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# Environmental Hf–Nd isotopic decoupling in World river clays

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ABSTRACT

The hafnium and neodymium radiogenic isotope systems behave differently during Earth surface processes, causing a wide dispersion of Hf and Nd isotopic compositions in sediments and other sedimentary rocks. The decoupling between Hf and Nd isotopes in sediments is generally attributed to a combination of preferential sorting of zircon during sediment transport and incongruent weathering processes on continents. In this study, we analysed size-fractions of sediment samples collected near the mouth of 53 rivers worldwide to better understand the factors controlling the distribution of Hf and Nd isotopes in sediments. Our results for rivers draining old cratonic areas and volcanic provinces demonstrate that both granite and basalt weathering can lead to significant grain-size dependent Hf isotopic variability. While silt-size fractions mainly plot along the Terrestrial Array, World river clays are systematically shifted towards more radiogenic Hf isotopic compositions, defining together with published data a new Clay Array ($\varepsilon_{Hf} = 0.78 \times \varepsilon_{Nd} + 5.23$). The Hf–Nd isotope decoupling observed in volcanogenic sediments is best explained by selective alteration of Lu-rich mineral phases (e.g. olivine) and preferential enrichment of resistant unradiogenic minerals, such as spinel and ilmenite, in silt fractions. We also show that the extent to which World river clays deviate from the Clay Array ($\Delta\varepsilon_{Hf}$ clay) is not linked to the presence of zircons. Instead, it correlates positively with weathering indices and climatic parameters (temperature, rainfall) of the corresponding drainage basins. Overall, these findings demonstrate that the distribution of Hf–Nd isotopes in clay-size sediments is related to a large extent to weathering conditions on continents, although the precise mechanisms controlling this relationship remain unclear. We finally propose that the Hf–Nd isotope pair proxy could be used in palaeoenvironmental studies to provide semi-quantitative information on past climates.

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## 1. Introduction

Weathering processes progressively lead, with time, to the disintegration of rocks on continents and development of soil sequences. Most silicate minerals are generally unstable at the Earth's surface, and typically weather to form clays (e.g. Velde, 1995). Upon formation in soils, clays incorporate a substantial fraction of the elements released during chemical weathering. Some of these elements are commonly referred to as immobile (e.g. Al, Ti, Zr), as opposed to other more mobile elements (e.g. Na, K), which often remain in solution and are exported away by freshwaters and/or bio-assimilated (e.g. Garrels and MacKenzie, 1971). Due to their small grain-size, clays and other fine-grained erosion products such as silts are efficiently removed from soils during erosion, which also implies that they can be delivered to the ocean via rivers with presumably minimum transfer times compared to coarser sedimentary particles.

Over the past decades, studies of fine-grained sediments and river particulates have provided a wealth of information on both the composition of the exposed continental crust and chemical weathering processes (e.g. Taylor and McLennan, 1985; Gaillardet et al., 1999a). The abundance and isotopic composition of immobile elements are often used as tools for assessing the provenance of sedimentary rocks. Amongst these, rare earth elements (REE) and neodymium (Nd) isotopes have received particular attention over the years (e.g. Goldstein et al., 1984; McLennan, 1989). Detrital sediments are thought to retain the Nd isotopic composition of their source rocks during continental weathering, sedimentary and post-depositional processes (e.g. Goldstein et al., 1984). As a consequence, Nd isotopes are often used for tracing the geographical provenance of sediments (e.g. Goldstein and Hemming, 2003).

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In contrast, the degree of chemical weathering of soils and associated source rocks is generally evaluated using indices of the relative abundance of immobile versus mobile elements, such as the widely used chemical index of alteration (CIA; Nesbitt and Young, 1982). Radiogenic isotope systems other than Nd (e.g. Pb, Sr, Os) have also proven to be particularly useful for tracing weathering processes (e.g. Erel et al., 1994). Their application to sedimentary rocks is based on evidence that incongruent dissolution of silicate rocks during chemical weathering leads to secondary products having distinctive radiogenic isotopic compositions. Recently, the emergence of non-traditional stable isotope geochemistry (e.g. Li, Si, Mg) has also led to promising perspectives for further understanding the links between clay mineral formation and the surrounding bio- and hydro-spheres (e.g. Opfergelt et al., 2010; von Strandmann et al., 2012).

In addition to the various proxies listed above, hafnium (Hf) isotopes also represent interesting tracers of silicate weathering, in particular when their measurement is combined with Nd isotopes. Despite behaving relatively similarly during magmatic processes (Vervoort et al., 1999), the Lu–Hf and Sm–Nd radiogenic isotopic systems are strongly decoupled by Earth surface processes (e.g. van de Flierdt et al., 2007). A substantial fraction of the Hf budget in rocks and sediments is indeed hosted in zircons, a mineral characterised by very unradiogenic isotopic compositions (i.e. low $^{176}Hf/^{177}Hf$ ratios or $\varepsilon_{Hf}$ values). Zircons are highly resistant to weathering and preferentially sorted into coarse-grained fractions during sediment transport (Patchett et al., 1984). In addition to this 'zircon effect', silicate weathering also leads to preferential dissolution of Lu-rich mineral phases such as apatite and sphene, which releases radiogenic Hf (i.e. high $^{176}Hf/^{177}Hf$ ratios or $\varepsilon_{Hf}$ values) to river waters and presumably to seawater (Bayon et al., 2006; Godfrey et al., 2007). The observed decoupling between Hf and Nd isotopes during Earth surface processes is clearly illustrated in the $\varepsilon_{Hf}$ vs. $\varepsilon_{Nd}$ diagram, where fine-grained sediments display a wide range of Hf–Nd isotopic compositions between the Terrestrial Array (Vervoort et al., 2011) and the Seawater Array (Albarède et al., 1998), which both refer to the broad correlations defined by most terrestrial rocks and seawater/marine precipitates, respectively (Fig. 1). Collectively, analyses of marine sediments (Vervoort et al., 1999, 2011; Pettke et al., 2002; Vlastelic et al., 2005; Prytulak et al., 2006; van de Flierdt et al., 2007; Bayon et al., 2009a; Carpentier et al., 2009, 2014), river particulates, bedloads and zircon grains (Bayon et al., 2006; Chen et al., 2011; Rickli et al., 2013; Garçon et al., 2013, 2014; Garçon and Chauvel, 2014), loess deposits (Chen et al., 2013; Chauvel et al., 2014) and aeolian dust (Lupker et al., 2010; Rickli et al., 2010; Aarons et al., 2013; Chen et al., 2013; Pourmand et al., 2014; Zhao et al., 2014) all indicate that coarse-grained (and/or zircon-rich) sediments typically fall along or below the Terrestrial Array, while clay-size (and/or zircon-poor) fractions generally display more radiogenic Hf signatures (Fig. 1). To a large extent, the observed decoupling can be explained by mineralogical sorting processes that occur during sediment transport (e.g. van de Flierdt et al., 2007; Aarons et al., 2013; Garçon et al., 2013). However, recent investigations of Late Quaternary sediments from the Congo fan area also led to the suggestion that the distribution of Hf–Nd isotopes in fine-grained sediments could be controlled instead by chemical weathering intensity on continents (Bayon et al., 2009a; Bayon et al., 2012).

In view of the above consideration, the main aim of this study was to further evaluate the relative role of mineralogical versus weathering processes in explaining the observed large dispersion of Hf and Nd isotopic ratios in fine-grained sediments. To this purpose, we have analysed a large set of sediments deposited near the mouth of rivers worldwide, for which we report $^{176}Hf/^{177}Hf$ and $^{143}Nd/^{144}Nd$ ratios on both silt (2–63 μm) and clay (<2 μm) size-fractions. These results allow us to identify new key parameters (temperature, rainfall) that control the distribution of Hf–Nd isotopes in clay-size detrital fractions.

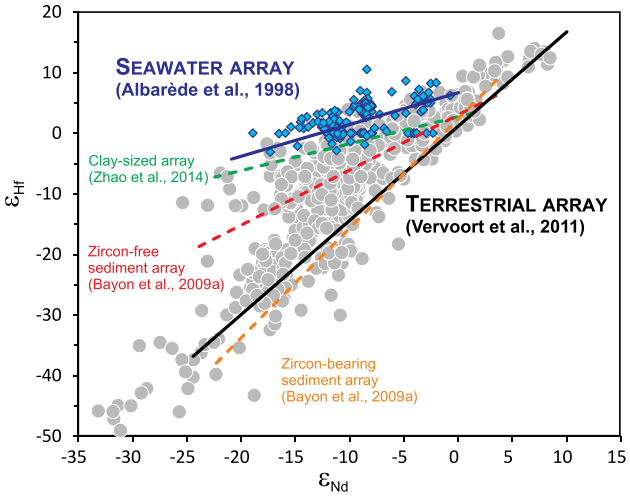


**Fig. 1.** A compilation of published Hf and Nd isotopic compositions for fine-grained sediments and other sedimentary rocks. The sediment data (grey circles) include present-day $\varepsilon_{Hf}$ and $\varepsilon_{Nd}$ values for: marine sediments (Vervoort et al., 1999, 2011; Pettke et al., 2002; Vlastelic et al., 2005; Prytulak et al., 2006; van de Flierdt et al., 2007; Bayon et al., 2009a; Carpentier et al., 2009, 2014), river particulates and bedloads (Bayon et al., 2006; Rickli et al., 2013; Garçon et al., 2013, 2014; Garçon and Chauvel, 2014), loess deposits (Chen et al., 2013; Chauvel et al., 2014) and aeolian dust (Lupker et al., 2010; Rickli et al., 2010; Aarons et al., 2013; Pourmand et al., 2014; Zhao et al., 2014). The Seawater Array ($\varepsilon_{Hf} = 0.55 \times \varepsilon_{Nd} + 7.1$; Albarède et al., 1998) and the present-day Terrestrial Array ($\varepsilon_{Hf} = 1.55 \times \varepsilon_{Nd} + 1.21$; Vervoort et al., 2011) are shown for comparison, together with 3 other correlations identified in previous studies for fine-grained sediments (the 'zircon-free sediment array'; $\varepsilon_{Hf} = 0.91 \times \varepsilon_{Nd} + 3.10$; Bayon et al., 2009a), coarse-grained sediments (the 'zircon-bearing sediment array'; $\varepsilon_{Hf} = 1.80 \times \varepsilon_{Nd} + 2.35$; Bayon et al., 2009a), and Mongolian and Chinese dust clays (the 'clay-sized array'; $\varepsilon_{Hf} = 0.45 \times \varepsilon_{Nd} + 2.85$; Zhao et al., 2014).

## 2. Samples and methods

### 2.1. River-borne sediments and corresponding basin characteristics

The sediment samples analysed during the course of this study were collected near the mouth of rivers (Fig. 2). They correspond to either marine core-top (or sub-surface) or river bank sediments, both from major river systems and rivers draining basins with particular geological and climatic contexts (Table S1). For clarity, studied samples were organised into four groups (Table 1): 1) Major river systems with watersheds larger than 100,000 km$^2$ (e.g. Amazon, Congo, Mississippi, Nile); 2) Rivers draining sedimentary basins and/or various lithologies, but with drainage areas smaller than 100,000 km$^2$; 3) Rivers draining igneous/metamorphic terranes, such as the Proterozoic cratonic regions of Fennoscandia and Northern South America; 4) Rivers draining volcanic provinces (e.g. Kamtchatka peninsula, New Zealand, Réunion Island). Several minor rivers were sampled in North-West and Northern Ireland (Fig. 2C), draining a large variety of mono-lithological formations (i.e. Paleocene basaltic rocks, Paleozoic sedimentary formations, Proterozoic metamorphic rocks). These rivers are characterised by a similar climatic setting, and hence are ideal to investigate separately the role of lithology in controlling the Hf–Nd isotopic distribution in sediments. In addition, a total of seven sediment samples were collected along a 50-km transect along the flow path of the Loire River estuary, corresponding to various depositional environments. These latter samples were used to assess the analytical uncertainty associated with sediment sampling and preparation. The mean annual air temperatures (MAT) and precipitations

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