



Lithium distribution and isotopic fractionation during chemical weathering and soil formation in a loess profile



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ABSTRACT

Lithium (Li) is a fluid-mobile element and $\delta^7\text{Li}$ in secondary deposits represents an excellent proxy for silicate weathering and authigenic mineral formation. The soil samples from 1205 to 1295 cm in the Weinan profile, one of the best developed loess-paleosol sequences covering the last glacial-interglacial climatic cycle, were collected and chemically separated into detritus and carbonate fractions for subsequent analyses of Li, $\delta^7\text{Li}$, major and trace elements. Other desert specimens (i.e., Qaidam Desert, Tengger Desert, Badain Juran Desert and Taklimakan Desert) near the Chinese Loess Plateau (CLP) and various standard clays were analyzed for assisting provenance determination. The Li and $\delta^7\text{Li}$ distributions in the detritus are rather homogeneous, 1.4–2.0 $\mu\text{g/g}$ and +2.5‰ to +4.7‰, respectively, compared with the carbonate fraction. The detrital $\delta^7\text{Li}$ varies systematically with magnetic susceptibility and grain size changes, reflecting significant Li isotopic variation associated with sources and mineralogy of detrital material. On the other hand, Li and $\delta^7\text{Li}$ in carbonates show large changes, 781–963 ng/g and –4.1‰ to +10.2‰, respectively. These carbonate $\delta^7\text{Li}$ correlated well with the estimated index of chemical weathering, as a result of Li mobilization and soil formation during chemical weathering.

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1. Introduction

The Chinese Loess Plateau (CLP) is one of the largest eolian dusts suppliers in the world. During the winter monsoon season, fine dusts from major deserts in northwest China were transported and re-deposited in southeast China. These deposits were further transformed by pedogenesis under various local climatic and ecological conditions. The chemical compositions of these loess/paleosols were affected mainly by: (i) Weathering intensity in the source areas; (ii) Pedogenesis intensity after re-deposition; (iii) Geological and ecological conditions in the re-deposition areas; and (iv) Climatic changes in both the original and re-depositional areas (Jia et al., 2007). In general, the loess/paleosol layer represents a sensitive archive for recording the weathering and erosions condition in the upper continental crust. The physical/chemical properties and isotopic compositions of these loess/paleosol layers can, therefore, provide critical information on provincial sources, and environmental and climatic variation in the deposition regions. The Chinese loess deposits are mainly distributed in arid and

semi-arid regions, an East–West trending belt between 33–40°N and 101–115°E. It is characterized by its great thickness and good continuity. Loess deposits in CLP were transported by wind from the inland deserts in northern and northwestern China (Liu and Ding, 1998). These sequences typically contain alternating loess (L) and paleosols (S) layers. The L units are characterized by a yellow color, massive structure, and high carbonate contents, whereas the S layers exhibit a brownish or reddish color and pedogenic features of high organic matter, clay and/or Fe–Mn coatings, and carbonate pseudomycellae and/or nodules (Guo et al., 1996). The loess beds are considered to have been deposited during arid glacial periods. In contrast, paleosols are developed under relatively humid climatic conditions.

Li isotopic geochemistry is a burgeoning field of research and has been developed rapidly in recent years due to advances in mass spectrometric techniques (Tang et al., 2007). Li has two natural stable isotopes, ^6Li and ^7Li , with relative abundances of 7.5% and 92.5%, respectively. Owing to its large relative mass difference (~15%) between the two isotopes, significant Li isotope fractionations (>60‰) were reported in terrestrial systems and have provided important information on crust–mantle cycling (Tomascak et al., 1999; Zack et al., 2003), oceanic crust alteration (Chan et al., 1994) and continental weathering (Huh et al., 2001). The Li

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isotopic system has several advantages as a tracer of chemical weathering. Being a fluid-mobile element, Li is widely distributed in the Earth's crust. Similar to other alkali metals, Li only has a mono-valent state and its isotopic distribution is not affected by redox reaction (Rudnick et al., 2004). Moreover Li is not a nutrient element and its isotopic composition is probably not influenced by biological processes (Rudnick et al., 2004). As a result, Li has served as a proxy for silicate weathering (Huh et al., 1998; Pistiner and Henderson, 2003). During chemical weathering, ^7Li is preferentially released from solid phases and ^6Li is retained in the alteration products, such as clays and soils (Kisakürek et al., 2004). These characteristics make Li isotopes useful tracers for near surface fluid/rock interactions. Pistiner and Henderson (2003) highlighted that significant isotopic fractionation occurred during Li incorporation (via stronger bonds, chemical adsorption) into minerals where the degree of fractionation was highly mineralogically dependent. Various minerals have displayed different Li isotopic fractionation factors where heterogeneous weathering has produced significant isotope shifts in the dissolved load (Huh, 2004; Pistiner and Henderson, 2003). On the other hand, physical adsorption is likely to have little isotopic fractionation (Pistiner and Henderson, 2003). Li surface sorption involved chemical bond formation between the adsorbate and adsorbent can cause significant isotopic fractionation where light Li isotopes were preferentially taken. Similarly, secondary mineral formation during continental weathering may significantly fractionate Li isotopic ratios (Pistiner and Henderson, 2003).

A fraction of minerals were dissolved by alteration reagents during weathering processes, such as river water or groundwater. At the sediment/water interfaces, chemical properties may greatly change, from fresh to rather high ionic strength. Clay formation resulted from the incongruent dissolution of silicate during weathering, where the predominant effect is the dissolution of various minerals. The chemical compositions of primary clay are closely regulated by ambient geological environments in which the temperature and solution chemistry are two key factors. The rock types and water/rock ratios also play an important role in determining the clay composition. Significant Li isotopic fractionation has been documented, while secondary minerals or clays formed at various temperatures (Chan et al., 1994; Williams and Hervig, 2005; Zhang et al., 1998). These reported materials are mainly mixed clays or sediments. To evaluate whether $\delta^7\text{Li}$ in secondary minerals can be used to archive chemical weathering processes, it is necessary to determine the degree of Li isotope fractionation linked with the formation of single phase or specific clays (Vigier et al., 2008). Various crystalline clays with Li sites available were reported to influence $\delta^{11}\text{B}$ in clayey sediments (You et al., 1995).

Li was released from primary silicate minerals and then incorporated into secondary clays during pedogenesis. It is not always enriched in clays. The highest Li enrichments can be linked with Mg substitution in minerals, indicating the role of Mg-bearing smectites and chlorites in the Li geochemical cycle (Stoffynegli and Mackenzie, 1984). There are two different sites in clay structures: interlayer and octahedral sites. Most exchangeable Li were adsorbed at the interlayer sites; however only Mg^{2+} substitution in the octahedral sites will result in significant isotopic fractionation. The latter emphasizes the importance of secondary minerals and Li/Mg substitution. Li is highly enriched in silicates and depleted in carbonates; therefore $\delta^7\text{Li}$ can serve as a potential proxy for silicate weathering intensity.

This study aims to establish a low blank and high-precision $\delta^7\text{Li}$ technique for investigating Li and Li isotope distribution in loess specimens. We have analyzed major/trace and $\delta^7\text{Li}$ in both detritus and carbonate fractions of specimens collected from Weinan, China. These results were used to delineate (i) the dust sources, (ii) the dust transport pathways, and (iii) the climatic environments in

southern CLP during the glacial/interglacial transition period. Five desert specimens near the CLP source region and 10 clay standards were measured for a better understanding of Li and $\delta^7\text{Li}$ in clays, as well as the associated Li isotopic fractionation factor.

2. Sampling location and site description

2.1. Sample location

The loess/paleosol profile at Weinan (34.21°N, 109.31°E) is located in southern CLP (Fig. 1b). It lies within the southeast monsoonal zone in East Asia, with semi-humid climatic conditions. The annual mean temperature is about 12.0–13.6 °C. The average annual precipitation is 570–620 mm, and mainly occurs in the summer season. This region is characterized by a warm-humid summer and a cold-dry winter season (Wen et al., 1997).

The Weinan section contains a sequence of 33 loess-paleosol levels with a total thickness of ~174 m. This is one of the best loess-paleosol sections, which has a good outcrop and well preserved paleo-environmental information from layers S5 to S0. The S0 layer represents the Holocene soil, L1 is the last glacial loess, S1 is the last interglacial soil, the penultimate glacial loess is named the L2 layer, and the rest may be deduced by analogy. The units are numbered in the order of increasing age. This loess-paleosol sequence represents a well-developed and continuous paleo-climatic record covering the last glacial-interglacial cycle. The locations of the Weinan profile and the major deserts near CLP are shown in Fig. 1a, where a detailed stratigraphy study was conducted Kukla and An (1989), and the Weinan section belongs to a clayed loess zone (Fig. 1b, Vandenberghe et al., 1997; Yokoo et al., 2004).

2.2. Source and mineralogy of loess

The origin of the Chinese loess was first reported by Richthofen (1882). Sun (2002) suggested that the dusts derived from the two inland basins of the Qaidam Desert and Taklimakan Desert were not the main source of CLP. The stated reasons were (i) the Qaidam Basin is surrounded by high mountains, and dust entrained from this basin is impeded by the downwind extensive high mountains. Field investigations also pointed out that dust was mainly accumulated in the southeastern piedmont areas of the basin. (ii) Areas to the north, west, and south of the Taklimakan Desert are surrounded by high mountains; an open area exists only in the east margin. The East Asia monsoon systems and the westerlies have long been regarded as the dominant wind regimes for loess transportation in Asia (An et al., 1991; Liu, 1985). Nevertheless, dust in the Taklimakan Desert cannot move out of the desert under the easterly winds. For the loess in CLP there are many issues that remain controversial, including its origin, transport pathway and source area.

Owing to the various stages of formation, transportation, deposition and diagenesis of loess materials, clay minerals have mostly altered their chemical compositions and crystal structures. The clays in loess and paleosols are mainly composed of illite, kaolinite, montmorillonite, chlorite and vermiculite, where the latter is more abundant in paleosols (Liu, 1988). These mineralogy of clays reflect information regarding the temperature and humidity in the past (Williams and Hervig, 2005). Changes in environmental conditions lead to the migration and deposition of mobile elements in the clays, which incurs transformation of the crystal structures. Williams and Hervig (2005) suggested that the variable abundances of Li during the diagenetic reaction printed particular isotopic signatures in the diagenetic clays, which allow the original sources to be traced.

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