

Assessment and fluxes of Rare Earth Elements in the Garonne River (SW France, 2003-2017): increasing Gd anomaly?

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New and rapidly developing technologies imply the emission of emerging potentially toxic contaminants such as Rare Earth Elements (REE). The emerging presence of REEs coming from large river systems and the impact of urban areas on annual REE fluxes in fluvio-estuarine systems remains widely unknown. The Garonne River ($Q=650 \text{ m}^3 \cdot \text{s}^{-1}$; $A=55,000 \text{ km}^2$), main tributary of the Gironde Estuary, drains water from ~20% of the French landsurface hosting about 5,200,000 inhabitants and two major metropolitan areas (Toulouse and Bordeaux). Based on long-term monitoring (2003-2017) of water discharges and dissolved REEs concentrations (analyzed with a Triple Quadrupole ICP-MS) at the outlet of the Garonne Watershed upstream Bordeaux, this study aims at assessing REE anomalies and evaluating temporal evolution of annual dissolved REE fluxes into the estuary. Additionally, we analyzed potential urban sources (e.g. domestic, medical, run-off) in the urban area of Bordeaux (1,200,000 inhab.) to evaluate their respective contributions.

Gadolinium (Gd) showed clear anomalies with annual average anthropogenic concentrations ranging from 2.5 to 9.3 ng.L⁻¹. If variations in annual Gd fluxes depend on hydrology, anthropogenic Gd fluxes have shown an overall increasing trend from 36 kg.yr⁻¹ in 2003 to 75 kg.yr⁻¹ in 2017. Sewer waters from the third hospital complex of France contributed 30% of the incoming daily flux to Bordeaux major Waste Water Treatment Plant (WWTP), as Gd is used as contrast agent for magnetic resonance imaging (MRI). Due to weak removal efficiency in the WWTP, Bordeaux significantly contributes (> 16 kg.yr⁻¹) to Gd fluxes in the Garonne River. The temporal evolution of anthropogenic Gd fluxes in the Garonne River may be related with the growing regional population and the increasing number of MRI instruments highlighting the impact of new high-tech applications in urban areas and their potential harmful effects on fluvio-estuarine systems in the future. Control of urban pollutants is a major issue in a context of rapidly growing urbanization.