



Behavior of lithium isotopes in the Changjiang River system: Sources effects and response to weathering and erosion

Qi-Lian Wang^a, Benjamin Chetelat^{a,*}, Zhi-Qi Zhao^{a,*}, Hu Ding^a, Si-Liang Li^a,
Bao-Li Wang^a, Jun Li^b, Xiao-Long Liu^b

^a State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, Guizhou 550002, China

^b Tianjin Key Laboratory of Water Resources and Environment, Tianjin Normal University, Tianjin 300387, China

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Abstract

Lithium (Li) concentrations and Li isotopic compositions of the main channel of the Changjiang River and its main tributaries were measured to better understand the geochemical behavior of Li and its isotopic fractionation during weathering and erosion. The Li concentrations of the dissolved loads of the main channel range from 475 to 4570 nmol/l and decrease from the upper reaches to the lower reaches; values are clearly higher than those reported for many of the world's largest rivers. Samples with very high Li concentrations are affected by the dissolution of evaporites in the headwater region. The $\delta^7\text{Li}$ values of the particulate material has a narrow range (-4.7‰ to $+0.7\text{‰}$), whereas the dissolved load has very variable $\delta^7\text{Li}$ ($+7.6\text{‰}$ to $+42.1\text{‰}$), with values increasing from the upper reaches to the lower reaches. After correction for contributions other than silicate weathering, we show that the Li isotopic compositions of the dissolved load reflect the balance between dissolution of primary minerals and preferential incorporation of the light isotope into secondary products of erosion which enrich the solution in ^7Li . The evolution of the isotopic composition from the Upper Reaches to the Lower Reaches is mainly controlled by the fraction of Li incorporated/adsorbed in/onto secondary minerals, and in fine by probably the difference in weathering regimes (i.e. kinetic limited vs transport limited). We also show that the magnitude of the apparent isotopic fractionation between the bedrock and dissolved Li for example are not always only due to variation of the isotopic fractionation factor but might also be due to composition (Li content of the bedrock) and mineralogical controls.

The composition of the eroded silicate crust estimated by a steady state mass balance reveals the important contribution of sedimentary rocks (shales) and highlights the effects of sedimentary recycling on the composition of the continental crust.

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1. INTRODUCTION

Silicate weathering of the Earth's surface is a crucial process influencing the transportation of elements through geological reservoirs, controlling landform evolution and soil formation, providing nutrients for the sus-

tainable development of ecosystems. In addition, it has the potential to regulate the Earth's climate on geologic time scales through its effect on atmospheric CO_2 levels (Berner et al., 1983; Kump et al., 2000; Berner and Kothavala, 2001). Investigations of the dissolved and suspended loads of rivers can provide important information on chemical and physical weathering processes at the basin scale (Gaillardet et al., 1999a; Dalai et al., 2002; Wu et al., 2005; Chetelat et al., 2008, and references therein).

* Corresponding authors. Fax: +86 851 5891609.

E-mail addresses: benjamin@vip.skleg.cn, b.chetelat@yahoo.com (B. Chetelat), zhaozhiqi@vip.skleg.cn (Z.-Q. Zhao).

Several geochemical proxies have been used to study silicate weathering, such as the strontium and osmium isotopic ratios (Krishnaswami et al., 1992; Raymo and Ruddiman, 1992; Derry and France-Lanord, 1996). The recently developed non-traditional isotope (e.g. B, Si, Mg and Ca) method has been proved to have the potential for studying silicate weathering (Rose et al., 2000; Georg et al., 2006; Tipper et al., 2006; Chetelat et al., 2009). Lithium isotopes are one of the most promising proxies for studies of continental silicate weathering (Huh et al., 1998, 2001, 2004; Kısakürek et al., 2004, 2005; Pistiner and Henderson, 2003; Rudnick et al., 2004; Hathorne and James, 2006; Vigier et al., 2009; Lemarchand et al., 2010; Millot et al., 2010a; Wimpenny et al., 2010; Yoon, 2010; Tipper et al., 2012), but further studies are required to understand the Li isotopes behaviour during continental weathering.

Lithium has two stable isotopes (^6Li and ^7Li) with a relative mass difference of $\sim 16\%$. The Li isotopic composition of the river dissolved load is mainly controlled by fractionation during weathering of silicate minerals: ^6Li is retained in the solid phase, while ^7Li preferentially goes into the solution (Huh et al., 1998, 2001). The degree of fractionation between solid and liquid phases is dependent on the nature of the solid in/on which Li is incorporated/adsorbed and the weathering intensity while the influence of bed-rock composition is minor (Huh et al., 1998, 2001; Vigier et al., 2002; Kısakürek et al., 2005; Pogge von Strandmann et al., 2006; Millot et al., 2010a). These results were then used to investigate the temporal variation of past silicate weathering rates and intensity as well as the influence of continental weathering on the oceanic chemical composition (Hathorne and James, 2006; Misra and Froelich, 2012; Paytan, 2012). However, up to now our understanding of Li isotope behavior in large river systems is limited. For example, why does the dissolved Li isotopic composition reflects the weathering rates in Icelandic rivers (Vigier et al., 2009), but not in the Mackenzie rivers (Millot et al., 2010a)? Is that the dissolved Li isotopic composition always negatively correlated to the chemical weathering rates in all drainage basins? What is the contribution of evaporite to the dissolved lithium of large rivers (Yoon, 2010; Liu et al., 2011)? Thus, there are many uncertainties on Li sources and fractionation processes during erosion such as possible contribution from evaporite and different fractionation mechanisms (Vigier et al., 2009; Yoon, 2010; Millot et al., 2010a,b). In this study, the lithium concentrations and Li isotopic compositions for both dissolved and suspended loads in the Changjiang main channel and its main tributaries were analysed to evaluate the Li budget and isotopic fractionation processes during weathering. Because the Changjiang drains a large portion of the continental crust covered by various lithologies and because the basin is characterised by different silicate weathering rates (Chetelat et al., 2008), this river is suitable for exploring these questions. These data have already been published in Wang et al. (2008), but they are discussed in more detail in this contribution.

Furthermore, Bouchez et al. (2013) used a modeling approach to investigate the relationship between different

geomorphic regimes and the Li isotopic composition of the ocean during the Cenozoic. As the Changjiang basin is characterized by a very variable topography, with a fall of about 5400 m from the Qinghai-Tibet Plateau to the East China Sea, the dependence of the Li isotopic composition of rivers with different geomorphic regimes predicted by Bouchez et al. (2013) can be tested.

2. GEOLOGICAL SETTING

The Changjiang River is the third longest river in the world. It originates from the Tibetan Plateau, and flows through 11 provinces or municipalities of China, and discharges into the East China Sea around the Chongming Island. The whole drainage basin is situated between $24^{\circ}27' - 35^{\circ}54'N$ and $90^{\circ}13' - 122^{\circ}19'E$, and covers about one-fifth of the area of China. The major part of the drainage basin is characterized by a humid sub-tropical climate with an annual mean temperature between 5 and 15 °C. The annual mean precipitation in the basin is 1100 mm, more than 60% of which falls during the flood season from April to October (Changjiang Water Resource Committee, 1999).

The drainage basin is characterized by complex bedrock lithologies, including Precambrian to Quaternary sedimentary layers mainly composed of marine carbonates, clastic and evaporitic rocks, red sandstone, and continental deposits. Carbonate rocks are widely spread throughout the drainage basin, and are particularly abundant in the southern region of the middle reaches (Yunnan, Guizhou and the western Hunan Provinces), and in the upper/middle reaches of the Hanjiang River. The Jurassic red sandstone is widely spread in the Sichuan Basin. Clastic and evaporitic rocks are mainly found in the upper reach of the Changjiang River (Fig. 1).

3. SAMPLING AND ANALYTICAL METHODS

3.1. Sample collection

Samples were collected in August 2006 along the Changjiang main channel and its main tributaries (Fig. 1). Between 10 and 20 L of surface water were filtered in the field through 0.22 μm cellulose acetate filters, and stored in acid-washed polypropylene bottles. The filters were saved for analysis of the suspended loads. Water samples for analysis of Li concentrations and Li isotopes were immediately acidified to pH 2 with thermally distilled (TD) HNO_3 . After return to the laboratory, the suspended material was recovered from the filters, dried at 60 °C, and ground using an agate mortar.

3.2. Analytical methods

The Li content of all of the samples was determined by atomic absorption spectrometry (AAS) with a precision of $\pm 5\%$. The suspended material was digested in a mixture of concentrated HF, HNO_3 and HClO_4 and refluxed with concentrated HNO_3 . Lithium was separated from the sample matrix by ion exchange chromatography before

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