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Seasonal variations in dissolved neodymium isotope composition in the Bay of Bengal

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ABSTRACT

Constraining the dissolved neodymium (Nd) cycle in the ocean is paramount for using Nd isotopic composition (ε_{Nd}) as a tracer to reconstruct deep-sea paleocirculations or continental weathering on different time scales. Dissolved ε_{Nd} has been measured in seawater samples from six hydrological stations collected along ~89°E North–South transect in the Bay of Bengal (BoB) in order to assess the impact of seasonal freshwater and sediment discharges from the continental river systems. Seawater samples collected in this study during June 2012 reveal more radiogenic ε_{Nd} (a difference of ~2 Epsilon units for the upper 2000 m, and ~0.5 Epsilon unit below 2000 m) and ~3–8 pmol/kg lower Nd concentrations than the reported values of nearby seawater samples collected in November 2008. These observations are most plausibly explained by a seasonal variations in dissolved Nd concentrations and ε_{Nd} in the BoB, induced by seasonal variations in the freshwater and sediment discharges from the Ganges–Brahmaputra (G–B) river system. However, we cannot entirely exclude the possibility of spatial differences given that the water stations collected in this study are not exactly the same positions collected in November 2008. A two-box model suggests, (1) the particulate Nd inputs from the G–B river system mainly control the seasonal shift of ε_{Nd} observed in the BoB seawater, and (2) a very rapid Nd exchange exists between lithogenic particles and seawater (at least on the scale of a few months). Seasonal changes in seawater ε_{Nd} may also occur in other marginal seas and in the outflows of major rivers, and these need to be taken into account when using the ε_{Nd} proxy in the ocean.

1. Introduction

Dissolved neodymium (Nd) isotopic composition (expressed as εNd) is not homogeneous in oceans due to its shorter residence time (Frank, 2002; Siddall et al., 2008; Tachikawa et al., 2003) compared to the global turnover time ~1500 yr of deep waters (Broecker and Peng, 1982). Nd in oceans is mainly derived from lithogenic inputs by rivers and dust transport, and "Boundary Exchange (BE)" processes that occur at the continental margin and dissolution/scavenging of lithogenic particles in the water column (Frank, 2002; Goldstein and Hemming, 2003; Lacan and Jeandel, 2005; Piepgras et al., 1979). ε_{Nd} has been widely used as a tracer for modern (Goldstein and Hemming, 2003; Jeandel, 1993) and past oceanic circulation, and continental weathering on different times scales (Burton and Vance, 2000; Colin et al., 2010; van de Flierdt and Frank, 2010; von Blanckenburg, 1999). However, the Nd cycling and the time evolution of ε_{Nd} in oceans, particularly during climatic shifts, are still under debate (Copard et al., 2010; Lacan et al., 2012; Rousseau et al., 2015; van de Flierdt et al., 2016).

Several studies (Bertram and Elderfield, 1993; Jeandel et al., 1995; Lacan and Jeandel, 2005), in particular for the BE process, have indicated that reversible scavenging and interaction between dissolved and particulate phases control oceanic ε_{Nd} distribution.

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Compilation of ε_{Nd} data, as well as modeling studies (Arsouze et al., 2009; Jeandel Arsouze et al., 2007), have revealed that lithogenic Nd flux from the BE process, which occurs at the continental margin (1.1×10^{10} g/yr), is much higher than both dissolved river discharge (2.6×10^{8} g/yr) and atmospheric inputs (1.0×10^{8} g/yr) and may constitute the dominant Nd source to the ocean. On a regional scale, several studies have been conducted over the past decade in marginal seas and sites adjacent to major river mouths such as the Bay of Bengal (BoB), the Arabian Sea, the South China Sea, the Baltic Sea, the Caribbean Sea and the Amazon Estuary, with the aim of constraining specific processes controlling dissolved Nd concentration and ε_{Nd} distribution (Chen et al., 2013; Goswami et al., 2014; Osborne et al., 2014; Rousseau et al., 2015; Singh et al., 2012; Wu et al., 2015a). These studies highlight the fact that high river sediment discharge contributes significantly to the Nd budget, and modifies the spatial distribution of ε_{Nd} at least in the upper water masses of the ocean. The adsorption and desorption of Nd from fluvial particles were highlighted earlier as crucial processes effecting the dissolved Nd concentration and ε_{Nd} in seawater (Nozaki and Alibo, 2003; Rousseau et al., 2015; Sholkovitz et al., 1994). Hence, seawater ε_{Nd} could also be strongly influenced by variations in freshwater and sediment discharges from the nearby rivers in marginal seas and offshore waters close to major river estuaries. However, at the present time, the potential effect of seasonal variations in river inputs on ocean ε_{Nd} has not been measured, thus limiting our understanding of modern Nd cycle and the application of ε_{Nd} for the determination of water mass mixing and continental weathering inputs.

As ε_{Nd} values of the Ganges–Brahmaputra (G–B) river sediments ($\sim -16.0 \pm 2.0$; $n = 28$) (Galy and France-Lanord, 2001; Goldstein et al., 1984; Lupker et al., 2013; Singh and France-Lanord, 2002) contrast strongly with those of water masses from the Northern Indian Ocean (~ -9 for surface northern Indian water and ~ -8 for the bottom water of the southern BoB) (Amakawa et al., 2000; Jeandel et al., 1998), seawater ε_{Nd} of the BoB can be used to assess Nd lithogenic input from the Himalayan system. The dissolved Nd concentrations and ε_{Nd} have been investigated by Singh et al. (2012) in many shallow and deep profiles sampled in November 2008 along a North–South transect in the BoB (at about 87°E): a major objective was to establish the effect of water mass mixing and Nd release from lithogenic sediment mostly delivered by the G–B river system. Singh et al. (2012), based on an inverse model approach, highlighted the fact that Nd contributions from all of the different water masses in the BoB are inadequate to balance the measured Nd abundances. They proposed that the release of unradiogenic Nd (-16) from particulate phases supplied by the G–B river system needs to be an additional source. Moreover, sediment traps deployed in the BoB have revealed monsoon-induced seasonal variations in lithogenic sediment fluxes (Winter: $\sim$20 mg/m^2/d, Summer: $\sim$60 mg/m^2/d) (Unger et al., 2003). Taking into consideration the fact that exchange of Nd between particles and seawater could be rapid (Rousseau et al., 2015), such seasonal change could also have an influence on the spatial distribution of dissolved Nd concentrations and ε_{Nd} in the BoB. Consequently, the BoB constitutes an ideal geographical area to constrain the influence of seasonal river discharge from the vast G–B river basin on the dissolved Nd concentration and ε_{Nd} distribution, and to examine its potential implications to use the ε_{Nd} proxy for water mass mixing and continental weathering reconstructions.

In this study, dissolved ε_{Nd} has been analyzed for six hydrological stations sampled along the $\sim$89°E meridian in the BoB. Major goals of this study are (i) to determine the spatial distribution of dissolved ε_{Nd} in June 2012 at a period of the year corresponding to the beginning of rainfall in the Southwest monsoon (SWM) the G–B river system; (ii) to compare the spatial distribution of dissolved ε_{Nd} in June 2012 to those reported earlier for seawater samples collected in November 2008 (Singh et al., 2012) in order to assess the possible impact of the seasonal discharge of the G–B river system on the geochemical cycle of Nd in the BoB. Such results are in turn important for re-evaluating past seawater ε_{Nd} investigations in terms of continental weathering and past ocean circulation reconstruction in sediment from marginal seas and sites adjacent to major river mouths (van de Flierdt et al., 2016; Wu et al., 2015b).

2. Hydrological setting

Freshwater and sediment inputs to the BoB are dominated by the G–B river system with annual freshwater and sediments supplies of 1.0×10^{12} m^3/yr and 1.0–1.5 $\times$ 10^9 t/yr, respectively (Milliman and Meade, 1983). This river system drains the Himalayas, characterized by one of the world's highest rates of physical and chemical erosion (Milliman and Meade, 1983). Most of the annual G–B river discharge (90%) is transported to the BoB during the SWM period (Shetye et al., 1996; Wyrtki, 1973). Rivers originating in the Indo-Burman ranges and the Irrawaddy River contribute freshwater input of 0.4×10^{12} m^3/yr and a total sediment input of 2.3 to 3.3×10^8 t/yr into the BoB (Robinson et al., 2007), whereas the Indian peninsular rivers in the west of the BoB supply a total freshwater input of 0.2×10^{12} m^3/yr and sediment input of 2.4×10^8 t/yr (Tripathy et al., 2011). The latter discharges are, therefore, much smaller than those of the G–B river system (Curray and Moore, 1971; Milliman and Meade, 1983).

The Indian monsoon drives a seasonal reversal of the surface circulation in the BoB. Surface water masses in the top 100 m include the Eastern Indian Ocean Surface Water (EIOW), Arabian Sea High Salinity Water (ASHS) in the southern BoB, and the BoB Lower Salinity Water (BoBLS) in the northern BoB (Fig. 1b). The intermediate waters at depths of 100–1500 m, are mainly BoB Intermediate Water (BoBIW) in the northern BoB and North Indian Intermediate Water (NIIW) in the southern BoB (Shankar et al., 2002; Wyrtki, 1973). The bottom waters of the BoB (below 2000 m) are strongly influenced by the Antarctic Bottom Water (AABW) (Kolla et al., 1976; Wyrtki, 1973).

3. Materials and methods

89 seawater samples distributed over six stations (MONO01, 02, 03, 4b, 06, 09) were collected in the middle of the BoB along the 89°E meridian from $\sim$18°N to $\sim$8°N during the MONOPOL cruise, onboard R/V *Marion Dufresne*, in June, 2012 (Fig. 1). Seawater samples and the relevant hydrographic parameters (potential temperature and salinity) were collected using a Sea-Bird SBE 911plus CTD-Rosette system equipped with 24 Niskin bottles. All seawater samples were filtered on-board using AcroPak 500 capsule filters (0.8–0.45 µm) and transferred to $\sim$10–12 L acid-cleaned (1 M HCl) cubitainers before being acidified to a pH lower than 2 with suprapur 6 N HCl. Nd was purified from seawater samples following the analytical procedures described in detail by Lacan and Jeandel (2001) and Wu et al. (2015a). In brief, seawater REEs were pre-concentrated using SepPak Classic C18 cartridges loaded with a HDEHP/H_2MEHP complexing agent in a land based clean lab in Laboratoire GEOsciences Paris-Sud (GEOPS), University of Paris-Saclay. Solutions were then passed through a cationic resin (AG50W-X8) and finally Nd was extracted and purified using an Eichrom Ln-Spec resin following the method described in detail by Copard et al. (2010).

The ^{143}Nd/^{144}Nd ratios of all purified Nd fractions were analyzed using the ThermoScientific Neptune^{Plus} Multi-Collector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS) hosted at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE) in Gif-sur-Yvette. The Nd isotopic composition was analyzed