



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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ARTICLE INFO

Article history:

Received 15 November 2011

Received in revised form 17 May 2013

Accepted 17 May 2013

Keywords:

Surface snow

Tien Shan

Major elements

Dissolved fraction percentage (DFP)

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) × 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 semi-quantitatively 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^+ , Mg^{2+} , and Ca^{2+} 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 semi-quantitatively describe the elution sequence.

2. Field sampling and method

2.1. Field sampling

The sampling site ($43^{\circ}06' \text{ N}$, $86^{\circ}49' \text{ E}$, 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 stream 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 $0.45 \mu\text{m}$. 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 $\text{HNO}_3 - \text{HF}$ at $150-190^{\circ} \text{ C}$ 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 ($18.2 \text{ M}\Omega$, Milli – Q Element, Millipore Corporation), were also digested and measured following the same procedures as for the dust samples. The

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