



Stable hydrogen and oxygen isotopes in mineral-bound water and the indication for chemical weathering intensity



Chengfan Yang, Shouye Yang*, Ni Su

State key Laboratory of Marine Geology, Tongji University, 1239 Siping Road, Shanghai 200092, PR China

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ABSTRACT

Chemical weathering plays a key role in the long-term climate change and global biogeochemical cycle. In this study, a 5.1 m-long basalt-developed weathering profile from Hunan Province, China, was selected for the analyses of major elements, grain size, pH, total organic carbon, thermogravimetric (TG) and differential thermogravimetric properties (DTG), as well as stable isotopes (δD and $\delta^{18}O$) in mineral-bound waters. The major purpose is to distinguish different types of mineral-bound water and to verify whether hydrogen and oxygen isotopes of mineral-bound water can indicate chemical weathering intensity. Based on the TG and DTG results, different types of mineral-bound water were extracted by a sequential heating extraction technique (40–120 °C, 120–300 °C, 300–600 °C and 600–900 °C). The mineral-bound water lost below 120 °C is absorbed water, a mixture of interlayer and crystal waters occurs at 120–300 °C, and above 300 °C is mostly constitution water. Hydrogen and oxygen isotopic ratios at top ~2 m are subject to the influences of eolian and pedogenic processes. The absorbed water has been exchanged and reached isotopic equilibrium with ambient soil water, while the crystal water and interlayer water cannot be separated by heating method in this study. Hydrogen and oxygen isotopes of constitution water can indicate chemical weathering, showing positive relationships with weathering intensity. Although the mechanism for their indication to chemical weathering remains further investigation, we suggest that the isotopic fractionation between the formation of clay minerals and surface water should be taken into account. This research sheds new light on mineral-bound water phase and a potential proxy for tracing the process of chemical weathering.

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1. Introduction

Chemical weathering of silicates plays an important role in governing earth surface processes (Nesbitt and Young, 1989; Nesbitt and Markovics, 1997; Brantley et al., 2007; Dixon et al., 2012; Maher and Chamberlain, 2014), crustal evolution (Taylor and McLennan, 1995; Liu and Rudnick, 2011), soil erosion (Lal, 2003; Rad et al., 2013), and climate change (Raymo and Ruddiman, 1992; Godd eris et al., 2013). Chemical weathering also significantly influences surface biogeochemical cycle as a result of elemental redistribution among Earth's reservoirs and material exchange between land and ocean (Gaillardet et al., 1997; Bouchez et al., 2012).

The prerequisite of chemical weathering study is to establish suitable proxies for the intensity and rate of chemical weathering at multiple spatial and time scales. The widely-used geochemical proxies such as chemical index of alternation (CIA), chemical index of weathering

(CIW), weathering index of Parker (WIP), weathering (mobility) index of αMe (Me indicates specific elements), are based on the mobility of major elements during chemical weathering (Parker, 1970; Nesbitt and Young, 1982; Fedo et al., 1995; Gaillardet et al., 1999; Ohta and Arai, 2007; Garzanti et al., 2013; Babechuk et al., 2014). The ratios of major and trace elements such as Ra/Sr, Sr/K and Ca/Zr have been also applied for the estimation of weathering intensity (Nesbitt and Markovics, 1980; Bokhorst et al., 2009). In addition, traditional radiogenic (e.g. Sr, Hf, Nd, U-Th) and stable isotopes (e.g. Li, Mg, Ca, Si, stable Sr, O) are increasingly applied as weathering indices because of their environmental sensitivity (Bird et al., 1993; Pistiner and Henderson, 2003; Misra and Froelich, 2012; Opfergelt et al., 2012; Fantle and Tipper, 2014; Bayon et al., 2016). Apart from geochemical parameters, other proxies including abrasion pH, loss on ignition (LOI), mineral assemblages and environmental magnetic parameters of soils and weathered sediments were also proposed by previous studies (Grant, 1969; Sueoka, 1988; Su et al., 2015).

Over the last decade, the indications and their applicability of the above chemical weathering indices have been revisited by some studies (Duzgoren-Aydin et al., 2002; Price and Velbel, 2003; Li and Yang, 2010;

* Corresponding author.

E-mail address: syyang@tongji.edu.cn (S. Yang).

Buggle et al., 2011; Shao et al., 2012; Bi et al., 2015). These studies suggest that most of these chemical indices (e.g., CIA, CIW, WIP and chemical proxy of alteration (CPA)) have their limitations in practical application, mostly because they can only indicate integrated weathering history registered in the siliciclastic sediments, and hydrodynamic sorting and mineral differentiation during sedimentary processes may also influence the application of these weathering proxies. Thus, it should be cautious in using these geochemical proxies directly to indicate weathering intensity and rates. To our knowledge, the essential of chemical weathering is the interaction between water and rocks and their weathering residues under the surface environment (Nesbitt and Young, 1989). Water as the most important agent for promoting chemical weathering, however, was poorly considered in previous studies, although hydrogen isotope of the hydroxyl groups has recently been used to evaluate chemical weathering (e.g., Lupker et al., 2012, 2013).

Nevertheless, the mineral-bound water contains different binding phases in various rock-forming and secondary minerals. One is by adsorption forces on different surfaces of the mineral structure (Ding and Frost, 2004), and another is by stronger coordination forces around certain cations of the lattice (Földvári et al., 1988). For the former, the water attached to the outer surface is called absorbed water, and the water bound to the internal surface is interlayer water (Maftuleac et al., 2002). The latter is so-called hydrate water, which can be further classified as crystal water and constitution water (Falk and Knop, 1973). Besides, organic water, existing as H^+ or OH^- in organic matter, also has a significantly influence on isotopic analysis of mineral-bound water (Sheppard and Gilg, 1996). Whether the hydrogen and oxygen isotopes of these different types of mineral-bound water can indicate chemical weathering intensity has to be clarified further.

Thus, this study will examine different binding ways of mineral-bound water extracted from a 5.1 m-long basalt-developed weathering profile from central China (Hunan Province). By measuring stable isotopes (δD and $\delta^{18}O$), together with mineral assemblage, major elements, grain size, total organic carbon (TOC), pH, thermogravimetric (TG) and differential thermogravimetric properties (DTG), we aim to recognize and discriminate different types of mineral-bound water, and further to reveal the implication of mineral-water hydrogen and oxygen isotopes for chemical weathering intensity.

2. Geological and geographical settings

The study site is located in the Youxian County of Hunan Province ($113^{\circ}21.799'E$, $27^{\circ}11.233'N$), southern of Dongting Lake (a sub-basin of the Changjiang catchment) (Fig. 1). Hunan Province is located in central China, and its northwest is situated on the Yangtze Craton, and the middle and southern parts belong to the Cathaysia Block (Pan et al., 2009). Six tectonic and magmatic movements characterize the geologic evolution in the study area, including Wuling Movement, Xuefeng Movement, Caledonia Movement, Indosinian Movement, and Yanshan Movement and Himalayan Movement (He, 1986). A variety of Mesoproterozoic to Cenozoic rocks occur in the province, with the dominance of sedimentary rocks. The igneous rocks cover an area of 8.3% of the whole province, and mainly include hypogene rocks, with volcanic rocks distributed sporadically (Bureau of Geology and Mineral Resources of Hunan Province, 1988). The hypogene rocks are dominated by acid rock, while volcanic rocks are mainly calc-alkaline and alkaline rocks. In this study, the bedrock that the weathering profile is developed is Cretaceous cala-alkaline basalt (Ma et al., 2002).

Geographically, Hunan Province is dominated by subtropical humid monsoon climate, which has a strong influence on chemical weathering in this area (Yang et al., 2004; Bo et al., 2011; Shao and Yang, 2012; Guo et al., 2013). Affected by monsoon climate, the evergreen broad-leaf forest is the dominated vegetation type, and soil laterization is widely observed, with an area of 31% of the Dongting Basin (Zhang et al., 2007). Besides, the annual mean precipitation is about 1200–1700 mm, of which about 60–85% is generated by the East Asia summer monsoon. The mean annual temperature is 16–18 °C, with the highest temperature in July and the lowest in January (Yang and Xue, 2001).

3. Samples and methods

The soil and sediment samples from a basalt weathering profile were collected in April 2014. The thickness of this weathering profile is about 15 m (Fig. 1). A total of 34 soil and rock samples were collected, among which 30 samples were used in this study. Soil samples were collected at 10–30 cm intervals in the upper 5.1 m, and the rock sample was taken from the 15 m depth. The detailed description of this weathering

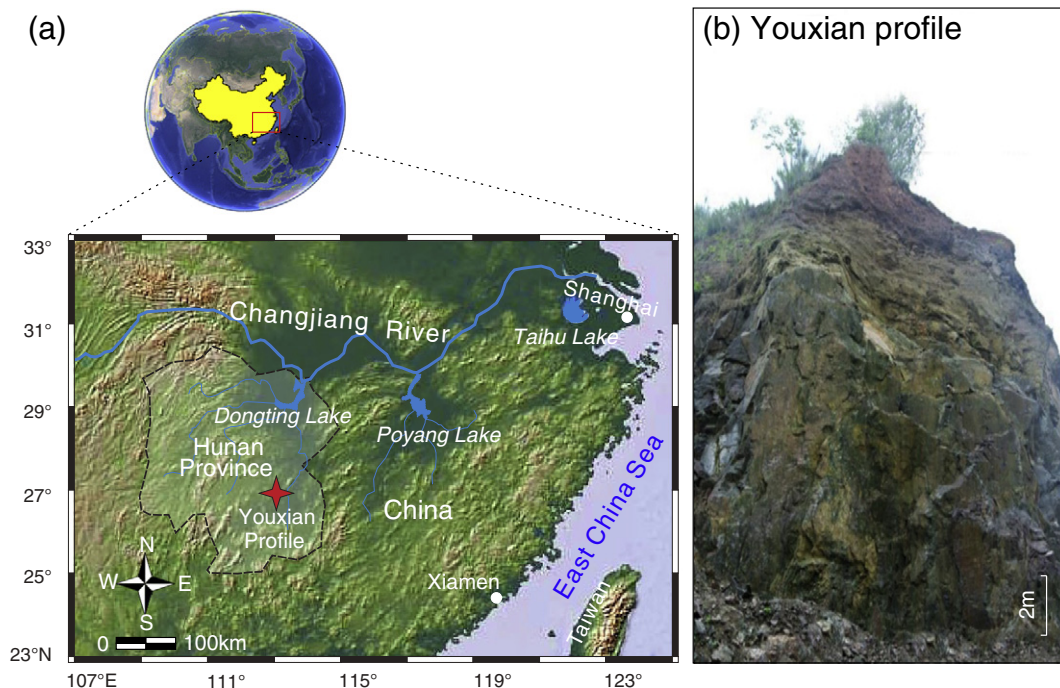


Fig. 1. A map showing the study area (a) and the basalt weathering profile (b) in Hunan Province, China.

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