10	2	740	7		
7	0.43-0.65	0.9										1660			
BU	0.43>	1.3													
Beijing no.003: 2002/4/8 9:30 - 21:00, total flow = 1.95 m³															
0	>11.0	9.3	2	0.3	1	0.1	1	0.1	1	3	0.3	30	0.3	4	1
1	7.0-11.0	5.1	1	0.2	1	0.1	0.5	0.1	1	2	0.2	30	0.5	3	0.6
2	4.7-7.0	4.2								3					
3	3.3-4.7	2.3	2	0.5	1	0.2	1	0.3	2	6	0.5	60	0.7	7	1
4	2.1-3.3	2.6	1	0.2	0.6	0.1	0.4	0.1	1	2	0.2	40	1	4	0.6
5	1.1-2.1	2.0	0.2		2	0.0		0.03		1		30	0.2	2	0.2
6	0.65-1.1	0.6								8		180	1	4	
7	0.43-0.65	1.3										250			
BU	0.43>	1.2										220			
Beijing no.008: 2002/4/17-4/19, total flow = 81.5 m³															
0	>11.0	5.1	2	0.3	1	0.1	1	0.1	2	3	0.6	90	0.5	5	1
1	7.0-11.0	3.4	1	0.2	0.7	0.1	0.6	0.1	2	3	0.5	80	0.6	6	1
2	4.7-7.0	4.2	2	0.3	1	0.2	1	0.1	2	4	1	130	1	6	1
3	3.3-4.7	3.3	1	0.2	0.6	0.1	0.4	0.1	1	5	1	120	1	4	1
4	2.1-3.3	1.6	1	0.3	1	0.1	0.7	0.2	2	3	3	600	6	6	2
5	1.1-2.1	1.0	0.6		0.3			0.04	2	20	4	1430	10	4	0.7
6	0.65-1.1	1.2									4	1740	20	2	0.3
7	0.43-0.65	1.8										1200			
BU	0.43>	2.0										1400			
Hefei no.03: 2002/3/30- 4/5, total flow = 214 m³															
0	>11.0	6.7	2	0.4	1	0.1	1	0.1	3	3	1	100	2	6	2
1	7.0-11.0	3.6	4	0.6	2	0.3	2	0.2	4	4	1	240	4	10	3
2	4.7-7.0	9.7	2	0.4	1	0.2	1	0.2	2	2	1	160	3	6	2
3	3.3-4.7	9.8													
4	2.1-3.3	4.6	3	0.6	2	0.3	2	0.3	3	5	2	920	20	10	3
5	1.1-2.1	3.4													
6	0.65-1.1	5.3	0.4	0.1	0.2	0.0	0.3	0.02	0.6	4	2	2380	50	2	0.7
7	0.43-0.65	3.0										2770			
BU	0.43>	7.9										600			
Hefei no.04: 2002/4/12- 4/19, total flow = 264 m³															
0	>11.0	3.9	2	0.4	1	0.2	1	0.1	3	3	0.5	120	2	7	2
1	7.0-11.0	2.4	2	0.3	1	0.1	1	0.1	2	3	0.6	220	4	6	2
2	4.7-7.0	4.1	2	0.4	1	0.1	1	0.2	2	4	1	240	5	6	2
3	3.3-4.7	5.9	2	0.4	1	0.2	1	0.1	2	3	0.7	240	5	6	2
4	2.1-3.3	5.9	2	0.3	1	0.2	1	0.1	2	4	1	480	10	5	2
5	1.1-2.1	3.6	1	0.3	1	0.1	1	0.1	3	5	2	1850	40	6	2
6	0.65-1.1	4.0	0.4	0.1	0.3	0.0	0.3	0.1	1	4	2	2510	50	2	0.7
7	0.43-0.65	3.1										2530			
BU	0.43>	1.6										1690			
Qingdao no.022: 2002/3/20-3/23, total flow = 121 m³															
0	>11.0	0.0													
1	7.0-11.0	6.2	5	0.9	3	0.4	3	0.4	4	4	1	150	2	10	4
2	4.7-7.0	13.9	3	0.6	2	0.3	2	0.2	3	2	1	110	1	9	3
3	3.3-4.7	12.9	3	0.7	2	0.3	2	0.2	3	2	1	150	2	10	3
4	2.1-3.3	15.2	2	0.4	1	0.2	1	0.1	2	2	0.6	160	2	5	2
5	1.1-2.1	4.2	3	0.6	2	0.3	2	0.2	2	5		1380	20	8	3
6	0.65-1.1	2.2	1	0.2	0.7	0.1	0.7	0.1	2	10		3960	50	3	2
7	0.43-0.65	1.9													
BU	0.43>	1.0													
Qingdao no.023: 2002/4/1-4/4, total flow = 121 m³															
0	>11.0	2.8	2	0.5	1	0.2	1	0.2	1	8	0.5	150	1	5	2
1	7.0-11.0	1.6	2	0.4	1	0.2	1	0.1	1	10	0.5	230	1	5	2
2	4.7-7.0	3.7	3	0.6	2	0.2	2	0.2	2	4	0.7	220	2	8	2
3	3.3-4.7	4.4	3	0.5	1	0.2	2	0.2	2	5	0.7	160	2	7	2
4	2.1-3.3	4.0	3	0.6	2	0.2	2	0.2	2	5	1	200	2	7	2
5	1.1-2.1	3.2	2	0.4	1	0.2	1	0.2	1	8	1	420	7	5	2
6	0.65-1.1	1.9	1	0.2	0.4	0.1	1	0.1	2	10	1	1650	20	2	1
7	0.43-0.65	2.9										1390			
BU	0.43>	2.4										1030			

Italic type indicates that the value is semiquantitative.