Mobility of U-series nuclides during basalt weathering: An example from the Deccan Traps (India)

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Abstract

U-series disequilibria have been measured by TIMS and MC-ICP-MS in dissolved phases and suspended sediments of the main basaltic rivers from the Deccan Trap region (India). For dissolved phases ($^{234}U/^{238}U$), ($^{230}Th/^{238}U$), and ($^{226}Ra/^{238}U$) range between 1.11 and 1.28, between 0.03×10^{-2} and 1.35×10^{-2}, and between 0.02 and 0.16 respectively. For suspended particles, ($^{234}U/^{238}U$), ($^{230}Th/^{238}U$), and ($^{226}Ra/^{238}U$) range from 0.95 to 1.1, from 1.23 to 1.59 and from 0.1 to 0.47 respectively. $^{230}Th–^{238}U$ and $^{234}U–^{238}U$ systems show direct links with indices of weathering. The data also indicate possible redistributions between the solid and aqueous phases and indicate that erosion processes do not presently operate at steady-state because present-day physical erosion rates are higher than expected (100–400 mm/ka versus 40 mm/ka for a steady-state process) for the Narmada and the Tapti basins. Models assuming either discrete or continuous particle leaching yield timescales for chemical erosion of 40–90 ka. In contrast with basins located in Northern latitudes, erosion in the Deccan basins does not appear to have been significantly disturbed during the last glaciation.

$^{234}U–^{238}U$ disequilibria in the dissolved phase of large rivers world-wide are also explored and two key factors are highlighted in this study: the dissolution of carbonates, releasing dissolved U at secular equilibrium and, the silicate weathering rate estimated at the scale of the watershed. In particular, a positive correlation is found between dissolved ($^{234}U/^{238}U$) and basaltic weathering rates for the Deccan that could reflect a key role of physical erosion since, for silicate monolithological basins, chemical erosion rates are primarily controlled by physical erosion rates. For basins with mixed lithology, dissolved organic matter also plays a key role in the preferential release of ^{234}U from silicate minerals. ($^{234}U/^{238}U$) ratios in rivers

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therefore reflect a mixture between U released by carbonate dissolution and ^{234}U preferentially released from silicate minerals located in high physical erosion and peat areas.

1. Introduction

Geochemical studies of large rivers and watersheds have recently been developed in order to constrain better continental erosion rates, the role of silicate weathering on atmospheric CO_2 consumption, and to quantify chemical fluxes to the oceans (Stallard and Edmond, 1981; Meybeck, 1997; Gaillardet et al., 1999; Dessert et al., 2001). Because rivers carry the products of continental erosion either as dissolved species, or as suspended particles, they can provide information on mechanical and chemical erosion rates at the scale of a watershed. Nevertheless, the time-scales over which these calculated erosion rates and the corresponding fluxes to the ocean apply is still poorly known and is expected to be variable from one area to another. For example, it has been shown that erosion rates have not remained constant over the past 10 ka in the Mackenzie basin in NW Canada, and must have been significantly disturbed by the last deglaciation (Vigier et al., 2001). Thus, the chemical and physical weathering flux estimates are highly dependent on the timescales of observation.

U-series nuclides (in particular ^{238}U, ^{234}U, ^{230}Th, and ^{226}Ra) provide powerful tools for studying chemical erosion because of their short half-lives – 1.6 ka to 250 ka – and because of their contrasting behaviour during weathering processes (Moore, 1967; Sarin et al., 1990; Andersson et al., 1995). For example, Th is widely found to be less mobile than U and Ra during mineral leaching (Latham and Schwarcz, 1987b), and therefore Th contents in filtered river waters are very low relative to U and Ra. Additionally, alpha-recoil effects during radio-active decay result in the preferential release of ^{234}U relative to ^{238}U into aqueous phase (Cherdyntsev, 1955; Rosholt et al., 1966, Kigoshi, 1971; Fleischer, 1980). More generally, estimating the timescales for chemical weathering requires (1) assessment of the open system behaviour of minerals in soils that are in constant or periodic contact with waters, and (2)

knowing the initial conditions, i.e. the chemical composition of source rocks prior to the inception of weathering. These two parameters are particularly difficult to constrain when studying erosion at a large spatial scale.

At the scale of a watershed, rivers can transport different populations of weathered particles derived from various locations within the basin, each of which may have variable lithologies and local weathering histories. However, for ^{238}U-decay series, the initial composition of fresh rocks (if they formed more than 1 Ma ago) is constrained by the condition of 'secular equilibrium', where activities of all nuclides are equal. In a watershed where the mean age of rocks is much greater than 1 Ma, this strongly constrains the initial composition of all source rocks drained by waters (carbonates and silicates). Moreover, the combined study of several nuclide pairs from the U-series chains is now possible in river samples mainly because Th isotopes can be measured precisely in low-level samples such as in the dissolved phases, using MC-ICP-MS (Turner et al., 2001; Pietruszka et al., 2002).

Here we investigate weathering processes of a basaltic region, the Deccan Traps, located in Central India. These monolithological watersheds allow us to explore the role of parameters other than lithology endmembers on the relative release of U-series nuclides to water, and to infer the role of erosion rates and the timescales of weathering in this area. Based on this data set and on a literature compilation, we have also examined the range in (^{234}U/^{238}U) ratios (where parentheses denote activity ratios) observed for silicate and mixed lithology basins.

2. Sampling location

The Deccan traps are located at the centre-west of the Indian peninsula (Fig. 1) and erupted about 65.6 ± 0.3 million years ago (Courtillot et al., 1986; Duncan and Pyle, 1988; Allègre et al., 1999), before