

87Sr/86Sr and 143Nd/144Nd for disentangling anthropogenic and natural REE contributions in river water during flood events.

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The sustainable management of water resources is one of the greatest challenges of the 21st century. Water is a vital resource that is increasingly put under pressure from multiple perspectives. While the global population is on the rise, socio-economic development makes equally rapid progress – eventually compromising access to clean water bodies. Multiple pollution sources constitute an immediate threat to aquatic ecosystems and are likely to cause long lasting contaminations of water bodies that are critical for drinking and/or irrigation water production. There is a pressing need for an adequate quantification of anthropogenic impacts on the critical zone of river basins and the identification of the temporal dynamics of these impacts. As an example, despite the work done to assess the environmental impact of REE pollutions in larger river systems, we are still lacking information on the dynamics of these anthropogenic compounds in relation to rapid hydrological changes. Filling these knowledge gaps is a pre-requisite for the design and implementation of sustainable water resources management strategies.

In order to better constrain the relative contributions of both anthropogenic and geogenic trace element sources we propose using a multitracer approach combining elemental and $87\text{Sr}/86\text{Sr}$, $143\text{Nd}/144\text{Nd}$, and $206\text{Pb}/207\text{Pb}$ isotopic ratios. The use of these three separate isotopic systems together with REE concentrations is new in the field of anthropogenic source identification in river systems.

We observed enrichments in Anthropogenic Rare Earth Elements (AREE) for dissolved Gd and suspended Nd loads of river water. With increasing discharge, AREE anomalies progressively disappeared and gave way to the geogenic chemical signature of the basin in both dissolved and suspended loads. The isotopic data confirm these observations and shed new light on the trace elements sources. On the one hand, dissolved loads have peculiar isotopic characteristics and carry mainly limestone-derived and anthropogenic Sr and Nd as well as significant amounts of anthropogenic Pb. On the other hand, the results clearly indicate that anthropogenic contributions impact the suspended loads in all hydrological conditions.

This study demonstrates that (i) the composition of the AREE pool is characteristic of a given river basin and controlled by the different anthropogenic contributions located in a specific study area and (ii) the anthropogenic contributions to the river may change not only Pb, but also Sr and Nd isotopic compositions in both dissolved and suspended loads. This is of importance for future provenance studies in the critical zone of polluted river basins.