

## Trace, REE and Sr isotopic investigation in the core top sediments from Bay of Bengal

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Trace and rare earth element (REE) data obtained using ICP-MS and Sr isotopic composition determined by TIMS was used for the detailed study of geochemistry and transport pathways of the sediments from Bay of Bengal. These terrigenous sediments are derived from: a) the Irrawaddy-Salween rivers b) Peninsular Indian rivers, c) Ganges-Brahmaputra (G-B) rivers, and d) erosional products from the Indo-Burman ranges. The objective of the present study is to understand the geochemistry of sediments and identify their provenance in Bay of Bengal using  $^{87}\text{Sr}/^{86}\text{Sr}$  and trace elemental proxies. The results from core SK-157-27 (Lat:  $15^{\circ}48'N$ , Long:  $82^{\circ}8'E$ , water-depth: 1723m) show a high concentration of REE (~153 ppm) with high  $^{87}\text{Sr}/^{86}\text{Sr}$  ( $0.723726 \pm 20$ ), which may be due the source rock characteristics in the drainage basins of the Krishna and Godavari rivers. The core was retrieved from a region that represents mainly the terrigenous sediments of Godavari and Krishna rivers. The remaining three cores were collected from the northern part of the Ninetyeast Ridge. All these cores consist of fine grained sediments. The  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio ( $0.713689 \pm 12$ ) from core SK-157-14 (Lat:  $5^{\circ}11.56'N$ , Long:  $90^{\circ}5.97'E$ , water depth: 3306 m) suggest that the main source at this location is Irrawaddy-derived sediments with minor contributions from the Ganges-Brahmaputra rivers. High Ba and high Th/Ta ratio characterize the sediments from this core. The other two cores SK-157-15 (Lat:  $7^{\circ}48.55'N$ , Long:  $90^{\circ}15.92'E$ , water depth: 2870 m) and SK-157-16 (Lat:  $8^{\circ}46.86'N$ , Long:  $90^{\circ}18.42'E$ , water depth: 2920m) from the northern part of the Ninetyeast Ridge have relatively higher  $^{87}\text{Sr}/^{86}\text{Sr}$  values ( $0.716995 \pm 12$  and  $0.716670 \pm 24$ ) suggesting a greater contribution from the Ganges-Brahmaputra sediments. Sediments from these two cores have high Sr, high Ba and relatively low concentration of redox sensitive elements. The decrease in the redox sensitive elements is attributed to the oxic deep-water environment. Our results clearly suggest that the  $^{87}\text{Sr}/^{86}\text{Sr}$ , REE and trace elemental characteristics in the Bay of Bengal sediments are highly variable and depend on the source and pathways of sediments.

### References

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## Interrelations between Fe(II)/Fe(III) and U(IV)/U(VI) redox couples

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The mobility of uranium in natural environments is strongly affected by changes in its redox state. Reduction of U(VI) to U(IV) typically leads to an immobilization of uranium due to the low solubility of U(IV) minerals such as uraninite. Hence, initiating uranium reduction by stimulating microbial iron reduction has been proposed as a remediation strategy for aquifers contaminated with U(VI). Indications for U(VI) reduction concomitant to dissimilatory iron reduction have been obtained in several studies. Uranium might become directly reduced by iron reducing bacteria or indirectly by microbially produced Fe(II). It has been demonstrated that Fe(II) containing iron minerals such as magnetite and green-rust or Fe(II) adsorbed onto iron oxide surface can act as reductants for U(VI). However, also the oxidation of U(IV) by iron oxides has been reported. Consequently, it seems that both, uranium reduction and oxidation, can be coupled to Fe(II)/Fe(III) redox transformations. Here we want to discuss possible reasons for these apparently contradictory observations. Thermodynamic considerations show that the energetically favourable direction of a coupled U-Fe redox reaction depends on U and Fe concentrations and their speciation in the system. In addition to the speciation in solution, the thermodynamic stabilities of the solid phases, which are involved in the reactions, are of great importance. In particular, reduction of U(VI) becomes less favourable if the  $U^{4+}$  activity is not controlled by the solubility of crystalline uraninite but by an U(IV) phase with higher solubility.

In a series of biotic and abiotic incubation experiments involving the metal reducing organism *Shewanella putrefaciens* and nanoparticulate hematite occurrence and extend of uranium reduction was investigated. Results from X-ray absorption spectroscopy reveal that, in the presence of hematite, U(VI) was not or only partially reduced. We propose that formation of crystalline uraninite is hindered in the experiments and by this complete reduction of U(VI) is inhibited. This hypothesis is supported by extended X-ray absorption fine structure (EXAFS) analyses which indicate that uraninite is not the primary product of microbial U(VI) reduction. Microbially produced U(IV) is either bound in an amorphous precipitate or remains adsorbed at the bacterial cell wall. It was further found that reoxidation of "labile" U(IV) by addition of hematite is possible. Occurrence or inhibition of uraninite formation might therefore explain the contradictory observations regarding the coupling of U and Fe redox reactions. That is, oxidation of U(IV) by iron oxides is feasible and only partial reduction of U(VI) can be achieved when uraninite formation is inhibited.