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Distribution of major elements between the dissolved and insoluble fractions in surface snow at Urumqi Glacier No. 1, Eastern Tien Shan

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ABSTRACT

A total of sixty-three surface snow samples collected at Urumqi Glacier No. 1 (UG1), Eastern Tien Shan between March 2006 and December 2007 were analyzed for the concentrations of Na, K, Mg, and Ca in the dissolved and insoluble fractions by ion chromatography (IC) and inductively coupled plasma-mass spectrometry (ICP-MS), respectively. The dissolved and insoluble concentrations of these four major elements showed significantly positive correlation (coefficients 0.7 ~ 1.0, p < 0.0001) with the concentration of insoluble Al (a reference element of the upper continental crust), suggesting that the four elements were mainly derived from crustal materials. The median dissolved fraction percentage (DFP, calculated as dissolved/(dissolved + insoluble) \times 100%) was 67.1% and 35.7% for Ca and Na, respectively, but only 16.8% and 8.9% for Mg and K, respectively. This suggests that Ca and Na were more readily released into the meltwater than were Mg and K from mineral dust in the UG1 snow. The elution sequence through the surface snow was determined to be Ca > Na > Mg > K by the change of DFP. The temporal variability of DFP was attributed to different processes, such as dry/wet deposition in the atmosphere and the post-depositional melting and elution in snow. The DFPs were relatively stable during the dry/cold season (December through March) due to predominantly dry deposition and infrequent elution, whereas they varied significantly during the wet/warm season (April through November) because DFP was modified by wet deposition (which will increase DFP) and the elution process (which will reduce DFP). Our results reveal that DFP can provide a new insight into the understanding and explanation of ice core records, and basic information for evaluating the impact of atmospheric dust on biogeochemical cycles.

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1. Introduction

Airborne particulate matter plays a vital role in climate change by directly affecting the Earth's radiation balance and indirectly through ice cloud–aerosol interactions (e.g., Sokolik and Toon, 1996; Tegen et al., 1996). Atmospheric dust is also a major source of trace elements and nutrients in the remote oceans, playing an important role in global biogeochemical cycles (e.g., Duce and Tindale, 1991; Zhuang et al., 1992; Duvall et al., 2008). Only the dissolved fractions of these nutrient elements, rather than the total amount, control biological availability. Since the solubility of the elements show significant variability (e.g., Fe: ~1%–40%, Fan et al., 2006), it is difficult to assess accurate deposition fluxes of bioavailable nutrients (such as iron) to oceans (e.g., Duce and Tindale, 1991; Jickells et al., 2005).

It has been suggested that the solubility of elements is mainly controlled by the pH of the aqueous phase and the mineralogical composition of the aerosols (Spokes and Jickells, 1996; Desboeufs et al., 1999, 2001; Cizmecioglu and Muezzinoglu, 2008; Okubo et al., 2013). The composition

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establishes the chemistry of the aqueous solution that will define several subsequent atmospheric chemical processes (Desboeufs et al., 1999; Okubo et al., 2013). For example, mineral dust can buffer the pH of precipitation (e.g., Zhang et al., 2003 and references cited therein; Anatolaki and Tsitouridou, 2009), which plays a dominant role in controlling the solubility of trace elements such as Fe, Pb, and Cu in precipitation (e.g., Chester et al., 2000; Vuai and Tokuyama. 2011). Alkaline and alkaline-earth elements (e.g. Na, K, Mg, and Ca) in mineral dust dissolve more rapidly than other metals in dissolution experiments (Desboeufs et al., 1999 and references cited therein). Moreover, the dissolved fractions of these elements are ubiquitous and dominant cations in natural waters (e.g., Drever, 1988; Rastogi and Sarin, 2005), and hence their dissolution might actually affect subsequent dissolution of the nutrient elements (e.g., Drever, 1988; Desboeufs et al., 1999; Cizmecioglu and Muezzinoglu, 2008). Despite the obvious importance of dissolution for Na, K, Mg, and Ca in aeolian dust in biogeochemical cycles, field and experimental works on this subject are scarce. Ground dust is predominantly carried by near-surface winds and thus might differ from dust carried a long distance in the mid- and high-level troposphere. Atmospheric dust in the mid-upper troposphere, which has experienced mixing and gravitational sorting during uplift, and suffered physico-chemical modification during subsequent long-range transport, can influence the global radiation budget by stimulating cirrus cloud formation and marine ecosystems by supplying nutrients to the open ocean (Uno et al., 2009). However, knowledge of the solubility of alkaline and alkaline-earth elements of mineral dust in the upper troposphere, especially that over source regions (which is the basic information for evaluating the impact of atmospheric dust on global biogeochemical cycles), is still unclear at present. In addition, post-depositional distortion (such as melting and elution) of geochemical items is still a serious obstacle to the explanation of ice core records in the mid- and low-latitudes (e.g., Eichler et al., 2001). Using a new method besides the ion ratio (e.g., Eichler et al., 2001; Li et al., 2006) to qualitatively or semiquantitatively describe the elution sequence is therefore of significant importance.

High-altitude glaciers on the Tien Shan (above 4000 m asl) receive dust from the surrounding deserts by wet and dry deposition processes, and provide an opportunity to characterize the physical and geochemical properties of Asian dust in the upper troposphere (Wu et al., 2010). In recent years, the dissolved impurities, including the concentrations of Na⁺, K⁺, Mg²⁺, and Ca²⁺ in aerosol, precipitation, snow, and firn/ice cores, have been well studied at Urumqi Glacier No. 1 (UG1), Eastern Tien Shan (Wake et al., 1992; Sun et al., 1998; Li et al., 2006, 2008; Zhao et al., 2008). The insoluble dust particles were also determined for their elemental composition, mass/number concentrations and size distribution (Wu et al., 2009; Dong et al., 2011). However, no studies were carried out that simultaneously quantified both the dissolved and insoluble fractions of Na, K, Mg, and Ca of dust in snow samples at this locality. In this study, we collected surface snow samples at UG1 between March 2006 and December 2007. Both the dissolved and insoluble concentrations of Na, K, Mg, and Ca in these samples were quantified, and the temporal variations of their distribution between the

two fractions as well as the impacting factors were discussed. The purpose of this study is to provide the basic characteristics of the dissolved and insoluble fractions of the four major elements and to develop a new method to semiquantitatively describe the elution sequence.

2. Field sampling and method

2.1. Field sampling

The sampling site (43°06′ N, 86°49′ E, 4130 m asl) is located in a percolation zone of the east branch of Glacier No. 1 at Urumqi riverhead in eastern Tien Shan, and is surrounded by Taklimakan Desert to the south, Gurbantungut Desert to the north, and Gobi Desert to the east (Fig. 1). The mean annual air temperature and precipitation at the sampling site are about -9.1 °C and 700 mm, respectively. The glacier's annual equilibrium-line altitude has averaged approximately 4055 m asl from 1959 to 2003 (Li et al., 2006). The high-level Westerly jet steam prevails across the high mountains throughout the year (Sun et al., 1998). According to the meteorological records from 1954 to 2001, dust storm events occurred most frequently between April and August in the study regions.

Efforts were made to collect fresh surface snow samples weekly or biweekly with protocols previously described (Li et al., 2006). Acid-cleaned wide-mouth low-density polyethylene (LDPE) Nalgene bottles were used both as sample scoops and containers. During the winter (or dry) season when there was little snowfall, the top 3 cm of the glacier surface was sampled. However, if a snow event occurred prior to sampling, the topmost snow as thin as 1 cm was collected. During the summer (or wet) season, there was usually sufficient fresh snow available, and snow samples not older than 2 days were collected from the top 5 cm. A total of 63 samples were collected from March 2006 to December 2007. After collecting, each LDPE bottle was packed in polyethylene bags and kept frozen during transportation to the laboratory before filtration.

2.2. Analysis method

All the samples were weighed and melted at room temperature before being filtered on LCR hydrophilic PTFE membrane filters (Millipore Corporation) with a diameter of 47 mm and a pore size of 0.45 µm. The filtrations were conducted in a class 1000 clean room at the State Key Laboratory of Cryospheric Sciences, Cold and Arid Regions Environment and Engineering Research Institute (Lanzhou), Chinese Academy of Sciences (CAS). After weighing, the dust particles on each filter were completely digested with super-pure HNO₃ – HF at 150–190 °C in PTFE screw-top jars. Quantitative elemental concentrations of the insoluble fraction (filters) for sodium, potassium, magnesium, calcium, and aluminum were performed by inductively coupled plasma-mass spectrometry (ICP-MS, X - 7, Thermo -Elemental Corporation) with detection limits (given as 3σ of the blank filter) of tens to hundreds pg g⁻¹ for major elements. The laboratory blank filters, which were filtrated using ultra-pure water (18.2 M Ω , Milli – Q Element, Millipore Corporation), were also digested and measured following the same procedures as for the dust samples. The



Fig. 1. Location of Urumqi Glacier No. 1 sampling site, eastern Tien Shan.

digestion procedures and accuracy of ICP-MS measurements have been previously described in detail (Wu et al., 2010). The concentrations of the investigated elements in the blank filters were at least one order of magnitude lower than the lowest concentration sample and have been subtracted from the measured result.

The concentrations of dissolved fractions for the major elements (sodium, potassium, magnesium and calcium) were measured by a Dionex ICS-2000 ion chromatograph as cations, with detection limits for all ions of less than 1 ng g⁻¹. The electrical conductivity (EC) and pH were measured with a digital conductivity meter (DDB-303A, with the theoretical precision 0.01 μ s cm⁻¹) and a pH meter (PHS-3E, with the precision ~0.03 pH unit), respectively. All the analyses were conducted at the Key Laboratory of Tibetan Environment Changes and Land Surface Processes, Institute of Tibetan Plateau Research, Chinese Academy of Sciences (CAS).

2.3. Calculation of dissolved fraction percentage (DFP)

Dust with a diameter <0.4 μ m was thought to be the soluble fraction (Duce and Tindale, 1991; Zhuang et al., 1992). In this study, the 0.45 μ m pore filter was used to separate the dissolved and insoluble fractions. Therefore, the filtrate passing through the 0.45 μ m pore size is the dissolved fraction, while the residue on the filter is the insoluble fraction. The concentrations of the insoluble fraction (dust particles on LCR filters, ng g⁻¹ snow) and the dissolved fraction (filtrate, ng g⁻¹) in snow samples were used to determine the total concentrations for a given element.

The dissolved fraction percentage (DFP) is used to describe the proportion of the dissolved fraction to the total amount for each element in UG1 snow. The DFP of a given element X was calculated by the following equation: DFP

 $(\%) = X_{\text{dis}}/(X_{\text{dis}} + X_{\text{insol}}) \times 100\%$, where X_{dis} and X_{insol} refer to the dissolved and insoluble concentration of a given element X in snow (and hereafter), respectively.

3. Results and discussion

3.1. Chemical characteristics of Na, K, Mg, and Ca in dissolved and insoluble fractions

The concentrations of Na, K, Mg, and Ca in dissolved and insoluble fractions, as well as the pH and EC values, are presented in Fig. 2, and their statistical summaries are listed in Table 1. As shown in Fig. 2, the concentrations of the four dissolved elements (cations) and the values of pH and EC are generally consistent with those of the insoluble fractions and Al (a reference element of upper continental crust) during the two-year sampling period. In order to clearly illustrate the variation in detail, the data sets were further subdivided into two groups according to the sampling date: dust season (April through August) and non-dust season (September through March). The concentration of insoluble mineral dust, as indicated by insoluble Al, frequently had higher levels during the dust season.

The average values of pH and EC were 6.70 and 13.19 μ s cm⁻¹, respectively, which were similar to previous results of 6.66 and 12.74 μ s cm⁻¹ for UG1 snow samples from September 2002 to September 2004 (Li et al., 2007). In UG1 snow samples during November 2002 and October 2005 (Li et al., 2008), Ca²⁺ had the highest concentration, with an average value of 1.50 μ g g⁻¹, comparable (*t*-test, *p* > 0.10) to our results of 1.80 μ g g⁻¹ (0.13 to 9.59 μ g g⁻¹). There were also no significant differences between the previous study (Li et al., 2008) and this study in average concentrations of Na⁺ (0.14 μ g g⁻¹ vs. 0.34 μ g g⁻¹), Mg²⁺ (0.11 μ g g⁻¹



Fig. 2. Temporal variation of the dissolved (black dots) and insoluble (grey dots) concentrations of Na, K, Mg, and Ca in UG1 snow samples, as well as pH and EC (electrical conductivity), together with the air temperature and amount of precipitation (black bars) at Daxigou station. The shaded areas indicate the dust season.

vs. 0.11 μ g g⁻¹), and K⁺ (0.04 μ g g⁻¹ vs. 0.06 μ g g⁻¹). The conformity of our results with previous studies (Li et al., 2007, 2008) suggests that the snow samples from UG1 have been geochemically homogeneous for a long time.

Fig. 3 shows that the concentrations of the insoluble fractions for Na, Mg, K, and Ca were positively ($r^2 = 0.9 \sim 1.0$, p < 0.0001) correlated with dust (insoluble Al) concentrations. The slopes and the intercepts of linear regression equations

Table 1

Summary of the concentrations and dissolved fraction percentage (DFP) of the major elements, together with pH and EC (electrical conductivity) in surface snow from March 2006 to December 2007. Please note that the mean (maximum, minimum) DFP% in this table is the average (maximum, minimum) of all the total 63 samples, but not the ratio of the average (maximum, minimum) dissolved to the average (maximum, minimum) total.

Species	Na			K			Mg			Ca			pН	EC
	dissolved	insoluble	DFP	dissolved	insoluble	DFP	dissolved	insoluble	DFP	dissolved	insoluble	DFP		
	$\mu g \ g^{-1}$	$\mu g \ g^{-1}$	%	$\mu g \ g^{-1}$	$\mu g \; g^{-1}$	%	$\mu g \; g^{-1}$	$\mu g \; g^{-1}$	%	$\mu g \; g^{-1}$	$\mu g \; g^{-1}$	%		$\mu s \ cm^{-1}$
Mean	0.34	0.96	35.7	0.06	1.75	8.9	0.11	1.53	16.8	1.80	1.36	67.1	6.70	13.19
Maximum	6.21	22.93	85.5	0.42	38.87	36.9	0.69	30.27	53.9	9.59	16.28	91.8	7.58	81.50
Minimum	0.01	0.01	0.1	0.01	0.01	0.1	0.02	0.03	0.5	0.13	0.04	3.2	5.53	1.52
Median	0.11	0.35	36.5	0.04	0.63	6.4	0.08	0.57	14.2	1.19	0.54	71.1	6.73	9.75
Std. Dev.	0.87	2.92	20.5	0.07	5.00	8.6	0.11	3.96	11.2	2.01	2.82	18.9	0.41	13.90



Fig. 3. Relationship between soluble, insoluble, and total fraction of major elements (Na, K, Mg, and Ca) and dust loading (using insoluble Al concentration) in surface snow. Seven samples that have experienced obvious elution were excluded.

established in dust and non-dust seasons did not differ significantly (ANCOVA, p > 0.10), suggesting that the insoluble fraction of dust in UG1 snow had a rather uniform composition throughout the two-year sampling period and that these elements might have a very similar source, as shown by the elemental composition of the insoluble dust particles (Wu et al., 2010). The enrichment factors (EF = (X/AI)_{sample}/(X/AI)_{crust}, using Al as the reference element) of these given elements X (Na, K, Mg, and Ca) were close to 1 (less than 1.8) for the insoluble fraction, indicating that they were not significantly distorted by human contribution. Therefore, the insoluble fractions of the major elements were mainly derived from crustal materials.

A strong correlation was also found among the studied elements in soluble and insoluble fractions. The dissolved (as cations) and total concentrations of Na, K, Mg, and Ca showed significantly positive correlation ($r^2 = 0.7 \sim 1.0$, p < 0.0001) with the concentration of insoluble Al (a reference element of the upper continental crust) in Fig. 3, suggesting that the four elements in the dissolved fraction might have common source origins with that of Al. Aeolian dust was a major component of the chemical content in the samples of aerosol, precipitation and snowpack from this study area (Sun et al., 1998; Zhao et al., 2008). The highest ion burden in the snowpack occurred in layers associated with visible debris and high microparticle (insoluble fraction) content, suggesting that the ion concentration was related to the input of desert dust to the Tien Shan region (Wake et al., 1992). The Na⁺, K⁺, and, especially Mg^{2+} and Ca^{2+} , were previously suggested to come from dust (Sun et al., 1998; Li et al., 2006). Sea salts contribute only a small amount of the chemical content in UG1 snow (Williams et al., 1992). Therefore, we can assume that the dissolved fractions of Na, K, Mg, and Ca also originated mainly from crustal materials.

Based on the above analysis, we can conclude that both the dissolved and insoluble fractions of the four major elements (Na, K, Mg, and Ca) in the snow samples originated mainly from crustal materials.

3.2. Dissolved fraction percentage (DFP) of Na, K, Mg, and Ca

The temporal variation profiles of DFP for Na, K, Mg, and Ca are shown in Fig. 4, and the DFP values are summarized in Table 1. The average DFP values of Ca, Na, Mg, and K listed in Table 1 are $67.1\% \pm 18.9\%$, $35.7\% \pm 20.5\%$, $16.8\% \pm 11.2\%$, and $8.9\% \pm 8.6\%$, respectively. It is evident that Ca and Na were more readily released into the meltwater than were Mg and K from the mineral dust in the snow samples. As shown in Fig. 4, the absolute DFP values of each element varied from sample to sample during the two-year sampling period, while the DFP variations for these elements display a similar trend. In order to clearly illustrate the detailed variability, the data sets were further subdivided into two groups according to the meteorological conditions: dry and cold season (December through March) and wet season (April though November). During the dry and cold season (see Fig. 4), the DFP values of these major elements remained relatively stable. For example, the standard deviation of DFPs for Na, K, Mg, and Ca were 15.2%, 2.1%, 6.1%, and 9.3%, respectively.



Fig. 4. Temporal variation of dissolved fraction percentage (DFP) of Na, K, Mg, and Ca in UG1 snow samples, daily air temperature and amount of precipitation (black bars) at Daxigou station. The shaded areas indicate the wet season. The arrows and dashed lines indicate snow samples with peak and anomalously low DFP values of the major elements, respectively.

During the wet and warm season (shaded area in Fig. 4), the DFP values showed significant variability, with the standard deviation of 21.4%, 9.1%, 11.8%, and 20.1% for Na, K, Mg, and Ca, respectively. The peak DFPs (indicated by the arrows) were often consistent with precipitation events, while the anomalously low ones (indicated by dashed lines) often occurred during days characterized by high temperatures.

During the cold and dry season (December through March), the dust in UG1 snow was mainly deposited by dry deposition processes because there was little precipitation at the study area (see Fig. 4). Therefore, possible dissolution of mineral dust by chemical actions during precipitation and scavenging seldom occurs. In addition, the air temperature was much lower than freezing point of 0 °C at UG1, and thus snowmelt events seldom occurred. Post-depositional processes, such as sublimation, blowing and floating snow, were also found to slightly alter the chemistry of the snow during these periods (Wang et al., 2006). Therefore, the DFP of Na, K, Mg, and Ca in snow samples collected during the dry season could actually reveal the initial dissolution of these elements from dust in snow-meltwater over the UG1 area. Here we define the DFP during this dry season as the typical unmelted DFP, with average values for Ca, Na, Mg, and K being 71.8%, 32.2%, 13.0%, and 4.9%, respectively.

During the wet season (April through November) with frequent precipitation events, thick clouds often occurred around the study region. During dust transport in the atmosphere, aerosols typically undergo around 10 condensation/evaporation cycles (Pruppacher and Jaenicke, 1995), which can dissolve part of dust particles since the pH in the cloud is low (Spokes and Jickells, 1996) and then subsequently contribute to the dissolved fraction in surface snow samples. Additionally, precipitation was an effective scavenging process for the dissolved components in the UG1 area (Li et al., 2008; Zhao et al., 2008). This could partly contribute DFP peaks that often occurred with precipitation events, as shown by the arrows in Fig. 4. However, several dry days could still occur between precipitation events, and dust particles from surrounding deserts and dry saline lakes could be blown into the atmosphere by strong winds. Dry deposition might be also an important process depositing atmospheric dust onto the glacier under such dusty atmospheric conditions during the warm/wet season (Kulshrestha et al., 2009).

Under elevated air temperature, snowmelt events occasionally occurred at the glacier surface during the warm and wet season (Li et al., 2006). Meltwater can leach the dissolved matter downwards and may drastically reduce its concentration in surface snow. Elution processes are inevitable obstacles to the reconstruction of paleoclimate information preserved in ice cores recovered from temperate glaciers at mid-low latitude mountains (e.g., Eichler et al., 2001). The influence of elution on the geochemical composition of natural snowpacks has been addressed in several field and laboratory studies (e.g., lizuka et al., 2002). These pioneering studies revealed that the major ions had preferential elution sequences in snow samples by comparing the concentration ratios between different ions to the reference one (e.g., Li et al., 2006). However, the reference ion itself was also influenced by elution. Therefore, it is still an open question to quantitatively or semi-quantitatively characterize the degree of elution. Insoluble particles, in contrast, can accumulate at the snow surface instead of moving down with the meltwater. The proportion of the dissolved fraction to the total content of each element in the surface snow can decrease considerably due to the elution process, and thus the DFP of the elements might provide a new index to semi-quantitatively determine the degree of elution in the snowpacks.

The elution process was mainly driven by air temperature (Li et al., 2006). Comparisons between the DFP values of the four elements (Na, K, Mg, and Ca) and the corresponding daily air temperature from the nearby Daxigou meteorological station (43°06'N, 86°50'E, 3539 m asl, 3.5 km east of the UG1 sampling site) are shown in Fig. 5. The DFP values of the four elements are generally within one standard deviation of the average value (lower and upper dashed lines in Fig. 5). However, a small number of samples have anomalously low DFP (under the lower dashed lines) and mostly correspond to temperatures higher than 6.5 °C (above 2.9 °C at the sampling site calculated by the lapse rate of 0.60 °C/100 m, which was much higher than the elution starting point of -3.6 °C (Li et al., 2006)). Therefore, it is likely that these anomalously low DFP samples were modified by the elution process. These samples are also indicated by the dashed lines in Fig. 4.

To describe the degree of elution for these dissolved elements, the DFP during the period 26 July to 10 August 2006 was defined as the typical melted DFP, with average values for Ca, Na, Mg, and K of 16.8%, 1.1%, 2.9%, and 0.4%, respectively. The DFP differences between the typical melted and un-melted periods (as discussed above) were 55.0%, 31.1%, 10.1%, and 4.5%, for Ca, Na, Mg and K, respectively, as listed in Table 2. Calcium displayed the most pronounced decrease in DFP, followed by Na, Mg, and K in that order. This indicates that

Ca was preferentially eluted compared to the others. Potassium showed a very small variability between the two typical periods, and was the most immobile of the four species. The elution sequence of the dissolved elements calculated by the DFP was almost the same as that obtained by comparing the concentration ratios between different cations (Ca²⁺ > Na⁺ > K⁺ > Mg²⁺, using Mg²⁺ as the reference species) (Li et al., 2006) in UG1 snow. Indeed, in the cation elution sequence, the difference in the elution trend between K⁺ and Mg²⁺ was very small. Therefore, we can conclude that there is no substantial difference between the cation elution sequence and our DFP order, and that the temporal variation of DFP could indicate post-depositional processes.

3.3. The factors influencing dissolution of Na, K, Mg, and Ca

The dissolution of minerals is usually a consequence of reactions occurring at their surfaces. Several factors might influence the dissolution. The composition of minerals, the pH of the aqueous phase and the dust loading were recognized as the major factors (Spokes and Jickells, 1996; Desboeufs et al., 2001; Baker and Jickells, 2006). Dust loading was represented by Al concentration. Previous studies indicated that the dust particle gain size increased with dust concentration (Wu et al., 2009). The surface area to volume ratio (A/V = 3/r with assumption of spherical particles, where *r* is the particle radius) of dust particles,



Fig. 5. Comparison of DFP values of Na, K, Mg, and Ca with air temperature from Daxigou station. The horizontal solid and dashed lines indicate the mean DFP values of the species and one standard deviation (σ). The vertical solid lines denote the air temperature value of 6.5 °C.

Table 2

Comparison of dissolved fraction percentage (DFP) of the major elements between typical samples.

	Na	K	Mg	Ca
	%	%	%	%
Un-melted ^a	32.2	4.9	13.0	71.8
Melted ^b	1.1	0.4	2.9	16.8
Difference ^c	32.1	4.5	10.1	55.0

^a Indicates the average DFP of samples collected during dry seasons.

^b Indicates the average DFP of samples between 26 July and 10 August 2006.

^c Indicates the difference in DFP between the typical melted periods and un-melted periods.

decreased with dust concentration. The higher A/V ratio is though to be a factor of increased DFP of dust (Baker and Jickells, 2006).

The pH values and dust loading of snow samples were compared with the DFP values of Na, K, Mg, and Ca in Figs. 6 and 7, respectively. To avoid (at least partly) the influence of the elution process on the DFP, the thirteen samples collected during high temperature (higher than 6.5 °C) and with extremely high Al concentrations (greater than 4.5 μ g g⁻¹) were excluded in Figs. 6 and 7.

Previous observation indicates that Ca-rich aerosol particles over the UG1 area are mostly composed of calcite, gypsum and their mixture (Li et al., 2011), which are readily dissolved in natural waters (Stumm and Wollast, 1990). This might explain that why the DFP of Ca shows no relationship with pH or dust loading.

The DFP of Na showed no correlation with pH but a weak correlation ($r^2 = 0.15$, p < 0.001) with dust loading, indicating that the dissolution of Na was not significantly dependent on the pH of meltwater (5.5 ~ 7.6) but was partly impacted by the surface character of dust particles in the meltwater. It could be expected that the Na-containing aeolian dust particles from Asian desert regions were mostly composed of evaporite minerals (halite and thenardite) (Okada and Kai, 2004; Yokoo et al., 2004) and feldspar (Li et al., 2011). Additionally, halite and thenardite are readily and rapidly dissolved, yielding upon dissolution the same stoichiometric proportions in solution as the proportions in the dissolving mineral without forming new solid phases (Stumm and Wollast, 1990), while dissolution of feldspar is a fundamental surface process.

The DFP of Mg and K showed significantly negative correlation with pH ($r^2 = 0.48$, p < 0.0001; $r^2 = 0.28$, p < 0.0001; respectively) and dust loading ($r^2 = 0.80$,



Fig. 6. Correlations of DFP versus pH of surface snow for Na, K, Mg, and Ca. Thirteen that collected during temperature higher than 6.5 °C and extremely high Al concentration (greater than 4.5 μ g g⁻¹) were excluded.



Fig. 7. Correlations of DFP versus dust (insoluble Al) concentration for Na, K, Mg, and Ca. Thirteen that collected during temperature higher than 6.5 $^{\circ}$ C and extremely high Al concentration (greater than 4.5 μ g g⁻¹) were excluded.

p < 0.0001; $r^2 = 0.43$, p < 0.02; respectively), suggesting that the dissolution of Mg and K was likely affected by the pH of meltwater and the A/V ratio of dust particles. When dust loading increased, the pH of meltwater increased due to the input of soluble Na-rich and Ca-rich minerals from the arid and semi-arid regions, and A/V ratio of dust particles decreased since the dust particle gain size increased with dust concentration (Baker and Jickells, 2006; Wu et al., 2010). Preferential re-adsorption of dissolved K and Mg onto the solid particles increased (e.g., Spokes and Jickells, 1996) in the samples. This could be confirmed by previous results which showed that K and Mg in aerosol particles over Asian desert regions were mainly present as feldspar and chlorite (Yokoo et al., 2004; Li et al., 2011), whose dissolution was mainly the surface-processes.

The mechanism of dust dissolution is complex and beyond the scope of this paper, and our explanation for the factors that influence the DFP of those elements is tentative.

4. Conclusions

This paper provides the dissolved and insoluble concentrations of the elements Na, K, Mg, and Ca in surface snow collected from March 2006 to December 2007 at Urumqi Glacier No. 1 (UG1), Eastern Tien Shan. Based on the above discussions, the following conclusions can be derived:

- (1) The dissolved and insoluble fractions of Na, K, Mg, and Ca in the snow samples originated mainly from crustal materials.
- (2) The average dissolved fraction percentage (DFP) was 67.1%, 35.7%, 16.8%, and 8.9% for Ca, Na, Mg, and K, respectively, suggesting that Ca and Na were more readily released into the meltwater from the mineral dust in the UG1 snow than Mg and K. The elution sequence through the surface snow was determined to be Ca > Na > Mg > K.
- (3) The temporal variability of DFP for each element was influenced by dry deposition, wet deposition and post-depositional elution processes in the study area.
- (4) The solubility of these major elements in aeolian mineral dust over the UG1 area was controlled mainly by the composition of minerals, the pH of the aqueous phase and the dust loading.

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