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## Neodymium budget in the modern ocean and paleo-oceanographic implications

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[1] The oceanic Nd budget is calculated using a steady state 10-box model and a compilation of field data. This is the first attempt to propose consistent estimates of the Nd fluxes entering the ocean, as well as indicating possible Nd sources and the proportion of Nd fluxes exchanged between dissolved and particulate fractions. With presently available Nd data the best estimates give a total Nd influx of  $9 \times 10^9$  g/yr, which leads to an oceanic Nd residence time of 500 years. From modeling tests we suggest that the authigenic Nd scavenged by particulates is 100% remineralized in the deep ocean. The total exchanged Nd flux may be as high as  $2 \times 10^{10}$  g/yr. The  $\epsilon_{\text{Nd}(0)}$  values of the influxes are  $-22$ ,  $-11$ ,  $+1$ , and  $-4$  for the North Atlantic, surface Atlantic, North Pacific, and surface Indo-Pacific regions, respectively. Atmospheric and riverine Nd fluxes are insufficient to explain the magnitude and regional variability of calculated Nd influxes and  $\epsilon_{\text{Nd}(0)}$ . We propose continental margins as an additional source supplying Nd to the ocean. Using the model calibrated for Nd, we examine the sensitivity of deep water  $\epsilon_{\text{Nd}(0)}$  to variations of Nd inputs to the ocean. Deep water Nd concentrations and  $\epsilon_{\text{Nd}(0)}$  vary with the changes in Nd influxes and their  $\epsilon_{\text{Nd}(0)}$ . As Nd sources to the ocean may change during glacial/interglacial periods, the  $\epsilon_{\text{Nd}(0)}$  shifts recorded in ferromanganese nodules and crusts do not necessarily reflect changes in paleoceanic circulation. The effects of continental erosion should be considered in reconstructing patterns of ocean circulation using Nd isotopes. *INDEX TERMS:* 1040

Geochemistry: Isotopic composition/chemistry; 1050 Geochemistry: Marine geochemistry (4835, 4850); 4267 Oceanography: General: Paleoceanography; *KEYWORDS:* neodymium, isotopes, residence time, budget

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### 1. Introduction

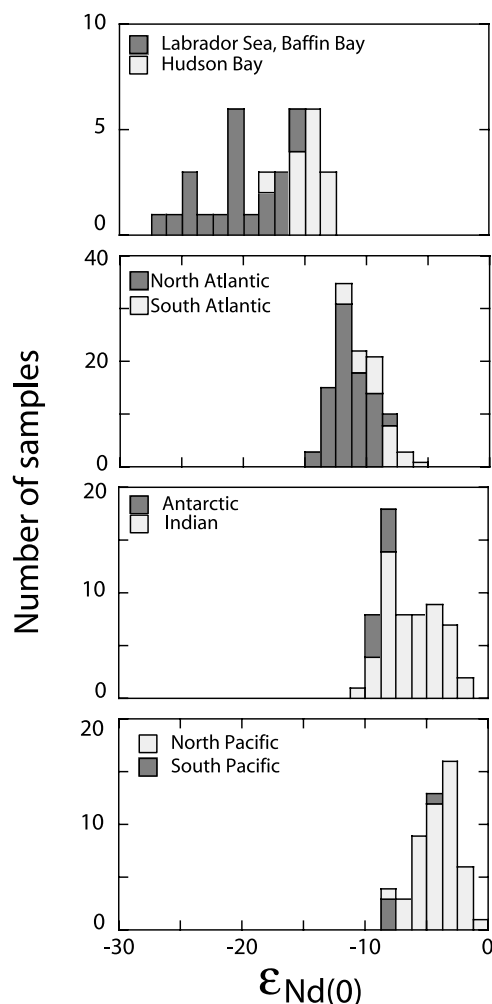
[2] In the modern ocean, seawater Nd isotopic ratio ( $^{143}\text{Nd}/^{144}\text{Nd}$ ) varies according to the water masses (Figure 1). Since Nd isotopic signatures are preserved in ferromanganese nodules and crusts [Albarède and Goldstein, 1992; Albarède et al., 1997; Burton et al., 1997; O'Nions et al., 1998; Abouchami et al., 1999], foraminiferal tests [Burton and Vance, 2000] and Fe-Mn oxide coatings of sediments [Rutherg et al., 2000], Nd isotopes could be used to trace past oceanic circulation patterns. However, the Nd budget in the modern ocean is not yet precisely constrained. Before undertaking paleo-oceanographic studies, we need to improve our knowledge of the Nd inventory in the modern ocean.

[3] The  $\epsilon_{\text{Nd}(0)}$  values ( $\epsilon_{\text{Nd}(0)} = [({}^{143}\text{Nd}/{}^{144}\text{Nd})_{\text{sample}} / ({}^{143}\text{Nd}/{}^{144}\text{Nd})_{\text{reference}} - 1] \times 10^4$ ) of the major water masses range from  $-26$  to  $0$  (Figure 1). Principal Nd sources to the ocean are expected to be continental materials because the flux from the depleted mantle (hydrothermal Nd flux) is

negligible [Elderfield, 1988, and references therein]. Negative values of  $\epsilon_{\text{Nd}(0)}$  in seawater imply the contribution of ancient continental materials to the ocean, whereas values close to zero imply the contribution of young volcanogenic materials [Faure, 1986]. Hence the  $\epsilon_{\text{Nd}(0)}$  gradient between the North Atlantic ( $\epsilon_{\text{Nd}(0)} = \sim -13$ ) and the North Pacific ( $\epsilon_{\text{Nd}(0)} = \sim -4$ ) is qualitatively explained by the age of the continents surrounding the ocean basins: the North Atlantic is encompassed by old cratonic continents such as Greenland ( $\epsilon_{\text{Nd}(0)} = \sim -19$ ) and the Canadian shield ( $\epsilon_{\text{Nd}(0)} = \sim -14/-32$ ) whereas the North Pacific is surrounded by volcanic rocks such as Japan ( $\epsilon_{\text{Nd}(0)} = \sim 0$ ) and Indonesia ( $\epsilon_{\text{Nd}(0)} = \sim +2$ ) [Goldstein and Jacobsen, 1987; Grousset et al., 1988; Albarède and Goldstein, 1992].

[4] Continental Nd enters the ocean in the form of dissolved and particulate material from the atmosphere and rivers [Goldstein and Jacobsen, 1987; Elderfield, 1988; Spivack and Wasserburg, 1988; Bertram and Elderfield, 1993; Greaves et al., 1994; Henry et al., 1994; Jeandel et al., 1995]. To date, net Nd fluxes from the continents are not precisely quantified. This is partly due to the fact that (i) the net removal of dissolved riverine Nd in estuaries is poorly constrained [Goldstein and Jacobsen, 1987; Elderfield et al., 1990; Sholkovitz, 1993; Byrne and Sholkovitz, 1996] and (ii)

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**Figure 1.** Histogram of seawater  $\epsilon_{Nd(0)}$  of the world ocean.  $\epsilon_{Nd(0)} = [({}^{143}Nd/{}^{144}Nd)_{sample}/({}^{143}Nd/{}^{144}Nd)_{reference} - 1] \times 10^4$ , where the reference is (Chondritic Uniform Reservoir (CHUR) representing the bulk earth  $\epsilon_{Nd(0)}$ ) [Wasserburg *et al.*, 1981]. Data are from Piepgras *et al.* [1979], Piepgras and Wasserburg [1980, 1982, 1983, 1987], Stordal and Wasserburg [1986], Piepgras and Jacobsen [1988], Spivack and Wasserburg [1988], Bertram and Elderfield [1993], Jeandel [1993], Shimizu *et al.* [1994], Jeandel *et al.* [1998], and Tachikawa *et al.* [1999].

the proportions of soluble Nd in atmospheric and riverine particles are highly variable. The net Nd flux from lithogenic (aluminosilicate) particles is the product of the bulk Nd flux and the proportion of soluble Nd. The estimated proportion of soluble Nd in dust varies from 2% to 50% [Greaves *et al.*, 1994; Henry *et al.*, 1994; Jeandel *et al.*, 1995; Tachikawa *et al.*, 1999].

[5] In the ocean, Nd is transported by water masses and marine particles. Dissolved Nd is removed from the surface water by adsorption onto marine particles (scavenging). In deeper waters, the scavenged authigenic Nd is released back to seawater (remobilization). We define the formation of authigenic particulate Nd and its subsequent remobilization as a “dissolved/particulate exchange”. The seawater  $\epsilon_{Nd(0)}$  is therefore determined by the mixing of water masses and the dissolved/particulate exchanges. Bertram

and Elderfield [1993] concluded that an exchanged Nd flux of  $4.2 \times 10^{10}$  g/yr is required to reproduce the distinct  $\epsilon_{Nd(0)}$  observed in the deep Atlantic ( $\epsilon_{Nd(0)} = \sim -13$ ), Indian ( $\epsilon_{Nd(0)} = \sim -8$ ) and Pacific ( $\epsilon_{Nd(0)} = \sim -4$ ). This estimate only holds for the Nd inventory in deep water. It is now necessary to establish a Nd inventory for the whole ocean, including the surface water. Indeed, continental Nd fluxes directly contribute to the surface water and determine the mean oceanic Nd residence time.

[6] The purpose of this study is to constrain the Nd inventory in the modern ocean using both a compilation of all the available Nd data and a numerical model [Broecker and Peng, 1987]. We propose to estimate the Nd fluxes from the continents (influxes), the  $\epsilon_{Nd(0)}$  values of these influxes, the mean Nd residence time and the influence of the dissolved/particulate Nd exchanges. During the last two decades, there has been a remarkable increase in the amount of data on dissolved Nd concentrations and  $\epsilon_{Nd(0)}$ . In this study, we use a compilation of Nd data to characterize the 10 boxes of the model [Broecker and Peng, 1987] and to improve constraints on the present-day situation of the ocean. The Nd inventory is then calculated using the box model. Finally, the model is calibrated for Nd and used to simulate the effects on deep water  $\epsilon_{Nd(0)}$  of varying Nd inputs to the ocean. To date, changes in deep water  $\epsilon_{Nd(0)}$  recorded in ferromanganese nodules and crusts have been explained in terms of changes in deep water circulation. However, it is well known that lithogenic element fluxes and sources to the ocean vary during glacial/interglacial periods because of several factors such as wind strength, aridity of continents and erosion by ice sheets [Williams *et al.*, 1998]. Our results suggest that deep water  $\epsilon_{Nd(0)}$  may change in response to Nd inputs, even though the circulation pattern remains unchanged.

## 2. Modeling

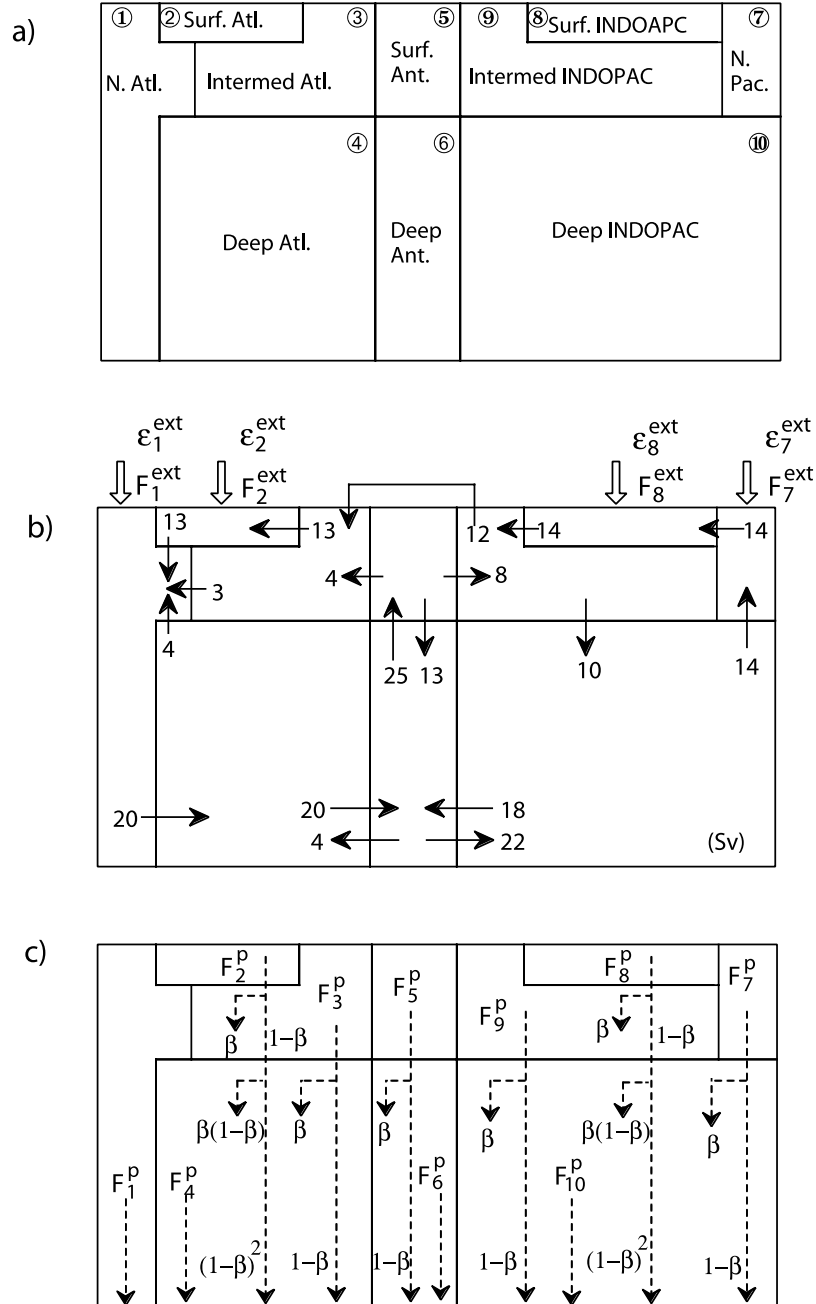
### 2.1. General Assumptions

[7] In the PANDORA model [Broecker and Peng, 1987], the ocean is described by 10 boxes whose volumes and surface areas are known (Figure 2a and Table 1). Each box is characterized by a dissolved Nd concentration, a dissolved  $\epsilon_{Nd(0)}$ , as well as dissolved and particulate Nd fluxes entering and leaving the box. Except for the North Pacific (box 7), the Indian and Pacific boxes are combined and referred as to INDOPAC boxes. The INDOPAC boxes are assumed to be composed of 70% of the Pacific Ocean and 30% of the Indian Ocean. Six hypotheses are applied to the Nd modeling.

[8] 1. Neodymium is thought to be at steady state in the modern ocean. This implies, first, that the total amount of Nd in each box  $i$  is constant with time, because the influxes are balanced by the outfluxes:

$$\frac{dQ_i}{dt} = V_i \frac{dC_i}{dt} = \sum_{j \neq i} F_{j \rightarrow i} - \sum_{j \neq i} F_{i \rightarrow j} = 0 \quad (1)$$

where  $F_{j \rightarrow i}$  indicates the Nd flux entering box  $i$  from box  $j$ , and  $F_{i \rightarrow j}$  indicates the Nd flux leaving box  $i$  to box  $j$ .  $Q_i$  is the total amount of Nd in box  $i$ .  $V_i$  and  $C_i$  are the volume and the dissolved Nd concentration of box  $i$ , respectively. We define the total amount of Nd contained in each box as follows:  $Q_i = V_i C_i$ .



**Figure 2.** PANDORA 10-box model [Broecker and Peng, 1987]. (a) Configuration of the model. Atl., Atlantic; Ant., Antarctic; Pac., Pacific; INDOPAC, Indian and Pacific; Surf., Surface; Intermed., Intermediate; and N, North. (b) Water fluxes (solid arrows) were calibrated by Broecker and Peng [1987] (Sv, Sverdrups =  $10^6$  m<sup>3</sup>/s). Four open arrows toward surface boxes indicate the Nd fluxes entering the ocean (Table 3). (c) Remineralization pattern of the particulate Nd fluxes. Remineralization rate is denoted as  $\beta$ . Note that our remineralization pattern is different from the original version [Broecker and Peng, 1987] for the North Atlantic box. There is no contribution of remineralized particulate flux from the Surface Atlantic to the North Atlantic box. This is necessary to avoid forming a negative flux in the North Atlantic box.

[9] The steady state hypothesis also means that the  $\varepsilon_{Nd(0)}$  of box  $i$  ( $\varepsilon_i$ ) does not change with time:

$$\frac{d\varepsilon_i}{dt} = \frac{1}{Q_i} \left[ \sum_{j \neq i} F_{j \rightarrow i} \cdot \varepsilon_j - \sum_{j \neq i} F_{i \rightarrow j} \cdot \varepsilon_i \right] = 0 \quad (2)$$

Hence  $\varepsilon_i$  is determined by the influxes entering box  $i$  as well as the  $\varepsilon_{Nd(0)}$  values of the influxes:

$$\varepsilon_i = \frac{\sum_{j \neq i} F_{j \rightarrow i} \cdot \varepsilon_j}{\sum_{j \neq i} F_{j \rightarrow i}} \quad (3)$$

**Table 1.** Nd Concentrations and Isotopic Ratios for the 10 PANDORA Boxes

No.	Name	Box		Field Data		Values Applied to the Model	
		Volume, 10 <sup>16</sup> m <sup>3</sup>	Area, <sup>a</sup> 10 <sup>13</sup> m <sup>2</sup>	Nd, pmol/kg	$\epsilon_{Nd(0)}$	Nd, pmol/kg C <sub>i</sub>	$\epsilon_{Nd(0)} \epsilon_i$
1	North Atlantic (>50°N)	8.2	1.1	20.8 ± 4.7 (124)	-13.9 ± 3.8 (21)	20.8	-13.9
2	Surface Atlantic	4.1	10.1	14.9 ± 5.9 (47)	-11.2 ± 1.1 (30)	14.9	-11.2
3	Intermediate Atlantic	4.1	1.4 10.5	13.8 ± 5.6 (40)	-10.1 ± 1.8 (23)	13.8	-10.1
4	Deep Atlantic	26.1	11.6	21.4 ± 8.0 (76)	-11.1 ± 2.1 (44)	21.4	-11.1
5	Surface Antarctic	1.4	1.4	11.6 ± 3.6 (4)	-9.1 ± 0.1 (3)	11.6	-8.2
6	Deep Antarctic	13.7	1.4	21.7 ± 7.9 (3)	-8.5 ± 0.4 (3)	21.7	-8.2
7	North Pacific (> 44°N)	1.4	1.1	22.1 ± 3.7 (5)	-4.0 ± 2.5 (6)	22.1	-4.0
8	Surface INDOPAC	8.3	19.5	8.2 ± 3.2 (151)	-4.3 ± 2.3 (43)	8.2	-4.3
9	Intermediate INDOPAC	16.5	1.4 20.9	14.0 ± 5.1 (52)	-4.6 ± 1.7 (26)	14.0	-4.6
10	Deep INDOPAC	53.5	22.0	26.9 ± 10.5 (107)	-5.6 ± 2.1 (64)	26.9	-5.6
	Whole ocean	137	36.1	4.2 × 10 <sup>12</sup> (g)		4.2 × 10 <sup>12</sup> (g)	

Numbers between the blankets indicate numbers of data.

<sup>a</sup>For two intermediate boxes (boxes 3 and 9), both surface and bottom areas are indicated (Figure 2a).

<sup>b</sup>The mean Nd concentration and  $\epsilon_{Nd}$  as well as their 2 $\sigma$  are calculated from the data [Piepgras et al., 1979; Piepgras et al., 1982, 1983, 1987; Elderfield and Greaves, 1982; De Baar et al., 1985, 1988; Stordal and Wasserburg, 1986; Spivack and Wasserburg, 1988; German and Elderfield, 1989, 1990; Greaves et al., 1991; Sholkovitz and Schneider, 1991; Westerlund and Ohman, 1992; Jeandel, 1993; Bertram and Elderfield, 1993; Zhang et al., 1994; Nozaki and Zhang, 1995; German et al., 1995; Zhang and Nozaki, 1996; Jeandel et al., 1998; Tachikawa et al., 1999; Amakawa et al., 2000; Lacan and Jeandel, 2001].

[10] 2. The continental Nd is assumed to enter the ocean via four surface boxes in the northern hemisphere: North Atlantic (i = 1), Surface Atlantic (i = 2), North Pacific (i = 7) and Surface INDOPAC (i = 8) (Figure 2b). The fluxes to the Southern Ocean are assumed to be negligible because of the long distances from the continents.

[11] Under this hypothesis, equation (1) for the four surface boxes (i = 1, 2, 7, 8) becomes as follows:

$$V_i \frac{dC_i}{dt} = \left( F_i^{ext} + \sum_{j \neq i} F_{j \rightarrow i}^{ad} \right) - \left( \sum_{j \neq i} F_{i \rightarrow j}^{ad} + F_i^p \right) = 0 \quad (4)$$

where  $F_i^{ext}$  is the Nd flux from external sources toward box i and  $F_i^p$  is the authigenic particulate Nd flux formed in box i.  $\sum F_{i \rightarrow j}^{ad}$  and  $\sum F_{j \rightarrow i}^{ad}$  are the sums of advective Nd fluxes entering and leaving box i, respectively. The advective Nd fluxes  $F_{i \rightarrow j}^{ad}$  (1 ≤ i ≤ 10, 1 ≤ j ≤ 10, i ≠ j) are the products of seawater Nd concentration ( $C_i$ ) and water flux ( $F_{i \rightarrow j}^{ad,sw}$ ), that is  $F_{i \rightarrow j}^{ad} = C_i \cdot F_{i \rightarrow j}^{ad,sw}$ . All the  $F_{i \rightarrow j}^{ad,sw}$  values have already been determined from the oceanic <sup>14</sup>C distribution by Broecker and Peng [1987] (Figure 2b).  $F_{i \rightarrow j}^{ad,sw}$  values are assumed not to vary with time.

[12] 3. A fixed proportion of Nd present in the authigenic particulate Nd flux is assumed to be remineralized in the deeper boxes in the Atlantic, Indian and Pacific Oceans (0% ≤ β ≤ 100%). This proportion, denoted here as β, is assumed to be constant with respect to ocean basin, water depth and time.

[13] When β is 0%, no remineralization occurs. When β is 100%, all the particulate authigenic Nd dissolves. The remineralization is schematically presented in Figure 2c. The Intermediate Atlantic, Deep Antarctic and Intermediate INDOPAC (i = 3, 6, 9) receive a remineralized authigenic Nd flux formed in the respective surface box. Hence equation (1) for these boxes (i = 3, 6, 9) is as follows:

$$V_i \frac{dC_i}{dt} = \left( \beta \cdot F_{i-1}^p + \sum_{j \neq i} F_{j \rightarrow i}^{ad} \right) - \left( \sum_{j \neq i} F_{i \rightarrow j}^{ad} + F_i^p \right) = 0 \quad (5)$$

The Deep Atlantic and Deep INDOPAC boxes (i = 4, 10) receive remineralized Nd fluxes formed in both surface and intermediate boxes:

$$V_i \frac{dC_i}{dt} = \left( \beta \cdot F_{i-1}^p + \delta_{i,10} \cdot (\beta \cdot F_7^p) + \beta \cdot (1 - \beta) \cdot F_{i-2}^p + \sum_{j \neq i} F_{j \rightarrow i}^{ad} \right) - \left( \sum_{j \neq i} F_{i \rightarrow j}^{ad} + F_i^p \right) = 0 \quad (6)$$

where  $\delta_{i,j}$  is a Kronecker symbol defined such that  $\delta_{i,j} = 0$  when  $i \neq j$  and  $\delta_{i,j} = 1$  when  $i = j$ .

[14] 4. Dissolved Nd concentrations ( $C_i$ ) and  $\epsilon_{Nd(0)}$  ( $\epsilon_i$ ) in the 10 boxes can be determined from field data. Table 1 compiles all the available data on seawater Nd concentrations and  $\epsilon_{Nd(0)}$ . The mean Nd concentrations ( $C_i$ ) were estimated taking into account the different Nd concentrations in the north and south basins. Volume-weighted averages of the north and south basins were used to determine  $C_i$  (Table 1). Similarly, seawater  $\epsilon_{Nd(0)}$  values are systematically different between the north and south basins (Figure 1). The mean  $\epsilon_{Nd(0)}$  values ( $\epsilon_i$ ) were estimated considering both volumes and Nd concentrations of the north and south basins (Table 1). Because of the limited amount of  $\epsilon_{Nd(0)}$  data (n = 3) for the Surface and Deep Antarctic boxes,  $\epsilon_5$  and  $\epsilon_6$  were determined using the  $\epsilon_{Nd(0)}$  equation at steady state (equation (3)). Equation (3) for the surface and deep Antarctic (i = 5, 6) becomes:

$$\epsilon_5 = \frac{F_{6 \rightarrow 5}^{ad} \cdot \epsilon_6}{F_{6 \rightarrow 5}^{ad}} = \epsilon_6 \quad (7a)$$

$$\epsilon_6 = \frac{\epsilon_4 \cdot F_{4 \rightarrow 6}^{ad} + \epsilon_5 \cdot F_{5 \rightarrow 6}^{ad} + \epsilon_{10} \cdot F_{10 \rightarrow 6}^{ad} + \epsilon_5 \cdot \beta \cdot F_5^p}{F_{4 \rightarrow 6}^{ad} + F_{5 \rightarrow 6}^{ad} + F_{10 \rightarrow 6}^{ad} + \beta \cdot F_5^p}$$

or

$$(\epsilon_6 - \epsilon_5) \cdot \beta \cdot F_5^p = (\epsilon_4 \cdot F_{4 \rightarrow 6}^{ad} + \epsilon_5 \cdot F_{5 \rightarrow 6}^{ad} + \epsilon_{10} \cdot F_{10 \rightarrow 6}^{ad}) - \epsilon_6 (F_{4 \rightarrow 6}^{ad} + F_{5 \rightarrow 6}^{ad} + F_{10 \rightarrow 6}^{ad}) \quad (7b)$$



The left-hand side of equation (7b) is zero because  $\varepsilon_5 = \varepsilon_6$  (equation (7a)). Consequently, equation (7b) can be rearranged:

$$\varepsilon_5 = \varepsilon_6 = \frac{\varepsilon_4 \cdot F_{4 \rightarrow 6}^{ad} + \varepsilon_{10} \cdot F_{10 \rightarrow 6}^{ad}}{F_{4 \rightarrow 6}^{ad} + F_{10 \rightarrow 6}^{ad}} \quad (7c)$$

Using  $\varepsilon_4$ ,  $\varepsilon_{10}$ ,  $C_4$  and  $C_{10}$  shown in Table 1,  $\varepsilon_5 = \varepsilon_6 = -8.2$ . This value is used for the model calculation.

[15] At present, all the Nd concentrations ( $C_i$ ) and all the  $\varepsilon_{Nd(0)}$  values ( $\varepsilon_i$ ) of the boxes are known. All the advective Nd fluxes ( $F_{i \rightarrow j}^{ad}$ ) then become known because both  $C_i$  and water fluxes ( $F_{i \rightarrow j}^{ad,sw}$ ) can be determined. Under the four hypotheses, the Nd inventory is described by 20 equations: 10 for the  $\varepsilon_{Nd(0)}$  values (equation (3)) and 10 for the Nd fluxes (equation (4), (5), or (6), depending upon box number). In this study, we aim to determine 19 unknowns including 4 Nd fluxes from the external sources ( $F_1^{ext}$ ,  $F_2^{ext}$ ,  $F_7^{ext}$  and  $F_8^{ext}$ ), 4  $\varepsilon_{Nd(0)}$  values of the external fluxes ( $\varepsilon_1^{ext}$ ,  $\varepsilon_2^{ext}$ ,  $\varepsilon_7^{ext}$  and  $\varepsilon_8^{ext}$ ), 10 authigenic particulate Nd fluxes formed in the boxes ( $F_i^p$ ) and 1 remineralization rate ( $\beta$ ). Among the 20 equations, only 17 effectively need to be considered. First, there are no unknown values in equation (3) for  $i = 5$  ( $\varepsilon_5 = \varepsilon_6$ ) and equation (7c) for  $i = 6$ . Second, the two equations shown below express the same relationship between  $F_2^p$ ,  $F_3^p$  and  $\beta$ :

$$\varepsilon_4 = \frac{\varepsilon_1 \cdot F_{1 \rightarrow 4}^{ad} + \varepsilon_6 \cdot F_{6 \rightarrow 4}^{ad} + \varepsilon_2 \cdot \beta \cdot (1 - \beta) \cdot F_2^p + \varepsilon_3 \cdot \beta \cdot F_3^p}{F_{1 \rightarrow 4}^{ad} + F_{6 \rightarrow 4}^{ad} + \beta \cdot (1 - \beta) \cdot F_2^p + \beta \cdot F_3^p} \quad (8a)$$

$$(F_{5 \rightarrow 3}^{ad} + F_{9 \rightarrow 3}^{ad} + \beta \cdot F_2^p) - (F_{3 \rightarrow 1}^{ad} + F_{3 \rightarrow 2}^{ad} + \beta \cdot F_3^p) = 0 \quad (8b)$$

Equation (8a) corresponds to equation (3) for  $i = 4$ , whereas equation (8b) is equation (4) for  $i = 3$ . To maintain consistency of the system, the expressions relating  $F_2^p$ ,  $F_3^p$  and  $\beta$  in equations (8a) and (8b) must be identical. This point is examined in section 2.4. In the model calculations, we use equation (8b) for the sake of simplicity. Since the number of unknowns ( $n = 19$ ) exceeds the number of equations ( $n = 17$ ), some further hypotheses are required.

[16] 5. The authigenic particulate Nd flux formed in the North Atlantic box ( $F_1^p$ ) is assumed to be known from the field data. The Nd inventory in the North Atlantic box ( $i = 1$ ) is described by equation (3) for  $\varepsilon_{Nd(0)}$  and equation (4) for Nd fluxes:

$$\varepsilon_1 = \frac{\varepsilon_1^{ext} \cdot F_1^{ext} + \varepsilon_2 \cdot F_{2 \rightarrow 1}^{ad} + \varepsilon_3 \cdot F_{3 \rightarrow 1}^{ad} + \varepsilon_4 \cdot F_{4 \rightarrow 1}^{ad}}{F_1^{ext} + F_{2 \rightarrow 1}^{ad} + F_{3 \rightarrow 1}^{ad} + F_{4 \rightarrow 1}^{ad}} \quad (9)$$

$$(F_1^{ext} + F_{2 \rightarrow 1}^{ad} + F_{3 \rightarrow 1}^{ad} + F_{4 \rightarrow 1}^{ad}) - (F_{1 \rightarrow 4}^{ad} + F_1^p) = 0 \quad (10)$$

As the two equations contain three unknowns ( $F_1^{ext}$ ,  $\varepsilon_1^{ext}$  and  $F_1^p$ ), we need to assume that one of the three unknowns is already determined. We propose that  $F_1^p$  is known from field data (see section 2.2). To check this hypothesis, we test the sensitivity of  $F_1^{ext}$  and  $\varepsilon_1^{ext}$  with respect to  $F_1^p$  (see section 2.3).

[17] 6. To a first approximation, the remineralization rate ( $\beta$ ) is assumed to be 100%. This hypothesis is based on the behavior of authigenic  $^{230}\text{Th}$  in the water column. The in situ decay of  $^{234}\text{U}$  produces  $^{230}\text{Th}$  which is scavenged from surface waters because of its high reactivity with marine particles. The interaction between the dissolved and particulate  $^{230}\text{Th}$  is considered to be totally reversible [Bacon and Anderson, 1982; Roy-Barman et al., 1996]. This implies that  $\beta$  for the authigenic  $^{230}\text{Th}$  can attain a value of 100% and that remineralization occurs rapidly. Since authigenic Nd is more labile than authigenic  $^{230}\text{Th}$  [Arraes-Mescoff et al., 2001],  $\beta$  for Nd is assumed to be 100%. This hypothesis is discussed further in section 2.4.

[18] According to the six hypotheses, the Nd inventory in the modern ocean can be described by 17 equations containing 17 unknowns. In the following section, we estimate the scavenged Nd fluxes using field data in order to determine  $F_1^p$  and to compare the modeled Nd fluxes with the field data.

## 2.2. Authigenic Particulate Nd Fluxes and Scavenged Nd Fluxes

[19] First, we define the relationship between the authigenic particulate Nd fluxes ( $F_i^p$ ) and scavenged Nd fluxes ( $F_i^{scav}$ ). For the five surface boxes (North Atlantic, Surface Atlantic, Surface Antarctic, North Pacific and Surface INDOPAC:  $i = 1, 2, 5, 7, 8$ ), the scavenged flux is equal to the authigenic flux (Figure 2c):

$$F_i^{scav} = F_i^p \quad (11a)$$

For the Intermediate Atlantic ( $i = 3$ ), Intermediate INDOPAC ( $i = 9$ ) and Deep Antarctic ( $i = 6$ ), the scavenged Nd flux is the sum of the remaining authigenic flux from the surface,  $(1 - \beta) \cdot F_{i-1}^p$ , and the particulate flux formed in the box ( $F_i^p$ ):

$$F_i^{scav} = (1 - \beta) \cdot F_{i-1}^p + F_i^p \quad (11b)$$

For the Deep Atlantic and INDOPAC ( $i = 4, 10$ ), we consider the authigenic fluxes formed in both the surface and intermediate boxes which remain after the remineralization:

$$F_i^{scav} = (1 - \beta)^2 \cdot F_{i-2}^p + (1 - \beta) \cdot F_{i-1}^p + F_i^p + \delta_{i,10} \cdot (1 - \beta) \cdot F_7^p \quad (11c)$$

[20] Field data on sediment-trapped materials give the range of scavenged Nd fluxes. The bulk trapped material contains both scavenged and lithogenic Nd. We used three methods to distinguish authigenic Nd from lithogenic Nd: model calculation [Murphy and Dymond, 1984], chemical leaching [Tachikawa, 1997; Lerche and Nozaki, 1998] and a subtraction of lithogenic Nd from bulk data using lithogenic Nd/Th or Nd/Al ratios [Taylor and McLennan, 1985; Masuzawa and Koyama, 1989; Tachikawa et al., 1997; Fagel et al., 1999]. Trap data used in this study come from the OMEX sites in the North Atlantic ( $49^\circ 11'N$ ,  $12^\circ 49'W$ ;  $49^\circ 05'N$ ,  $13^\circ 25'W$ ;  $49^\circ 02'N$ ,  $13^\circ 42'W$  [Fagel et al., 1999]), EUMELI sites in the subtropical Atlantic ( $18^\circ N$ ,  $21^\circ W$ ;  $21^\circ N$ ,  $31^\circ W$  [Tachikawa, 1997]), the Japan Sea ( $40^\circ 50'N$ ,  $138^\circ 40'E$  [Masuzawa and Koyama, 1989]), MANOP site H

**Table 2.** Authigenic Particulate Nd Fluxes and Scavenged Nd Fluxes Calculated From the Model Together With Scavenged Nd Fluxes Estimated From Field Data

Box	Authigenic Nd flux (g/yr)		Scavenged Nd flux ( $10^{-5}$ g/m <sup>2</sup> /yr)		
	Calculated ( $\beta = 100\%$ )		Calculated ( $\beta = 100\%$ )	Field data	Reference <sup>b</sup>
1. N. Atl. (>50°N)	$8.6 \times 10^7$		0.79	0.794 (n = 1)	1
2. Surf. Atl.	$4.2 \times 10^9$		4.16	$2.18 \pm 1.98$ (n = 2)	1, 2
3. Intermed. Atl.	$4.2 \times 10^9$		3.79	$15.7 \pm 19.2$ (n = 3)	1, 2
4. Deep Atl.	$4.1 \times 10^9$		3.55	$7.32 \pm 8.65$ (n = 4)	1, 2
5. Surf. Ant.	$1.2 \times 10^9$		7.97	n.d.	
6. Deep Ant.	$9.5 \times 10^8$		6.59	n.d.	
7. N. Pac. (>44°N)	$8.3 \times 10^8$		7.69	$5.28 \pm 3.33$ (n = 2) <sup>a</sup>	3
8. Surf. INDOPAC	$4.2 \times 10^9$		2.00	$2.51 \pm 3.44$ (n = 2)	3, 4
9. Intermed. INDOPAC	$3.7 \times 10^9$		1.78	$5.29 \pm 3.85$ (n = 6)	3, 4, 5
10. Deep INDOPAC	$3.4 \times 10^9$		1.57	$6.99 \pm 6.05$ (n = 7)	3, 4, 5

n.d. = not determined.

*italic* = pre-determined from field data (see text for detail).

<sup>a</sup>No data are available in the North Pacific (>44°N). Estimate from data of the Japan Sea (40°N50'; [Masuzawa and Koyama, 1989]).

<sup>b</sup>1 = [Fagel et al., 1999] (North Atlantic), 2 = [Tachikawa, 1997] (Subtropical Atlantic), 3 = [Masuzawa and Koyama, 1989] (Japan Sea), 4 = [Murphy and Dymond, 1984] (North Pacific) and 5 = [Lerche and Nozaki, 1998] (Japan Trench).

in the equatorial Pacific (6°5'N, 93°W [Murphy and Dymond, 1984]) and the Japan Trench (34°01'N, 141°50'E [Lerche and Nozaki, 1998]). No direct data are available for the Antarctic boxes (i = 5, 6) or the North Pacific (i = 7).

[21] Using the OMEX result,  $F_1^p$  is estimated at  $8.6 \times 10^7$  g/yr ( $0.78 \times 10^{-5}$  g/m<sup>2</sup>/yr). This estimate is used for the model calculation. All the scavenged Nd fluxes estimated from field data are compiled in Table 2.

### 2.3. Modeled $F_i^{\text{ext}}$ , $\varepsilon_i^{\text{ext}}$ , Oceanic Nd Residence Time, Scavenged Nd Fluxes, and Exchanged Nd Fluxes Assuming $\beta$ is 100%

[22] The total Nd flux from the external sources toward the ocean ( $F_1^{\text{ext}} + F_2^{\text{ext}} + F_7^{\text{ext}} + F_8^{\text{ext}}$ ) is estimated at  $8.6 \times 10^9$  g/yr (Table 3). Using this total Nd influx, the mean Nd residence time ( $\tau_{\text{Nd}}$ ) is determined by:

$$\tau_{\text{Nd}} = \frac{Q}{F_1^{\text{ext}} + F_2^{\text{ext}} + F_7^{\text{ext}} + F_8^{\text{ext}}} \quad (12)$$

Knowing the total amount of Nd in the ocean ( $Q = 4.2 \times 10^{12}$  g; Table 1),  $\tau_{\text{Nd}}$  is estimated at 487 yr. This value is consistent with the lower limit of previous estimates: 200 ~ 300 years [Piepgras and Wasserburg, 1983] and ~600 years [Tachikawa et al., 1999]. As mentioned in

section 2.1,  $F_1^{\text{ext}}$  depends on  $F_1^p$ , while  $F_1^p$  is assumed to be predetermined from field data. Provided that the uncertainty of the predetermined  $F_1^p$  does not exceed one order of magnitude, the estimated  $F_1^{\text{ext}}$  ranges between  $4.4 \times 10^8$  and  $1.3 \times 10^9$  g/yr. This leads to a  $\tau_{\text{Nd}}$  of between 490 and 450 years.

[23] Neodymium sources to the ocean are characterized by  $\varepsilon_i^{\text{ext}}$  values. The calculated  $\varepsilon_1^{\text{ext}}$ ,  $\varepsilon_2^{\text{ext}}$ ,  $\varepsilon_7^{\text{ext}}$  and  $\varepsilon_8^{\text{ext}}$  are  $-22.0$ ,  $-11.4$ ,  $+1.1$ , and  $-4.4$ , respectively (Table 3). These values are qualitatively consistent with nonradiogenic old materials contributing to the Atlantic and more radiogenic young materials contributing to the Pacific. In the same way as for  $F_1^{\text{ext}}$ ,  $\varepsilon_1^{\text{ext}}$  depends on  $F_1^p$ . Using the same uncertainty as with  $F_1^p$ , we obtain  $\varepsilon_1^{\text{ext}} = -20 \pm 3$ .

[24] We can now compare the modeled scavenged Nd fluxes ( $F_i^{\text{scav}}$ ) with the estimates from field data (Figure 3 and Table 2). The field data used for the North Pacific box (i = 7) are derived from data in the Japan Sea (40°50'N) because no direct measurements are available in the Pacific north of 44°N. All the calculated  $F_i^{\text{scav}}$  are in agreement with the field data within error (Figure 3). The calculated  $F_i^{\text{scav}}$  values are the highest in the surface boxes and decrease toward the respective intermediate and deep boxes (Table 2). This tendency is generally observed in the real ocean.

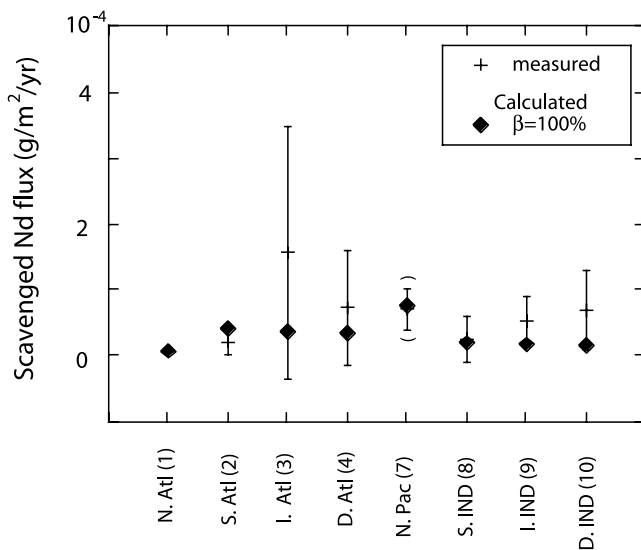
**Table 3.** Comparison of the Nd Fluxes (g/yr) and  $\varepsilon_{\text{Nd}(0)}$  Between the Calculated Influxes and Some Potential Sources

Box	Calculated influx		"Missing" <sup>a</sup>		Redissolution in estuaries <sup>b</sup>		30% of dust dissolution <sup>c</sup>	
	$F_i^{\text{ext}}$	$\varepsilon_i^{\text{ext}}$	Flux	$\varepsilon_{\text{Nd}(0)}$	Flux	$\varepsilon_{\text{Nd}(0)}$	Flux	$\varepsilon_{\text{Nd}(0)}$
1. N. Atl.	$5.2 \cdot 10^8$	-22.0	$4 \cdot 10^8$	-24	$3 \cdot 10^8$	-17	$3 \cdot 10^7$	-15
2. Surf. Atl.	$4.3 \cdot 10^9$	-11.4	$4 \cdot 10^9$	-11	$6 \cdot 10^8$	-11	$2 \cdot 10^9$	-12
7. N. Pac.	$5.3 \cdot 10^8$	+1.1	$5 \cdot 10^8$	+2	$4 \cdot 10^7$	-6	$4 \cdot 10^8$	-5
8. Surf. INDOPAC	$3.3 \cdot 10^9$	-4.4	$3 \cdot 10^9$	-4	$2 \cdot 10^8$	-4	$3 \cdot 10^9$	-7
Global	$8.6 \cdot 10^9$	-8.6	$8 \cdot 10^9$	-9	$1 \cdot 10^9$	-11	$6 \cdot 10^9$	-8

<sup>a</sup>The difference between the calculated global flux and the sum of dissolved river flux (70% of Nd removal in estuaries) and atmospheric flux (2% of Nd dissolution). See text for details.

<sup>b</sup>After Goldstein and Jacobsen [1987]. The fluxes given by these authors are redistributed to the four surface boxes for comparison with our estimates (uncertainty of the estimates is a factor of 2 ~ 3) [Goldstein and Jacobsen, 1987]. All Nd removed in estuaries (70%) are assumed to be released to the solution.

<sup>c</sup>The mineral aerosol flux is estimated from the map of Duce et al., [1991] and the Nd isotopic ratios are estimated from aerosols, sediments and source rocks [Goldstein et al., 1984; Goldstein and Jacobsen, 1987, 1988; Grousset et al., 1988, 1992; McLennan et al., 1990; Nakai et al., 1993; Jones et al., 1994].



**Figure 3.** Modeled scavenged Nd fluxes ( $\beta = 100\%$ ) compared with estimates from field data (Table 2). The large uncertainties in the field data are due to the scattering of data obtained from sites having completely different oceanic conditions. As direct field data are not available for the North Pacific box ( $>44^\circ\text{N}$ ), the flux for this box is derived from data in the Japan Sea ( $40^\circ\text{N}50^\circ$ ) and presented here between blankets. All the calculated fluxes are in agreement with the field data within error.

[25] Finally, we compare the modeled exchanged Nd flux with previous estimates [Bertram and Elderfield, 1993]. The exchanged Nd flux is defined as the part of the remineralized Nd flux that is balanced with the authigenic Nd flux formed in the same box. For instance, in the Intermediate INDOPAC ( $i = 9$ ), the remineralized Nd flux is  $4.2 \times 10^9$  g/yr and the authigenic Nd flux is  $3.7 \times 10^9$  g/yr (Table 2). Therefore  $3.7 \times 10^9$  g/yr of the remineralized Nd flux is balanced with the authigenic Nd flux (an excess of remineralized Nd flux equal to  $0.5 \times 10^9$  g/yr). Consequently, the exchanged Nd flux in this box is  $3.7 \times 10^9$  g/yr. In the same manner, the exchanged Nd flux in the whole ocean is estimated at  $1.6 \times 10^{10}$  g/yr. This value corresponds to 40% of the previous estimate ( $4.2 \times 10^{10}$  g/yr [Bertram and Elderfield, 1993]). These authors applied a seven-box model, using different dissolved Nd concentrations and  $\varepsilon_{\text{Nd}(0)}$  of the boxes compared with our values. Despite the differences in calculation, there is a satisfactory agreement within the orders of magnitude.

#### 2.4. Sensitivity of Calculated $\varepsilon_i^{\text{ext}}$ , Nd Residence Time ( $\tau_{\text{Nd}}$ ), and Scavenged Nd Fluxes ( $F_i^{\text{scav}}$ ) to Variations in Remineralization Rate ( $\beta$ )

[26] Changes in  $\beta$  influence the Nd influxes ( $F_i^{\text{ext}}$ ) as well as the  $\varepsilon_{\text{Nd}(0)}$  values ( $\varepsilon_i^{\text{ext}}$ ). Let us consider the case of the North Pacific ( $i = 7$ ). The authigenic particulate Nd flux formed in the box ( $F_7^{\text{p}}$ ) is remineralized in the Deep INDOPAC ( $i = 10$ , Figure 2c). Since  $\varepsilon_7$  ( $-4.0$ ) is higher than  $\varepsilon_{10}$  ( $-5.6$ ), this remineralization provides radiogenic Nd to box 10. The magnitude of remineralized Nd flux ( $\beta \cdot F_7^{\text{p}}$ ) is constrained by the balance of  $\varepsilon_{\text{Nd}(0)}$  and Nd fluxes (equations (3) and (4)). If we decrease  $\beta$ , the value of  $F_7^{\text{p}}$  increases to maintain the

required  $\beta \cdot F_7^{\text{p}}$ . The increase in  $F_7^{\text{p}}$  requires a higher value of  $F_7^{\text{ext}}$  to maintain a constant Nd concentration in box 7 (equation (4)). Thus the change in  $F_7^{\text{ext}}$  produces a modification of  $\varepsilon_i^{\text{ext}}$  (equation (3)). Moreover, the increase in the Nd influx ( $F_7^{\text{ext}}$ ) shortens the oceanic Nd residence time (equation (12)). The sensitivity of the calculated values with respect to  $\beta$  is variable from one box to another, depending upon the required remineralized Nd flux.

[27] The  $\varepsilon_i^{\text{ext}}$  values for the Surface Atlantic ( $i = 2$ ) and Surface INDOPAC ( $i = 8$ ) are not sensitive to  $\beta$ :  $\varepsilon_2^{\text{ext}}$  varies only from  $-11.2$  ( $\beta = 20\%$ ) to  $-11.4$  ( $\beta = 100\%$ ) and  $\varepsilon_8^{\text{ext}}$  from  $-4.3$  to  $-4.4$  when  $\beta$  ranges from 20% to 100% (Figure 4a). By contrast,  $\varepsilon_i^{\text{ext}}$  for the North Pacific ( $i = 7$ ) shows a significant variation with  $\beta$ :  $\varepsilon_7^{\text{ext}}$  is stable ( $-3.9$  to  $-3.4$ ) for  $\beta < 60\%$  and increases markedly from  $-3.4$  to  $+1.1$  for  $\beta > 60\%$  (Figure 4a).

[28] The calculated  $\tau_{\text{Nd}}$  value is very sensitive to  $\beta$  (Figure 4b). It decreases with  $\beta$ , reaching a value of 30 years at  $\beta = 20\%$ . Such a short  $\tau_{\text{Nd}}$  has never been reported for Nd, and is incompatible with the reactivity of this element with marine particles (residence time of Al which is more reactive than Nd, having a residence time is estimated at  $\sim 50$  yr). The shortest  $\tau_{\text{Nd}}$  previously reported for Nd is a few hundred of years [Piepgras and Wasserburg, 1983]. To obtain this range of  $\tau_{\text{Nd}}$ ,  $\beta > 60\%$  is required. The relationship between  $\tau_{\text{Nd}}$  and  $\beta$  favors a high remineralization rate.

[29] We compare the calculated scavenged Nd fluxes ( $F_i^{\text{scav}}$ ) with field data using  $\beta$  values of 60 and 80% (Figure 4c). In both cases,  $F_i^{\text{scav}}$  for the Surface Atlantic and the North Pacific ( $i = 2, 7$ ) are much higher than the field data. In particular,  $F_7^{\text{scav}}$  is surprisingly high, corresponding to a value  $\sim 10$  times higher than in the other INDOPAC boxes (Figure 4c). There is no geochemical reason for having such a high flux just in the North Pacific box. As the  $F_i^{\text{scav}}$  values decrease with increasing  $\beta$ , the relationship between  $F_i^{\text{scav}}$  and  $\beta$  favors a high remineralization rate.

[30] Finally, we can observe the consistency of equations (8a) and (8b) (section 2.1) with respect to varying  $\beta$ . The discrepancy in  $F_3^{\text{p}}$  calculated from the two equations decreases with increasing  $\beta$ . It becomes minimal when  $\beta = 100\%$ . Since both equations should define the same  $F_3^{\text{p}}$ , this relationship also supports a high  $\beta$  value.

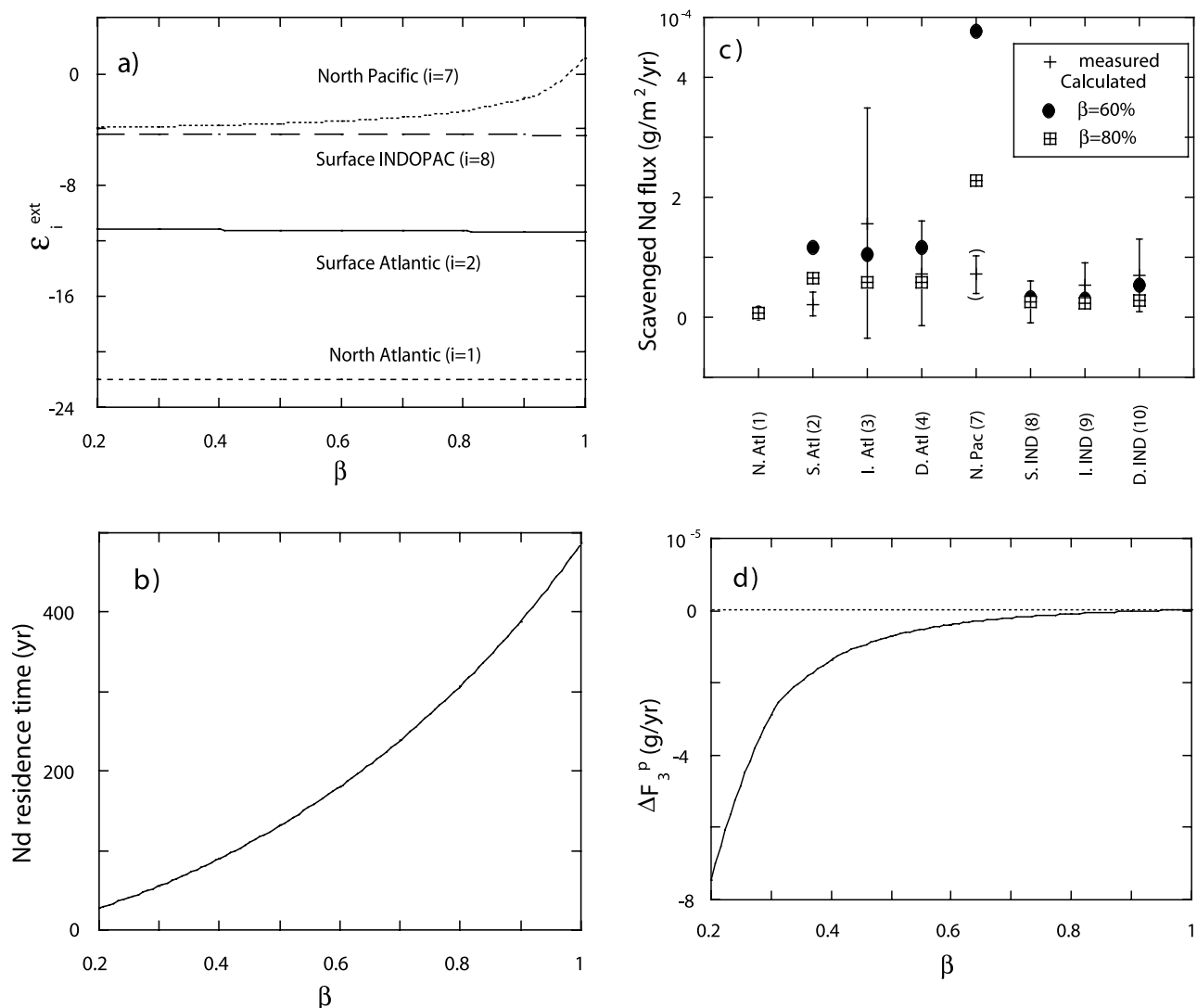
[31] We use a remineralization rate of 100% in the following discussion, since all the results suggest it is the most appropriate value consistent with the presently available data set.

### 3. Discussion

#### 3.1. Neodymium Sources to the Ocean

[32] We compare the estimated Nd influxes to the ocean with the sum of riverine and atmospheric inputs. Taking into account that 70% of the dissolved Nd brought in by rivers is removed in estuaries, the net Nd flux from the dissolved river load is  $5 \times 10^8$  g/yr [Goldstein and Jacobsen, 1987]. Using a total mineral aerosol flux of  $9.1 \times 10^{14}$  g/yr [Duce et al., 1991], a Nd concentration in the dust of  $20 \mu\text{g/g}$  [Goldstein et al., 1984; Grousset et al., 1988, 1998] and a soluble Nd proportion of 2% [Greaves et al., 1994], we obtain a net Nd flux from dust of  $4 \times 10^8$  g/yr. Hence the sum of Nd fluxes from rivers and dust is  $9 \times 10^8$  g/yr. This





**Figure 4.** (a) Sensitivities in calculated  $\epsilon_{Nd(0)}$  of external Nd sources ( $\epsilon_i^{ext}$ ) as a function of  $\beta$ ; (b) oceanic Nd residence time as a function of  $\beta$ ; (c) modeled scavenged Nd fluxes ( $\beta = 60$  or  $80\%$ ) compared with the estimates from field data; and (d) difference in authigenic Nd flux formed in the Intermediate Atlantic box ( $F_3^P$ ) determined from equations (8a) and (8b) as a function of  $\beta$ .

is about 10 times lower than the calculated total Nd influx, suggesting that other Nd sources may contribute to the oceanic Nd inventory.

[33] Potential Nd sources for the “missing flux” (the difference between the calculated total influx and the sum of dust and river fluxes,  $8 \times 10^9$  g/yr; Table 3) are as follows: (1) the release of particulate Nd from rivers by contact with seawater [Spivack and Wasserburg, 1988; Sholkovitz, 1993; Jones et al., 1994; Byrne and Sholkovitz, 1996; von Blanckenburg et al., 1996]; (2) a higher dissolution rate ( $>2\%$ ) of Nd from dust [Henry et al., 1994; Jeandel et al., 1995; Tachikawa et al., 1999]; and (3) sediment/seawater interactions at continental margins [German and Elderfield, 1990; Moran and Moore, 1991; von Blanckenburg et al., 1996; Jeandel et al., 1998; Amakawa et al., 2000; Lacan and Jeandel, 2001]. Another potential source is groundwater, which is a significant source for oceanic Ra and Ba [Moore, 1997]. However, since no data on Nd isotopic ratios are available, we are

unable to quantify the contribution of this source. In the following, we discuss the potential contribution of the first three possible sources only.

### 3.1.1. Release of Particulate Nd From Rivers by Contact With Seawater

[34] River inputs contribute to both dissolved and particulate Nd in the ocean. Although dissolved riverine Nd is removed from solution in estuaries at  $0 \sim 5$  psu, a redissolution of removed Nd is observed at higher salinity ( $5 \sim 35$ ) [Sholkovitz, 1993]. As the global river particle flux is huge ( $1.4 \times 10^{16}$  g/yr [Milliman and Meade, 1983] to  $2.3 \times 10^{16}$  g/yr [Holeman, 1968]), the partial dissolution of river particles could be an important Nd source. Assuming that the Nd concentration of river particles is  $\sim 30$   $\mu\text{g/g}$  [Goldstein et al., 1984], only 1 to 2% of partial Nd dissolution would be sufficient to obtain the missing flux (Table 3). The importance of this source is also suggested by  $^9\text{Be}$ , another tracer derived from continents [von Blanckenburg et al., 1996].

[35] To evaluate the contribution of “the redissolved Nd”, we can assume that all the Nd removed in estuaries goes back into solution. This provides us with an upper limit for the contribution. The total redissolved Nd flux is  $1 \times 10^9$  g/yr [after Goldstein and Jacobsen, 1987], which corresponds to 13% of the total “missing flux” (Table 3). On a regional scale, it represents 75%, 15%, 8% and 7% of the missing flux for the North Atlantic, Surface Atlantic, North Pacific and Surface INDOPAC, respectively.

[36] The  $\epsilon_{Nd(0)}$  of the redissolved Nd flux agrees closely with the value for the missing flux in the Surface Atlantic and Surface INDOPAC boxes (Table 3). By contrast, it differs by +7 and -8  $\epsilon$  units from the  $\epsilon_{Nd(0)}$  value of the missing fluxes in the North Atlantic and North Pacific boxes, respectively. The redissolved Nd flux may not be a prominent Nd source to the North Pacific because of its small contribution (8%) and the discrepancy in  $\epsilon_{Nd(0)}$ . We do not totally rule out the possibility of this source to the North Atlantic box because of the uncertainty of  $\epsilon_1^{ext}$  related to  $F_1^p$ . However, if  $\epsilon_1^{ext}$  is correctly estimated, the supplementary Nd source to this box would be characterized by a highly negative  $\epsilon_{Nd(0)}$  of -45, which is not plausible.

[37] River particles consist of several minerals having different  $\epsilon_{Nd(0)}$ . Provided that the  $\epsilon_{Nd(0)}$  of soluble river particles is the same as that of river water, we obtain the same  $\epsilon_{Nd(0)}$  distribution as found for redissolved Nd (Table 3). Consequently, river particles could contribute to the Surface Atlantic and Surface INDOPAC boxes. If the  $\epsilon_{Nd(0)}$  of soluble river particles is different from that of river water, river particles may contribute to the budget of Nd in other regions. Volcanic materials (high  $\epsilon_{Nd(0)}$ ) are suggested to be more soluble than old silicate materials (low  $\epsilon_{Nd(0)}$ ) [Albarède and Goldstein, 1992; Jones et al., 1994]. Therefore this source may contribute to the North Pacific box but probably not to the North Atlantic box.

### 3.1.2. Higher Dissolution of Nd From Atmospheric Fallout

[38] The proportion of soluble Nd in dust ( $\alpha$ ) is highly variable (2 ~ 50%) depending on estimation methods. The proportion obtained using Nd isotopes [Henry et al., 1994; Jeandel et al., 1995; Tachikawa et al., 1999] is systematically higher than that based on Nd concentration analysis [Greaves et al., 1994]. The discrepancy is likely related to dissolved/particulate Nd exchanges that can be detected by isotope ratios but not by elemental concentrations. Dust enters the ocean and releases Nd by partial dissolution. If seawater Nd is adsorbed onto dust particles after the dissolution of Nd from the particles, it is impossible to estimate the original proportion of dissolution from the change in Nd concentration (the Nd dissolution is partly canceled by the readsorption). By contrast,  $\epsilon_{Nd(0)}$  may indicate the original dissolution if the  $\epsilon_{Nd(0)}$  of seawater is different from that of the dust. Thus the discrepancy of the estimates may be due to Nd exchange between dust and seawater. Using  $\alpha = 30\%$ , we obtain a global Nd influx of  $6 \times 10^9$  g/yr, which almost agrees with the missing flux (Table 3).

[39] We can examine the dust contribution to the missing flux on the basis of  $\epsilon_{Nd(0)}$  assuming that  $\alpha = 30\%$ . The dust  $\epsilon_{Nd(0)}$  is evaluated from data on aerosols [Goldstein et al., 1984; Goldstein and Jacobsen, 1987, 1988; Grousset et al., 1988, 1992], marine sediments [Grousset et al., 1988, 1992;

Nakai et al., 1993; Jones et al., 1994] and combining the wind direction with the age of rocks in various source regions [Goldstein and Jacobsen, 1987; McLennan et al., 1990]. The dust  $\epsilon_{Nd(0)}$  are approximately -15, -12, -5, and -7 for the North and Surface Atlantic, North Pacific and Surface INDOPAC boxes, respectively (Table 3). The results suggest the lack of nonradiogenic Nd sources to the North Atlantic and radiogenic Nd sources to the North Pacific box (Table 3). In other words, if rivers and dust were the only Nd source for the missing flux, we would be unable to reproduce the regional  $\epsilon_{Nd(0)}$  variation of the sources. This means that additional local Nd sources provide a highly negative  $\epsilon_{Nd(0)}$  to the North Atlantic and a radiogenic Nd component to the North Pacific.

### 3.1.3. Sediment/Seawater Interaction at the Continental Margins

[40] The  $\epsilon_{Nd(0)}$  of the continental margin is highly variable from region to region: the highest value of the circum-Pacific margin ( $\epsilon_{Nd(0)} = -4.2 \pm 3.8$ : [Jones et al., 1994],  $\epsilon_{Nd(0)} = +2 \pm 6$ : von Blanckenburg, unpublished compilation) is as high as the estimated  $\epsilon_{Nd(0)}$  of the influx to the North Pacific box. On the other hand, values for Greenland and the Canadian shield ( $\epsilon_{Nd(0)} = -14/-32$  [Grousset et al., 1988]) are as low as the estimated  $\epsilon_{Nd(0)}$  influx to the North Atlantic box. Hence Nd fluxes from the continental margins could generate regional variations of  $\epsilon_{Nd(0)}$  in the influxes.

[41] The release of lithogenic elements from continental margins was originally suggested from Al analysis [Moran and Moore, 1991]. Laboratory and field studies (the deep NW Atlantic) suggest that the continuous resuspension and deposition of sediments within the nepheloid layer could promote the release of Al into the overlying water. Jeandel et al. [1998] suspected the release of Nd in an area of high hydraulic energy (~6 Sv) in the Indonesian Strait. Recently, Amakawa et al. [2000] showed that remobilization from coastal and shelf sediments may be an important Nd source to the eastern Indian Ocean and its adjacent seas. In the studied area, the riverine dissolved Nd flux appears to be relatively minor. In addition, surface seawater  $\epsilon_{Nd(0)}$  varies over a wide range from -11.4 to -1.3, suggesting a Nd contribution to surface waters from local sources such as old rocks in the Himalayas and from volcanic islands [Amakawa et al., 2000]. The global budget of Be isotopes also suggests that continental margins are of key importance for the  $^9\text{Be}$  inventory [von Blanckenburg et al., 1996]. The additional flux required for  $^9\text{Be}$  is  $5 \pm 3 \times 10^8$  g/yr [von Blanckenburg et al., 1996], which is about 10 times lower than that for Nd. Interestingly, this difference corresponds to the concentration ratio between these elements in the continental crust (Be and Nd concentrations in the upper continental crust are 3  $\mu\text{g/g}$  and 26  $\mu\text{g/g}$ , respectively [Taylor and McLennan, 1985]).

[42] The influence of continental margins on seawater  $\epsilon_{Nd(0)}$  is directly suggested by data from the Mauritanian upwelling region in the NE Atlantic [Tachikawa et al., 1999]. Seawater  $\epsilon_{Nd(0)}$  values off the African continent were analyzed at the E site (~200 km offshore, 20°N, 18°W) and the M site (~500 km offshore, 18°N, 21°W). Although the water mass structures are very similar at these sites, the seawater  $\epsilon_{Nd(0)}$  values are different.  $\epsilon_{Nd(0)}$  at the E site (-13.0 to -12.1) is closer to the African continent value (-13 to -12) [Grousset et al., 1992, 1988] than that at the

M site (−12.2 to −10.8). Since this trend is not limited to the surface layer, the partial dissolution of atmospheric dust could not be a major reason for the more negative values at the E site. In fact, the E site is located in the Mauritanian upwelling region where the organic flux is high. The high organic flux produces a reducing environment in the sediments [Legeleux *et al.*, 1994], which could promote a Nd release from coastal sediments during early diagenesis. Fluids containing diagenetic Nd derived from reducing coastal sediments have been observed in other regions [German and Elderfield, 1990]. It is noteworthy that the dissolved Nd concentration at the E site is no higher than that at the M site. This may be related to a higher particulate flux at the E site which could remove excess dissolved Nd by scavenging. The influence of margins on seawater  $\epsilon_{\text{Nd}(0)}$  is also suggested in the equatorial Pacific [Lacan and Jeandel, 2001]. The  $\epsilon_{\text{Nd}(0)}$  of the Antarctic Intermediate Water (AAIW) becomes more radiogenic as the AAIW flows along the highly radiogenic Papua New Guinea slope in the Pacific [Lacan and Jeandel, 2001]. We do not precisely understand yet how continental margins release terrigenous elements to the ocean. Further studies are required to identify the mechanisms and to quantify the fluxes from this potential source.

### 3.2. Sensitivity of Deep Water $\epsilon_{\text{Nd}(0)}$ With Nd Influxes: Paleo-Oceanographic Implications

[43] In this section, we examine the sensitivity of deep water Nd concentrations and  $\epsilon_{\text{Nd}(0)}$  values by varying the Nd inputs to the ocean. Changes in deep water  $\epsilon_{\text{Nd}(0)}$  recorded in ferromanganese crusts and nodules are often interpreted as resulting from modifications in paleoceanic circulation. However, we know that fluxes from the continents to the ocean underwent changes during glacial/interglacial periods. For instance, dust fluxes increased by a factor of 3 to 5 during glacial periods mainly because of an increase in continental aridity [Rea, 1994]. Abrasion of the continents by ice sheets could also accelerate the erosion [Williams *et al.*, 1998]. In addition to the changing magnitude of influxes, Nd sources to the ocean might vary with time because of changes in wind direction, aridity of continents, vegetation cover and ice sheets. In this section, we examine whether deep water  $\epsilon_{\text{Nd}(0)}$  might vary in response to changing Nd inputs to the ocean, even though oceanic circulation patterns remain unchanged.

[44] We calculate 10 Nd concentrations ( $C_i$ ) and 10  $\epsilon_{\text{Nd}(0)}$  values ( $\epsilon_i$ ) at nonsteady state following modifications in the external Nd fluxes ( $F_i^{\text{ext}}$ ) and their sources ( $\epsilon_i^{\text{ext}}$ ). Two cases are discussed. In case 1, all the present-day  $F_i^{\text{ext}}$  are multiplied by a factor of 4, whereas  $\epsilon_i^{\text{ext}}$  values remain at present-day values. The factor of 4 is chosen to mimic glacial situations with increased dust flux and accumulation rates of lithogenic components in sediments [Rea, 1994; Williams *et al.*, 1998]. In case 2, all the  $F_i^{\text{ext}}$  are taken at the present-day values, whereas all the  $\epsilon_i^{\text{ext}}$  values are increased by +1  $\epsilon$ -unit ( $\Delta\epsilon_i^{\text{ext}} = +1$ ) to mimic the change in external Nd sources. We use an arbitrary  $\Delta\epsilon_i^{\text{ext}} = +1$  to study the response of the deep water  $\epsilon_{\text{Nd}(0)}$ . The precise estimation of  $\epsilon_i^{\text{ext}}$  variability with time is beyond the scope of this study.

[45]  $C_i$  at nonsteady state is described with 10 differential equations (equation (4), (5), or (6), depending upon the

box). These three equations can be applied to any Nd isotope. Using  $R_i = {}^{143}\text{Nd}/{}^{144}\text{Nd}$ , equations (4), (5), and (6) can be written as follows:

$$V_i \frac{R_i \cdot C_i}{dt} = \left( R_i^{\text{ext}} \cdot F_i^{\text{ext}} + \sum_{j \neq i} R_j \cdot F_{j \rightarrow i}^{\text{ad}} \right) - \left( \sum_{j \neq i} R_i \cdot F_{i \rightarrow j}^{\text{ad}} + R_i \cdot F_i^{\text{p}} \right) \quad (13)$$

$$V_i \frac{R_i \cdot C_i}{dt} = \left( R_{i-1} \cdot \beta \cdot F_{i-1}^{\text{p}} + \sum_{j \neq i} R_j \cdot F_{j \rightarrow i}^{\text{ad}} \right) - \left( \sum_{j \neq i} R_i \cdot F_{i \rightarrow j}^{\text{ad}} + R_i \cdot F_i^{\text{p}} \right) \quad (14)$$

$$V_i \frac{R_i \cdot C_i}{dt} = \left( R_{i-1} \cdot \beta \cdot F_{i-1}^{\text{p}} + \delta_{i,10} \cdot (\beta \cdot F_7^{\text{p}}) + R_{i-2} \cdot \beta \cdot (1 - \beta) \cdot F_{i-2}^{\text{p}} + \sum_{j \neq i} R_j \cdot F_{j \rightarrow i}^{\text{ad}} \right) - \left( \sum_{j \neq i} R_i \cdot F_{i \rightarrow j}^{\text{ad}} + R_i \cdot F_i^{\text{p}} \right) \quad (15)$$

$R_i$  determines an  $\epsilon_{\text{Nd}(0)}$  value in box  $i$  ( $\epsilon_i$ ) by fixing  $\epsilon_{\text{Nd}(0)}$ . The variability in  $R_i$  of the 10 boxes is described by 10 differential equations based on equations (13), (14), or (15). Consequently, the Nd inventory at nonsteady state will be determined by 20 differential equations. We apply the following three hypotheses to the modeling. (1) The volume of the 10 boxes ( $V_i$ ) as well as the advective water fluxes ( $F_{i \rightarrow j}^{\text{ad,sw}}$ ) are known (Table 1 and Figure 2b) and do not change with time; (2) the authigenic particulate Nd flux ( $F_i^{\text{p}}$ ) is formed in proportion to the dissolved Nd concentration in box  $i$  ( $C_i$ ), with a constant  $f_i^{\text{p}}$ :

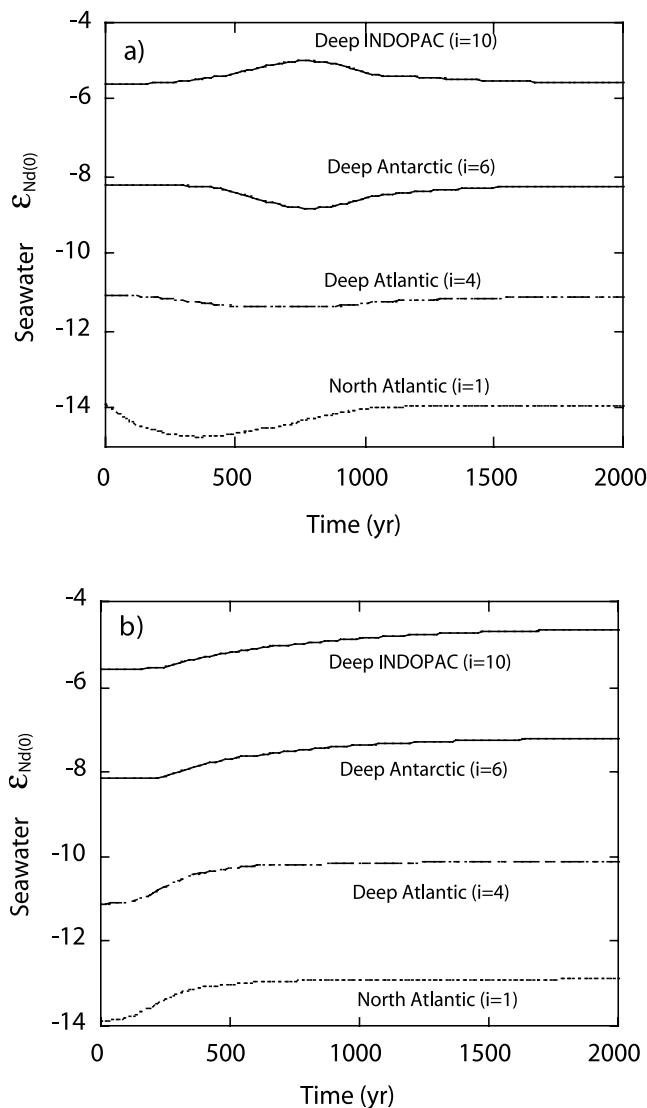
$$F_i^{\text{p}} = f_i^{\text{p}} \cdot C_i \quad (16a)$$

$$f_i^{\text{p}} = (F_i^{\text{p}})_{\text{present}} / (C_i)_{\text{present}}, \quad (16b)$$

where  $(F_i^{\text{p}})_{\text{present}}$  and  $(C_i)_{\text{present}}$  are present-day  $F_i^{\text{p}}$  (Table 2) and present-day  $C_i$  (Table 1).  $f_i^{\text{p}}$  is assumed not to vary with time; and (3)  $\beta$  is assumed to be 100%.

[46] Under these conditions, 20 differential equations are solved using Matlab. The present-day  $C_i$  and  $\epsilon_i$  ( $R_i$ ) are used as an initial condition (Table 1). The model is run for 2000 yr.

[47] We focus here on the Nd variability in the four deep boxes ( $i = 1, 4, 6$  and  $10$ ), because the deep water Nd signatures could be recorded in ferromanganese crusts and nodules. The results show that deep water Nd concentrations and  $\epsilon_{\text{Nd}(0)}$  change in response to the Nd inputs (Figure 5). In case 1, Nd concentrations in the deep boxes increase by a factor of four, which is proportional to the change in  $F_i^{\text{p}}$  (not shown in figure). The deep water  $\epsilon_{\text{Nd}(0)}$  values show transitional changes and revert to the initial values (Figure 5a). In case 2 ( $\Delta\epsilon_i^{\text{ext}} = +1$ ), the deep water  $\epsilon_{\text{Nd}(0)}$  values increase by +1  $\epsilon$  unit (Figure 5b). When  $\Delta\epsilon_i^{\text{ext}}$  is +2, the shift of deep water  $\epsilon_{\text{Nd}(0)}$  is also +2 (not shown in figure). Our results suggest that the change in deep water  $\epsilon_{\text{Nd}(0)}$  is



**Figure 5.** Variability in  $\epsilon_{Nd(0)}$  of the deep waters (boxes 1, 4, 6, and 10) (a) where the Nd influxes ( $F_i^{ext}$ ) are multiplied by a factor of 4 using the present-day  $\epsilon_i^{ext}$  and (b) where the present-day  $F_i^{ext}$  values are used with an increased  $\epsilon_i^{ext}$  ( $\Delta\epsilon_i^{ext} = +1$ ). The x axis shows the time since modified Nd influxes are introduced to the ocean.

proportional to  $\Delta\epsilon_i^{ext}$ . Note that these Nd concentrations and  $\epsilon_{Nd(0)}$  values in deep boxes are obtained under conditions where advective water fluxes remain unchanged.

[48] Up to now, changes in seawater  $\epsilon_{Nd(0)}$  recorded in Fe-Mn nodules, crusts and oxide components of sediments have been generally interpreted as reflecting a variability in paleoceanic circulation. For instance, an increase in  $\epsilon_{Nd(0)}$  in the Antarctic ( $i = 6$ ) and Indo-Pacific sector ( $i = 10$ ) could be explained by a decrease in NADW. We show here that a similar  $\epsilon_{Nd(0)}$  shift can be reproduced with no change in NADW (Figure 5b). We also examined the variability in deep water  $\epsilon_{Nd(0)}$  using a  $\beta$  of 70% (not shown in figure). The main trend feature of change is similar to that with  $\beta$  of 100%.

[49] Our results suggest that seawater  $\epsilon_{Nd(0)}$  may vary according to Nd inputs to the ocean as well as changes in

water circulation. Since oceanic Nd inputs may vary because of changing conditions of continental erosion, this effect should be considered, in some cases, as accounting for the variability of seawater  $\epsilon_{Nd(0)}$  in the paleo-ocean.

#### 4. Conclusions

[50] The oceanic Nd budget was calculated using a 10-box model (PANDORA) and a compilation of field data. With the currently available data, the best estimates give a Nd flux of  $9 \times 10^9$  g/yr entering the ocean, which yields an oceanic Nd residence time of  $\sim 500$  yr. Although neodymium sources to the ocean are highly variable from region to region ( $\epsilon_{Nd(0)}$  from  $-22$  to  $+1$ ), riverine and atmospheric fluxes cannot explain the variability. Therefore additional Nd sources must be considered. One of the potential Nd sources may be the continental margins.

[51] Using the model calibrated for Nd, we examined the sensitivity of deep water  $\epsilon_{Nd(0)}$  as a function of Nd inputs to the ocean. The deep water  $\epsilon_{Nd(0)}$  value varies in proportion to changes in the  $\epsilon_{Nd(0)}$  of external sources. This implies that shifts in deep water  $\epsilon_{Nd(0)}$  recorded in ferromanganese nodules and crusts do not necessarily reflect changes in oceanic circulation patterns. The deep water  $\epsilon_{Nd(0)}$  shifts could be interpreted in terms of changes in Nd inputs related to continental erosion.

[52] The calibration of our model is essentially based on field data. Although all the available literature data were used to establish the model, more data are required to improve the quality of calibration. In particular, we note that seawater Nd data are scarce in the Antarctic and the North Pacific (north of  $44^\circ N$ ). We also require the analysis of sediment-trapped material from the Indian Ocean.

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