Tracing wastewater input into the North Sea using anthropogenic Gd anomalies: Evidence for widespread MRI contrast agent contamination of river estuaries of Elbe, Ems and Weser and the southern North Sea including the German Bight

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Many trace elements gained societal and economic importance in recent decades due to their growing demand in hightechnology applications, including medicinal applications and renewable energies. Their increasing application in enabling technologies results in a tremendously growing and largely unconstrained input into the environment. Knowledge of the anthropogenic input, the environmental behavior and the toxicity of such compounds are for many of these elements still in their infancy.

The widespread use of the rare earth element gadolinium as a contrast agent in magnetic resonance imaging scans has led to a significant contamination of riverine and coastal environments worldwide. Due to their inert character, these compounds are not removed in most wastewater treatment plants and are introduced into the natural environment, including surface water, (soil) pore water and groundwater. Inevitably, it enters the food chain via drinking water and potential incorporation into plants. The concentrations that are observed today are probably non-toxic and current studies on the bioavailability of commercially available Gd-contrast agents indicate a very conservative behavior. However, the anthropogenic Gd anomalies in normalized Rare Earth Element and Yttrium (REY) patterns of natural waters are ideal far-field tracers capable to track the spatial and temporal distribution of potentially hazardous, chemically-inert wastewater-derived substances from source to sink

We analyzed >100 water samples that were collected in December 2020 during research cruise M169 "TRAM" ("Tracing geogenic and anthropogenic critical high-technology metals in the southern North Sea") with R/V Meteor. We present evidence for an increasingly wide-spread anthropogenic input and distribution of wastewater-derived Gd in the southern North Sea, including the World Natural Heritage area of the German Wadden Sea, and the Weser, Ems and Elbe estuaries (Fig. 1). Our new dataset indicates a roughly ten-fold increase in anthropogenic Gd concentrations in the estuaries and a circa three-fold increase in parts of the Southern North Sea since 2005. Complementary ultrafiltration and DGT passive sampling data show that the anthropogenic Gd exclusively occurs in the truly dissolved element pool of the discharged river and estuarine waters and that a major share of anthropogenic Gd is bound in stable, inert complexes that is likely not bioavailable.



Fig. 1: Anthropogenic Gd (in % of total Gd) in 0.2µm filtered water samples.