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Lead isotopic composition of insoluble particles from widespread mountain glaciers in western China: Natural *vs.* anthropogenic sources

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HIGHLIGHTS

- Snowpit samples were collected in mountain glaciers of Western China.
- Lead (Pb) isotopic compositions in insoluble particle of snow samples show geographic trend.
- The anthropogenic lead contributions in snow samples are observed and assessed.

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ABSTRACT

Stable lead (Pb) isotopic fingerprints provide opportunities to trace natural and anthropogenic Pb sources in the environment. In order to evaluate Pb deposition from different sources over mountainous areas of western China, Pb isotopic compositions were characterized from modern aeolian dust in 15 snowpit samples collected from 13 typical mountain glaciers between 2008 and 2010. Most of the snowpits sampled cover more than a whole year of accumulation and overlap with each other on deposition date. Pb isotopic variability among all the samples is small, varying in the range of 18.1399–18.9199 for $^{206}Pb/^{204}Pb$, 15.5979–15.8743 for $^{207}Pb/^{204}Pb$, 38.2272–39.9453 for $^{208}Pb/^{204}Pb$, 1.1605–1.2009 for $^{206}Pb/^{207}Pb$ and 2.4433–2.5182 for $^{208}Pb/^{207}Pb$. Three isotopic plots of the different Pb isotope ratios ($^{207}Pb/^{204}Pb$ *vs.* $^{206}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$ *vs.* $^{206}Pb/^{204}Pb$, and $^{208}Pb/^{207}Pb$ *vs.* $^{206}Pb/^{207}Pb$) in all the samples show identical geographic trends, with more radiogenic in the south and less in the north. This trend is consistent with the distribution of natural dust sources and supports the interpretation of a regional/local source for insoluble particles (IP) to snow/glaciers in this region. Comparison with the Pb isotope results from potential dust sources, however, it shows that the Pb isotopic compositions of IP samples in snow samples are relatively less radiogenic. Parts of these less-radiogenic Pb isotopes are comparable with the ice core results during recent decades, which are shown to be influenced by anthropogenic sources. At sites located along the periphery of western China, the Pb isotopic compositions are much closer to anthropogenic results. Natural and anthropogenic Pb sources are roughly assessed using a simple binary model. The sites with a high anthropogenic fraction are at lower elevations and are relatively close to population centers.

1. Introduction

Western China defined herein as the area encompassing the Qinghai-Xizang (Tibet) Plateau (QXP) and northwestern China, covers more than 4,500,000 km^2, almost half of China's land area. It is a region of widespread mountains and arid/semi-arid areas. The QXP, a vast highland surrounded by the earth's highest mountains including the Himalayas, Pamir, and Kunlun Shan, and sometimes called earth's "third pole", has an area of around 2,500,000 km^2 with an average elevation exceeding 4,500 m a.s.l. The region of northwestern China lies to the north of the QXP, and consists of two large deserts, the Taklimakan Desert and the Gurbantunggut Desert, which are separated by the Tienshan Mountains. The huge highland and mountainous areas of the QXP exert a profound

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influence on regional and global atmospheric circulation through thermal and dynamic effects (Wu and Zhang, 1998). The arid and semi-arid areas in this region are the second largest dust source on the planet, producing a large amount of dust and having a profound effect on the climate and environment. These effects include radiation forcing (Huang et al., 2006), the supply of important biological nutrients to lakes and remote seas (Okin et al., 2004), uptake of anthropogenic aerosols (Huang et al., 2010), and decreasing the albedo of snow and glaciers in this region (Fujita, 2007).

The snow and glaciers in the mountainous parts of this region are an important water source to downstream rivers such as the Yaze River, Yellow River, Indus, Ganges, and Brahmaputra, which are important water supplies for people residing in and around this area. Recently, the glaciers in the mountains of western China have undergone noticeable shrinkage (Yao et al., 2012), and the impurities present in the snow and glaciers, mainly composed of dust and anthropogenic particles, have been thought to be an important factor in accelerating glacier retreat (Xu et al., 2009a). The concentration of impurities in snow and glaciers of western China has a significant spatial variability, with a high concentration in northern regions and a low concentration in southern regions (Xu et al., in preparation). The concentration and geochemical characteristics of particles in snow and glaciers have been used to identify dust source areas and transport routes in central Asia (Wu et al., 2010; Xu et al., 2009b, 2012). In addition, the elevated levels of anthropogenic aerosols in mountain glaciers in recent years have led to concern about the role of dust uptake and the influence of anthropogenic aerosols across this wide area (Xu et al., 2009a). Although several observation stations have been built on and around the QXP (Bonasoni et al., 2008; Cong et al., 2007; Kivekäs et al., 2009), the anthropogenic aerosol source and its influence on this region remain poorly understood.

Insoluble particles (IP) in snow and glaciers are a direct indicator of ambient aerosols, and the Pb isotopic signatures of IP have been used to trace potential dust sources in the QXP (Lee et al., 2011; Xu et al., 2009b). An increased Pb concentration after the 1950s was observed in an ice core from Muztagata mountain (Li et al., 2006) and is suspected to appear in most glaciers in western China. Pb isotopic compositions in a Himalayan ice core were found to be primarily derived from natural sources until the mid-20th century (Lee et al., 2011). Pb concentrations in a Mt. Altai ice core remained low during the period from 1680 to 1935, but strongly increased after 1935 (Eichler et al., 2012). These combined results indicate an evident anthropogenic source influencing this region. In contrast to Pb concentration, the Pb isotopic signature is unaffected by physical or physicochemical processes associated with transport, deposition, and loss in the environment. This feature makes it an ideal tool for tracing sources of atmospheric pollution. For a mixed aerosol comprising natural and anthropogenic sources, the isotopic composition of Pb in the aerosol reflects the relative mixing of these sources. Source apportionment may be quantified if all potential sources of Pb are characterized and have specific defined ratios. Pb isotopic studies have therefore proved a convenient approach to studying and tracing the sources of Pb pollution in various environmental reservoirs (Komárek et al., 2008).

In this study, 15 snowpit samples were collected from widespread mountain glaciers in western China during field expeditions from 2008 to 2010. Insoluble particles were enriched and analyzed for Pb isotopes. The goal of this work was to determine Pb isotopic compositions and evaluate the relative effects of natural and anthropogenic sources of Pb in the mountainous areas of western China.

2. Sampling and methods

2.1. Sampling site description and snowpit collection

Between September 2008 and November 2010, 15 snowpit samples (Fig. 1) were collected from 13 typical mountain glaciers in western China. Details including location, sampling date, elevation, snowpit depth, and time periods covered by snowpit are presented in Table 1. All the containers and collecting tools were

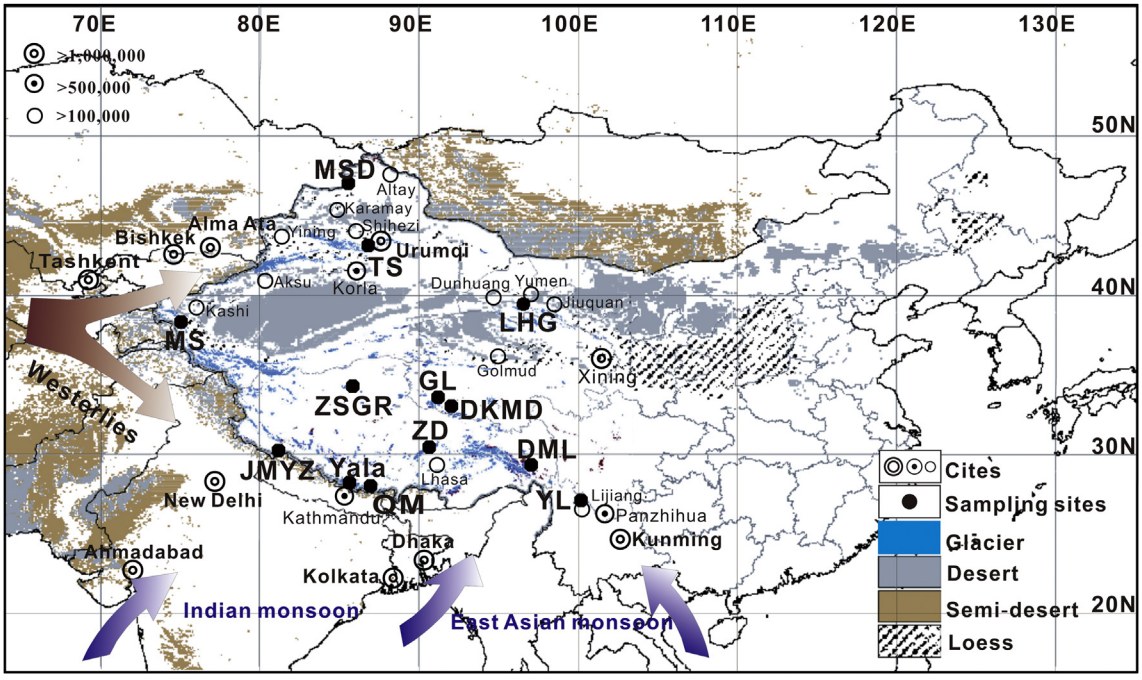

Fig. 1. Location map of snowpit sampling sites in western China and its vicinity. Black dots denote the locations of different sampling sites, whereas black hollow circles with different size mark up typical cities in and beyond the western China according to its urban population. The patterns of major wind circulation over the western China, which is dominated by the Indian and East Asian summer monsoon in summer, and westerly circulation during winter are also presented.