



Provenance of uranium in a sediment core from a natural reservoir, South China: Application of Pb stable isotope analysis

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HIGHLIGHTS

- High enrichment of U was found across the whole sediment core.
- Pb isotopic fingerprinting techniques allowed quantification of source inputs of the U contamination.
- A hidden geoenvironmental health hazard may arise from the natural-occurrence of U contamination.

ARTICLE INFO

Article history:

Received 30 August 2017

Received in revised form

7 November 2017

Accepted 22 November 2017

Available online 23 November 2017

Handling Editor: Martine Leermakers

Keywords:

Uranium migration

Pb isotopes

Source apportionment

Binary mixing

Radiogenic

ABSTRACT

As part of ongoing environmental investigations of U mining impacts, forty-two sediment samples of a nearly-half-meter-long sediment core retrieved from a natural reservoir near an active uranium (U) mining site, South China were analyzed to quantify the extent of U release and identify U release mechanism within the riverine catchment. Enrichment levels of U was dispersed not only in the surface sediments but also in deep sediments across the depth profile. Further analysis by SEM-EDS and XRD indicated that U partitioning in the depth profile was possibly controlled by complicated interplay of leaching and precipitation cycles of U-bearing minerals. Even with the relative complexity of U dispersal processes within the catchment, the Pb isotopic fingerprinting techniques allowed quantification of source inputs of the sediments by using a binary mixing model. The results revealed that along the depth profile, only 6%–50% of the sediment material is anthropogenically derived from the U ore tailing, with the other predominant proportions originated from geogenically natural weathering of granitic bedrocks. This study highlights the use of Pb isotopes as a powerful tool for quantitatively fingerprinting the sources of U dispersal in the sediment core, and natural-occurring U contamination that may become a hidden geoenvironmental health hazard in this area.

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1. Introduction

During the last five decades, extensive and widespread environmental degradation has occurred as a result of open pit U mining in China. Mining operations have generated numerous waste-rock dumps and slime dams, exposing U ore and tailings to weathering. As a consequence, U and other potentially harmful elements (e.g. Pb, Mn and the other daughter radioactive elements of U) are being released into the environment at rates exceeding

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those typically found in nature (Abdelouas et al., 1998; Kipp et al., 2009; Sun et al., 2014; Song et al., 2017). To date, the U mines in China have been rarely reclaimed, often leaving loose contaminated materials vulnerable to erosion and leaching, risking release and migration of U and other elements into the environment for decades. Understanding the impacts of U mine wastes has become an important part of environmental geochemistry (Kipp et al., 2009; Sun et al., 2014, 2016a; 2016b; Wu et al., 2013; Chen et al., 2017).

In 1957, specific granite beds in a county of the Northern Guangdong Province, China, were identified as uraniferous and uranium extraction commenced on a large scale, using heap leaching techniques with sulfuric acid. In this process, the disposal of solid and liquid waste constitutes a major environmental problem since these materials contain elevated levels of radioactive nuclides as well as harmful non-radioactive elements. A long-term failure to effectively control these materials has created a significant environmental hazard and may have health implications for the local population. Only very limited studies on environmental contamination in this area exist so far (Liu et al., 2015; Wang et al., 2012). Our previous investigation (Liu et al., 2015; Wang et al., 2012) showed that surface waters and sediments from local aquifers around the U industrial site were contaminated by U mining waste. Unexpected high enrichment of U, Mn and Th was even found in a natural reservoir downstream (Liu et al., 2015). However, the origin and dispersal mechanisms of U within this riverine environment remain largely unknown.

Lead isotope measurements are a well-established tool for determining contaminant provenance, transport pathways and source apportionment within atmospheric and marine environments (Monna et al., 1999; Chiaradia and Cupelin, 2000; Luck and Othman, 2002; Renberg et al., 2002; Baker et al., 2004; Kurkjian et al., 2004; Ettler et al., 2006). There are four stable isotopes of Pb (^{204}Pb , ^{206}Pb , ^{207}Pb and ^{208}Pb). Three of them are the stable end products of the radioactive series of U and Th: ^{206}Pb from ^{238}U , ^{207}Pb from ^{235}U , and ^{208}Pb from ^{232}Th . The fourth isotope, ^{204}Pb , is non-radiogenic having remained constant since the formation of the earth (Gulson et al., 1989; Komarek et al., 2008; Sun et al., 2011; Bosch et al., 2002; Chen et al., 2016), whereas the radiogenic Pb isotopes are continuously generated by the radioactive decay of U and Th. Consequently, the crustal Pb isotope ratios are changing over time (Cummings and Richards, 1975; Frostick et al., 2011). The present day average crustal (PDAC) $^{206}\text{Pb}/^{207}\text{Pb}$ and $^{208}\text{Pb}/^{207}\text{Pb}$ ratios are 1.20 and 2.48, respectively (Cummings and Richards, 1975; Frostick et al., 2011; Bollhöfer, 2012).

It is well-known that Pb from U ores is predominantly radiogenic. The Pb isotope fingerprint of a U ore body is markedly different from those found in other metal ore bodies (e.g., those rich in Zn, Cu, Pb) and background rocks. Due to the small relative mass differences between the isotopes, only minimal physical or chemical fractionation occurs during environmental and industrial processes, compared to the primary differences in the isotopic compositions of the sources (Lima et al., 2005). Therefore, Pb isotope ratios are ideally suited as a source tracer (Lima et al., 2005; Bird, 2011) and have been widely utilized for source fingerprinting of atmospheric and marine Pb pollution. However, application of Pb isotopes to establish contaminant provenance and source apportionment of U contamination is relatively under-studied worldwide, especially in China (Dean and Gulson, 1987; Gulson et al., 1989; Frostick et al., 2008, 2011; Kyser et al., 2015; Cuvier et al., 2016).

As a part of ongoing environmental investigations of U mining in the Northern Guangdong Province, China, a sediment core that may serve as sink and source of U contamination was sampled in the area of a natural water reservoir. For comparison, surface sediments

were sampled from the recipient stream (Stream 2) flowing through tailing footprints (thereby collecting mining waste) and from a natural stream (Stream 3) in the vicinity, which can be used as a background reference. The reservoir collects both U mining wastes and mountain run-off.

The aims of this paper are to (1) identify dispersal mechanisms and fate of U within the catchment; (2) quantify the relative contributions of contaminants from U mining waste and from natural bedrock. To achieve these aims, metal contents and isotopic compositions of Pb were also measured for the U ore tailing and for the background sediment around the U mining site.

2. Materials and methods

2.1. Study area and sampling site

The investigated uranium ore is an intra granitic deposit occurring in the eastern Guidong granite massif, a relatively pristine area remote from industrial centres in the Northern Guangdong Province, China (Chen et al., 1999; Wang et al., 2012). The deposit is associated with silicified fracture zones. The uraniferous veins are dominated by pitchblende, colloidal pyrite, pink microcrystalline quartz, hydrogoethite, and hematite (Wu et al., 2005; Wang et al., 2012; Liu et al., 2015; Franz, 2009). The rocks are mainly composed of Early Jurassic porphyritic, medium-grained biotite granite and Late Jurassic fine-grained muscovite granite (Wu et al., 2005; Franz, 2009). The mean grades of U vary in a range from 5 to 13 mg/kg, with original in-situ resources of 1500–5000 t (Wu et al., 2005; Liu et al., 2015). The U ore field extends over an area of approximately 400 km², which includes several villages with about 50,000 residents. This area is characterized by a subtropical-monsoonal climate with a mean annual precipitation of 1600 mm and an average temperature of 20 °C.

The sediment core studied here was collected from nearly the center of a natural water reservoir situated 5 km downstream of the U mining/milling site (Fig. 1) in July 2012, using a 1 m gravity corer. The reservoir covers an area around 2.5×10^4 m² with an average water depth of 10 m. The length of the core was measured in the field, and the core was then cut into sub-samples of 10 mm length using an acid-cleaned stainless steel knife. As shown in Fig. 1, several surface sediments (S5, S6, S7, S9, S10 and S11) and a surface sediment sample (S8) were also sampled from the recipient stream (Stream 2) flowing through U tailing footprints and from a nearby natural background stream (Stream 3), respectively. In total, 42 sub-samples from the sediment core and seven surface sediments from the adjacent streams were obtained, which were immediately placed in acid-cleaned polyethylene bags. After drying at room temperature, the samples were ground and sieved. The sediment fraction <100 μm was used for elemental analysis and Pb isotope analysis.

2.2. Trace metal analysis

Trace metal contents of the sediments were determined by inductively coupled plasma mass spectrometry (ICP-MS) using a Perkin-Elmer ELAN 6000 instrument. Sediment samples were digested in a nitric/hydrofluoric acid mixture and heated in a teflon vessel at 180 °C. Aliquots were evaporated to dryness and redissolved in 3% nitric acid before analysis. Duplicate samples and blank controls were prepared in the same way to ensure an adequate background correction. A certified reference material (GBW07405, soil) supplied by the National Center for Standard Reference Materials, China was used to control the quality of the metal analysis. The accuracy of the measurement was found to be within 5% RSD. The recoveries of the metals from the certified reference material

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