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Sensitivity of Nd isotopic composition in seawater to changes in Nd sources and paleoceanographic implications

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[1] It has been argued that past changes in the sources of Nd could hamper the use of the Nd isotopic composition (ϵ_{Nd}) as a proxy for past changes in the overturning of deep water masses. Here we reconsider uncertainties associated with ϵ_{Nd} in seawater due to potential regional to global scale changes in the sources of Nd by applying a modeling approach. For illustrative purposes we describe rather extreme changes in the magnitude of source fluxes, their isotopic composition or both. We find that the largest effects on ϵ_{Nd} result from changes in the boundary source. Considerable changes also result from variations in the magnitude or ϵ_{Nd} of dust and rivers but are largely constrained to depths shallower than 1 km, except if they occur in or upstream of regions where deep water masses are formed. From these results we conclude that changes in Nd sources have the potential to affect ϵ_{Nd}. However, substantial changes are required to generate large-scale changes in ϵ_{Nd} in deep water that are similar in magnitude to those that have been reconstructed from sediment cores or result from changes in meridional overturning circulation in model experiments. Hence, it appears that a shift in ϵ_{Nd} comparable to glacial-interglacial variations is difficult to obtain by changes in Nd sources alone, but that more subtle variations can be caused by such changes and must be interpreted with caution.

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1. Introduction

[2] The isotopic composition of neodymium (ϵ_{Nd}) is a novel and promising tracer of water mass distribution and mixing [*von Blanckenburg*, 1999; *Rutberg et al.*, 2000; *Frank*, 2002; *Goldstein and Hemming*, 2003]

$$\epsilon_{Nd} = \left(\frac{\left(^{143}Nd/^{144}Nd\right)_{sample}}{\left(^{143}Nd/^{144}Nd\right)_{std}} - 1 \right) \cdot 10^4, \qquad (1)$$

where $(^{143}Nd/^{144}Nd)_{std}$ corresponds to the "bulk earth" reference value of 0.512638 [*Jacobsen and Wasserburg*, 1980]. However, the use of ϵ_{Nd} as paleocirculation tracer is not without ambiguities. For example, *Tachikawa et al.* [2003] reported that variations in ϵ_{Nd} in seawater not only might reflect changes in ocean overturning circulation but also changes in the magnitude or the isotopic composition of Nd sources. Such effects would affect the use of ϵ_{Nd} as a paleoceanographic tracer as it would complicate the interpretation of variations in ϵ_{Nd} as changes in water mass distribution and mixing.

[3] The purpose of this study is to reconsider uncertainties associated with ϵ_{Nd} in seawater due to potential regional to global scale changes in the magnitude or the isotopic composition of Nd sources by applying the cost-efficient Bern3D Earth System Model of Intermediate Complexity. The model has been shown to simulate both Nd concentration and ϵ_{Nd} in reasonable agreement with observations [*Rempfer et al.*, 2011, 2012] when forced with Nd input at continental boundaries, from rivers and dust deposition. For illustrative purposes we describe rather extreme changes in the magnitude of source fluxes, in their isotopic composition or in both quantities for different regions and different source processes individually.

[4] In the modern ocean, particularly in the Atlantic basin, ϵ_{Nd} covaries with salinity and thus traces the distribution and mixing of water masses [*von Blanckenburg*, 1999; *Goldstein and Hemming*, 2003; *Rempfer et al.*, 2011]. Deep water ϵ_{Nd} is reliably recorded in archives such as benthic foraminifera, Fe-Mn coatings, fish teeth, and deep water corals [e.g., *Martin and Haley*, 2000; *Klevenz et al.*, 2008; *van de Flierdt et al.*, 2010; *Elmore et al.*, 2011] and variations in ϵ_{Nd} extracted from such archives are increasingly being interpreted as changes in the distribution of water masses [e.g., *Rutberg et al.*, 2000; *Frank*, 2002; *Piotrowski et al.*, 2004; *Roberts et al.*, 2010]. In a recent study, *Rempfer et al.* [2012]

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Table 1. List of Parameters, Corresponding Abbreviations, and Their Values or References

Variable	Symbol	CTRL	Unit
Nd dissolved concentration	$[Nd]_d$		pmol kg^{-1}
Nd dust source, total	f_{du}	$2.6 \cdot 10^8$	g Nd yr^{-1}
ϵ_{Nd} of dust source	$\epsilon_{Nd}(du)$	*Tachikawa et al.* [2003]	ϵ_{Nd}-units
Nd-concentration dust	$c_{du,Nd}$	20	μg g^{-1}
Nd release from dust	$\beta_{du,Nd}$	0.02	
Nd river source, total	f_{ri}	$3.4 \cdot 10^8$	g Nd yr^{-1}
Nd-concentration, river	$c_{ri,Nd}(\theta, \phi)$	*Goldstein and Jacobsen* [1987]	g Nd m^{-3}
Nd-removal, estuaries	$\gamma_{ri,Nd}$	0.7	
ϵ_{Nd} of river source	$\epsilon_{Nd}(ri)$	*Goldstein and Jacobsen* [1987]	ϵ_{Nd}-units
Nd boundary source, total	f_{bs}	$4.5 \cdot 10^9$	g Nd yr^{-1}
ϵ_{Nd} of boundary source	$\epsilon_{Nd}(bs)$	*Jeandel et al.* [2007]	ϵ_{Nd}-units
Nd source, total	f	$5.1 \cdot 10^9$	g Nd yr^{-1}

systematically examined the sensitivity of ϵ_{Nd} in seawater to changes in ocean overturning circulation and found that variations in ϵ_{Nd} reflect weakening and strengthening of the formation of North Atlantic Deep Water (NADW) and Antarctic Bottom Water (AABW). Apart from variations at depth, glacial-interglacial variations in ϵ_{Nd} in surface water were reconstructed in the North Indian Ocean [*Burton and Vance*, 2000; *Stoll et al.*, 2007; *Gourlan et al.*, 2010]. These variations were ascribed to changes in either the magnitude or the isotopic composition of river runoff and thus were interpreted as glacial-interglacial changes in monsoon circulation.

[5] For the interpretation of variations in ϵ_{Nd} as changes in overturning circulation it is important to be able to disentangle variations resulting from changes in Nd sources from variations that are due to changes in overturning circulation [e.g., *Wilson et al.*, 2012]. This in turn requires a good overall knowledge of the nature and magnitude of these sources. In this regard, considerable progress has been made in recent years [e.g., *Goldstein and Jacobsen*, 1987; *Jeandel et al.*, 1995; *Tachikawa et al.*, 1999, 2003; *van de Flierdt et al.*, 2004; *Lacan and Jeandel*, 2005; *van de Flierdt et al.*, 2007; *Siddall et al.*, 2008; *Arsouze et al.*, 2009; *Rempfer et al.*, 2011]. Nevertheless, processes contributing to the boundary source are still not well constrained [e.g., *Amakawa et al.*, 2000; *Johannesson and Burdige*, 2007; *Rickli et al.*, 2010], and therefore it is inherently difficult to estimate potential changes in the past. However, changes in the boundary source are likely to occur with changes in climate for example due to fluctuations in groundwater discharge [*Zektser and Loaiciga*, 1993; *Johannesson and Burdige*, 2007], changes in the distribution and extent of oxygen minimum zones affecting the flux of Rare Earth Elements (REEs) into the water column [*Haley et al.*, 2004], or changes in sediment source regions and thus its ϵ_{Nd}, due to changes in continental erosion through glaciers and ice sheets [*Tütken et al.*, 2002].

[6] Past changes in the magnitude [*Harris and Mix*, 1999; *Nürnberg and Tiedemann*, 2004; *Lèzine et al.*, 2005; *Rincón-Martínez et al.*, 2010] and origin [*Burton and Vance*, 2000; *Stoll et al.*, 2007; *Gourlan et al.*, 2010] (indicated by variations in ϵ_{Nd} of 3–4 ϵ_{Nd}-units) of river runoff have been reconstructed at different sites and have also been reported from climate model simulations [e.g., *Masson et al.*, 2000].

[7] Moreover, past changes in the magnitude [*Grousset et al.*, 1998; *Wolff et al.*, 2006] and origin [*Lupker et al.*, 2010] (indicated by variations in ϵ_{Nd} of more than 6 ϵ_{Nd}-units) of dust fluxes have been reported from observations and from models [*Mahowald et al.*, 1999, 2006]. For example, *Mahowald et al.* [1999, 2006] reported that dust fluxes during the LGM were increased by a factor up to 20, depending on the location, with a global mean change of about a factor 3.

[8] Effects of changes in the magnitude or the isotopic composition of Nd sources on ϵ_{Nd} have been examined by *Tachikawa et al.* [2003], using the PANDORA 10-box model. They found that ϵ_{Nd} in deep boxes is affected by changes in Nd sources and therefore concluded that such changes might complicate the interpretation of variations in ϵ_{Nd} as changes in overturning circulation. Nevertheless, although a sensitivity of ϵ_{Nd} at the seafloor to changes in the sources would have important implications for its use as a paleocirculation tracer, this has not been examined any further so far.

[9] Here, we address this gap by modifying either the magnitude of Nd source fluxes, or their Nd isotopic composition, or both in the Bern3D model. Using the 3-dimensional Bern3D ocean model allows us to examine the spatial structure of the effects of such modifications in much more detail than it was possible for *Tachikawa et al.* [2003] using a 10-box model.

2. Methods

[10] For our simulations we use the Bern3D ocean model of intermediate complexity [*Müller et al.*, 2006], coupled to an energy-moisture balance model [*Ritz et al.*, 2011]. The resolution of the ocean model is 36 × 36 grid cells in the horizontal, equidistant in longitude and in the sine of latitude. Spacing of the 32 depth layers is logarithmic, increasing with depth from 39 m in the uppermost to 397 m in the bottom layer. The ocean model also contains a biogeochemical module which allows the calculation of the export production of biogenic particles such as calcite ($CaCO_3$), opal, and particulate organic carbon (POC) from prognostic equations (for a detailed explanation, see *Tschumi et al.* [2008]).

[11] Isotopes of Nd (^{143}Nd, ^{144}Nd) have been included into the Bern3D model (see *Rempfer et al.* [2011] for a detailed description of the approach). Three sources of Nd isotopes are explicitly represented in the model; continental margins (the boundary source), river discharge and aeolian dust (see Table 1 for a list of parameters and abbreviations used in the text). The boundary source is represented by a globally uniform flux of Nd across the sediment-water interface at continental margins at depths between 0 and 3000 m and is one

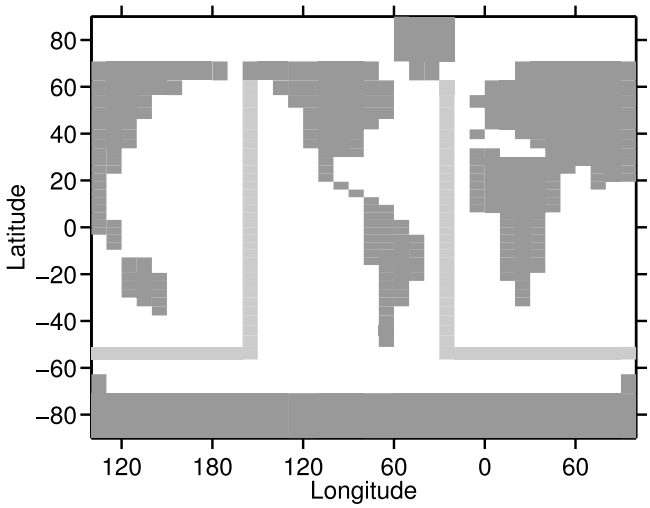

Figure 1. Global map and track of vertical sections as shown in Figures 2, 3, and 4.

of the major tuning parameters of the model [*Rempfer et al.*, 2011, 2012]. The isotopic composition of the boundary source is based on a global map of ϵ_{Nd} [*Jeandel et al.*, 2007]. A global map of atmospheric dust flux [*Luo et al.*, 2003] is prescribed at the sea surface and ϵ_{Nd} of dust is assumed to vary between different regions [see *Rempfer et al.*, 2011, for further details]. Nd concentration in dust ($c_{du,Nd}$, 20 μg g^{-1}) as well as the dissolution of Nd from dust ($\beta_{du,Nd}$, 2%) are assumed to be globally uniform. Values for discharge as well as Nd concentration ($c_{ri,Nd}(\theta, \phi)$) and ϵ_{Nd} ($\epsilon_{Nd}(ri)$) of major rivers are taken from *Goldstein and Jacobsen* [1987] and are prescribed at the sea surface at the corresponding geographic location. In order to account for removal of Nd in estuaries, only a fraction of 30% of the Nd dissolved in rivers is supplied to the ocean, i.e., 70% of the Nd are removed.

[12] In the ocean Nd isotopes are subject to internal cycling which is parameterized by a reversible scavenging approach. Reversible scavenging describes the physical process of adsorption onto and desorption from surfaces of aeolian dust and biogenic particles such as particulate organic carbon (POC), opal, and calcite ($CaCO_3$). Processes of adsorption and desorption are assumed to be fast and therefore particle-associated and dissolved concentration are in equilibrium. The partitioning between particle-associated and dissolved Nd is based on an "equilibrium scavenging coefficient" which is another major tuning parameter [*Rempfer et al.*, 2011, 2012]. Particles are subject to gravitational force and settle down the water column. Due to remineralization and redissolution of biogenic particles at depth and the release of associated Nd, the concentration of dissolved Nd ($[Nd]_d$, in pmol/kg) increases with depth and with the age of water masses, thus exhibiting a nutrient-like pattern. Overall, biogenic particles act as sink for dissolved Nd at shallow depths and as a source at greater depths. An overview of the performance of the model is given in *Rempfer et al.* [2012].

3. Overview of Experiments

[13] In this study we examine the sensitivity of ϵ_{Nd} in seawater to changes in the magnitude, the isotopic composition, or both of Nd sources in a number of experiments. All experiments start from steady state, i.e., from the CTRL experiment.

[14] First, and similar to *Tachikawa et al.* [2003], we globally increase the magnitude of all sources (dust, rivers, and boundary source) by a factor of 4. We then increase ϵ_{Nd} of all sources by 4 ϵ_{Nd}-units, and finally scale the magnitude and increase ϵ_{Nd} of Nd sources at the same time. Additionally, we consider only the major source, the boundary source, and apply each modification globally as well as regionally, i.e., in the North Atlantic (between 30° and 71°N), the North Pacific (between 46° and 71°N), and the Southern Ocean (between 56° and 71°S). The same modifications are then applied to the dust and river Nd sources on a global scale. Note that for a modification of the magnitude of the dust source we increase the release of Nd from dust instead of increasing the dust particle flux, thus not affecting the sink of Nd. Following the application of the corresponding changes the model was run into steady state again during another 10,000 model years.

[15] In additional experiments we apply the modifications to all sources for 100 and 1000 years. Following 100 and 1000 years, respectively, the magnitude of source fluxes as well as their isotopic composition are reset to original CTRL values and the model is run into steady state again during several thousand model years. These experiments indicate the transient response of ϵ_{Nd} in seawater for the most pronounced case considered here. Although this is not a likely scenario, it illustrates the dimension of modifications which is needed to affect ϵ_{Nd} to a similar extent as reconstructed, e.g., on glacial-interglacial timescales.

[16] $[Nd]_d$ and ϵ_{Nd} as obtained with the CTRL are shown along a transect from the North Atlantic to the North Pacific (the track of the transect is indicated in Figure 1) in Figure 2. Both $[Nd]_d$ and ϵ_{Nd} are simulated in reasonable agreement with observations. Characteristic numbers of the marine Nd cycle for the CTRL set up, such as the magnitude of the boundary source (f_{bs}), the ratio of particle-associated to dissolved Nd concentration ($[Nd]_p/[Nd]_d$), the mean residence time (τ_{Nd}) and the Nd global inventory (I_{Nd}), are within a reasonable range, as far as data-based constraints are available [*Rempfer et al.*, 2012].

4. Results and Discussion

4.1. Manipulating Sources of Nd

4.1.1. Changes in All Nd Sources

[17] In three experiments we modify the magnitude (f), the Nd isotopic composition (ϵ_{Nd}), or both, of Nd sources rivers (ri), dust (du), and continental margins (bs) at the same time. As shown in Figure 3a, modifying the magnitude of all sources simultaneously leaves the relationship between the individual sources unchanged and therefore does not affect ϵ_{Nd}. In contrast, a shift in ϵ_{Nd} of all sources causes a proportional shift in ϵ_{Nd} in the ocean (Figure 3b). Similarly, applying both modifications at the same time causes an overall shift in ϵ_{Nd} by 4 ϵ_{Nd}-units (Figure 3c).

[18] Besides, regarding the sign of the shift in ϵ_{Nd} we note that the magnitude of effects on ϵ_{Nd} in seawater is independent of the sign of modifications. We therefore do not show results from experiments where shifts to more negative ϵ_{Nd} are applied.

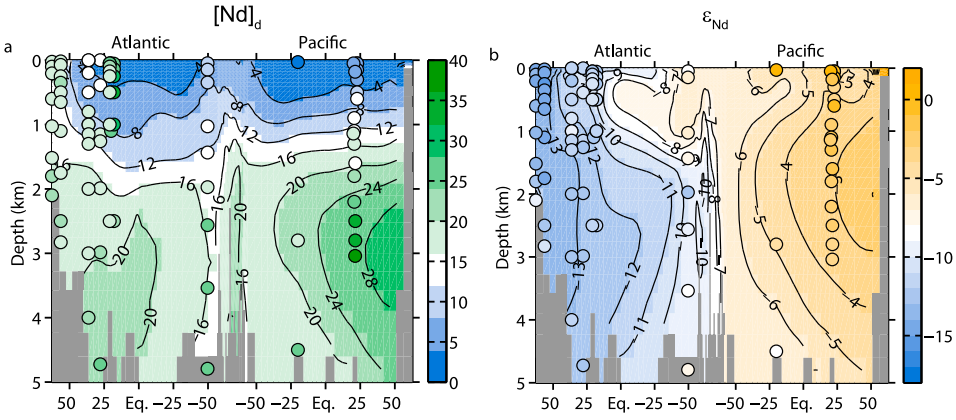

Figure 2. Simulated Nd dissolved concentration ($[Nd]_d$ in pmol/kg, left) and ϵ_{Nd} (right), obtained with CTRL. Results apply to steady-state and are provided in vertical sections along a track from the Atlantic to the Pacific (the course of the track is indicated in Figure 1). Observations are superimposed as colored circles, using the same color scale [see *Rempfer et al.*, 2012, for references].

[19] Results shown in Figures 3a and 3b are in agreement with findings of *Tachikawa et al.* [2003] who raised the question whether variations in ϵ_{Nd} as reconstructed from sediments not only reflect changes in oceanic overturning circulation, but also changes in Nd sources. On the one hand, no change in seawater ϵ_{Nd} following an increase in the magnitude of all sources ($4 \times f$), and a proportional shift in seawater ϵ_{Nd} following a shift in ϵ_{Nd} of all sources ($\epsilon_{Nd} + 4$), is not surprising but rather is to be expected. On the other hand, it seems not very likely that all sources were affected by changes of this magnitude and spatial extent at the same time in the recent past, e.g., on glacial-interglacial or even shorter timescales. In the following we therefore examine the effect of smaller scale changes in individual Nd sources on seawater ϵ_{Nd}.

4.1.2. Changes in the Nd Boundary Source

[20] Before continuing the discussion, we would like to emphasize that changing the source flux by a factor of four or the isotopic signature by 4 ϵ_{Nd}-units globally or within an entire ocean basin as done here corresponds to a rather extreme change for the most important Nd source, the flux from the continental boundary (f_{bs}).

[21] Scaling f_{bs} globally affects ϵ_{Nd} at depths shallower than about 1 km, particularly in the tropical and subtropical Atlantic (Figure 4a) where the relative influence of dust and rivers decreases. An increase in ϵ_{Nd} of the boundary source ($\epsilon_{Nd}(bs)$) by 4 ϵ_{Nd}-units results in an increase in ϵ_{Nd} in seawater by up to 3.5 ϵ_{Nd}-units. Effects on seawater ϵ_{Nd} in general are larger at greater depths, and are particularly small in shallow depths of the tropical and subtropical Atlantic, where contributions from dust and rivers are large (Figure 4b). A combination of both experiments results if both quantities are changed simultaneously (Figure 4c).

[22] If scaling of f_{bs} is applied on more regional scales (i.e., in the North Atlantic, the North Pacific, the Southern Ocean), effects on ϵ_{Nd} are generally more pronounced and range from −2.5 to +2.5 ϵ_{Nd}-units (Figures 4d, 4g, and 4j). Scaling f_{bs} in the North Atlantic leads to more positive ϵ_{Nd} by 1.5 ϵ_{Nd}-units in the North Atlantic and slightly more negative ϵ_{Nd} in the Pacific Ocean (Figure 4d). $\Delta\epsilon_{Nd}$ is positive throughout the transect shown in Figure 4g if scaling is applied to f_{bs} in the North Pacific Ocean only. Finally, $\Delta\epsilon_{Nd}$ is negative, particularly in the deep Pacific and in deep and

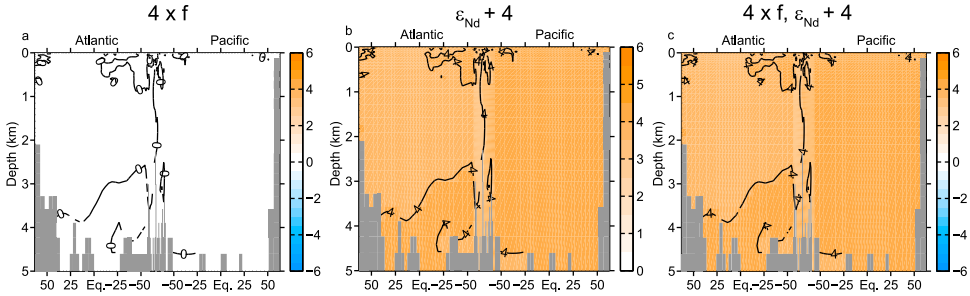

Figure 3. Differences in the ^{143}Nd to ^{144}Nd isotopic ratio (ϵ_{Nd}) between CTRL and experiments (left) where the magnitude of all sources are scaled by a factor of 4, (center) where ϵ_{Nd} of all sources was increased by 4 ϵ_{Nd}-units, and (right) where the magnitude and ϵ_{Nd} of all sources were scaled and increased. All changes are applied on a global scale. Results apply to steady-state and are provided along a track from the North Atlantic to the North Pacific traversing the Southern Ocean from west to east (the course of the track is indicated in Figure 1).

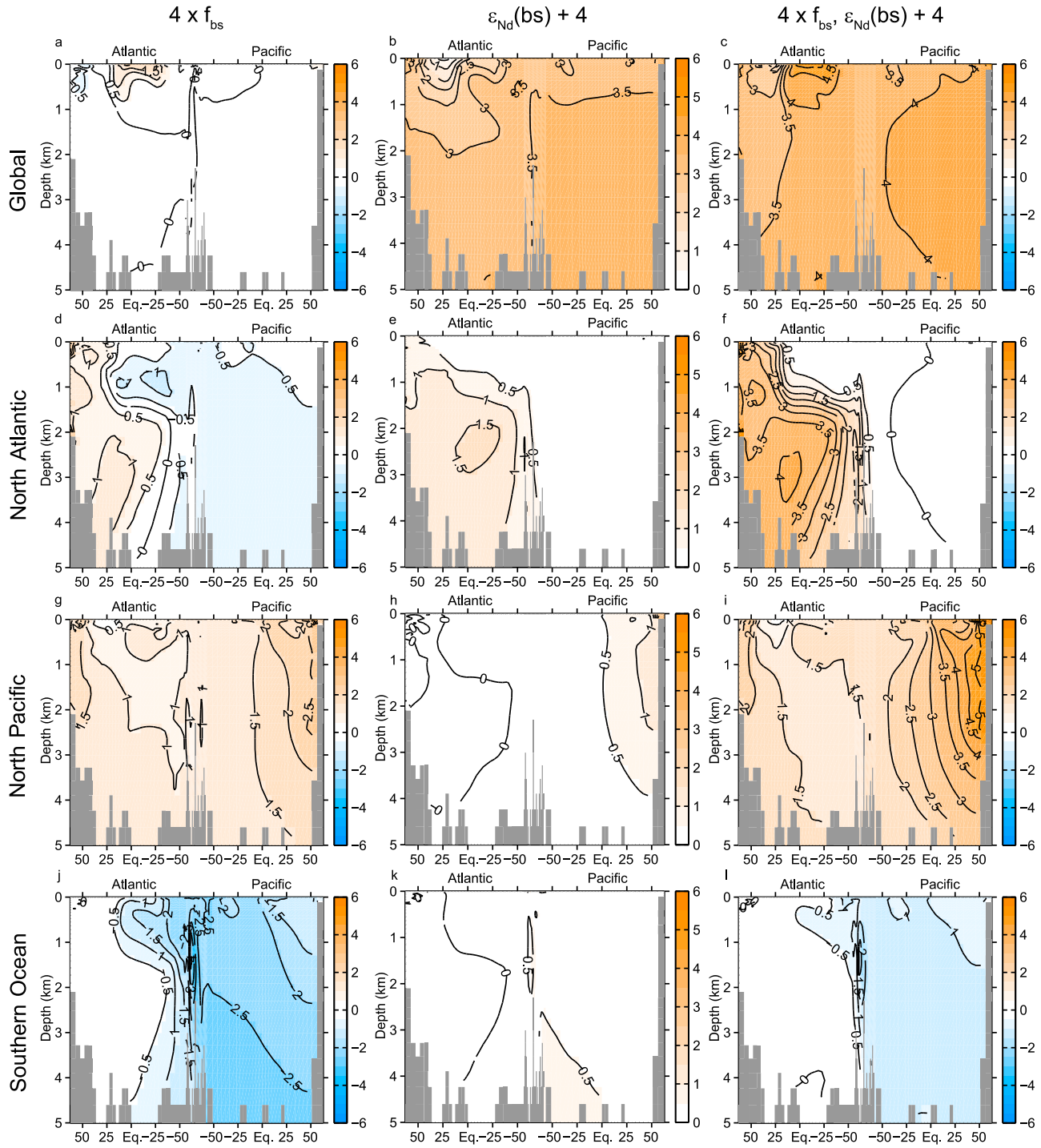

Figure 4. Differences in the ^{143}Nd to ^{144}Nd isotopic ratio (ϵ_{Nd}) between CTRL and experiments (left) where the magnitude of the Nd boundary source (f_{bs}) is scaled by a factor of 4, (center) where ϵ_{Nd} of the boundary source ($\epsilon_{Nd}(bs)$) is increased by 4 ϵ_{Nd}-units, and (right) where the magnitude and ϵ_{Nd} of the boundary source are scaled and increased. The corresponding changes were applied (a–c) globally, (d–f) in the North Atlantic, (g–i) in the North Pacific, and (j–l) in the Southern Ocean. Results apply to steady-state and are provided along a track from the North Atlantic to the North Pacific traversing the Southern Ocean from west to east (the course of the track is indicated in Figure 1).

in intermediate waters of the South Atlantic in case of a scaling of f_{bs} in the Southern Ocean (Figure 4j). In experiments where f_{bs} is increased, the sign of $\Delta\epsilon_{Nd}$ depends on ϵ_{Nd} in the region where the scaling is applied as well as on ϵ_{Nd} in the corresponding grid cell. For example, ϵ_{Nd} in the North Pacific compared to ϵ_{Nd} in seawater is relatively positive [*Jeandel et al.*, 2007]. A scaling of f_{bs} therefore causes an overall shift to more positive ϵ_{Nd} in seawater. In contrast, if f_{bs}

in the North Atlantic or the Southern Ocean is applied, the pattern is more complex. ϵ_{Nd} in Antarctica regarding to ϵ_{Nd} of main water masses NADW, AABW and Antarctic Intermediate Water (AAIW) is intermediate [*Jeandel et al.*, 2007]. Therefore, ϵ_{Nd} in AABW and AAIW becomes more negative, while ϵ_{Nd} in NADW becomes more positive. In case where f_{bs} is shifted in the North Atlantic, amongst others the imprint of positive ϵ_{Nd} from Iceland increases, thus shifting NADW to more positive but AAIW and AABW to more negative ϵ_{Nd}, as ϵ_{Nd} in NADW is still more negative than ϵ_{Nd} in AABW and AAIW.

[23] Increasing $\epsilon_{Nd}(bs)$ in the North Atlantic, the North Pacific, or the Southern Ocean, leads to more positive seawater ϵ_{Nd} by up to 1.5 ϵ_{Nd}-units in the corresponding region (Figures 4e, 4h, and 4k). The spatial extent of effects on ϵ_{Nd} is most pronounced if changes are applied in the North Atlantic region due to the export of NADW to the south. In this case, ϵ_{Nd} is more positive by up to 1.5 ϵ_{Nd}-units throughout the Atlantic part of the transect (Figure 4e). In contrast, if $\epsilon_{Nd}(bs)$ is increased in the North Pacific or the Southern Ocean, effects are distributed less and rather confined to the region where they are applied to (Figures 4h and 4k). Note, that effects of an increase in $\epsilon_{Nd}(bs)$ are generally less pronounced than if f_{bs} is increased. As already mentioned above, the effect on ϵ_{Nd} is a combination of results from experiments where magnitude or ϵ_{Nd} are increased, if both modifications are applied simultaneously (Figures 4c, 4f, 4i, and 4l).

[24] Regarding past changes in the boundary source, an evaluation of their effect on ϵ_{Nd} is complicated by the fact that little is known about processes involved in this source in general. Anyway, our results indicate that changes of considerable magnitude and spatial extent are required to affect ϵ_{Nd} in seawater in a manner comparable e.g., to the magnitude of variations of about 3 ϵ_{Nd}-unit that have been reconstructed from sediment records from the Cape Basin in the southeast Atlantic Ocean [*Rutberg et al.*, 2000; *Piotrowski et al.*, 2008] and the Indian Ocean [*Piotrowski et al.*, 2009]. Nevertheless, due to the fact that the boundary source is applied at the sediment surface between the sea surface and 3000 m depth, effects on ϵ_{Nd} emerge throughout the water column.

4.1.3. Changes in the Nd Dust Source

[25] Scaling of f_{du} affects ϵ_{Nd} at the surface in certain regions of the Atlantic, the Indian and the Pacific (Figure 5a). However, ϵ_{Nd} at the seafloor is not affected at any place in open ocean regions (Figure 5d). Overall, $\Delta\epsilon_{Nd}$ does not exceed 0.5 ϵ_{Nd}-units and is confined to depths shallower than 1 km (Figure 5g,j).

[26] A shift in ϵ_{Nd} in dust causes ϵ_{Nd} at the surface to increase, particularly in regions of the tropical and subtropical Atlantic and Indian oceans. Some minor effects are also observed in the North and South Pacific (up to 0.5 ϵ_{Nd}-units, Figure 5b). Again, ϵ_{Nd} is not affected at the seafloor in open ocean regions and effects on ϵ_{Nd} in Atlantic and Indian transects are confined largely to depths shallower than 1 km (Figures 5e, 5h, and 5k).

[27] If both modifications are applied simultaneously, effects on seawater ϵ_{Nd} are larger than in the former two cases. Again, $\Delta\epsilon_{Nd}$ is relatively large in surface waters of the tropical and subtropical Atlantic and Indian Oceans (Figure 5c), but $\Delta\epsilon_{Nd}$ up to 1.5 ϵ_{Nd}-units can also be observed at the seafloor in regions close to the continents (Figure 5f). Indian and Atlantic transects show that ϵ_{Nd} is affected throughout the water column to some extent, particularly in the Atlantic Ocean where $\Delta\epsilon_{Nd}$ below 1 km depth reaches values up to 1 ϵ_{Nd}-unit and where the signal is advected southward via NADW (Figures 5i and 5l).

[28] Even more than in experiments where the boundary source is changed at a regional scale, effects on ϵ_{Nd} are of limited spatial extent if manipulations are applied to the Nd dust source. Large effects are observed in regions where major dust flux occurs, i.e., below dust plumes in the Atlantic and the Indian Oceans. Some minor effect are also observed in the North Pacific, around the continent of Australia, and in the South Atlantic off South America. Furthermore, as Nd is released from dust in the uppermost layer of the ocean, effects are largely confined to shallow depths. A transport to greater depths can be observed almost exclusively in the Atlantic, and particularly if both the magnitude and the isotopic composition are affected. This indicates the importance of convection and thus formation of deep water masses for a more pronounced distribution of the effects.

4.1.4. Changes in the Nd River Source

[29] Scaling of f_{ri} affects ϵ_{Nd} at the surface in various regions of the Atlantic, the Indian and the Pacific Oceans (Figure 6a). In contrast, hardly any effect is observed at the seafloor in open ocean regions (Figure 6d) and Indian and Atlantic transects indicate that effects are largely confined to depths shallower than 1 km (Figures 6g and 6j).

[30] Similarly, a shift in ϵ_{Nd} in the river source leads to more positive ϵ_{Nd} in certain regions of the Atlantic, the Indian and the Pacific surface oceans (Figure 6b). No effect on ϵ_{Nd} at the seafloor is observed, except some small changes in the Atlantic (Figure 6e). Indian and Atlantic transects indicate that effects on ϵ_{Nd} are largely confined to depths shallower than about 1 km, again with the exception of the Atlantic where $\Delta\epsilon_{Nd}$ is more positive by about 0.5 ϵ_{Nd}-units even below 1 km from the North Atlantic until 40°S (Figures 6h and 6k).

[31] As in previous experiments, effects are larger in magnitude and spatial extent, both at the surface and at the seafloor, if both the magnitude and the isotopic composition are modified at the same time (Figures 6c and 6f). Besides, the signal of modifications in the Nd river source enters deep water layers in the North Atlantic, where NADW is formed and exported southward (Figures 6i and 6l).

[32] Note, that the Nd river source in our model is based on data of *Goldstein and Jacobsen* [1987] which cover primarily large rivers. Consequently, effects of changes in the Nd river source are probably underestimated in our experiments. In any case, transects indicate that a transport of effects to greater depths primarily occurs in regions where deep water is formed through deep convection.

4.1.5. Temporal Evolution of ϵ_{Nd} Following Modifications in All Nd Sources

[33] The temporal evolution of ϵ_{Nd} resulting from experiments where the magnitude or/and the isotopic composition of all sources is modified, as described in section 4.1.1, as well as from experiments where the same changes are globally applied for 100 and 1000 yr, respectively, are shown in Figure 7. As mentioned above, effects are relatively small in experiments where the magnitude of Nd sources is increased (Figure 7a), more pronounced in experiments where ϵ_{Nd} is increased (Figure 7b), and largest in experiments where both modifications are applied at the same time (Figure 7c). Besides, it is indicated that the magnitude of $\Delta\epsilon_{Nd}$ strongly depends on the duration of the time period during which the

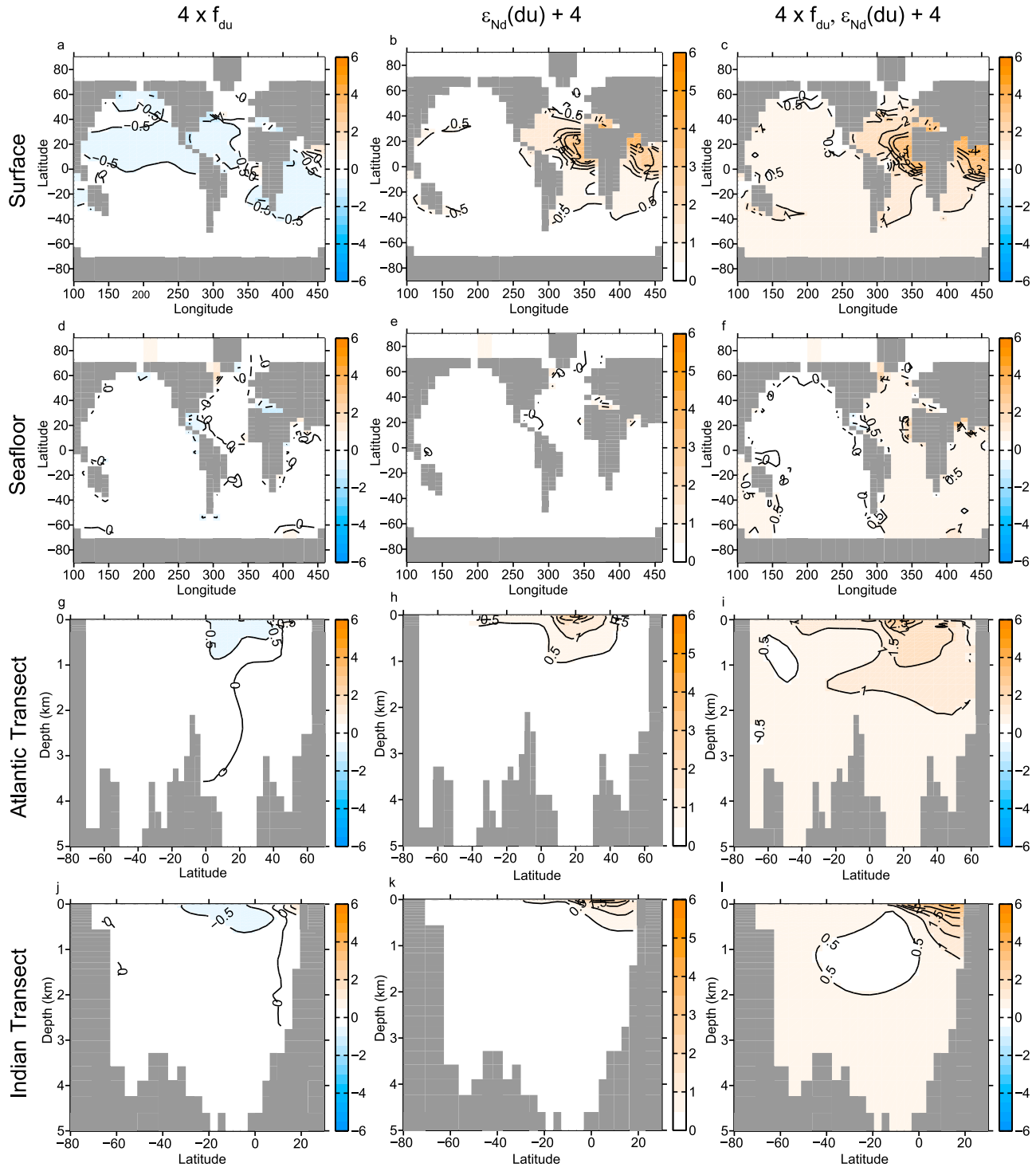

Figure 5. Differences in the ^{143}Nd to ^{144}Nd isotopic ratio (ϵ_{Nd}) between CTRL and experiments (left) where the magnitude of the dust Nd source (f_{du}) is scaled by a factor of 4, (center) where ϵ_{Nd} in the dust source ($\epsilon_{Nd}(du)$) is increased by 4 ϵ_{Nd}-units, and (right) where the magnitude and ϵ_{Nd} of the dust Nd source are scaled and increased. All changes are applied on a global scale. Results apply to steady-state and are from (a–c) the sea surface, (d–f) the seafloor, and along transects through (g–i) the Atlantic (30°–40°W) and (j–l) the Indian Ocean (80°–90°E).

modification is applied. If a specific change is applied for a certain period only, ϵ_{Nd} in seawater approaches the original steady state within a few thousand years, depending on the type of modification, the magnitude of $\Delta\epsilon_{Nd}$ and the basin of interest. For example, in experiments where ϵ_{Nd} is increased during 1000 yr and reset to CTRL values thereafter, time until a new steady state is approached is longest in the Pacific Ocean (>2000 yr) and much shorter in the Atlantic (a few

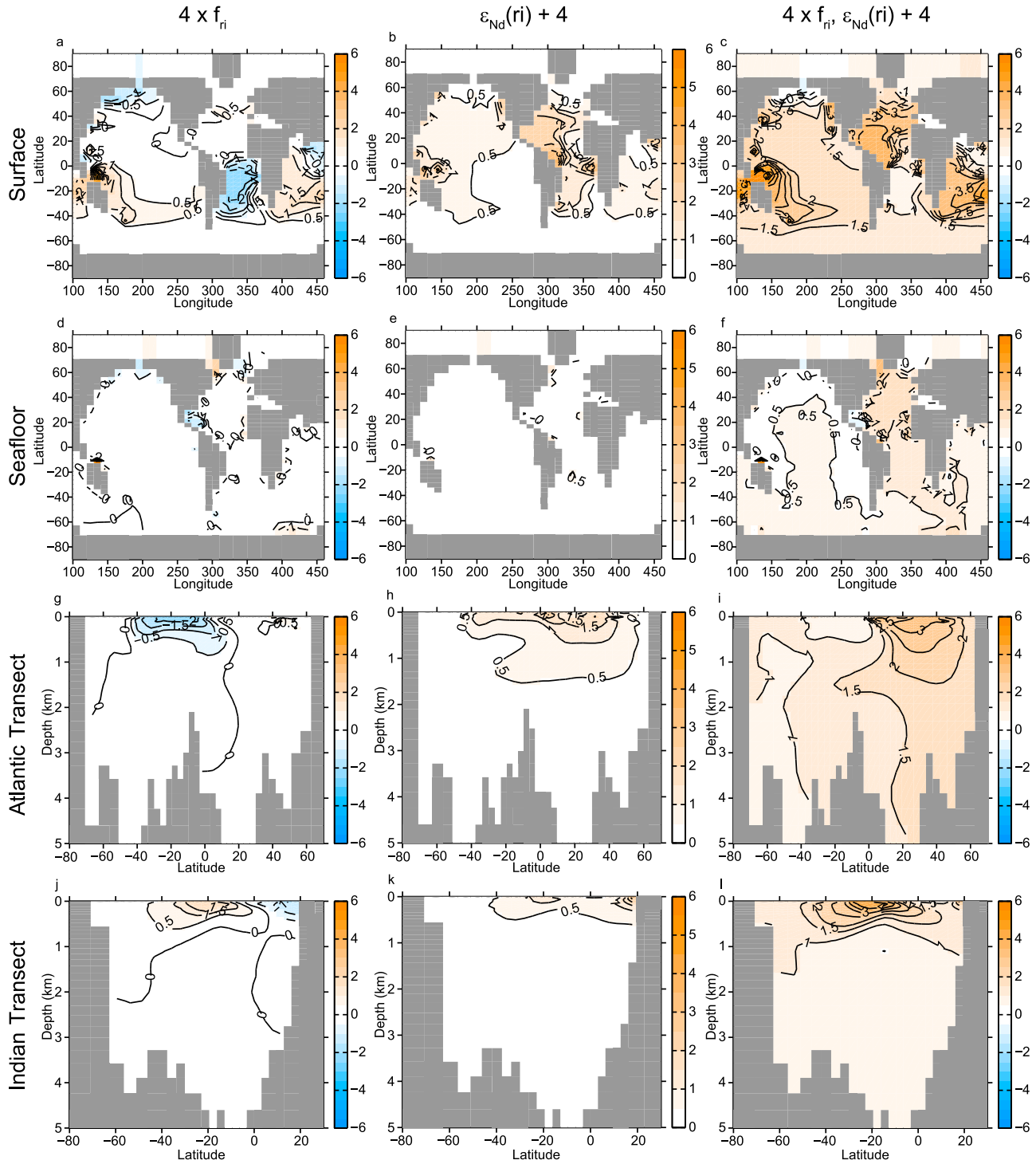

Figure 6. Differences in the ^{143}Nd to ^{144}Nd isotopic ratio (ϵ_{Nd}) between CTRL and experiments (left) where the magnitude of the river Nd source (f_{ri}) is scaled by a factor of 4, (center) where ϵ_{Nd} in the river source ($\epsilon_{Nd}(ri)$) is increased by 4 ϵ_{Nd}-units, and (right) where the magnitude and ϵ_{Nd} of the river source is scaled and increased. All changes are applied on a global scale. Results apply to steady-state and are from (a–c) the sea surface, (d–f) the seafloor, and along transects through (g–i) the Atlantic (30°–40°W) and (j–l) the Indian Ocean (80°–90°E).

hundred years). On the other hand, in experiments where the magnitude of source fluxes and ϵ_{Nd} are modified simultaneously during 1000 yr, time until a new steady state is approached is similar in all basins (about 4000 yr).

4.2. Overall Discussion

[34] On the one hand, our experiments indicate that changes in the magnitude, the isotopic composition, or both of Nd

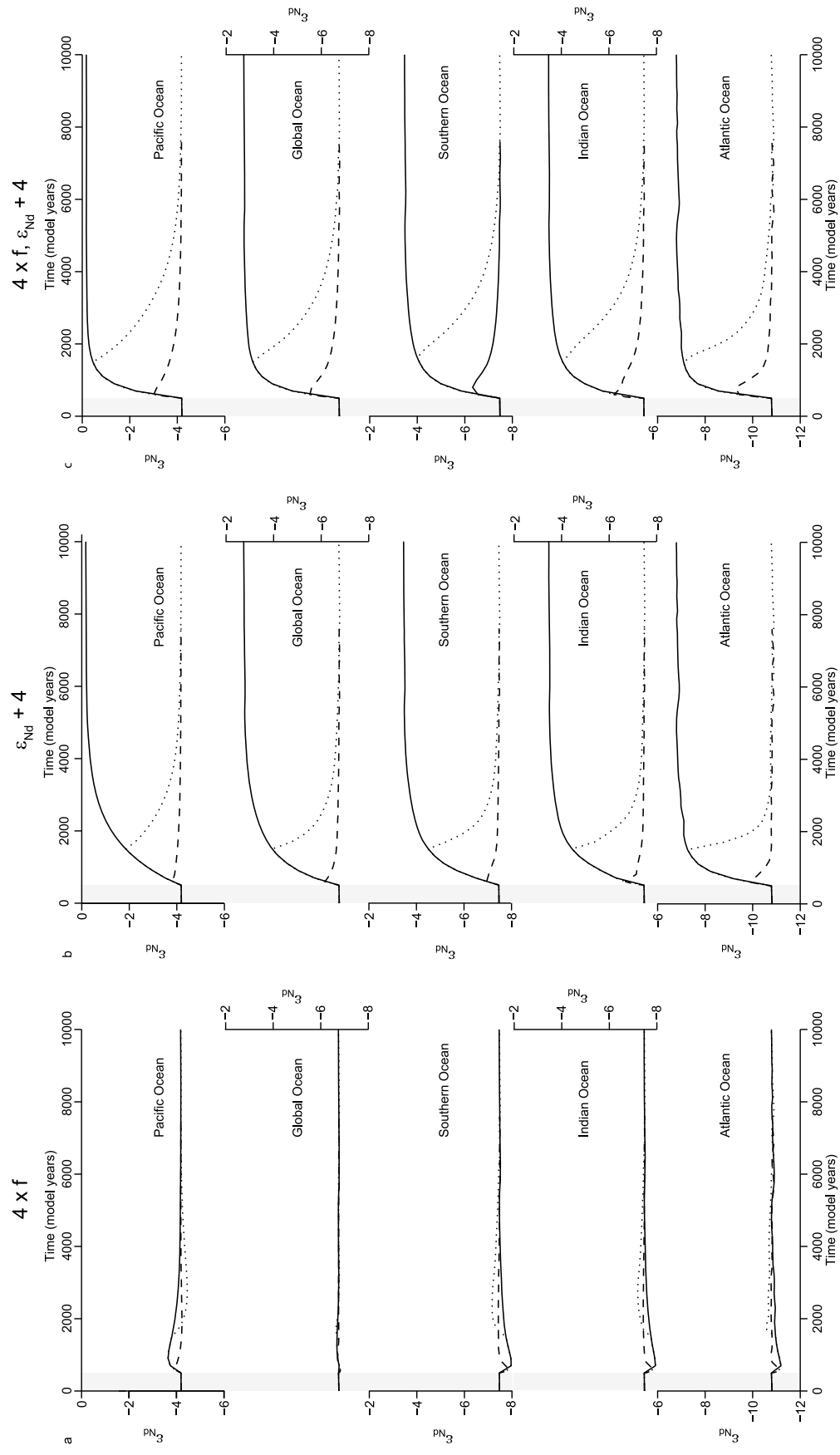

Figure 7. Temporal evolution of mean ϵ_{Nd} in the global ocean, the Atlantic, the Pacific, the Indian, and the Southern Ocean (see details on panels). Results are from experiments where the (a) magnitude, (b) isotopic composition, or (c) both of all Nd sources is changed for 100 yr (dashed lines), 1000 yr (dotted lines), as well as during the entire experiment (solid lines). Note, that the latter corresponds to experiments discussed above. Each simulation starts from the steady-state CTRL, indicated by a gray band.

sources affect ϵ_{Nd} in seawater to some extent, and thus partly confirm concerns raised by *Tachikawa et al.* [2003]. In particular, the effect of changes in the boundary source, which is located below the sea surface, can be observed throughout the water column. In contrast, Nd from dust and rivers enters the ocean at the surface, and major changes are therefore mainly confined to shallow depths. Nevertheless, if changes are located upstream or close to regions of deep water formation the signal may also be transported to greater depth. On the other hand, our results also indicate that modifications of considerable spatial extent and magnitude are required in order to affect seawater ϵ_{Nd} on large spatial scale and magnitude. Besides, our results emphasize the need for a 3-dimensional model for an adequate evaluation of such effects.

[35] The stability of ϵ_{Nd} in the North Atlantic end-member is an important prerequisite for the interpretation of downstream ϵ_{Nd} records in terms of circulation changes [e.g., *Piotrowski et al.*, 2004]. The temporal evolution of ϵ_{Nd} in the North Atlantic has been examined by *van de Flierdt et al.* [2006] and *Foster et al.* [2007]. Both studies revealed remarkable temporal stability of ϵ_{Nd} from the Holocene to the last glacial [*van de Flierdt et al.*, 2006] and during the last 500 kyr, respectively [*Foster et al.*, 2007] and thus indicate that ϵ_{Nd} of the North Atlantic end-member has not undergone substantial variability during this time.

[36] In light of the results presented in this study, this could indicate that no change occurred in the most important boundary source in the North Atlantic Ocean during time intervals covered by *van de Flierdt et al.* [2006] and *Foster et al.* [2007]. However, it should be noted that the study of *van de Flierdt et al.* [2006] does not cover the entire glacial period but rather certain time slices, not including e.g., the LGM, and that the temporal resolution of reconstructions reported by *Foster et al.* [2007] is on the order of 30–40 kyr. It is therefore not possible to exclude variations on shorter timescales, or during periods that are not resolved by the reconstructions based on these data. Overall, the relative stability found by *van de Flierdt et al.* [2006] and *Foster et al.* [2007] does not rule out changes in the Nd dust (such as reported, e.g., by *Mahowald et al.* [1999, 2006]) and river sources as according to our results effects on ϵ_{Nd} are largely confined to the surface ocean in these cases. *Gutjahr et al.* [2008] indicated that ϵ_{Nd} in GNAIW may have been more positive than modern NADW by 3.5–4 ϵ_{Nd}-units during the LGM. On the one hand, such changes in ϵ_{Nd} of GNAIW potentially can be explained by changes in the composition of GNAIW compared to NADW, and *Gutjahr et al.* [2008] argued that the more positive character of GNAIW could for example be due to a missing contribution of Labrador Seawater to GNAIW. On the other hand, part of these changes could also well be due to changes in either the magnitude, or the isotopic composition, or both of the boundary source. Note, that we do not take into account changes in the composition of water masses in the experiments presented in this study and therefore are not in the position to make a final conclusion on this but leave it as an open question. Instead, we simply point to the fact that in our experiments a combined increase and shift in the North Atlantic boundary source is required in order to generate $\Delta\epsilon_{Nd}$ of 3.5–4 ϵ_{Nd}-units in the North Atlantic (Figure 4f).

[37] Substantial variations in ϵ_{Nd} in surface waters have been reported from the North Indian Ocean on glacial-interglacial timescales (up to about 4 ϵ_{Nd}-units) and are assumed to be due to changes in monsoon circulation [*Burton and Vance*, 2000; *Stoll et al.*, 2007; *Gourlan et al.*, 2010]. In our experiments, modifications of Nd dust and river sources generate changes of similar magnitude in shallow waters of the North Indian and are thus generally in line with the interpretation of these studies. As mentioned above, an important characteristic of such changes is that they are largely confined to shallow depths, except if they are located upstream or close to regions of deep water formation, thus hardly affecting the interpretation of reconstructions from greater depth in terms of water mass distribution and mixing.

[38] Major limitations of our study are caused by uncertainties associated with the nature of sources, i.e., to the generally limited understanding of the marine Nd cycle [e.g., *Rempfer et al.*, 2011]. A better understanding of processes involved, for example, in the boundary source would facilitate the evaluation of the probability of past changes in this source. New insights into the marine Nd cycle will be provided in the near future by the GEOTRACES program [*SCOR Working Group*, 2007]. Another effect, that has the potential to further complicate the interpretation of ϵ_{Nd} as reconstructed from sediments, and which is not considered in this study, is post depositional reallocation of sediment material [*McCave*, 2002; *Gutjahr et al.*, 2008] and leaching of volcanic ash from bulk sediments [*Roberts et al.*, 2010; *Elmore et al.*, 2011].

5. Summary and Conclusions

[39] In this study we reconsider one of the main conclusions of *Tachikawa et al.* [2003] which indicates that variations of ϵ_{Nd} as reconstructed from sediment cores may reflect not only changes in overturning circulation but also in Nd sources. Understanding the impact of such changes is of importance for the interpretation of variations in ϵ_{Nd} as past changes in Meridional Overturning Circulation.

[40] Major findings of our study are as follows: First, not surprisingly and similar to *Tachikawa et al.* [2003], we find that an increase in the magnitude of all Nd sources simultaneously does not affect ϵ_{Nd} in seawater and that a shift in ϵ_{Nd} of all sources at the same time causes a proportional shift in ϵ_{Nd}. Second, in experiments where more subtle changes are applied to individual sources, largest effects on ϵ_{Nd} in deep water masses result from changes in the boundary source, i.e., the major Nd source. Considerable changes also result from changes in dust and riverine sources but are largely confined to shallow depths. In these cases, ϵ_{Nd} of deep water masses is only affected if changes occur in or upstream of regions where deep water masses are formed by deep convection. Third, the temporal evolution of ϵ_{Nd} in experiments where sources were changed during 100 and 1000 years, indicates that the extent to which ϵ_{Nd} is affected depends on the type of modification, its duration, and differs between sites.

[41] Regarding concerns raised by *Tachikawa et al.* [2003] we find that changes in Nd sources indeed have the potential to affect ϵ_{Nd} in seawater. However, considerable changes in the magnitude, the isotopic composition, or both, are required to generate large-scale changes in ϵ_{Nd} in deep water that are similar in magnitude to those that have been reconstructed from sediment cores on glacial-interglacial timescales [e.g., *Rutberg et al.*, 2000; *Piotrowski et al.*, 2008, 2009] or to those that result from changes in overturning circulation in model experiments [*Rempfer et al.*, 2012]. Based on results

presented in this study we therefore conclude that a shift in ϵ_{Nd} comparable to glacial-interglacial variations is difficult to obtain by changes in Nd sources alone. However, more subtle variations indeed can be caused by such changes and must be interpreted with caution. Therefore, our results emphasize the need to constrain past changes in Nd sources to allow a reliable application as paleocirculation tracer.

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