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Which minerals control the Nd–Hf–Sr–Pb isotopic compositions of river sediments?

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ABSTRACT

River sediments naturally sample and average large areas of eroded continental crust. They are ideal targets not only for provenance studies based on isotopic compositions, but also can be used to establish average continental crust isotopic values. In large fluvial systems, however, mineral sorting processes significantly modify the mineralogy, and thus the geochemistry of the transported sediments. We still do not know, in any quantitative way, to what extent mineral sorting affects and fractionates the isotopic compositions of river sediments. Here, we focus on this issue and try to decipher the role of each mineral species in the bulk isotopic compositions of bedloads and suspended loads sampled at the outflow of the Ganga River that drains the Himalayan mountain range.

We analyzed Nd, Hf, Sr, and Pb isotopic compositions as well as trace element contents of a large number of pure mineral fractions (K-feldspar, plagioclase, muscovite, biotite, magnetite, zircon, titanite, apatite, monazite/ allanite, amphibole, epidote, garnet, carbonate and clay) separated from bedload and bank sediments. We combine these data with mineral proportions typical of the Ganga sediments to perform Monte Carlo simulations that quantify the contributions of individual mineral species to the Nd, Hf, Sr, and Pb isotopic budgets of bedloads and suspended loads.

The isotopic systematics of river sediments are buffered by very few minerals. Despite their extremely low proportions in sediments, zircon and monazite/allanite control Hf and Nd isotopes, respectively. Feldspars, epidote, and carbonate buffer the Sr isotopic budget while clay, feldspars, and heavy minerals dominate Pb isotopes. Hafnium, Sr, and Pb isotopic differences between bedloads and suspended loads are well explained by their different mineral compositions. This confirms that Hf, Sr and Pb isotopic compositions of sediments are strongly biased by mineral sorting processes during fluvial transport; hence they do not always constitute good proxies for provenance studies. In addition, we anticipate that fractionation of the isotopic systems continues at the river/ocean interface to deliver sediments to the deep ocean that are not necessarily similar to their crustal precursors, creating a systematic bias between the compositions of crustal sources and oceanic sediments.

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1. Introduction

Because they integrate large portions of continental crust exposed to weathering, river sediments from worldwide fluvial systems have been widely used to trace sediment provenance, estimate the average isotopic composition of the upper continental crust, and constrain its evolution through time (Goldstein et al., 1984; Goldstein and Jacobsen, 1988; Asmerom and Jacobsen, 1993; Allègre et al., 1996; Clift et al., 2002; Singh and France-Lanord, 2002; Millot et al., 2004; Kamber et al., 2005; Richards et al., 2005; Roddaz et al., 2005; Singh et al., 2008; Cina et al., 2009; Belousova et al., 2010; Hawkesworth et al., 2010; Wu et al., 2010; Dhuime et al., 2011; Padoan et al., 2011). Most studies

assume that the isotopic compositions of river sediments reflect that of the drained continental area. However, depending on its size, shape, and density, detrital material transported by rivers is segregated within the water column by hydrodynamic processes: fast-settling coarse and heavy minerals concentrate in bedloads while fine, platy and light minerals are preferentially transported in suspension, and at different depths in the water column depending on their settling velocities (Singh and France-Lanord, 2002; Komar, 2007; Galy et al., 2008; Garzanti et al., 2008). Because chemical elements are carried in different amounts by the various minerals, it is now well described that mineral sorting processes lead to large chemical variability between bedload and suspended load sediments (Garzanti et al., 2010; Bouchez et al., 2011; Garzanti et al., 2011; Lupker et al., 2011, 2012). In contrast, our knowledge of how such processes affect the Nd, Hf, Sr and Pb isotopic compositions of sediments is relatively limited. While several studies have pointed out Nd, Hf, Sr and Pb isotopic fractionations between fine and coarse-grained sediments due to mineral sorting processes





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(Patchett et al., 1984; McLennan et al., 1989; Revel et al., 1996; Eisenhauer et al., 1999; Singh and France-Lanord, 2002; Chauvel et al., 2008; Bayon et al., 2009; Carpentier et al., 2009; Chauvel et al., 2009; Vervoort et al., 2011; Garçon et al., 2013a, 2013b), the influence of each mineral species on the isotopic composition of sediments has never been thoroughly quantified.

In this paper, we report trace element contents and Nd, Hf, Sr and Pb isotopic compositions for a large number of pure mineral fractions separated from a bedload and a bank sediment sampled at the outflow of the Ganga fluvial system in the Bangladesh delta. Using Monte Carlo simulations, we combine these data with mineral proportions reported by Garzanti et al. (2010, 2011) and Lupker et al. (2012) in river sediments sampled at the same location to (1) evaluate the individual contribution of each mineral species to the Nd, Hf, Sr and Pb isotopic budgets of bedloads and suspended loads and (2) determine if the known mineralogy of bedloads and suspended loads accounts for the observed difference in isotopic compositions. Finally, we discuss the implications of our results for sediment provenance studies based on isotope data and for large-scale isotopic partitioning between continental and oceanic sediments.

2. Studied area and samples

The studied mineral fractions were separated from two samples (bank sediment BGP 6 and bedload BR 717) collected at the outflow of the Ganga River, before the confluence with the Brahmaputra River (Fig. 1). The sampling site was selected because it is located down-stream, far away from the mountain range. As a consequence, the sediments integrate all materials transported by the Ganga River and its tributaries. These rivers belong to one of the largest fluvial systems on Earth (Milliman and Meade, 1983), stretch over more than 2500 km and deliver about four hundred million tons of sediments to the ocean each year (Lupker et al., 2011). Such sediments are dominantly (>95%)



Fig. 1. Map of the Bangladesh delta showing sampling sites for river sediments.

derived from the Himalayan mountain range with minor contributions from the Deccan Trapps and Indian craton supplied by the southern tributaries of the Ganga (Galy and France-Lanord, 2001; Singh et al., 2008; Lupker et al., 2011). Himalayan materials mainly consist of sedimentary series and crystalline metamorphic rocks (e.g., Le Fort, 1975). Because of the large lithological and chemical diversity of Himalayan rocks, the Ganga basin can be considered as a representative portion of the upper continental crust. All minerals analyzed in our study are thus ubiquitous in the erosion products of any upper continental crust exposed to weathering (McLennan, 1989; Taylor and McLennan, 1995; McLennan, 2001) and our results should be relevant for river sediments sampled in other worldwide fluvial systems draining large continental areas.

The second major advantage of the selected site is that these sediments were extensively studied for their mineralogy by Garzanti et al. (2010, 2011) and Lupker et al. (2012) (Supplementary Table 1). These authors have shown that bedloads and suspended loads share quite similar types of minerals but in different proportions. Surface suspended loads are generally finer and relatively rich in micas and clays while bedloads tend to be coarser and richer in quartz and heavy minerals. Among the heavy minerals that are commonly found in the Ganga sediments, amphibole, epidote and garnet are the most abundant but many others are also present: titanite, zircon, rutile, apatite, monazite, tourmaline and allanite for example (Garzanti et al., 2010, 2011). Detailed mineral proportions in the Ganga bedloads and suspended loads are listed in Supplementary Table 1.

Finally, several studies have reported major and trace element contents, as well as Nd, Hf, and Pb isotopic compositions, of bulk suspended loads and bedloads from this sampling site (Galy and France-Lanord, 2001; Lupker et al., 2011; Garçon, 2012; Lupker et al., 2012; Garçon et al., 2013a, 2013b; see Supplementary Table 2). We used these data to evaluate the role of individual minerals in the bulk isotopic budget of river sediments. Because no Sr isotopic data exist in the literature for unleached bedloads and suspended loads, we analyzed the bulk Sr isotopic compositions of two bedloads (BR 717 and BR 418), one bank sediment (BGP 6) and 2 suspended loads (BR 414 and BR 412) sampled in the Bangladesh delta (Fig. 1). The trace element data and the Nd, Hf, and Pb isotopic compositions of these five samples are reported in Supplementary Table 2. The detailed sampling methods and the major element data can be found in Galy and France-Lanord (2001), Lupker et al. (2011, 2012) and Garcon et al. (2013a, 2013b). We also analyzed the Sr isotopic compositions of several grainsize fractions separated from a suspended load from the Ganga River (sample BR 522, Fig. 1) to help us in interpreting the Sr results of the Monte Carlo simulations.

3. Methods

3.1. Mineral separation

The starting material consists of several kilos of sediment collected during the monsoon season. With the exception of the clay fraction separated from bank sediment BGP 6, all mineral fractions were separated from bedload BR 717 dredged in the main channel of the Ganga River. This sample was first washed and dried, then divided into 3 grain-size fractions by sieving: 63–100 μ m, 125–250 μ m, and >250 μ m. For each fraction, we used successive centrifugations in heavy liquids (sodium metatungstate and methylene iodide) to separate minerals and partial freezing in liquid nitrogen to recover them. After this step, some of the minerals were isolated using a hand magnet or the Frantz isodynamic magnetic separator. Biotite, muscovite, plagioclase, quartz, and K-feldspar were isolated from the >250 µm fraction; magnetite, amphibole, apatite, garnet, epidote, titanite, monazite, allanite, and zircon from the 63-100 µm and/or the 125-250 µm fractions. Each mineral fraction was finally carefully purified under the binocular and polarizing microscopes to remove impurities.

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