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Rare earth element distributions in the West Pacific: Trace element sources and conservative vs. non-conservative behavior



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ABSTRACT

Recent studies suggest that transport and water mass mixing may play a dominant role in controlling the distribution of dissolved rare earth element concentrations ([REE]) at least in parts of the North and South Atlantic and the Pacific Southern Ocean. Here we report vertically and spatially high-resolution profiles of dissolved REE concentrations ([REE]) along a NW-SE transect in the West Pacific and examine the processes affecting the [REE] distributions in this area. Surface water REE patterns reveal sources of trace element (TE) input near South Korea and in the tropical equatorial West Pacific. Positive europium anomalies and middle REE enrichments in surface and subsurface waters are indicative of TE input from volcanic islands and fingerprint in detail small-scale equatorial zonal eastward transport of TEs to the iron-limited tropical East Pacific. The low [REE] of North and South Pacific Tropical Waters and Antarctic Intermediate Water are a long-range (i.e., preformed) laterally advected signal, whereas increasing [REE] with depth within North Pacific Intermediate Water result from release from particles. Optimum multiparameter analysis of deep to bottom waters indicates a dominant control of lateral transport and mixing on [REE] at the depth of Lower Circumpolar Deep Water (>3000 m water depth; \sim 75–100% explained by water mass mixing), allowing the northward tracing of LCDW to \sim 28°N in the Northwest Pacific. In contrast, scavenging in the hydrothermal plumes of the Lau Basin and Tonga-Fiji area at 1500-2000 m water depth leads to [REE] deficits (~40-60% removal) and marked REE fractionation in the tropical West Pacific. Overall, our data provide evidence for active trace element input both near South Korea and Papua New Guinea, and for a strong lateral transport component in the distribution of dissolved REEs in large parts of the West Pacific.

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1. Introduction

Dissolved rare earth element concentrations ([REE]) in seawater are used to trace sources of micronutrients and other trace elements (TE) to the ocean by exploiting systematic changes and differences in their behavior (e.g. Lacan and Jeandel, 2001; Grenier et al., 2013). The chemically coherent group of REEs shows a characteristic light (LREE) over heavy (HREE) REE fractionation in seawater that is due to preferential adsorption of LREE onto particles and stronger complexation of HREE by carbonate ions (e.g., Byrne and Kim, 1990). Elevated surface water [REE] are typical for

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local continental input of TEs to the ocean via coastal aquifers, margin sediments, dust, or rivers (e.g., Sholkovitz et al., 1999; Duncan and Shaw, 2003; Jeandel et al., 2013; Grenier et al., 2013; Fröllje et al., 2016). Post Archean Australian Shale (PAAS) (Taylor and McLennan, 1985) normalized positive europium (Eu) anomalies in seawater have been shown to be indicative of input or exchange with basaltic rocks or sediments (e.g., Grenier et al., 2013). Pore fluids from bottom sediments have recently been hypothesized as an REE source to deep waters in some areas of the ocean (Abbott et al., 2015) but not in others (Yang and Halev. 2016). The long-standing interpretation of the distribution of REEs in seawater, with generally low concentrations at the surface and increasing concentrations with depth, is therefore that of a dominance of vertical processes (input, scavenging, release) (e.g., Elderfield and Greaves, 1982). Several recent studies using highresolution transects of [REE] through the ocean, have suggested an

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additional lateral control on REE distributions. The evidence came from nearly constant vertical neodymium concentrations ([Nd]) at the depth of vigorously flowing North Atlantic Deep Water (e.g., Stichel et al., 2015), and the upwelling of high [Nd] with deep waters in the southern South Pacific and Atlantic (Basak et al., 2015; Hathorne et al., 2015). Using a zonal [REE] section in the South Atlantic, Zheng et al. (2016) provided evidence for a dominance of water mass mixing processes on the distribution of REEs. Due to the potential of dissolved seawater [REE] to identify sources and processes associated with TE input, scavenging, and transport, REEs are a diagnostic tool to better understand TE cycling in the ocean (e.g., Lacan and Jeandel, 2001; Grenier et al., 2013; Hathorne et al., 2015).

Trace element cycling in the West Pacific is of particular interest as this key study area is surrounded by volcanic island margins that are thought to be an important source of micronutrients and other TEs to the ocean. In addition, the West Pacific is connected to the iron-limited high-nutrient, low-chlorophyll (HNLC) area in the East Pacific through the equatorial zonal current system that may act as a conduit of micronutrients and other TEs from the tropical West Pacific to the HNLC area in the East Pacific (e.g., Lacan and Jeandel, 2001; Radic et al., 2011; Grenier et al., 2013).

Here, we present the first vertically and spatially high-resolution transect of dissolved [REE] from the West Pacific between South Korea and Fiji (Fig. 1(a)). We classify the relative importance of the different sources and processes that control the seawater REE cycling in the West Pacific and highlight the small-scale equatorial zonal and long-range lateral transport of [REE].

Our previous study using [Nd] and Nd isotope compositions (expressed as ε_{Nd}) from the same samples suggested input and/or modification of the seawater ε_{Nd} signal and a dominance of water mass mixing on [Nd] (Behrens et al., 2018), here we quantify the component of Nd controlled by water mass mixing and transport relative to that influenced by non-conservative Nd addition or removal.

2. Study area and circulation

The study area and circulation are presented in detail in Behrens et al. (2018). Briefly, the NW–SE transect from South Korea to Fiji comprises twelve full water column stations that were sampled during cruise SO223T (TransGeoBioOc) with R/V *Sonne* (GEOTRACES Process Study GPpr04) from September to October 2012. We divide the transect into two sections: the South Korea to open ocean section (SKO) that includes stations GeoB17001 to GeoB17005 (referred to as stations 01–05), and the tropical West Pacific section (TWP) that includes stations GeoB17011 to GeoB17019 (referred to as stations 11–19) (Figs. 1(a), (b)).

The surface (upper 50 m water depth) and subsurface circulation is dominated by the North Equatorial Current (NEC) and the Kuroshio Current (KC) in the study area of the SKO section (stations 03–05), and by currents of the complex equatorial zonal current system in the TWP (stations 15–19): the North Equatorial Countercurrent (NECC), surface currents (SC), the Equatorial Undercurrent (EUC), the Equatorial Intermediate Current (EIC), the South Equatorial Countercurrent (SECC), and the South Equatorial Current (SEC) (e.g., Wyrtki and Kilonsky, 1984; Fine et al., 1994; Reid, 1997; Tomczak and Godfrey, 2003; Talley et al., 2011; SC: determined from real time ocean surface current analyses for the time of the SO223T cruise, http://www.oscar.noaa.gov/) (Fig. 1(a)).

Subsurface waters are North and South Pacific Tropical Water (NPTW and SPTW) at 61–250 m and 101–200 m water depth in our study area, respectively. NPTW differs from SPTW by its less pronounced subsurface salinity maximum (e.g., Qu et al., 1999). It originates in the central subtropical North Pacific and is transported by the NEC and the KC towards stations 03–05 (e.g., Fine

et al., 1994; Qu et al., 1999) and the Mindanao Current (MC), that feeds the NECC, towards stations 11 and 14 (e.g., Fine et al., 1994). SPTW originates in the oligotrophic South Pacific Subtropical Gyre (e.g., Qu et al., 2013). It flows from its formation region westwards within the SEC into our study area (stations 17–19), and further within the New Guinea Coastal Undercurrent (NGCU) along the coast of PNG and finally turns eastward within the EUC (stations 15, 16) (e.g., Fine et al., 1994).

North Pacific Intermediate Water (NPIW) flows along the North Pacific Subtropical Gyre and is found within the NEC and the KC (stations 02–05, at 498–1004 m water depth) in our study area (e.g., Qu et al., 1999) (Figs. 1(a), (b)). Antarctic Intermediate Water (AAIW), originating in the Southeast Pacific, is entrained into the South Pacific Subtropical Gyre and flows within the SEC into our study area (stations 18, 19, at 501–893 m water depth), where it reaches the equatorial TWP via the NGCU (stations 15–17) (Figs. 1(a), (b)). In the equatorial TWP, AAIW is transported eastward and then returns westwards to the northern TWP (stations 11, 14) (e.g., Kawabe and Fujio, 2010) (Fig. 1(b)).

The deep water circulation in our study area is marked by the northward flow of Southern Ocean sourced Upper and Lower Circumpolar Deep Water (UCDW and LCDW, respectively) and the southward flow of North Pacific Deep Water (NPDW) (e.g., Kawabe and Fujio, 2010) (Fig. 1(b)).

3. Materials and methods

3.1. Onboard procedures

Seawater was sampled for the determination of dissolved nutrient (phosphate, nitrate, silicate) and REE concentrations at twelve full water column stations. Details of onboard seawater sampling followed established GEOTRACES protocols (Pahnke et al., 2012; http://www.geotraces.org/images/stories/documents/ intercalibration/Cookbook.pdf) and are described in Behrens et al. (2018). All reagents used were of ultra-clean quality (Fisher Scientific optima grade). Briefly, seawater samples were filtered directly from Niskin bottles (or the ship's contamination-free seawater intake in the lab for the collection of surface water) through AcroPak500 (0.8/0.2 µm pore size) filter cartridges into previously acid-cleaned 20 mL HDPE bottles (for nutrient analysis) or 250 mL LDPE bottles (for REE analysis) using Teflon-lined Tygon tubing and PP fittings. Seawater samples were poisoned with 3.5% mercury chloride for nutrient analysis or acidified to pH ≤ 2 using 6 N HCl for REE analysis. All samples were stored onboard for further processing in the home laboratory at the ICBM (Institute for Chemistry and Biology of the Marine Environment). Total procedural onboard blanks of (1) onboard MilliQ water (18.2 M Ω cm), (2) MilliQ water brought onto the ship from a Millipore system at the ICBM, and (3) MilliQ water half-and-half from onboard and ICBM were also acidified to pH <2 using 6 N HCl and stored onboard.

3.2. Nutrient and REE analysis

At the ICBM, nutrients were measured photometrically using a Multiscan GO Microplate Spectrophotometer (Thermo Scientific) and an EasyChem Plus discrete analyzer (AMS-SYSTEA). Nitrate (NO_3^-) was determined from nitrite and NO_x^- , following the method described by Schnetger and Lehners (2014). Reference solutions of known concentrations were used to check for precision and accuracy (<10%).

Seawater REEs were pre-concentrated and purified using the automated seaFAST-pico system in offline mode (Elemental Scientific Inc., Nebraska, USA) and quantified by isotope dilution (ID) analysis as described in Behrens et al. (2016). Briefly, depending on the expected [REE], seawater samples (pH \leq 2) of ~11 mL, or

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