Gondwana Research 36 (2016) 503–522

Contents lists available at ScienceDirect

Gondwana Research

journal homepage: www.elsevier.com/locate/gr

# Evolution of neodymium isotopic signature of seawater during the Late Cretaceous: Implications for intermediate and deep circulation

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## ARTICLE INFO

*Article history:*
Received 26 February 2015
Received in revised form 1 August 2015
Accepted 8 August 2015
Available online 9 September 2015

Handling Editor: R.D. Nance

*Keywords:*
Cretaceous
Neodymium isotopes
Ocean circulation
Southern Ocean

## ABSTRACT

Neodymium isotopic compositions ($\varepsilon_{Nd}$) have been largely used for the last fifty years as a tracer of past ocean circulation, and more intensively during the last decade to investigate ocean circulation during the Cretaceous period. Despite a growing set of data, circulation patterns still remain unclear during this period. In particular, the identification of the deep-water masses and their spatial extension within the different oceanic basins are poorly constrained. In this study we present new deep-water $\varepsilon_{Nd}$ data inferred from the Nd isotope composition of fish remains and Fe–Mn oxyhydroxide coatings on foraminifera tests, along with new $\varepsilon_{Nd}$ data of residual (partly detrital) fraction recovered from DSDP Sites 152 (Nicaraguan Rise), 258 (Naturaliste Plateau), 323 (Bellinghausen Abyssal Plain), and ODP Sites 690 (Maud Rise) and 700 (East Georgia Basin, South Atlantic). The presence of abundant authigenic minerals in the sediments at Sites 152 and 690 detected by XRD analyses may explain both middle rare earth element enrichments in the spectra of the residual fraction and the evolution of residual fraction $\varepsilon_{Nd}$ that mirror that of the bottom waters at the two sites. The results point towards a close correspondence between the bottom water $\varepsilon_{Nd}$ values of Sites 258 and 700 from the late Turonian to the Santonian. Since the deep-water Nd isotope values at these two sites are also similar to those at other proto-Indian sites, we propose the existence of a common intermediate to deep-water mass as early as the mid-Cretaceous. The water mass would have extended from the central part of the South Atlantic to the eastern part of proto-Indian ocean sites, beyond the Kerguelen Plateau. Furthermore, data from south and north of the Rio Grande Rise–Walvis Ridge complex (Sites 700 and 530) are indistinguishable from the Turonian to Campanian, suggesting a common water mass since the Turonian at least. This view is supported by a reconstruction of the Rio Grande Rise–Walvis Ridge complex during the Turonian, highlighting the likely existence of a deep breach between the Rio Grande Rise and the proto-Walvis Ridge at that time. Thus deep-water circulation may have been possible between the different austral basins as early as the Turonian, despite the presence of potential oceanic barriers. Comparison of new seawater and residue $\varepsilon_{Nd}$ data on Nicaraguan Rise suggests a westward circulation of intermediate waters through the Caribbean Seaway during the Maastrichtian and Palaeocene from the North Atlantic to the Pacific. This westward circulation reduced the Pacific water influence in the Atlantic, and was likely responsible for more uniform, less radiogenic $\varepsilon_{Nd}$ values in the North Atlantic after 80 Ma. Additionally, our data document an increasing trend observed in several oceanic basins during the Maastrichtian and the Palaeocene, which is more pronounced in the North Pacific. Although the origin of this increase still remains unclear, it might be explained by an increase in the contribution of radiogenic material to upper ocean waters in the northern Pacific. By sinking to depth, these waters may have redistributed to some extent more radiogenic signatures to other ocean basins through deep-water exchanges.

## 1. Introduction

Climatic variations affecting the Cretaceous have been extensively documented through palaeontological records, isotopic analyses and model simulations (Hallam, 1985; Barron et al., 1995; Huber et al., 1995; Poulsen et al., 2001; Otto-Bliesner et al., 2002; Pucéat et al.,

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http://dx.doi.org/10.1016/j.gr.2015.08.005

2003; Donnadieu et al., 2006; Hay, 2008; Craggs et al., 2012). Amongst the main features of this time period, climate reconstructions show the occurrence of extreme warmth during the Cenomanian–Turonian interval, followed by a long-term cooling during the Late Cretaceous, particularly accelerated during the Campanian (Huber et al., 1995; Pucéat et al., 2003; Steuber et al., 2005; Friedrich et al., 2012). The Late Cretaceous is also marked by a deepening of the Central Atlantic Gateway, the enlargement of the North and South Atlantic basins, and the first stages of Tethyan Ocean narrowing (Jones et al., 1995; Sewall et al., 2007; Cramer et al., 2009; Friedrich et al., 2012). It has been suggested that these changes in climate and palaeogeography during the Late Cretaceous may have caused substantial reorganisation of ocean circulation modes, possibly related in turn to the disappearance of the Oceanic Anoxic Events (OAEs) after the Cretaceous (e.g. Robinson et al., 2010; Robinson and Vance, 2012).

In order to investigate the ocean circulation patterns and their evolution during the Late Cretaceous, neodymium isotope data have been recently generated for this period of time (e.g. Pucéat et al., 2005; Soudry et al., 2005; MacLeod et al., 2008; Robinson et al., 2010; Martin et al., 2012; Murphy and Thomas, 2013; Voigt et al., 2013). The neodymium isotopic composition ($^{143}$Nd/$^{144}$Nd ratio, expressed as $\varepsilon_{Nd}$) of past seawater, typically reconstructed from that of fish remains or authigenic Fe–Mn marine oxides, has demonstrated a potential for tracking ocean circulation in both modern and ancient oceans (e.g. Frank, 2002; Goldstein and Hemming, 2003; Thomas, 2004; Piotrowski et al., 2008; Robinson et al., 2010). The distribution of neodymium isotopes in seawater is mainly controlled by dissolved Nd inputs, provided by subaerially exposed rocks on the continents, and exported to the oceans through weathering and fluvial transport (Piepgras et al., 1979; Goldstein and O'Nions, 1981; Frank, 2002; Tachikawa et al., 2003). Differences in mean age and geochemical composition (crustal vs. mantellic) of the rocks surrounding the oceanic basins strongly influence the isotopic composition of nearby surface seawaters. This particular isotopic signature is subsequently exported from upper ocean waters to depth in areas of deep-water formation, or during particle settling through the water column. The estimated oceanic residence time of neodymium (~300 to 600 years) is shorter than the oceanic mixing rate of about 1500 years (Broeker and Peng, 1982; Frank, 2002; Tachikawa et al., 2003; Arsouze et al., 2009). These characteristics lead to interbasinal heterogeneities in the $\varepsilon_{Nd}$ composition of water masses, with Nd isotopic signatures inherited from the geographical provenance (Piepgras and Wasserburg, 1982). Hence in modern deep-water circulation, the unradiogenic signature of North Atlantic Deep Water (NADW; $\varepsilon_{Nd} \sim -13.5$ ε-units) is derived from the contribution of Nd from Archean and Proterozoic continental rocks in northern Canada and Greenland (Piepgras and Wasserburg, 1987; Lacan and Jeandel, 2005a,b). In contrast, the Pacific Ocean has a more radiogenic composition ($\varepsilon_{Nd} = 0$ to $-5$) reflecting the weathering of young island arc volcanic materials (Piepgras and Jacobsen, 1988; Shimizu et al., 1994; Amakawa et al., 2004, 2009). These distinct signatures are subsequently exported by deep currents and modified through water mass exchange, as shown in the Southern Ocean where the mixing of both Atlantic and Pacific waters by the vigorous Antarctic Circumpolar Current (ACC) generates intermediate values ($\varepsilon_{Nd} \sim -9/-8$) (Piepgras and Wasserburg, 1982; Jeandel, 1993; Rickli et al., 2009; Carter et al., 2012).

Despite the growing set of recently published palaeoceanographic data, the deep-water circulation modes and their evolution during the Cretaceous period remain disputed (e.g. Frank et al., 2005; MacLeod et al., 2008, 2011; Friedrich et al., 2012; Martin et al., 2012; Murphy and Thomas, 2012; Robinson et al., 2010; Robinson and Vance, 2012; Murphy and Thomas, 2013; Voigt et al., 2013). During the Late Cretaceous, a long-term decrease in intermediate and deep-water $\varepsilon_{Nd(t)}$ has been depicted at different sites of the Southern Ocean, the South Atlantic and the North Atlantic, from values of $\sim -5/-7$ ε-units in the Turonian–Santonian interval to $\sim -8/-10$ ε-units in the Campanian–Turonian interval. This trend towards lower $\varepsilon_{Nd}$ values has been interpreted as the consequence of the onset or intensification of deep-water production of Southern Component Water (SCW) in the southern Atlantic or Indian Ocean (Robinson et al., 2010; Robinson and Vance, 2012; Murphy and Thomas, 2013; Voigt et al., 2013). Propagation of this unradiogenic signal into the different oceanic basins would then have been favoured by better communications between oceanic basins due to the subsidence of tectonic barriers and a more vigorous ocean circulation (Robinson et al., 2010; Robinson and Vance, 2012; Murphy and Thomas, 2013; Voigt et al., 2013). Previous studies have also mentioned the progressive subsidence of Large Igneous Provinces (LIPs) and hot spots in the South Atlantic and Indian oceans during the Late Cretaceous to explain the decreasing trend in Nd isotope compositions (Murphy and Thomas, 2013). Indeed, the drowning of mantle-derived material below sea-level would have reduced the subaerial weathering of radiogenic Nd in the Southern Ocean. Alternatively, initiation or intensification of unradiogenic deep-water production in the northern Atlantic has also been proposed to explain the observed Nd isotope trends (MacLeod et al., 2011; Martin et al., 2012). Amongst other potential sources of deep-waters in the Late Cretaceous and Palaeogene, the North and South Pacific areas have been proposed in different studies based on Nd isotope data and on model simulations (Thomas, 2004; Frank et al., 2005; Hague et al., 2012; Murphy and Thomas, 2012; Thomas et al., 2014). Alternatively, water sinking to abyssal depth has been suggested to occur in the equatorial Atlantic near Demerara Rise during most of the Late Cretaceous based on neodymium isotope studies (MacLeod et al., 2008, 2011).

Therefore, there is still no consensus on the origin of the deep-waters filling the different oceanic basins and the circulation changes during the Late Cretaceous. Part of these uncertainties arises from the still insufficient spatial distribution of deep-water Nd isotopic data in the South Atlantic, Indian Ocean, and in the southern Pacific that has been suggested as a potential area of deep-water production. Key passages like the Caribbean Seaway, which potentially linked the North Atlantic and Pacific Oceans, are also devoid of any Cretaceous $\varepsilon_{Nd}$ data. Here we present new early Turonian to late Palaeocene (~93 to 58 Ma) seawater Nd isotope records derived from fish debris and foraminifera tests from different Deep Sea Drilling Project (DSDP) and Oceanic Drilling Project (ODP) sites, to reconstruct the Nd isotopic composition of intermediate (500–2000 m) to deep (beneath 2000 m) water masses in the South Hemisphere and in the Caribbean Seaway. We targeted sites from the South Atlantic and proto-Indian Ocean (DSDP Site 258 and ODP Sites 690 and 700), some of which (Sites 258 and 700) had never been investigated for Nd isotope studies, to (i) constrain the extension of the SCW and its communication between the Late Cretaceous oceanic basins, and (ii) discuss the possible influence of oceanic barriers such as the Walvis Ridge–Rio Grande Rise system on deep-water circulation. Additional $\varepsilon_{Nd}$ data were provided from a southern Pacific site (DSDP Site 323) to decipher potential areas of deep-water formation in this region. Finally, we present the first Nd isotope data from the Caribbean region (Site 152) to track the water exchange between the North Atlantic and Pacific oceans during the Late Cretaceous–Early Palaeocene. These results are then compared to published $\varepsilon_{Nd}$ data from the literature.

## 2. Site selection and description

For all sites, the age models are presented in the Supplementary information. DSDP Leg 15 Hole 152 is located on the lower flanks of the Nicaragua Rise, and was drilled at a modern depth of 3899 m (Fig. 1). The good preservation of calcareous fossils suggests that the sediments at Site 152 were deposited at a bathyal depth, well above the calcium carbonate compensation depth (CCD). The samples were collected in the interval 204.51–453.91 m below sea floor (mbsf) (core sections 6-3/50–52 cm to 21-1/90–92 cm), consisting of white micritic chalk (Edgar et al., 1973). These sediments are rich in foraminifera and nannofossils, which indicate ages from late Campanian to Palaeocene