

Sr, Nd and Pb stable isotopes of surface dust on Ürümqi glacier No. 1 in western China

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ABSTRACT. Stable-isotopic ratios of strontium (Sr), neodymium (Nd) and lead (Pb) provide a means of identifying a geological source of substances and are used as tracers of elements in biological and geochemical processes. We analyzed these isotopic ratios of surface dust (cryoconite) collected on Ürümqi glacier No. 1, Tien Shan, China. The dust was separated chemically into five fractions (four minerals and organic matter), and the isotopic ratios of each fraction were measured. The Sr and Nd isotopic ratios in the fractions extracted with ultrapure water (saline minerals), hydrogen peroxide solution (organic matter) and acetic acid (carbonate minerals) were low and invariable, whereas those extracted by hydrochloric acid (phosphate minerals) and the residual fraction (silicate minerals) were higher. The difference was likely due to the original source of each fraction. The isotopic ratios of the surface dust collected from different sites showed no significant difference, suggesting that they were spatially uniform across the glacier. The isotopic ratios of the silicate fraction were closer to those of desert sand reported in China than those of the soil and bedrock around the glacier. This suggests that the silicate minerals on the glacier were derived from distant deserts. The isotopic ratios in saline, carbonate and phosphate fractions were also close to those of evaporites and apatite in that desert region, suggesting that these minerals were also derived from that source. The Sr isotopic ratios in the organic fraction were closer to ratios in the saline and carbonate fractions rather than the silicate or phosphate fractions and may therefore reflect the isotopic ratios of the elements when they are incorporated into living microbes on the glacier.

INTRODUCTION

Snow and ice on glaciers contain various impurities, such as soluble ions, mineral particles, pollen and microbes. It is important to understand the transportation process and dynamics of these impurities, since they can affect the melting of glaciers and are also indicative of various environments surrounding them. For example, impurities deposited on the glacier surface can reduce surface albedo and accelerate glacial melting, thus affecting the glacial mass balance (e.g. Takeuchi and others, 2001, 2006). Moreover, impurities preserved in annual snow layers of glaciers have been used in ice-core studies as indicators of previous glacial environments (e.g. Grousset and others, 1992; Biscaye and others, 1997).

Stable-isotopic ratios of strontium (Sr), neodymium (Nd) and lead (Pb) can be very helpful in understanding the transportation process and dynamics of such impurities on glaciers. Sr, Nd and Pb are commonly contained in natural substances such as rocks, water and organisms, although they are present at trace levels. Stable-isotopic ratios of these elements, especially $^{87}\text{Sr}/^{86}\text{Sr}$, $^{143}\text{Nd}/^{144}\text{Nd}$, $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$, vary significantly based on their geological origin, since they rarely change during transport. Since their isotopic ratios are not susceptible to either physical or chemical conditions, the ratios rarely change during transport in the atmosphere or after deposition as sediments. Thus, isotopic ratios can be used to determine the provenance of various substances and have

also been commonly used in loess or sediment studies, as well as in recent ice-core studies. For example, an analysis of the isotopic ratios of dust in a Greenland ice core revealed that its provenance was neither the Sahara Desert nor the North American continent but most likely East Asia (Biscaye and others, 1997). Furthermore, Grousset and others (1992) analyzed Sr and Nd isotopic ratios of dust in Antarctic ice cores that revealed its main provenance to be Patagonia. However, such isotopic analyses of polar ice cores generally proved very difficult and able to reveal only limited information on surface dust provenance since their concentration of impurities was very low (Svensson and others, 2000).

The analyses of these isotopes seem much easier on Asian mountain glaciers because the levels of impurities are greater there compared with those on polar glaciers, with one of their major components being desert sand. Huge amounts of mineral dust are blown from vast Asian deserts, such as Taklamakan, Junggar and Gobi, and are deposited on mountain glaciers in this region (Wake and others, 1994). Desert dust usually consists of several mineral components, each of which has a different geological origin. Yokoo (2000) chemically separated loess and desert sands in China into four fractions (saline, carbonate, phosphate and silicate minerals) and showed that each fraction had different Sr, Nd and Pb isotopic ratios. Thus, it is important to analyze the isotopic ratios of each mineral fraction to precisely identify their provenance.

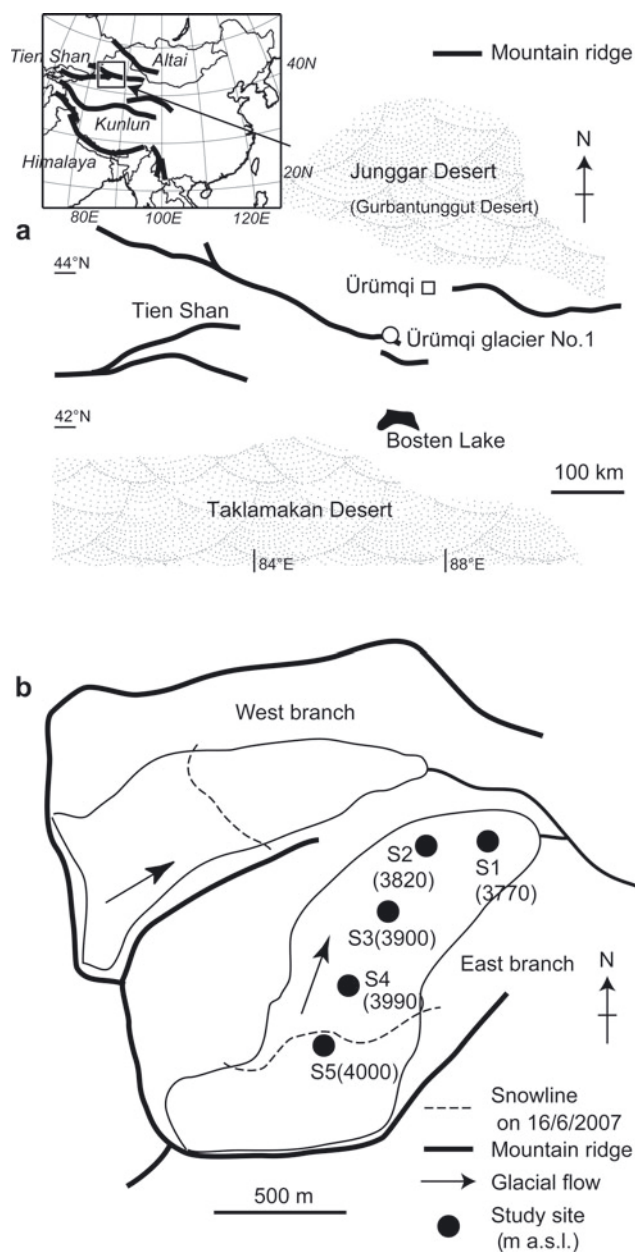


Fig. 1. (a) Location of Ürümqi glacier No. 1 in the Tien Shan, China, and (b) map of the glacier showing sites of sample collection. Date is day/month/year.

Another component of impurities on Asian glaciers is biogenic material derived from microbes living on the glaciers (e.g. Takeuchi and others, 2006). It has been reported that there is an abundance of diverse organisms living on Asian glaciers (e.g. snow algae, copepods, tardigrades, insects and bacteria). They include special organisms that readily adapt to cold environments and produce organic matter on the glacier surface (Takeuchi and others, 2001, 2005, 2006; Takeuchi, 2002). The isotopic ratios of such organic matter may reflect those of the mineral sources used as nutrients by the microbes. For example, the Sr isotope has been used as a tracer of Ca^{2+} in studies of geochemical process since its chemical characteristics are similar to those of Ca^{2+} . Since Ca is one of the elements essential for living organisms, it is incorporated together with Sr into their bodies. Therefore, the Sr isotopic ratios of organic matter derived from living organisms on the glacial



Fig. 2. Photograph of surface dust (cryoconite) on Ürümqi glacier No. 1.

surface may reveal relationships between such organisms and their mineral sources.

In this study, we analyzed Sr, Nd and Pb isotopic ratios of surface dust (cryoconite) collected on Ürümqi glacier No. 1, Tien Shan, China. The surface dust was separated chemically into five fractions (four mineral fractions and organic matter) using different acids, and the isotopic ratios of each fraction were measured. We also analyzed the isotopic ratios of soil and bedrock around the glacier to evaluate their contribution to the impurities. Variations in isotopic ratios are discussed in terms of the sources of each mineral and biological process on the glaciers.

STUDY SITE AND SAMPLE COLLECTION

Ürümqi glacier No. 1 (43°06' N, 86°48' E) is located on the eastern side of the Tien Shan in the Xinjiang Uygur autonomous region of China (Fig. 1). The area of this glacier is surrounded by vast deserts: Taklamakan to the south, Junggar to the north and Gobi to the east. Dust storms frequently occur in spring, depositing wind-blown dust on the glacier. The glacier is a north-facing valley glacier covering approximately 1.73 km² and lies between 3740 and 4486 m a.s.l. It comprises two branches (east and west) that became separated in 1994 due to glacial shrinkage (Zhang and others, 1985; Ye and others, 2005). Our sample collection was carried out on the east branch.

Abundant surface dust (cryoconite) has been reported on this glacier (Fig. 2). The amount of surface dust ranges from 86 to 1113 g m⁻² in dry weight (Takeuchi and Li, 2008; Takeuchi and others, 2010). The surface dust contains biogenic organic matter including microbes living on the glacier. The organisms are mainly composed of snow algae and cyanobacteria.

Sample collections were carried out in August 2006 and June 2007 on the glacier (Table 1). Collections of the surface dust were carried out at five sites, ranging from 3770 (S1) to 4000 m a.s.l. (S5; Fig. 1). Samples at site S3 were collected on 4 August 2006 and those at sites S1, S2, S4 and S5 were collected on 24–26 June 2007. The surface condition of sites S1, S2, S3 and S4 was bare ice, while that of site S5 was snow.

Soil and rocks around the glacier were also collected to evaluate their contribution to the surface dust. One sample of soil was collected from the ground surface located approximately 500 m away from the glacial terminus. One

sample of rock was also collected from a rock cliff at the side of the glacier (Fig. 1).

All these samples were collected with a stainless-steel scoop, stored in clean 30 mL polyethylene bottles and dried at 60°C for 24 hours.

ANALYTICAL PROCEDURE

Microscopic observation of surface dust

The surface dust was observed with optical microscopes (Olympus BX51 and Leica MZ-12). The size of the constituents (organic and mineral particles) was measured manually on digital photographs with an image-processing application (Image J, National Institutes of Health, USA). 200 particles of each constituent were randomly chosen from the photographs, and their longest diameter was measured.

X-ray diffraction (XRD) analysis

The mineralogical composition of surface dust, soil and bedrock was identified by XRD analysis using RIGAKU Geigerflex RAD 11-B at Chiba University. XRD analysis is an efficient means with which to identify and quantify minerals and is used in loess studies and provenance identification of dust in ice cores (e.g. Svensson and others, 2000). In XRD analysis each mineral shows its specific diffraction spectrum, thus enabling mineral identification by comparing the peak position and peak intensity of measured minerals with that of a standard mineral.

Samples were powdered with an agate mortar. The powdered samples were mounted on low-background support aluminum specimen holders and irradiated with a monochromatic Cu $K\alpha$ source ($\lambda=1.54\text{ \AA}$) at 40 kV and 25 mA, after which the diffraction spectra of minerals in the samples were obtained. The minerals were identified with a Macintosh computer using XRD analysis software (MacDiff). The analysis was done for one sample at each study site.

Chemical separation (sequential leaching) and isotope analysis

To measure the Sr, Nd and Pb isotopic ratios of mineral and organic components in surface dust, the samples were chemically separated into the following five fractions using four different acid solutions: (1) ultrapure water (H_2O); (2) hydrogen peroxide solution (H_2O_2); (3) acetic acid (HOAc); (4) hydrochloric acid (HCl); and (5) HCl residual fraction. Fractions (1), (2), (3) and (4) were extracted in the following sequence: for 5 min with H_2O at room temperature, for 24 hours with 10% H_2O_2 at 70°C, for 2 hours with 5% HOAc at 75°C, and for 45 min with 20% HCl at 100°C, respectively. All extractions were conducted in a Teflon airproof system. The residual of HCl was digested with 38% hydrofluoric acid (HF), 70% perchloric acid (HClO_4) and 68% nitric acid (HNO_3) at 200°C, and the HCl residual fraction (5) was then obtained. Based on the same extractions of Chinese desert sand and soil by Yokoo (2000), each fraction corresponded to: (1) saline minerals, (2) organic matter, (3) carbonate minerals, (4) phosphate minerals and (5) silicate minerals. The amount of sample used in each extraction was 1.0 g in dry weight. Teflon beakers pre-washed with HCl and HNO_3 were used for the analysis. These sequential extractions and isotopic measurements were performed in a class 100 clean laboratory at the

Table 1. Descriptions of sample of surface dust, soil and bedrock on and near Ürümqi glacier No. 1 (43°06' N, 86°48' E)

Sample	Site	Sample type	Sampling date	Elevation m a.s.l.
UM1	S1	Surface dust (ice)	24 June 2007	3770
UM2	S2	Surface dust (ice)	24 June 2007	3820
UM3	S3	Surface dust (ice)	4 August 2006	3900
UM4	S4	Surface dust (ice)	25 June 2007	3990
UM5	S5	Surface dust (snow)	26 June 2007	4000
Soil		Soil	24 June 2007	3785
Bedrock		Bedrock	26 June 2007	3830

Research Institute for Humanity and Nature (RIHN), Kyoto. More detailed analytical procedures of these sequential extractions are described in Yokoo (2000).

After the extractions, Sr, Nd and Pb were separated chromatographically from each fraction using a cation-exchange column. $^{87}\text{Sr}/^{86}\text{Sr}$, $^{143}\text{Nd}/^{144}\text{Nd}$, $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, $^{208}\text{Pb}/^{204}\text{Pb}$, $^{206}\text{Pb}/^{207}\text{Pb}$ and $^{208}\text{Pb}/^{206}\text{Pb}$ ratios were then determined with a TRITON thermal ionization mass spectrometer (Thermo Fisher Scientific K.K.) at RIHN. Sr isotopes were measured for all five fractions, Nd isotopes were measured for HCl-extracted and residual fractions, and Pb isotopes for H_2O - and H_2O_2 -extracted fractions and the HCl residual fraction. The measured $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ values were normalized to a $^{86}\text{Sr}/^{88}\text{Sr}$ value of 0.1194 and a $^{146}\text{Nd}/^{144}\text{Nd}$ value of 0.7219, respectively. The mean $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of NIST-SRM987 and the mean $^{143}\text{Nd}/^{144}\text{Nd}$ ratios of JNdi-1 (the international standard specimen for Sr and Nd isotopes determined during this study) were 0.710256 ± 0.000003 (mean \pm standard deviation (SD), $n=11$) and 0.512126 ± 0.000003 (mean \pm SD, $n=7$). The mean $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ values of NIST-SRM981 (the international standard specimen for Pb isotopes determined during this study) were 16.9166 ± 0.0021 (mean \pm SD, $n=6$), 15.4640 ± 0.0028 (mean \pm SD, $n=6$) and 36.6155 ± 0.0089 (mean \pm SD, $n=6$), respectively. The measured Sr, Nd and Pb isotopic ratios of each standard specimen and sample were determined by repeated measurements (140 times). Internal precisions of Sr and Nd isotope ratios were better than 0.000005 during the course of this study. For convenience, the $^{143}\text{Nd}/^{144}\text{Nd}$ ratios were normalized and denoted as $\varepsilon\text{Nd}(0) = [(^{143}\text{Nd}/^{144}\text{Nd})/0.512636 - 1] \times 10^4$.

RESULTS

Components of surface dust

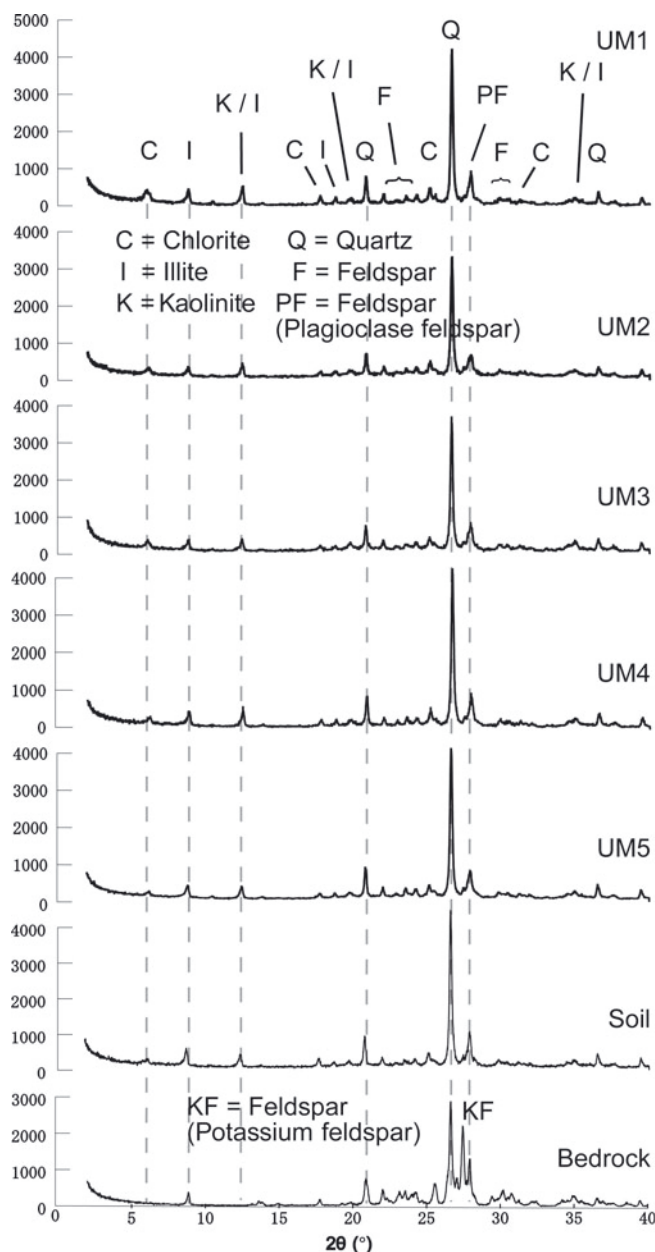
Microscopic observations showed that the dust on Ürümqi glacier No. 1 consisted mainly of mineral particles, organic matter, snow algae and cyanobacteria, as reported by Takeuchi and Li (2008). On the ice surface (S1–S4), these constituents formed spherical cryoconite granules, as described by Takeuchi and others (2010). The mineral particles in the dust were brown, reddish brown, black, green or red. The maximum size of the particles was 166 μm and the mean size was 29.5 μm .

The cryoconite granules were brown and spherical and contained filamentous cyanobacteria and mineral particles.

Table 2. Sample weight (dry weight), amounts of extracted fraction and weight percentage of extracted fraction to analyzed weight of surface dust, soil and bedrock

Sample	Weight g	H ₂ O-extracted fraction g (%)	H ₂ O ₂ -extracted fraction g (%)	HOAc-extracted fraction g (%)	HCl-extracted fraction g (%)	HCl residual fraction g (%)
UM1	0.998	0.002 (0.2)	0.009 (0.8)	0.018 (1.6)	0.279 (25.0)	0.808 (72.3)
UM2	0.996	0.004 (0.4)	0.020 (1.8)	0.019 (1.7)	0.285 (25.7)	0.776 (70.3)
UM3	1.007	0.006 (0.6)	0.022 (2.3)	0.012 (1.2)	0.171 (17.5)	0.764 (78.3)
UM4	0.999	0.041 (0.4)	0.019 (1.7)	0.019 (1.7)	0.240 (22.1)	0.803 (74.0)
UM5	0.126	NA	0.002 (1.8)	0.005 (4.0)	0.024 (18.5)	0.099 (76.4)
Soil	1.016	0.003 (0.3)	0.008 (0.7)	0.011 (1.0)	0.288 (24.7)	0.857 (73.4)
Bedrock	1.099	0.001 (0.1)	0.007 (0.7)	0.001 (0.1)	0.974 (1.9)	1.003 (97.1)

NA: not available.

**Fig. 3.** XRD spectra of surface dust, soil and bedrock near Ürümqi glacier No. 1.

The maximum size of cryoconite granules was 3.4 mm and the mean size was 1.4 mm. Granules in the size range 1.3–2.0 mm were most abundant. There were no significant differences in the size of cryoconite granules and mineral particles among the study sites.

The results of chemical extractions of the surface dust showed that the amounts of each fraction were relatively close among the samples (Table 2). The mass fraction of the HCl residual (silicate minerals) was the largest of all samples (70.3–78.3%), whereas the H₂O-, H₂O₂- and HOAc-extracted fractions (saline minerals, organic matter and carbonate minerals) were relatively smaller. The proportions of each fraction of the soil sample were similar to those in the surface dust (Table 2). However, the proportion in the bedrock sample was significantly different from those in the surface dust, especially in the HCl-extracted and residual fractions (1.9% vs 17.5–25.7% and 97.1% vs 70.3–78.3%).

XRD analysis

The XRD spectra of the surface dust showed peaks of several silicate minerals (Fig. 3). The observed peaks were identified as quartz (26.7°), plagioclase (28.2°), chlorite (6.2°, 12.5°) and clay minerals such as illite (8.9°) and kaolinite (12.5°, 25.2°). They were observed in all the surface dust samples with intensities that were also similar among the samples.

The XRD spectra of the soil samples were similar to those of surface dust on the glacier (Fig. 3), indicating that the silicate minerals contained in the soil were the same as those in surface dust. The soil spectrum showed the peaks of quartz, plagioclase, chlorite and clay minerals, including illite and kaolinite. The peak intensities of quartz, plagioclase and illite in the soil spectrum were relatively higher than those of the surface dust.

The spectrum of the bedrock sample differed slightly from those of the soil and the surface dust on the glacier (Fig. 3). The bedrock spectrum showed peaks of quartz, plagioclase, chlorite, illite and potassium feldspar (27.2°). The peaks of quartz and illite in the bedrock were of lower intensity than those of the surface dust. In contrast, the intensity of plagioclase in the bedrock was higher than that of the surface dust.

Sr, Nd and Pb isotopic ratios of surface dust on the glacier

The Sr isotopic ratios (⁸⁷Sr/⁸⁶Sr) of surface dust on the glacier varied significantly among the fractions (Table 3; Fig. 4). The

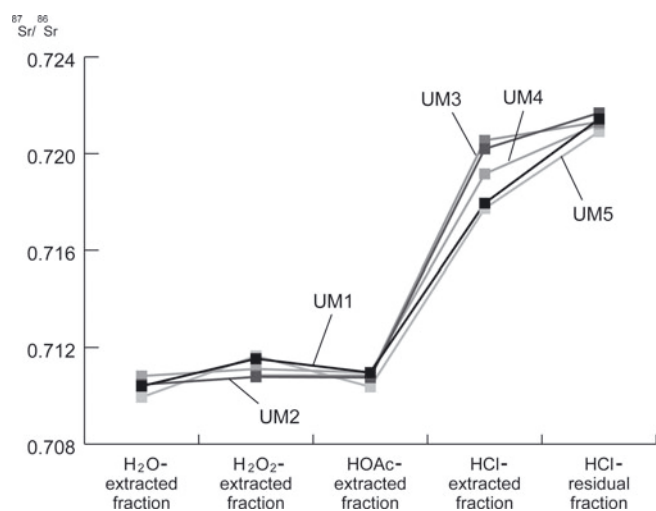


Fig. 4. Sr isotopic ratios of five fractions of surface dust on Ürümqi glacier No. 1.

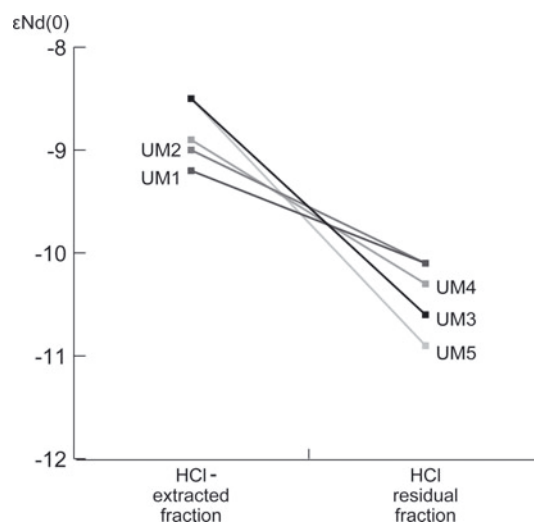


Fig. 5. Nd isotopic ratios of two fractions of surface dust on Ürümqi glacier No. 1.

Sr isotopic ratios of all the fractions of surface dust ranged from 0.710399 to 0.721685. The ratios of the H₂O-, H₂O₂- and HOAc-extracted fractions were relatively low and invariable (0.710404 ± 0.000003, 0.711184 ± 0.000004 and 0.710777 ± 0.000004, mean ± SD), while those of the HCl-extracted and residual fractions were higher (0.719115 ± 0.000003 and 0.721312 ± 0.000003, mean ± SD) than the others.

Variations in the Sr isotopic ratio among the samples collected from five different sites on the glacier were smaller than the variations among the fractions. For example, HCl

residual fractions ranged from 0.720908 to 0.721685, while the SDs of the isotopic values of each fraction were <0.001276.

The Nd isotopic values (εNd(0)) also differed significantly among the fractions (Table 4; Fig. 5). The isotope of the HCl-extracted fraction was higher than that of the HCl residual fraction (−8.8 vs −10.4, mean) in all samples. The isotopic ratio showed little difference among the samples.

The Pb isotopic ratios (²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb and ²⁰⁸Pb/²⁰⁴Pb) of each fraction showed that the ratios in the HCl residual fraction were markedly distinct from those in

Table 3. Sr isotopic ratio (⁸⁷Sr/⁸⁶Sr, mean ± (SD × 10⁶)) of five fractions of surface dust, soil and bedrock on and near Ürümqi glacier No. 1

Sample	H ₂ O-extracted fraction	H ₂ O ₂ -extracted fraction	HOAc-extracted fraction	HCl-extracted fraction	HCl residual fraction
UM1	0.710461 ± 03	0.710783 ± 04	0.710757 ± 03	0.720195 ± 04	0.721685 ± 03
UM2	NA	0.710844 ± 04	0.710838 ± 06	0.720547 ± 04	0.721309 ± 03
UM3	0.710399 ± 03	0.711533 ± 04	0.710960 ± 04	0.717947 ± 03	0.721433 ± 03
UM4	0.710818 ± 03	0.711110 ± 03	0.710959 ± 04	0.719160 ± 03	0.721225 ± 03
UM5	0.709940 ± 03	0.711648 ± 03	0.710372 ± 03	0.717727 ± 03	0.720908 ± 03
Soil	0.712297 ± 03	0.712531 ± 04	0.712547 ± 04	0.740487 ± 03	0.728641 ± 04
Bedrock	0.774755 ± 03	0.771920 ± 03	0.761039 ± 04	0.914682 ± 03	1.086115 ± 04

Note: NA: not available.

Table 4. Nd isotopic ratio (¹⁴³Nd/¹⁴⁴Nd, mean ± (SD × 10⁶)) and εNd(0) of five fractions of surface dust, soil and bedrock on and near Ürümqi glacier No. 1

Sample	HCl-extracted fraction		HCl residual fraction	
	¹⁴³ Nd/ ¹⁴⁴ Nd	εNd(0)	¹⁴³ Nd/ ¹⁴⁴ Nd	εNd(0)
UM1	0.512168 ± 03	−9.2	0.512120 ± 03	−10.1
UM2	0.512179 ± 03	−9.0	0.512121 ± 03	−10.1
UM3	0.512200 ± 02	−8.5	0.512093 ± 02	−10.6
UM4	0.512182 ± 04	−8.9	0.512108 ± 03	−10.3
UM5	0.512200 ± 03	−8.5	0.512080 ± 03	−10.9
Soil	0.512087 ± 04	−10.7	0.512111 ± 03	−10.3
Bedrock	0.512068 ± 03	−11.1	0.512064 ± 03	−11.2

Table 5. Pb isotopic ratio ($^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$, mean \pm SD) of five fractions of surface dust, soil and bedrock on and near Ürümqi glacier No. 1

	Sample	H ₂ O-extracted fraction	H ₂ O ₂ -extracted fraction	HCl residual fraction
$^{206}\text{Pb}/^{204}\text{Pb}$	UM1	18.445 \pm 0.010	18.507 \pm 0.003	19.070 \pm 0.001
	UM2	18.391 \pm 0.002	18.447 \pm 0.001	19.088 \pm 0.002
	UM3	18.392 \pm 0.002	18.433 \pm 0.000	19.054 \pm 0.000
	UM4	18.360 \pm 0.004	18.409 \pm 0.000	19.071 \pm 0.007
	UM5	18.365 \pm 0.001	18.412 \pm 0.004	19.028 \pm 0.000
	Soil	18.859 \pm 0.002	NA	19.236 \pm 0.001
	Bedrock	20.762 \pm 0.001	NA	20.052 \pm 0.000
$^{207}\text{Pb}/^{204}\text{Pb}$	UM1	15.619 \pm 0.008	15.612 \pm 0.003	15.671 \pm 0.001
	UM2	15.595 \pm 0.002	15.595 \pm 0.001	15.673 \pm 0.001
	UM3	15.625 \pm 0.002	15.617 \pm 0.000	15.680 \pm 0.000
	UM4	15.595 \pm 0.003	15.594 \pm 0.000	15.668 \pm 0.006
	UM5	15.598 \pm 0.001	15.631 \pm 0.003	15.644 \pm 0.000
	Soil	15.631 \pm 0.001	NA	15.663 \pm 0.000
	Bedrock	15.763 \pm 0.001	NA	15.752 \pm 0.000
$^{208}\text{Pb}/^{204}\text{Pb}$	UM1	38.418 \pm 0.020	38.472 \pm 0.007	39.065 \pm 0.002
	UM2	38.311 \pm 0.004	38.385 \pm 0.001	39.093 \pm 0.003
	UM3	38.449 \pm 0.004	38.469 \pm 0.000	39.241 \pm 0.001
	UM4	38.294 \pm 0.008	38.353 \pm 0.001	39.092 \pm 0.014
	UM5	38.238 \pm 0.003	38.345 \pm 0.000	39.041 \pm 0.001
	Soil	38.693 \pm 0.003	NA	39.019 \pm 0.001
	Bedrock	38.819 \pm 0.002	NA	38.433 \pm 0.001

Note: NA: not available.

the H₂O- and H₂O₂-extracted fractions (Table 5; Fig. 6). The ratio of $^{206}\text{Pb}/^{204}\text{Pb}$ in the HCl residual fraction was higher than those in the H₂O- and H₂O₂-extracted fractions (19.062 \pm 0.002 vs 18.391 \pm 0.004 and 18.442 \pm 0.002), whereas the ratios of $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$ in the HCl residual fraction were lower than those in the H₂O- and H₂O₂-extracted fractions (15.667 \pm 0.002 vs 15.606 \pm 0.003 and 15.610 \pm 0.001; 39.107 \pm 0.002 vs 38.342 \pm 0.008 and 38.405 \pm 0.002, respectively, mean \pm SD).

The Sr isotopic ratios in the soil sample also varied among the fractions, exhibiting a trend similar to those of surface dust on the glacier (Table 3; Fig. 7). The ratios were highest in the HCl residual fraction and were similar to each other in the H₂O-, H₂O₂- and HOAc-extracted fractions. The isotopic values for all fractions in the soil were generally higher than those of surface dust. In particular, the difference was greater in the HCl-extracted and residual fractions (Table 3). The Nd isotopic ratios in the soil also differed between the HCl-extracted and HCl residual fractions (Table 4; Fig. 5). The ratios were higher in the HCl residual than in the HCl-extracted fractions (0.512111 vs 0.512087).

The Sr and Nd isotopic ratios of the bedrock differed significantly from those of the surface dust and soil (Tables 3 and 4; Fig. 7). Each fraction had a significantly higher Sr isotope (more than 0.048491) and lower Nd isotopic values (over 0.00100) compared with those of the surface dust and soil.

DISCUSSION

Variations in Sr, Nd and Pb isotopic ratios of surface dust

The Sr, Nd and Pb isotopic ratios of the surface dust varied significantly among the fractions, indicating that mineral components in the surface dust have different isotopic ratios. According to the analyses of Chinese desert sand (Yokoo,

2000), the H₂O-, HOAc- and HCl-extracted fractions and HCl residual fraction corresponded to saline, carbonate, phosphate and silicate minerals, respectively. Our results showed that the Sr isotope was highest for the HCl residual fraction (mean: 0.721312), and lower for the H₂O-, HOAc- and HCl-extracted fractions (mean: 0.710404, 0.710777 and 0.719115). This trend was similar to that of the Chinese desert sand. Fractions of the surface dust are probably the same minerals as those in the desert sand. Our results indicate that the geological origins of each mineral component are different.

In contrast to the variation among the fractions, the Sr, Nd and Pb isotopic ratios of the surface dust showed little variation among the samples collected from different sites on the glacier. The difference (range) of Sr isotopic ratio was, respectively, 0.000878 (0.709940–0.710818), 0.000588 (0.710372–0.710960), 0.002821 (0.717727–0.720547) and 0.000777 (0.720908–0.721685) for H₂O-, HOAc- and HCl-extracted fractions and the HCl residual fraction. This finding indicates that there is little spatial variation among the isotopic ratios on the glacier. The proportion of each extracted fraction also revealed no significant difference among the samples. Furthermore, XRD analyses of the surface dust also exhibited almost the same spectrum in the samples. These findings indicate that the surface dust on the glacier was spatially uniform in terms of its isotopic ratios and mineral composition.

The isotopic ratios of the UM5 sample collected from site S5 on the snow surface at the upper part of the glacier differed slightly from that of the other dust (UM1–UM4) collected on the ice surface. The Sr isotopic ratios of the H₂O- and HOAc-extracted fractions and the HCl residual fraction in UM5 were slightly lower than those from other sites (Table 3). There was little difference in the mineralogical composition between UM5 and other surface dust samples (UM1–UM4), except for the lower weight percentage of the

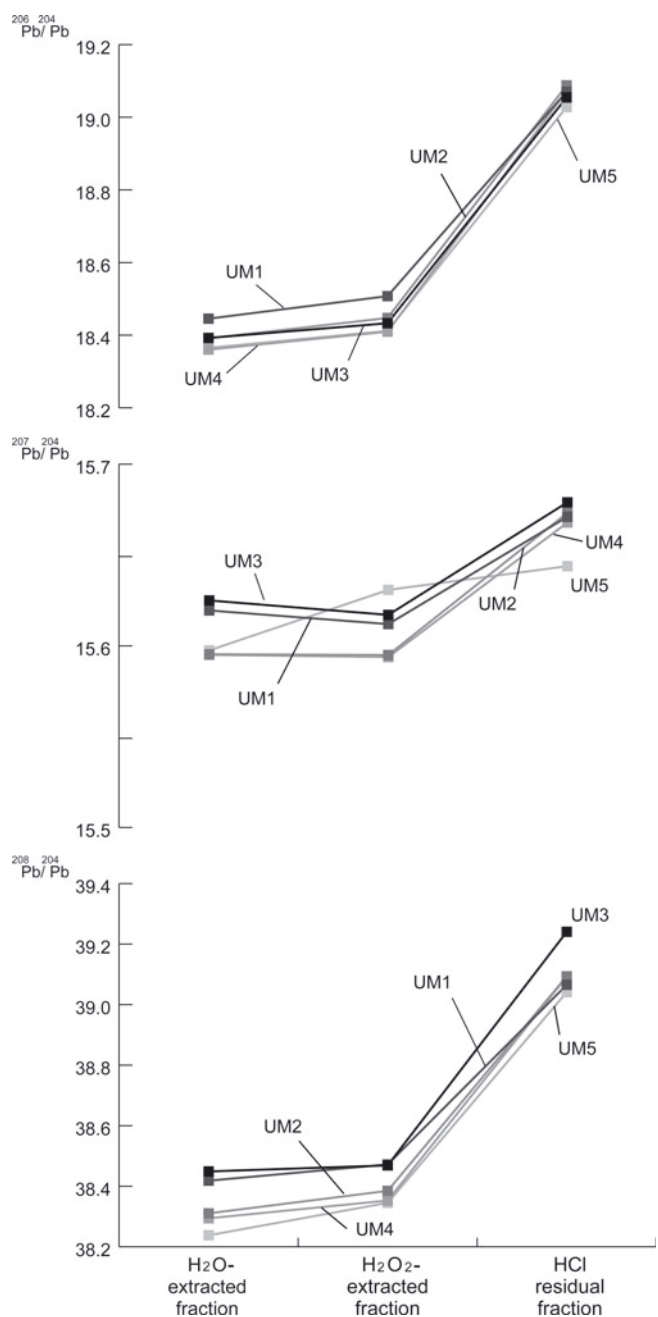


Fig. 6. Pb isotopic ratios ($^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{208}\text{Pb}/^{204}\text{Pb}$) of three fractions of surface dust on Ürümqi glacier No. 1.

HCl-extracted fraction in UM5 (Table 2). Although the dust on both the snow and ice surfaces of the glacier originates from aeolian dust, their dust provenance may vary with the wind direction. The dust on the snow surface was deposited between the previous winter and the following summer. On the other hand, the dust on the ice surface was deposited over longer periods of time, probably several to 100 years, since the surface is ablation ice of the glacier and lies exposed every melting season. The dust forms cryoconite granules, which repeatedly grew and disintegrated over a cycle of several years on the glacier (Takeuchi and others, 2010). This process can homogenize the isotope composition of the dust over these longer timescales. Because of the difference in retention times on the glacier, the dusts on the snow and ice surface may contain minerals of different provenances, a fact that may account for the distinct isotope ratio of dust on the snow surface.

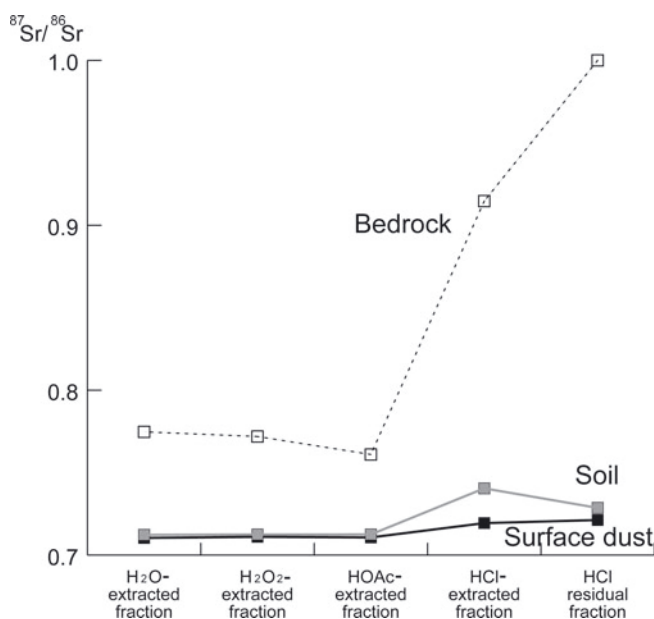


Fig. 7. Comparison of Sr isotopic ratios of five fractions among surface dust (mean of five sites), soil and bedrock on and near Ürümqi glacier No. 1.

Isotopic ratios of silicate minerals in surface dust and their provenance

The most dominant fraction in the surface dust on the glacier was the HCl residual fraction, which corresponds to silicate minerals. XRD analysis revealed that the surface dust mainly contains five silicate minerals: quartz, feldspar, chlorite, illite and kaolinite. These minerals are common in aeolian dust. Yokoo and others (2004) showed that silicate minerals in loess from the Central Loess Plateau in China mainly comprised quartz, plagioclase and clay minerals such as illite, kaolinite, chlorite, smectite, amphibole and pyroxenes, suggesting that the silicate minerals in the surface dust are most likely derived from such desert sand and loess.

The silicate mineral composition of the soil samples resembled that of surface dust on the glacier, suggesting that the soil around the glacier is a potential source of such surface dust. However, the Sr isotopic ratio of silicate minerals in the soil was significantly higher than that of surface dust (0.728641 vs 0.720908–0.721685, Table 3). This difference indicates that silicate minerals in the surface dust have a different geological origin from those in the soil.

The mineral composition and isotopic ratios of the bedrock sample differed markedly from those of surface dust on the glacier. XRD analysis showed the difference in mineralogical composition between the surface dust and bedrock. While potassium feldspar was found only in the bedrock, chlorite and kaolinite were found exclusively in the surface dust. Furthermore, the Sr isotopic ratios of bedrock were significantly higher than that of surface dust (a difference of >0.05 in all fractions; Tables 3 and 4; Fig. 7). This indicates that the bedrock around the glacier is not the main source of surface dust on the glacier.

Compared with the isotopic ratios of silicate minerals of desert or loess reported over China, those of the surface dust on this glacier were closer to those in sands from the Taklamakan Desert, southern Gobi Desert and Loess Plateau (Fig. 8). Previous studies have revealed that the provenance of loess, soil or desert sand can be identified in detail when

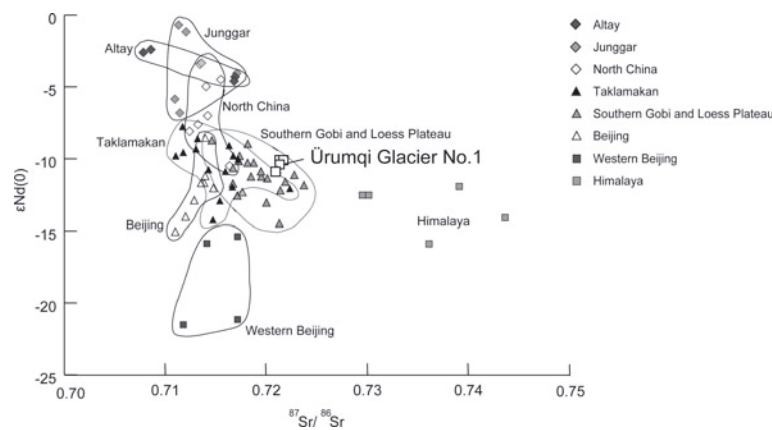


Fig. 8. Sr–Nd isotopic ratios of HCl-extracted fractions (silicate minerals) of glacial surface dust and those of loess, sand and stream sediment reported in the Asian region.

the Sr and Nd isotopic ratios are plotted in combination (Biscaye and others, 1997; Honda and others, 2004; Nakano and others, 2004; Yokoo and others, 2004; Chen and others, 2007). When the Sr isotopic ratios and $\epsilon\text{Nd}(0)$ values of silicate minerals in the surface dust are compared with those of loess, desert sand and river sediments reported in this area, the surface dust is plotted close to the Taklamakan Desert, southern Gobi Desert and Loess Plateau (Fig. 8; Altay: Zhu and others, 2006; Junggar: Honda and others, 2004; Chen and others, 2007; Taklamakan: Honda and others, 2004; Nakano and others, 2004; north China, southern Gobi, Loess Plateau, Beijing and western Beijing: Nakano and others, 2004; Himalaya: Singh and France-Lanord, 2002). However, since the predominant wind direction over the middle latitudes of eastern China is northwesterly (Sun, 2002), the southern Gobi Desert and Loess Plateau located in the downwind region of the glacier are unlikely source areas of the surface dust. According to the predominant wind direction, the Junggar Desert is the most likely source area, although our isotopic ratios were clearly distinct from those collected in the Junggar Desert. Further, Sun (2002) showed that the atmospheric circulation in the Taklamakan Desert is more complicated than in other areas and so it is possible that dust from the Taklamakan Desert can be entrained to elevations above 5000 m and then transported for long distances by the westerly jet stream. The surface dust on Ürümqi glacier No. 1 was therefore probably transported by such winds from the Taklamakan Desert.

Pb isotopic ratios of silicate minerals in the surface dust were also close to those reported from the Asian desert sand and loess. The $^{206}\text{Pb}/^{207}\text{Pb}$ and $^{208}\text{Pb}/^{207}\text{Pb}$ of silicate minerals in Chinese desert sand have been reported to range from 1.206 to 1.217 and from 2.500 to 2.509, respectively (Jones and others, 2000). These ratios generally matched those of silicate minerals in the surface dust on the glacier (Fig. 9).

Isotopic ratios of saline, carbonate and phosphate minerals

Lower Sr isotopic ratios in the H_2O -, HOAc - and HCl-extracted fractions (saline, carbonate and phosphate minerals) compared with those in the HCl residual fraction (silicate minerals) indicate that these minerals in the surface dust have different geological origins from those of silicate minerals. The Sr isotopic ratios of saline and carbonate minerals were similar to each other, suggesting that these two minerals share the same origin. On the other hand, the Sr isotopic ratios in phosphate minerals were relatively higher than those of saline and carbonate minerals, implying that the geological origin of the phosphate minerals differed from that of the other two minerals.

Saline, carbonate and phosphate minerals were not detected in the XRD analysis, probably due to the smaller amounts of these fractions compared with those of silicate minerals. Asian desert sand has also been reported to contain smaller amounts of these minerals. Using XRD analysis, Shao and others (2007) showed that airborne

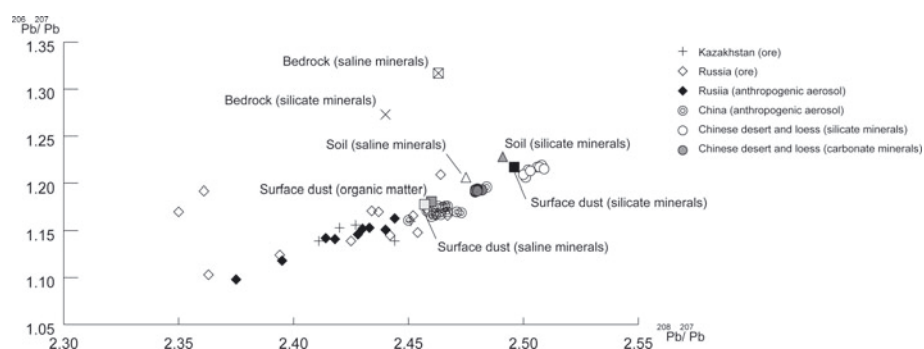


Fig. 9. Comparison of Pb isotope ratios of H_2O - and H_2O_2 -extracted fraction (saline minerals and organic matter) and HCl residual fraction (silicate minerals) in the surface dust and those in silicate and carbonate minerals, ore and aerosol influenced by anthropogenic activities.

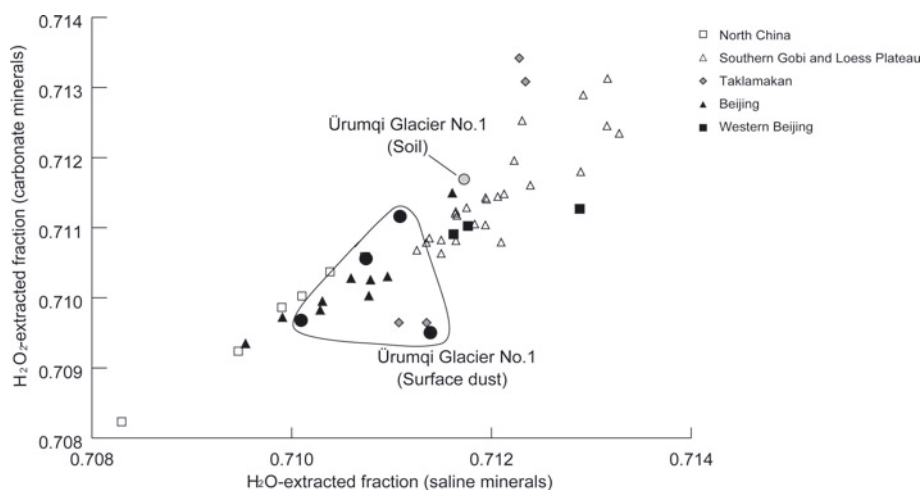


Fig. 10. Comparison of Sr isotope ratios of H₂O- and HOAc-extracted fraction (saline and carbonate minerals) in the surface dust and those in loess and sand reported by Nakano and others (2004).

particles collected during a severe dust storm in Beijing comprised about 90% silicate minerals, 5% carbonate minerals and 1% saline minerals. The low contents of saline and carbonate minerals in surface dust on the glacier are almost identical to that in Asian dust.

The Sr and Nd isotopic ratios of the three fractions of soil differed from those of surface dust, especially that of phosphate minerals (Tables 3 and 4; Fig. 7), although the proportions of saline, carbonate and phosphate minerals in the soil were generally similar (Table 2). The differences in Sr isotopic ratios were more than 0.001479 for saline minerals, 0.001587 for carbonate minerals and 0.019940 for phosphate minerals, which were greater than the SD of Sr isotopic ratios in the surface dust (0.000360, 0.000242 and 0.001276). Moreover, the difference of Nd isotopic ratios in phosphate minerals between the surface dust and soil was >0.000081, which also exceeded the SD of Nd isotopic ratios in surface dust (0.000014). The difference in isotopic ratios between the soil and surface dust indicates that the soil around the glacier is not the only source of saline, carbonate and phosphate minerals.

When the Sr isotopic values of saline and carbonate minerals in the surface dust were plotted with data from various regions in Asia (Nakano and others, 2004), the isotopic ratios of surface dust on the glacier were close to those of north China (Fig. 10). This suggests that the saline and carbonate minerals on the glacier originated from evaporites in the northern Chinese desert. Although sea water might be a source of saline and carbonate minerals, the Sr isotopic ratios in the surface dust on the glacier (0.710372–0.710960) are significantly higher than those in sea water (0.7092).

The Sr isotopic ratios of phosphate minerals analyzed in this study were similar to those of phosphate minerals in sand and loess from the Loess Plateau (Yokoo and others, 2004). The phosphate minerals in the deserts were reported to be apatite, the Sr isotope ratio of which is higher than those of saline and carbonate minerals and lower than those of silicate minerals (Yokoo and others, 2004). This suggests that phosphate minerals on the glacier may also be apatite derived from such deserts.

The Pb isotopic ratios of saline minerals in the surface dust were significantly different from those of silicate

minerals, indicating their disparate geological origins. Compared with various Pb isotopes reported in China, the ²⁰⁶Pb/²⁰⁷Pb and ²⁰⁸Pb/²⁰⁷Pb of saline minerals in the surface dust are similar to anthropogenic aerosol (Fig. 9; Kazakhstan and China: Mukai and others, 2001; China anthropogenic: Duzgoren-Aydin, 2007; Chinese desert sand and loess: Jones and others, 2000). This suggests that the H₂O-extracted fraction contained not only saline minerals but also anthropogenic chemicals dissolved in meltwater on the glacier. The Pb isotopic ratios in saline minerals (H₂O-extracted fraction) are likely to reflect not only those of their mineralogical origin, but also those of anthropogenic material on the glacier.

Isotopic ratios of organic matter in surface dust

The Sr isotopic ratios of the H₂O₂-extracted fraction (organic matter) of surface dust were lower than those of phosphate and silicate minerals, but were almost the same as those of saline and carbonate minerals. This suggests that the saline and carbonate minerals were the source of Sr in organic matter in the surface dust, which may have been wind-blown soil organic matter from the ground surface surrounding the glacier and/or organic matter derived from microbes living on the glacier. Microscopy revealed that organic particles in the surface dust were mainly cryoconite granules that contained abundant filamentous cyanobacteria, which produce organic matter photosynthetically on the glacier surface. Thus, most organic matter in the surface dust seems to be derived from microbes living on the glacier.

Although Sr is not a nutrient source for microbes, it is randomly taken up by microbes along with Ca from the environment, since its chemical characteristics are similar to those of Ca. Therefore, the Sr isotopic ratios in organisms show values similar to the source of Ca entrained within them (Capo and others, 1998; Yokoo and Nakano, 2001). The similar Sr isotope ratio of organic matter to those of saline and carbonate minerals suggests that these minerals constitute a major source of Ca for microbes living on the glacier.

The cryoconite ecosystem is known for being phosphorus limited (Mindl and others, 2007). The wind-blown phosphate minerals may therefore fertilize the ecosystem on the glacier, in which case the isotopic ratios of organic matter would be close to those of phosphate minerals. However,

the Sr isotopic ratios differed between organic matter (H₂O₂-extracted fraction) and phosphate minerals (HCl-extracted fractions) (Table 3; Fig. 4). This suggests that the nutrient sources of microbes are not only phosphate minerals but also other minerals (saline and carbonate minerals) or allochthonous organic matter.

The Pb isotopic ratios of organic matter were close to those of saline minerals in surface dust (Table 5; Fig. 6); Pb may also be entrained by microbes on the glacier. This finding suggests that the Pb in organic matter was originally from saline minerals rather than from silicate minerals.

CONCLUSIONS

Analyses of the Sr, Nd and Pb isotopic ratios of surface dust on the glacier showed significant variations among mineral fractions, indicating that these ratios differ in their mineral components from surface dust. On the other hand, the isotopic ratios and mineral composition showed almost no differences among the study sites, confirming that they were spatially uniform on the glacier.

Although the mineral composition of soil outside the glacier was similar to that of surface dust on the glacier, the Sr isotopic ratio in the soil differed from that in the surface dust. Furthermore, the isotopic ratios and composition of bedrock along the side of the glacier were considerably different from those in surface dust, showing that mineral particles in the surface dust were not derived from those in soil or bedrock around the glacier.

The Sr and Nd isotopic ratios of silicate mineral fractions in the surface dust were close to the values from the Taklamakan Desert, southern Gobi Desert and Loess Plateau. This finding suggests that such deserts may have been provenances of the surface dust. Based on the predominant wind direction in this area, the Taklamakan Desert is most likely to be the main provenance.

The Sr isotopic ratios of saline, carbonate and phosphate minerals in the surface dust were close to those of evaporite and apatite in Chinese desert sand. The Pb isotopic ratios of saline minerals were similar to that of anthropogenic aerosol in China, suggesting that the ratios are also likely to reflect anthropogenic material on the glacier.

The isotopic ratios of organic matter were almost the same as those of saline or carbonate minerals, suggesting that the two minerals may have been entrained by microbes on the glacier.

Our study shows that the Sr, Nd and Pb isotopic ratios of surface dust on an Asian mountain glacier can be used to trace the provenance of mineral particles deposited on the glacier. Furthermore, the isotopic ratios of organic matter might be used to trace the elements in biogeochemical process on the glacier and might help us to better understand the ecology of microbes living there. In this study, since the isotopic ratios in minerals were measured without regard to size, we are unable to identify the contribution from several provenances. As previous studies have shown that the Sr isotopic ratios of silicate minerals vary with their particle size (e.g. Biscaye and others, 1997; Kanayama and others, 2005), analyses of the separated particle sizes of surface dust may enable us to determine its detailed provenance. Furthermore, since the surface is actually a combination of several provenances, in order to reveal the ideal mixing proportion of dust from several origins, a more accurate quantification of Sr and Nd in the samples will also be necessary in a future study.

ACKNOWLEDGEMENTS

We thank the staff and students of the Cold and Arid Regions Environment and Engineering Research Institute of the Chinese Academy of Science in Lanzhou, China, for logistical support during the fieldwork. We also thank Mr Kobayashi and Dr Saitou for their valuable assistance in the isotopic analysis. Finally, we thank two anonymous reviewers for many helpful suggestions which improved the manuscript. This study was financially supported by a Grant-in-Aid for Young Scientists (A, No. 21681003) of the Japan Society for the Promotion of Science (JSPS) and by the Ili Project funded by the Research Institute for Humanity and Nature, Japan.

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