



# Evolution of the neodymium isotopic signature of neritic seawater on a northwestern Pacific margin: new constraints on possible end-members for the composition of deep-water masses in the Late Cretaceous ocean

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## ABSTRACT

The Neodymium (Nd) isotope composition of fish remains has been widely used to track past changes in oceanic circulation. Although the number of published Nd isotope data for the Cretaceous has markedly increased in the last years, no consensus has been reached on the structure of the oceanic circulation and its evolution during the Late Cretaceous. Yet this period is characterised by major geodynamical and climatic changes and marked by the disappearance of global oceanic anoxic events in which changes in oceanic circulation modes may have played a significant role.

In this study we present the first record of Nd isotopic composition of fish remains from continental margin environments on the northwestern Pacific margin (Yezo Group in the Hokkaid o area, Northern Japan) for the Late Cretaceous period. This record, interpreted in terms of Nd isotopic composition of local neritic seafloor seawater, is characterised by relatively radiogenic Nd isotope compositions and presents variations of several  $\epsilon$ -units from the Turonian to the Campanian ranging from  $\sim -5.5$  to  $\sim 0.5$   $\epsilon$ -units, although most values remain in the  $\sim -1$  to  $\sim -3$  range. Conversely, the local detrital fraction remains more constant and around  $-4$   $\epsilon$ -units on the studied interval. This new set of seawater Nd data contains some of the most radiogenic values for the Cretaceous published yet. The more radiogenic seawater Nd isotope values compared to that of the sediments points to an input of radiogenic seawater in the studied area by surface currents during the Late Cretaceous. Similarly to the modern configuration, these radiogenic waters could have been conveyed in the studied area by a southward current comparable to the modern Oyashio current bathing the Hokkaid o area. Our data are then consistent with the presence in the northern Pacific of highly radiogenic seawater, and support the northern and northwestern Pacific as a possible radiogenic source for the deep parts of the basin. As such this work represents a first step toward a better characterisation of the various end-members that could have contributed to the Nd isotopic signature of the deep-water masses filling the Cretaceous oceans.

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## 1. Introduction

After the extreme warmth of the Cenomanian–Turonian interval, the climate of the Late Cretaceous was characterised by a long-term cooling, which intensified during the Campanian (Huber et al., 1995; Puc at et al., 2003; Steuber et al., 2005; Friedrich et al., 2012). This long-term decrease of temperatures is concomitant to the termination of the occurrence of worldwide oceanic anoxic events (OAEs) in the sedimentary records (Jenkyns, 2010). The widening of the Atlantic Ocean, variations in the Panama Strait depth and the first stages of Tethyan Ocean

closure strongly affected the palaeogeography during the Late Cretaceous, and may have induced major changes in the oceanic circulation. Yet the oceanic circulation modes and their evolution during the Cretaceous period remain unclear, despite the growing set of palaeoceanographic data recently published (e.g. Frank et al., 2005; MacLeod et al., 2008, 2011; Robinson et al., 2010; Friedrich et al., 2012; Martin et al., 2012; Murphy and Thomas, 2012; Robinson and Vance, 2012).

In the past decades, the neodymium isotopic composition ( $^{143}\text{Nd}/^{144}\text{Nd}$  ratio, expressed as  $\epsilon_{\text{Nd}}$ ) has been increasingly used to track oceanic 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). Neodymium is exported to the oceans through

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weathering and drainage of subaerially exposed rocks on the continents (Piepgras et al., 1979; Goldstein and O'Nions, 1981; Frank, 2002; Tachikawa et al., 2003). The short oceanic residence time of Nd (~300 to ~600 years) (Frank, 2002; Tachikawa et al., 2003; Arsouze et al., 2009) relative to the oceanic mixing rate of about 1500 years (Broecker and Peng, 1982) and differences in general age and composition of the rocks surrounding oceanic basins give rise to distinct interbasinal differences in the  $\epsilon_{\text{Nd}}$  values of the water masses. Each deep-water mass has a characteristic Nd isotopic signature derived from the composition of Nd that is discharged into the source regions (Piepgras and Wasserburg, 1982). At present, the unradiogenic signature of North Atlantic Deep Water ( $\epsilon_{\text{Nd}} = -13.5 \pm 0.5$ ) derives from the contribution of Nd from Archean and Proterozoic continental rocks in northern Canada and Greenland (Piepgras and Wasserburg, 1987; Lacan and Jeandel, 2005a, 2005b). In contrast, the Pacific Ocean has a more radiogenic composition ( $\epsilon_{\text{Nd}} = 0$  to  $-5$ ) that reflects the weathering of island arc material (Piepgras and Jacobsen, 1988; Shimizu et al., 1994; Amakawa et al., 2004b, 2009).

During the latest Cretaceous (Campanian–Maastrichtian), a long-term decrease in deep or intermediate water  $\epsilon_{\text{Nd}}$  to values of about  $-11$  depicted at different sites of the Southern Ocean has been interpreted as reflecting the onset of deep-water production in the Southern Ocean and a more vigorous oceanic circulation (Robinson et al., 2010; Robinson and Vance, 2012). Conversely, an increase in the  $\epsilon_{\text{Nd}}$  of bottom waters to values of about  $-11$  as well, depicted in North Atlantic sites during the latest Cretaceous (Maastrichtian) has been interpreted as the result of the initiation of deep-water production in the northern Atlantic (MacLeod et al., 2011). Deep-water production has also been suggested to occur at low latitude sites of the Atlantic during the Late Cretaceous (Friedrich et al., 2008; MacLeod et al., 2008, 2011). Based on modeling experiments (Poulsen et al., 2001; Otto-Bliesner et al., 2002; Pucéat et al., 2005b), on benthic and planktonic foraminifera carbon or oxygen isotopic composition (Barrera et al., 1997), and on  $\epsilon_{\text{Nd}}$  values (Thomas, 2004; Hague et al., 2012), high latitudes of the North and South Pacific have additionally been suggested as potential sites of deep-water production.

Therefore no consensus has yet emerged on the origin of the deep-waters bathing the different oceanic basins and the nature of the oceanic circulation changes that occurred during the Late Cretaceous. Part of these uncertainties arises from the still insufficient spatial distribution of deep-water Nd isotopic data and from the lack of surface water  $\epsilon_{\text{Nd}}$  data in the potential areas of deep-water formation (Pucéat, 2008). As variations depicted in the  $\epsilon_{\text{Nd}}$  of a deep-water mass can reflect either a change in oceanic circulation (mixing with different water masses or changes in the location of deep-water sinking) or a change in the composition of the continents eroded around the source area, surface water  $\epsilon_{\text{Nd}}$  data around possible areas of water convection may help to interpret existing deep-water signals.

The Nd isotope composition of upper ocean waters can be reconstructed in neritic environments by the analysis of fish remain  $\epsilon_{\text{Nd}}$  deposited on continental shelves (Grandjean et al., 1987; Pucéat et al., 2005a; Soudry et al., 2005, 2006; Charbonnier et al., 2012). Fluorapatite of fossil fish remains (teeth, bones, and scales) is characterised by relatively high Nd concentrations (100–1000 ppm) (e.g., Wright et al., 1984; Shaw and Wasserburg, 1985; Staudigel et al., 1985) and acquire their Nd isotope composition during early diagenesis, at the sediment–water interface. Consequently their  $\epsilon_{\text{Nd}}$  reflects that of seawater at the base of the water column. In neritic environments, where the sediment–water interface remains at shallow depths and where seawater is well-mixed by wave motion and strong currents, fish remain  $\epsilon_{\text{Nd}}$  allows to access the Nd isotope composition of these shallow, neritic seawaters. Importantly, interactions between sediments and seawater are known to occur on continental margins and on shelves (termed 'boundary exchanges') (Lacan and Jeandel, 2005a, 2005b; Jeandel et al., 2007; Arsouze et al., 2009; Carter et al., 2012). As a result, Nd isotope composition of seawater in neritic environments is

likely to differ from that of surface pelagic seawater. Yet especially because the composition of seawater flowing along margins and on shelves is more impacted by changes in the composition of nearby erosional inputs, their evolution around the areas of surface water sinking should help to discuss the evolution of  $\epsilon_{\text{Nd}}$  depicted in deep-waters (oceanic circulation changes vs. changes in erosional inputs in the source area).

Here we focus on the margin of one of the potential areas of deep or intermediate water production during the Late Cretaceous, the North-West Pacific (Poulsen et al., 2001; Otto-Bliesner et al., 2002), for which no data currently exist. We collected fish debris from samples of Cenomanian to Campanian marine sediments deposited in shelf to slope environments of two sections of the Hokkaidō area (northern Japan), and analysed them as well as the detrital fraction for their Nd isotope composition in order to reconstruct and discuss the evolution of the surface waters  $\epsilon_{\text{Nd}(t)}$  during the Late Cretaceous at this site on the palaeo-margin of North Japan. Although other sites on the northern Pacific margin are required to obtain a more complete view of the Nd isotopic composition of the upper ocean waters and their evolution due to changes in erosional inputs in this area, this study represents the first step to improve the spatial and temporal set of data available in this region for the Cretaceous.

## 2. Geological setting

The sections studied in this work are exposed in the Yezo Group from Hokkaidō, northern Japan (Fig. 1). The Yezo Group conformably overlies the Sorachi Group, a volcano-sedimentary sequence corresponding to the Jurassic oceanic arc phase (Takashima et al., 2002). During the Late Jurassic, the main subduction zone responsible for the original Japanese continental arc was shifted further offshore to form other oceanic arcs. Consequently, the major island arc volcanism ceased in the northern Japanese area during the Early Cretaceous. No back arc basin existed behind the future Japanese islands during the Cretaceous, as the rifting of the Sea of Japan only started during the Oligocene to form the modern back arc basin (Ingle, 1992; Tamaki et al., 1992). The Japanese islands formed an eastern margin of the Asian continent to which they were still attached. The Cretaceous sediments collected in this study were thus deposited along the Asian continental margin and represent forearc deposits (Takashima et al., 2002).

The Cretaceous forearc sediments of the Yezo Group comprise thick sandstone and mudstone sequences, with abundant intercalations of volcanic tuff, turbiditic sandstone beds and frequent hiatuses, related to a regressive sequence from the Early Aptian to Early Maastrichtian (Fig. 2). Facies studies suggest an eastward deepening of depositional environments, from coastal plain to continental slope. Cenozoic fluvial and shallow marine sediments disrupt the sedimentary series in its upper part (Takashima et al., 2004).

The studied sections encompass the upper Hikagenosawa (Lower Cenomanian), Saku (Upper Cenomanian–Late Turonian), and Haborogawa (Uppermost Turonian–Early Maastrichtian) Formations, in the Tomomae area of the Yezo Group (Fig. 2). These sediments were deposited from Cenomanian to Campanian in outer to inner shelf environments (Takashima et al., 2004). Due to the presence of many coarse-grained gravity flows deposits, the selected samples were carefully extracted in intervals of fine grain-sized sediments, with minimal evidence of bioturbation, slump or turbiditic deposits. X-ray powder diffraction analyses were performed on the bulk sediments samples analysed in this work for their Nd isotope composition and REE patterns, along with 12 additional bulk sediments samples, in order to characterise their mineralogical composition (see Supplementary information). These analyses reveal that the mineralogical composition of the selected samples is quite similar, with a dominance of quartz and smaller amounts of clay minerals (mainly illite/smectite mixed-layers, illite and kaolinite), plagioclases and micas.

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