



Invited review

Neodymium associated with foraminiferal carbonate as a recorder of seawater isotopic signatures

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ABSTRACT

Neodymium isotopic ratios in marine environments have been used as a tracer of water masses and exchange processes between dissolved and particulate phases. The interest in this tracer has been growing with improvement of our knowledge on its chemical behaviour in the modern ocean and the identification of sedimentary phases that preserve past seawater ϵ_{Nd} values. In the last few decades the Nd isotopic composition measured on Fe–Mn crusts, sediment leachates, bulk carbonate fraction, corals and fish teeth have been increasingly interpreted in the context of understanding the role of the ocean in paleoclimate changes. In particular, calcareous foraminiferal tests (shells) have acquired increasing attention as an archive of seawater Nd isotopic signatures, because it allows continuous high-resolution records to be measured and directly compared to other proxies including stable isotopes and trace metals. The main challenge of interpreting the Nd isotopic composition of foraminifera is determining the origin of the Nd preserved within them. In this review, we present an overview of methodological progress including that of bulk foraminifera and microanalyses within foraminiferal tests, as well as geochemical meaning of extracted Nd concentrations and isotopic compositions. The growing body of evidence suggests that Nd isotopic signatures of sedimentary planktonic foraminifera correspond to bottom water values rather than surface water ones. The Nd-rich phases associated with sedimentary foraminifera are adhesive nano-scale particles of Mn and Fe oxides and hydroxides, and Mn-rich carbonates formed within layers of foraminiferal calcite. Mechanical cleaning to remove clay minerals is likely to be sufficient in most cases to reconstruct past bottom water circulations. Unresolved issues include the potential influence of pore water Nd on ϵ_{Nd} values extracted from sedimentary foraminiferal tests under different sedimentological and oceanographic conditions.

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1. Introduction

Neodymium is a member of the rare earth elements (REEs), and its major sources to the ocean are continental inputs, including dissolved and particulate river loads, atmospheric inputs and continental margin sediments (Piepgras et al., 1979; Goldstein and Jacobsen, 1987; von Blanckenburg and Igel, 1999; Frank, 2002; Goldstein and Hemming, 2003; Tachikawa et al., 2003; Jeandel et al., 2011). The isotope ^{143}Nd , roughly one quarter of the abundance of the seven stable isotopes of Nd, is produced by radiogenic decay by α -decay of ^{147}Sm , another member of REEs. Because of similar chemical and physical properties of Nd and Sm, the Nd isotopic composition ($^{143}\text{Nd}/^{144}\text{Nd}$ or $\epsilon_{\text{Nd}} = [(^{143}\text{Nd}/^{144}\text{Nd})_{\text{sample}}/$

$(^{143}\text{Nd}/^{144}\text{Nd})_{\text{CHUR}} - 1] \times 10^4$, where CHUR is chondritic uniform reservoir representing the bulk Earth $^{143}\text{Nd}/^{144}\text{Nd}$ value: Jacobsen and Wasserburg, 1980) of rocks varies with its Sm/Nd ratio and the age. For instance, old cratons surrounding the North Atlantic have ϵ_{Nd} values as low as -30 whereas young volcanic mantle-derived rocks abundant in the Pacific basin have positive ϵ_{Nd} values (Jeandel et al., 2007).

The present-day seawater ϵ_{Nd} distribution is heterogeneous, reflecting the oceanic Nd residence time of about 200–1000 years (Tachikawa et al., 1999) that is shorter than mean mixing time of deep ocean (about 1500 years) (Broecker and Peng, 1982). The three main factors controlling seawater ϵ_{Nd} distribution in the modern ocean (and presumably over the late Quaternary period) are circulation of water masses including both advection and diffusion, interaction between dissolved and particulate Nd in the ocean, and to lesser extent, changes in geological sources of Nd to the ocean (Piepgras et al., 1979; Bertram and Elderfield, 1993; Lacan and

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Jeandel, 2001; Tachikawa et al., 2003; Lacan and Jeandel, 2005; Jeandel et al., 2011; Rempfer et al., 2012). Dissolved Nd in seawater is moderately reactive with particles via processes of adsorption/desorption and dissolution/precipitation (Bertram and Elderfield, 1993; Sholkovitz, 1993; Tachikawa et al., 1999; Jones et al., 2012). Oxides and oxyhydroxides of Mn and Fe are known to adsorb Nd in solution because of the active surfaces (Koeppenkastrup and De Carlo, 1992; Sholkovitz, 1993). Complexation with organic ligands also contributes to dissolved-particulate interaction (Johannesson et al., 2004; Tang and Johannesson, 2010) although Nd is generally present as a carbonate ion in seawater. These interplays could affect quasi-conservative behaviour of Nd isotopic ratios in high-particle concentration regions. In contrast, the ϵ_{Nd} values in the open ocean can be used as water mass tracers (Piepgras and Wasserburg, 1983, 1987; Jeandel, 1993; Jeandel et al., 1998; Goldstein and Hemming, 2003; Lacan and Jeandel, 2004a,b). Indeed, seawater Nd isotopic signatures of intermediate, deep and bottom waters vary during thermohaline circulation from low values ($\epsilon_{\text{Nd}} = -11.5 \pm 2.7$, $n = 171$; Lacan et al., 2012 and references therein) in the North Atlantic to higher values in the North Pacific ($\epsilon_{\text{Nd}} = -4.1 \pm 1.3$, $n = 111$; Lacan et al., 2012 and references therein). Modelling studies also support that the Nd isotopic ratios are quasi-conservative for deep waters far from the continental margins (Jones et al., 2008).

The interest in Nd isotopic compositions in marine environments has been growing because of the possibility to trace past circulation of deep water masses (Goldstein and Hemming, 2003) using various fractions such as Fe–Mn crusts (Frank, 2002 and references therein), sediment leachates (Rutberg et al., 2000; Bayon et al., 2002, 2004; Piotrowski et al., 2004; Piotrowski et al., 2005; Gutjahr et al., 2007, 2008; Piotrowski et al., 2009; Haley and Polyak, 2013), bulk carbonate fraction (Gourlan et al., 2010), corals (van de Flierdt et al., 2010; Robinson et al., 2013 and references therein) and fish teeth (Martin and Haley, 2000; Martin and Scher, 2004; Horikawa et al., 2011). The isotopic compositions recorded in Fe–Mn nodules and crusts have been widely used to reconstruct the oceanic circulation on long geological timescales (Piepgras et al., 1979; von Blanckenburg and Igel, 1999; Frank, 2002 and references therein). More recently, the use of Nd preserved in dispersed Fe–Mn oxides reductively leached from bulk sediments has been developed to investigate bottom water circulation during the late Quaternary with sufficient temporal resolution to resolve glacial-interglacial and millennial changes (Rutberg et al., 2000; Piotrowski et al., 2004, 2005; Gutjahr et al., 2008; Pahnke et al., 2008; Haley et al., 2008a,b; Piotrowski et al., 2009; Crockett et al., 2011; Gutjahr and Lippold, 2011; Chen et al., 2012; Xie et al., 2012; Maccali et al., 2013; Werner et al., 2013). Nonetheless, several studies have revealed that the chemical leaching procedures generally used for extracting the oxides can partially dissolve labile terrigenous fractions (e.g. volcanogenic matter and pre-formed Fe–Mn coatings) and consequently might bias leachate ϵ_{Nd} values from bottom water signatures in some locations (Bayon et al., 2004; Roberts et al., 2010; Elmore et al., 2011; Piotrowski et al., 2012; Wilson et al., 2013; Kraft et al., 2013). To overcome this difficulty, foraminiferal tests have been recently proposed as a more robust and reliable phase to access the authigenic coatings which preserve past seawater Nd isotopic compositions (Roberts et al., 2010; Elmore et al., 2011; Piotrowski et al., 2012).

Calcareous foraminifera are unicellular organisms that form calcium carbonate tests (shells) recording physical and chemical properties of the ocean. The advantages of this archive are the ubiquitous occurrence of the foraminifera in marine environments and at sufficiently high temporal resolution in sediment cores to study late Quaternary climate variability (Vance and Burton, 1999; Burton and Vance, 2000; Scrivner et al., 2004; Klevenz et al., 2008;

Osborne et al., 2008; Roberts et al., 2010; Piotrowski et al., 2012; Pena et al., 2013). Accurate interpretation of this tracer relies on precise determination of the origin of Nd preserved in foraminifera. If Nd is biologically incorporated in tests, it is logical to assume that ϵ_{Nd} values correspond to the seawater signals at calcification depths (Vance et al., 2004; Martinez-Boti et al., 2009; Pena et al., 2013). However, sedimentary foraminifera contain various Nd-rich phases, such as clay minerals, organic matter, Fe–Mn oxides and oxyhydroxides, and other authigenic and diagenetic minerals like Mn-rich carbonates (Palmer, 1985; Vance et al., 2004; Roberts et al., 2012; Tachikawa et al., 2013). If Nd extracted from foraminifera is derived from authigenic and/or diagenetic phases, then measured Nd isotopic signatures would instead correspond to bottom and/or pore waters, independent of the calcification depths of organisms.

In order to determine the origin of Nd recorded in foraminifera, a series of studies have carried out analysis of Nd content and isotopic compositions using different cleaning techniques for sedimentary foraminiferal tests, trapped material and plankton tow samples (Burton and Vance, 2000; Pomiès et al., 2002; Vance et al., 2004; Haley et al., 2005; Martinez-Boti et al., 2009; Roberts et al., 2010; Elmore et al., 2011; Piotrowski et al., 2012; Roberts et al., 2012; Kraft et al., 2013). Comparison of seawater and foraminiferal ϵ_{Nd} values from sites where surface, intermediate and bottom waters have significantly different Nd isotopic composition offers fundamental constraints on the origin of Nd in foraminifera (Roberts et al., 2010, 2012; Tachikawa et al., 2013). Microanalyses provide further information of Nd origins by direct determination of Nd-rich phases. In particular, micro-scale Nd-rich phases can be detected using laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) (Roberts et al., 2012) and nano secondary ion mass spectrometry (NanoSIMS) (Tachikawa et al., 2013).

In this review, we present an overview of the progress of our knowledge about Nd associated with foraminifera. In light of new results, the geochemical meaning of Nd isotopic compositions recorded in sedimentary planktonic foraminiferal tests will be discussed.

2. Oxidative–reductive cleaning effects on Nd in foraminiferal tests

The typical multi-step cleaning of the sedimentary foraminifera consists of shell crushing to open all internal chambers, mechanical cleaning using ultrasonication to remove adhesive clay minerals, an oxidative step to remove organic matter, and finally a reductive step to eliminate Fe–Mn oxides coatings (Boyle and Keigwin, 1985/1986). This full procedure is referred to as “cleaned” throughout this paper, while “uncleaned” refers to samples which were only mechanically cleaned of sediment and there was no attempt at chemically removing authigenic phases. Based on comparison of Nd concentrations between uncleaned and reductively cleaned tests, a pioneering work showed that 90% of Nd associated with uncleaned planktonic foraminiferal tests is related to the oxide coatings (Palmer, 1985). Considering that the Fe–Mn oxides record bottom water Nd isotopic signatures, uncleaned foraminifera were proposed to represent bottom water ϵ_{Nd} values (Palmer and Elderfield, 1985). In other words, if the cleaning permits obtaining pristine foraminiferal tests, then planktonic foraminifera could be used as an indicator of the surface water ϵ_{Nd} changes. Founded on this idea, fully cleaned planktonic foraminifera were applied to tracking the past (sub)surface water ϵ_{Nd} variability (Vance and Burton, 1999; Burton and Vance, 2000; Scrivner et al., 2004; Stoll et al., 2007; Osborne et al., 2008; Pena et al., 2013). However, this application is in conflict with the fact that cleaning does not significantly modify foraminiferal ϵ_{Nd} values (Fig. 1). Uncleaned and fully cleaned foraminiferal tests indicate identical Nd isotopic

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