# Upper ocean vertical supply: A neglected primary factor controlling the distribution of neodymium concentrations of open ocean surface waters?

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[1] Neodymium (Nd) isotopes are an important geochemical tool to trace the present and past water mass mixing as well as continental inputs. The distribution of Nd concentrations in open ocean surface waters (0–100 m) is generally assumed to be controlled by lateral mixing of Nd from coastal surface currents and by removal through reversible particle scavenging. However, using $^{228}$Ra activity as an indicator of coastal water mass influence, surface water Nd concentration data available on key oceanic transects as a whole do not support the above scenario. From a global compilation of available data, we find that more stratified regions are generally associated with low surface Nd concentrations. This implies that upper ocean vertical supply may be an as yet neglected primary factor in determining the basin-scale variations of surface water Nd concentrations. Similar to the mechanism of nutrients supply, it is likely that stratification inhibits vertical supply of Nd from the subsurface thermocline waters and thus the magnitude of Nd flux to the surface layer. Consistently, the estimated required input flux of Nd to the surface layer to maintain the observed concentrations could be nearly two orders of magnitudes larger than riverine/dust flux, and also larger than the model-based estimation on shelf-derived coastal flux. In addition, preliminary results from modeling experiments reveal that the input from shallow boundary sources, riverine input, and release from dust are actually not the primary factors controlling Nd concentrations most notably in the Pacific and Southern Ocean surface waters.

## 1. Introduction

[2] The quasi-conservative isotopic composition of the rare earth element (REE) Nd in seawater is being widely used as a powerful tracer for modern and past water-mass mixing, as well as for continental inputs [e.g., *von Blanckenburg*, 1999; *Goldstein and Hemming.*, 2003; *Peucker-Ehrenbrink et al.*, 2010]. Although considerable efforts and progress have been made in the understanding of the modern marine Nd cycle in recent years [e.g., *Lacan and Jean-del*, 2005; *Arsouze et al.*, 2007, 2009; *Jones et al.*, 2008; *Siddall et al.*, 2008; *Andersson et al.*, 2008; *Porcelli et al.*, 2009; *Oka et al.*, 2009; *Amakawa et al.*, 2009; *Rickli et al.*, 2010; *Rempfer et al.*, 2011; *Carter et al.*, 2012; *Grasse et al.*, 2012; *Stichel et al.*, 2012; *Singh et al.*, 2012; *Grenier et al.*, 2013], some important issues, such as the nature and magnitude of Nd sources and the scavenging behavior of Nd by different particle types, remain unresolved. In addition, although both Nd-concentrations and Nd isotopic composition have recently been simulated in reasonable agreement with observations using three-dimensional ocean models, Nd concentrations at shallow depths of the open ocean are underestimated by these models [*Arsouze et al.*, 2009; *Rempfer et al.*, 2011]. The limited understanding of the Nd cycle at the ocean surface in turn complicates the reliable use of the Nd isotopic composition ($\varepsilon_{Nd}$) as a (paleo)circulation proxy [e.g., *Rempfer et al.*, 2012a, 2012b].

[3] With recent geochemical studies on Nd along important oceanic transects such as in the Southern Ocean (in particular south of 30°S as proposed by *Lacan et al.* [2012]), a more complete picture of the basin-scale variations of surface Nd concentrations has now become available for the first time. Based on these global distributions compiled in our study, we aim to evaluate the role of

different factors that control variations of Nd concentration in open ocean surface waters (0–100 m, with bottom depth >500 m).

[4] Neodymium concentrations in the surface ocean (0–100 m) are highly variable, with reported data ranging from 2.72 to 101.5 pmol/kg [*Lacan and Jeandel*, 2004; *Zhang and Nozaki*, 1996]. High values (>30 pmol/kg) are usually observed in oceanic regions close to continents such as in the Nordic seas [*Lacan and Jeandel*, 2004] and in the Mediterranean Sea [*Tachikawa et al.*, 2004]. Input of Nd from rivers, release of Nd from shelf sediments, as well as dissolution of Nd from dust have been shown to be important sources and input pathways to surface waters, particularly at the ocean margins [e.g., *Tachikawa et al.*, 2004; *Porcelli et al.*, 2009; *Rickli et al.*, 2010; *Singh et al.*, 2012]. These areas, however, only account for a small fraction of the total surface area of the global ocean.

[5] In general and independent of the location, Nd is ultimately and exclusively derived from continental inputs. Its sources include riverine inputs, dissolution of eolian dust, and the flux across the sediment water interface [e.g., *Lacan and Jeandel*, 2005; *Arsouze et al.*, 2007, 2009; *Rempfer et al.*, 2011; *Wilson et al.*, 2012; *Grenier et al.*, 2013]. The vertical distribution of Nd in the water column is characterized by reversible scavenging by particles [cf., *Siddall et al.*, 2008; *Oka et al.*, 2009]. Surface water Nd concentrations are therefore expected to be partly controlled by the intensity of particle scavenging. Open ocean surface water Nd concentrations have been considered to primarily reflect coastal supplies and contributions from atmospheric dust and subsequent mixing processes via surface currents coupled with removal through particle scavenging [e.g., *Bertram and Elderfield*, 1993; *Nozaki*, 2001; *Amakawa et al.*, 2000; *Arsouze et al.*, 2009].

[6] It is well known that the resupply of nutrients (e.g., iron [*de Baar et al.*, 1995]) from subsurface to surface waters is the major mechanism to maintain productivity in open ocean surface waters [*Sarmiento et al.*, 2004; *Palter et al.*, 2010]. Processes contributing to the flux of nutrients to the euphotic zone are for example, diapycnal diffusion from the thermocline, Ekman pumping and upwelling [*Williams and Follows*, 1998], isopycnal perturbation by mesoscale eddies [*McGillicuddy et al.*, 2007], and transport from the thermocline into the mixed layer during winter convection [*Williams et al.*, 2006]. In this respect, Nd (and the REEs) and nutrients share some similarities. They are removed by biogenic particles and released at depth through particle dissolution and remineralization. Therefore, it is likely that underlying mechanisms of exchange fluxes of Nd and nutrients between the subsurface and shallower depths are analogous.

[7] In this study, we argue that surface water Nd concentrations are influenced by all the above mentioned external and subsurface fluxes. In order to identify the most influential factor that drives the variations of surface water Nd concentrations, we will separately evaluate the potential influences of the fluxes mentioned above.

## 2. Methods

[8] To estimate the potential influence of coastal advection versus scavenging, we refer to a well-established tracer for surface coastal water mass mixing. $^{228}$Radium with a half-life of 5.75 years has been applied in many studies for tracing coastal supply and lateral mixing processes of the upper ocean [e.g., *Kaufman et al.*, 1973; *Nozaki et al.*, 1990, 1998]. $^{228}$Radium mainly originates from the $\alpha$-decay of $^{232}$Th in estuarine, coastal, and shelf sediments, and is released into the water column via diffusion from porewaters or submarine groundwaters [e.g., *Moore*, 1969]. It is then transported to the open ocean by lateral surface advection and mixing with an average decay residence time of about 8 years. Because of its radioactive decay along the transport path, it exhibits sharp gradients between the coast and the open ocean [e.g., *Kaufman et al.*, 1973; *Nozaki et al.*, 1990]. It is thus assumed that the surficial distribution of $^{228}$Ra provides the actual pattern of coastal water mass influence on open ocean surface waters [e.g., *Nozaki et al.*, 1990]. However, when $^{228}$Ra has already decayed away, it cannot be a sensitive tracer for coastal water mass influence anymore. Fortunately, $^{228}$Ra has a residence time (removed through radioactive decay) of about 8 years which is comparable with surface layer Nd scavenging residence time (1–4 years in the upper 100 m [*Amakawa et al.*, 2000; *Rickli et al.*, 2010]). Consequently, if $^{228}$Ra originating from coastal sources has decayed away, the Nd flux from the same coastal sources will have been removed by scavenging and the Nd concentrations will have been diminished to negligible levels in the surface layer.

[9] Similar to nutrients, the vertical supply of Nd from subsurface water may also depend on upper ocean stratification. A measure of upper ocean stratification is the "buoyancy frequency" [e.g., *Jenkins*, 2003] which is a function of fluid density and vertical density gradient. However, this approach needs complex data processing, and is beyond the scope of our study. Instead, for simplicity we tentatively use the potential density anomaly ($\sigma_0$, the potential density of a water parcel with reference to the ocean surface subtracted by 1000 kg/m$^3$) at the base of the euphotic layer ($\sim$200 m, Figure 1) as an index of upper ocean stratification (200–1000 m, assuming 1000 m water depth is the lower boundary of the upper ocean mixing regime [e.g., *Wunsch and Ferrari*, 2004]). Since the global oceans have more similar potential densities in the deep waters at a given water depth (e.g., 1000 m) than the surface layers, it is reasonable to assume that the lower the potential density anomaly near the surface layer, the larger the potential density range of the upper ocean (i.e., more stratified). A similar approach to define ocean stratification as potential density differences between the upper and lower boundaries of the studied waters can be found for example in *Capotondi et al.* [2012]. As an additional index for upper ocean stratification we introduce the potential density difference between 200 and 1000 m water depth ($\Delta\sigma_0$). $\Delta\sigma_0$ is thus a more direct measure of the density gradient in the upper ocean. For example, a larger $\Delta\sigma_0$ would indicate higher potential density contrast in the upper ocean (200–1000 m water depth) and thus stronger stratification.

[10] Our main assumption is that if a specific flux or mechanism cannot cause geographical variations of Nd concentration similar to what we observe in the ocean (Figure 1), it cannot be considered as a primary controlling factor. Besides, we will estimate the Nd fluxes to the surface waters required to achieve the observed concentrations
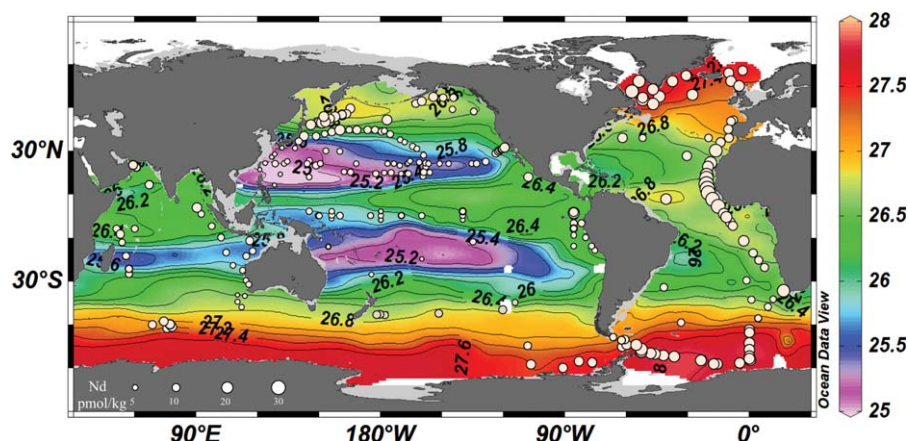
**Figure 1.** The distribution of Nd concentrations (circles) in the open ocean surface layer (shallower than 100 m). Also shown for comparison is the potential density anomaly $\sigma_0$ (contours) at the water depth of 200 m. The semienclosed basins or marginal seas (e.g., the Arctic [*Andersson et al.*, 2008; *Porcelli et al.*, 2009], the Bengal Bay [*Singh et al.*, 2012], the Nordic seas [*Lacan and Jeandel*, 2004], and the Mediterranean Sea [*Tachikawa et al.*, 2004]) are not compiled here. Some new data from the South Pacific reported in this study are marked by gray circles. See supporting information Table S1 for data collection.

to constrain our assumption. Finally, we will apply a three-dimensional model approach [*Rempfer et al.*, 2011, 2012b] to further test our arguments (see a brief description of the model in supporting information).

## 3. Discussion

### 3.1. The Influence of Coastal/Dust Flux and Particle Scavenging on Nd Concentrations in the Surface Layer

[11] Although Nd released from atmospheric dust is one of the important sources of the oceanic surface waters, previous studies have revealed that the distribution of Nd concentrations can hardly be explained by dust transport through the prevailing wind system [*Nozaki*, 2001]. Therefore, atmospheric dust deposition cannot be the dominant source of Nd for most of the global surface ocean with the exception of areas directly downwind of major dust plumes

such as the eastern tropical North Atlantic [*Rickli et al.*, 2010].

[12] To constrain the coastal water mass influence, we compiled the global surface seawater $^{228}$Ra activities (Figure 2). $^{228}$Radium is enriched in the continental margins and major surface currents, e.g., Kuroshio and its extension (Northwest and mid-North Pacific), or the Gulf stream (Northwest Atlantic), but strongly depleted in the southern Hemisphere oceans due to their remoteness from coastal influences. In contrast, high Nd concentrations are observed in high-latitude oceans of both hemispheres, while low Nd concentrations generally occur in subtropical gyres. In particular, in the Southern Ocean where $^{228}$Ra activities are relatively low, Nd concentrations are higher than in the middle and low latitudes of the North Pacific (Figure 1). Nevertheless, there are also areas showing comparatively high $^{228}$Ra activities in the Southern Ocean (e.g., Western Australia, off the tip of South America, South Africa).
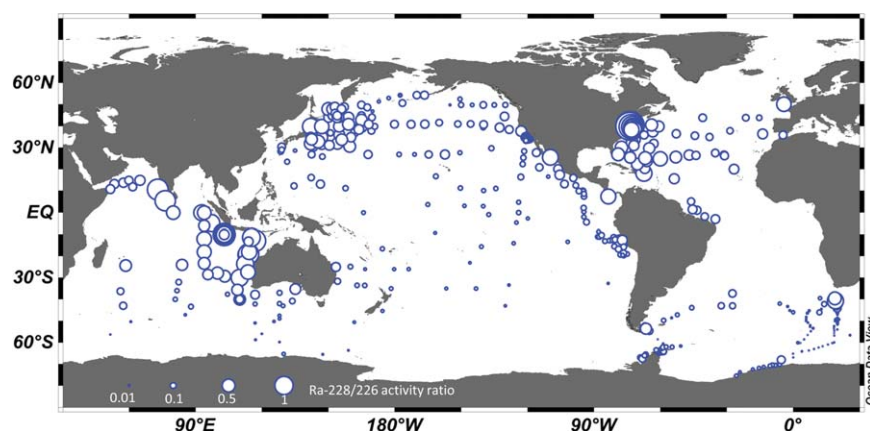


**Figure 2.** Compilation of $^{228}$Ra/$^{226}$Ra activity ratios of the surface ocean. To account for the potential particle scavenging effect, $^{226}$Ra activity was normalized by $^{226}$Ra [e.g., *Nozaki et al.* 1990]. Data sources: *Moore* [1969]; *Knauss et al.* [1978]; *Kaufman et al.* [1973]; *Nozaki et al.* [1990]; *Nozaki et al.* [1998]; *Nozaki and Yamamoto* [2001]; *Hanfland* [2002]; *Kawakami and Kusakabe* [2008].

Given these sites are mostly close to continents, it is likely that they have been considerably influenced by coastal fluxes, for which it is well known that they are characterized by high [228]Ra activities. These fundamental and systematic differences between Nd concentrations and [228]Ra activities indicate that surface water Nd concentrations must also be influenced by processes other than the coastal influence and which did not exert primary control on [228]Ra distribution. It is noteworthy that Nd concentrations in high-latitude surface waters of both hemispheres [*Siddall et al.*, 2008] are actually not lower than in subtropical areas where particle export fluxes are lower. This characteristic challenges the role of particle concentration as the major controlling factor of the global distribution of surface water Nd concentrations. Instead, we suggest that flux of Nd from the subsurface waters is an important source of Nd to surface waters and thus an important mechanism for driving basin-scale variations of Nd concentrations in the surface ocean.

### 3.2. The Supply of Nd From Subsurface Thermocline Waters to the Surface Layer

[13] Because Nd behaves similar to nutrients as introduced in section 1, a potentially important issue is the mechanism by which nutrients are transferred from the subsurface thermocline waters to the euphotic layer at different latitudes, which is still subject of considerable biogeochemical debate both in observational and modeling studies [e.g., *Oschlies and Garcon*, 1998; *McGillicuddy et al.*, 2007; *Sarmiento et al.*, 2004; *Palter et al.*, 2010].

[14] In high latitudes, where the surface ocean is generally less stratified than in low latitudes, nutrients are transferred from thermocline waters into the overlying deepened mixed layer by a combination of vertical and lateral advection (e.g., nutrient streams) [*Williams et al.*, 2006; *Marshall and Speer*, 2012]. Pronounced vertical mixing and upwelling of nutrient-rich waters in the Southern Ocean sustains export fluxes in large parts of the global ocean [*Sarmiento et al.*, 2004; *Palter et al.*, 2010]. The mixed layer depths become deeper toward high latitudes, and this is where increased supply occurs. In contrast, at low latitudes, water is preferentially subducted to the permanent thermocline (i.e., formation of mode waters [*Qiu and Huang*, 1995]). In this case, shallow Ekman advection most likely brings the major part of marginal nutrients to the gyre interior. In addition, there is growing evidence for mesoscale eddies driving the thermocline nutrients to the surface and a number of studies [*Oschlies and Garcon*, 1998; *McGillicuddy et al.*, 2007] have suggested that this mechanism may be important for maintaining primary surface water productivity of the subtropical ocean, although the quantitative significance of mesoscale eddies is still under debate. To summarize, exchange between thermocline and surface waters at different latitudes is controlled by different mechanisms. Overall, stratification inhibits vertical mixing within the water column and thus limits the magnitude of the flux of nutrients and probably also of Nd from subsurface to surface waters.

[15] The Pacific subtropical gyres which correspond to lowest surface concentrations of Nd, are significantly more stratified than the high latitudes where higher Nd concentrations prevail (Figure 1). Interestingly, a positive (negative) correlation between $\sigma_{0-200\ m}$ ($\Delta\sigma_0$) and surface water Nd concentrations is also observed (Figure 3). In contrast, there is virtually no relation between the [228]Ra/[226]Ra ratio and $\sigma_{0-200\ m}$ (Figure 4). Note however that the majority of observations for surface water [228]Ra concentrations available in the literature have been obtained from locations close to continental margins. Such a bias may result in the fact that Figure 4 is not fully representative of the global surface ocean. Given the already poor correlation presented in Figure 4, it seems rather unlikely that more data from the oceanic interior (expected to show low [228]Ra/[226]Ra ratios) would improve the correlation between the [228]Ra/[226]Ra ratios and the $\sigma_{0-200\ m}$. Moreover, the observation that Nd concentrations are generally lower in subtropical gyres than at high latitudes is robust (e.g., see Pacific data in Figure 1), despite Nd concentration data are sparse in most of the subtropical gyres compared to other locations.

[16] This leads us to hypothesize that upper ocean vertical supply, itself limited by density stratification, is an important factor in controlling variations of open ocean surface Nd concentrations. As mentioned above, the magnitude of Nd concentrations is modified by a number of processes at any location (e.g., dust dissolution, lateral advection, and scavenging/remineralization), thus complicating the quantitative interpretation of variations of surface Nd concentrations. In particular, lateral advection (e.g., within the Antarctic Circumpolar Current (ACC)) has the potential to effectively transport thermocline-derived Nd over long distances within surface currents. Moreover, efficient remineralization of sinking particles within subsurface layers may rapidly release the adsorbed Nd flux back to the water column. The magnitude of Nd flux being released this way at shallower depths may not only depend on the stratification of the upper ocean but also on the nature of the particles. Finally, in some local open ocean areas like the eastern equatorial Atlantic, dust and/or coastal sources may in fact be the major sources for Nd in the surface waters [*Rickli et al.*, 2010] (marked outliers in Figure 3). In the Southern Ocean southwest of Australia, Nd concentrations are unexpectedly low, and more observational studies are needed to evaluate the main controlling processes in this area. All the above factors contribute to the scatter of observations in Figure 3. In addition, the scatter could also simply be related to the uncertainties in estimating potential density anomalies for each sampling location.

### 3.3. Surface Nd Flux Estimation and Modeling

[17] The inventory of Nd in the uppermost 100 m of the global ocean excluding the Arctic is about $5.9 \times 10^{10}$ g (based on the compilation of *Tachikawa et al.* [2003]). To be consistent with a scavenging residence time of 1∼4 yr [*Amakawa et al.*, 2000; *Rickli et al.*, 2010], an influx between ∼$1.48 \times 10^{10}$ and $5.90 \times 10^{10}$ g/yr is needed. This is about two orders of magnitude larger than the riverine and dust fluxes of Nd (about $3.4 \times 10^8$ g Nd/yr and $2.6 \times 10^8$ g Nd/yr, respectively [*Rempfer et al.*, 2011]). Clearly, large additional influxes of Nd to the surface layer are required, as also noted in earlier studies [e.g., *Amakawa et al.*, 2000]. It has been proposed that the supply of Nd from the ocean margins via dissolution of and exchange with marine sediments [e.g., *Jeandel et al.*, 1998; *Amakawa et al.*, 2000; *Tachikawa et al.*, 2003; *Lacan and Jeandel*, 2005;
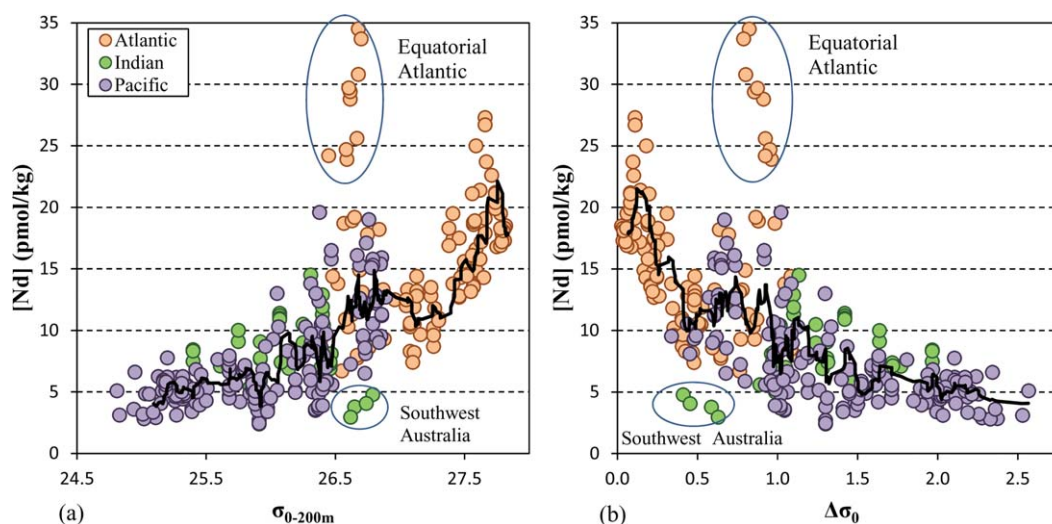
**Figure 3.** Surface water Nd concentrations ([Nd], 0–100 m water depth) versus potential density anomaly $\sigma_{0-200\text{ m}}$ at (a) 200 m water depth, and (b) density differences $\Delta\sigma_0$ (between 200 and 1000 m water depth). The data of $\sigma_{0-200\text{ m}}$ and $\sigma_{0-1000\text{ m}}$ have been extracted from the Electronic Atlas of WOCE (http://www.ewoce.org/). A depth of 200 m (which corresponds to the bottom of the euphotic layer) is chosen because such depth does not greatly suffer from seasonal disturbances, variability of particle concentrations as well as hydrographic changes, but still tracks the overall stratification status of the upper ocean as a first approximation. The lower boundary of the upper ocean mixing regime is taken as 1000 m depth in all ocean basins [e.g., *Wunsch and Ferrari*, 2004]. Deviations from the general trend are marked by ovals. Solid lines are the 10 points moving average of these data which have a (linear) correlation coefficient ($R^2$) of 0.65 between [Nd] and $\sigma_{0-200\text{ m}}$, and of 0.59 between [Nd] and $\Delta\sigma_0$ (excluding the circled outlier data).

*Arsouze et al.*, 2009; *Rempfer et al.*, 2011, 2012b] is an important, even overwhelming source of Nd to the global ocean. Since the exact nature of the boundary Nd source is still largely unknown, a precise estimation of shallow coastal Nd flux to the surface layer is difficult. Even if we assume that all the Nd from the boundary source ($5.5 \times 10^9$ g Nd/yr following *Rempfer et al.* [2011]) is supplied to the ocean via the surface layer, the total external flux from dust, river, and boundary source (summing up to $6.1 \times 10^9$ g Nd/yr) only



**Figure 4.** Surface water $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio versus potential density anomaly $\sigma_0$ at 200 m water depth. The data of $\sigma_{0-200\text{ m}}$ are from Electronic Atlas of WOCE, while data of $^{228}\text{Ra}/^{226}\text{Ra}$ are the same as in Figure 2.

accounts for up to 37% of the required surface flux. Moreover, it is important to keep in mind that extremely high oceanic surface productivity normally occurs in the continental shelf areas [*Falkowski et al.*, 1998]. It is thus likely that, due to the high particle concentration and associated scavenging on or near the shelves, a large fraction of shelf Nd cannot escape the shelf area to the open ocean via surface currents [*Grasse et al.*, 2012]. Thus, the shallow coastal Nd flux reaching open ocean surface waters is further reduced. The argument above is consistent with the study of *von Blanckenburg and Igel* [1999]. Using an idealized two-dimensional model, these authors showed that the distribution of particle reactive tracers (such as Be, Pb) would have high concentration gradients from the continental margin to the gyre interior, when only considering lateral advection and scavenging of the coastal inputs.

[18] Using a three-dimensional modeling approach, we examine the potential influence of shallow coastal/dust supply on surface Nd concentrations (see a brief description of the model in supporting information). Figures 5a and 5b show the Nd concentrations and $\varepsilon_{\text{Nd}}$, respectively, as obtained with the CTRL experiment of *Rempfer et al.* [2012a]. In an additional simulation, Nd inputs by dust and riverine discharge are omitted and the Nd boundary source is confined to depths between about 200 and 3000 m depth while keeping its magnitude unchanged (EXP1, Figure 5c for concentrations, Figure 5d for $\varepsilon_{\text{Nd}}$). In this experiment, no sources are applied at depths shallower than 200 m. The difference (CTRL-EXP1) between the two simulations is indicated in Figures 5e (concentrations) and 5f ($\varepsilon_{\text{Nd}}$).
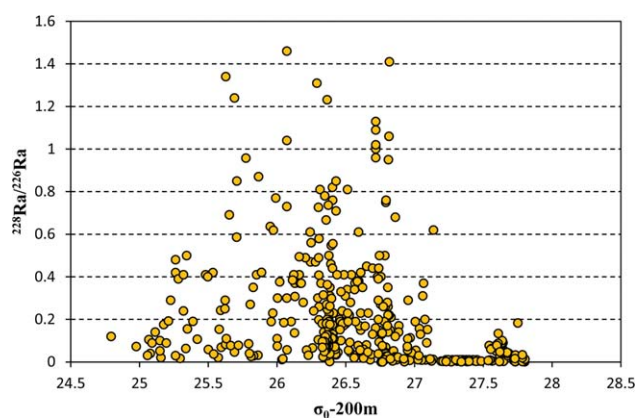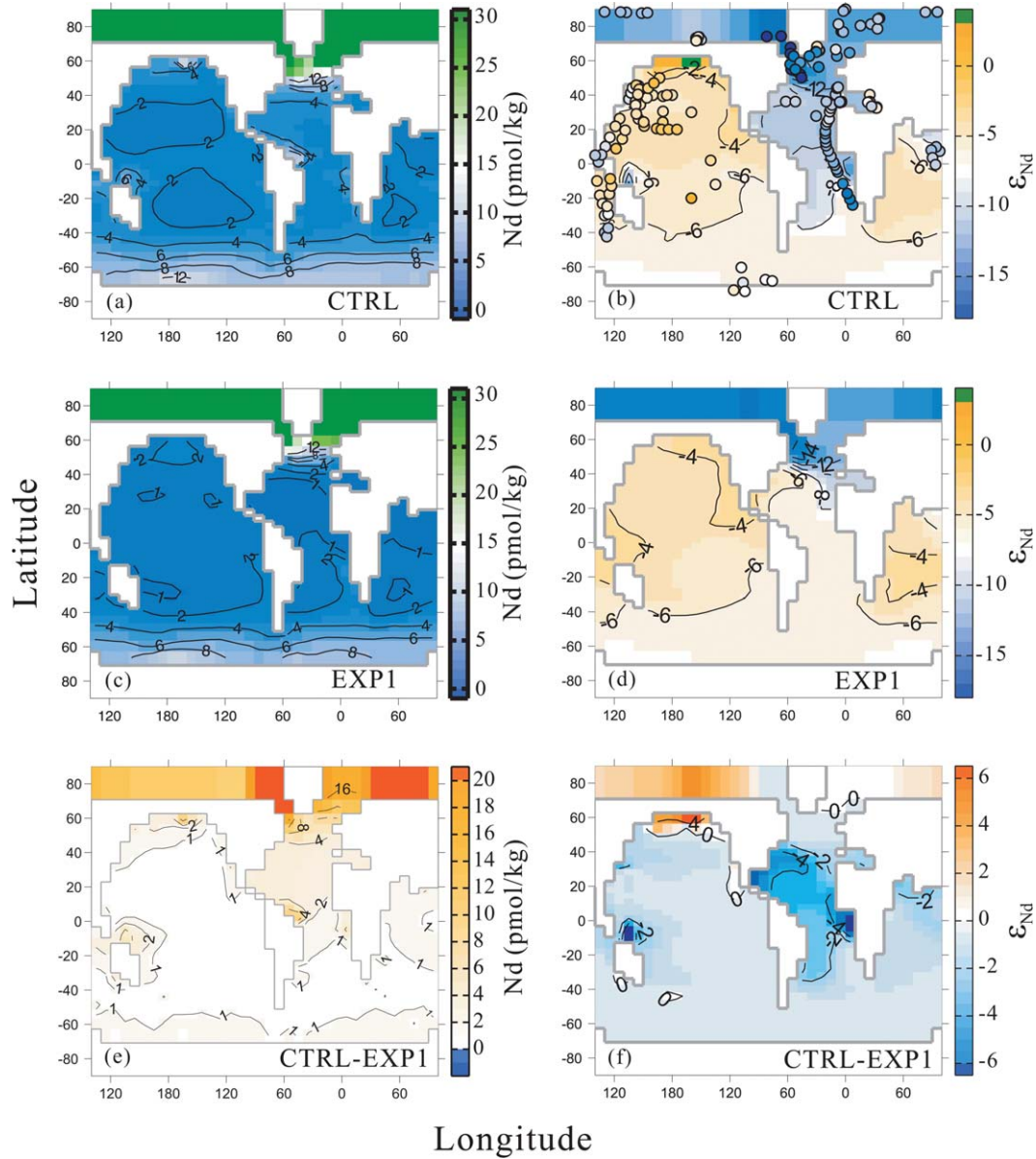
**Figure 5.** (a) Surface Nd concentration (pmol/kg) and (b) $\varepsilon_{Nd}$ obtained with the CTRL of *Rempfer et al.* [2012a]. Observations (circles) are superimposed using the same color coding [*Rempfer et al.*, 2012a]. (c) Nd concentration (pmol/kg) and (d) $\varepsilon_{Nd}$ from a simulation (EXP1) where all Nd sources at depths shallower than 200 m have been omitted. Note, in EXP1 the boundary source is confined to depths between 200 and 3000 m. The (e) concentration and (f) $\varepsilon_{Nd}$ differences between the two simulations (CTRL-EXP1).

[19] Interestingly, the simulated $\varepsilon_{Nd}$ of CRTL experiment is broadly consistent with observations (Figure 5b) and is similar to those of EXP1 (Figure 5d). In most of the Pacific and Southern Ocean, the differences between CTRL and EXP1 are less than 0.5–1.0 $\varepsilon_{Nd}$ units. Exceptions are the tropical and subtropical Atlantic, where CRTL-EXP1 is larger than 2 $\varepsilon_{Nd}$ units, implying riverine and dust inputs might have exerted a notable influence on the Nd isotope compositions of ocean surface waters in these areas, which was also noted in *Arsouze et al.* [2009]. On the other hand, the simulated surface water Nd concentrations in CTRL (Figures S1 and 5a) are 1–2 pmol/kg in most of the open Pacific, 2–10 pmol/kg in the open Atlantic, and 4–8 pmol/

kg in the ACC which is lower than available observations (e.g., 4–15 pmol/kg in the open Pacific, 10–30 pmol/kg in the open Atlantic, and 10–20 pmol/kg in the Antarctic Circumpolar Current, see also Figure 1). Nevertheless, the CTRL experiment initially presented *Rempfer et al.* [2012a] is an optimal parameterization designed for the simulation of both Nd concentration and $\varepsilon_{Nd}$ in the global ocean in agreement with available observations. First-order characteristics of distributions of Nd concentrations in the surface ocean are clearly captured by the model. For example, simulated Nd concentrations are lower in the Pacific than in the Atlantic and the ACC. Nd concentrations are also lower in subtropical gyres than in high latitudes where

Nd concentrations become higher toward the polar regions. Given that the vertical transport of Nd was simulated by the model, we can obtain information on the relative importance of this vertical supply in different oceanic areas relative to the contributions from the boundary sources, rivers and dust.

[20] Overall, the effect of cutting off external Nd-input into the upper 200 m on Nd concentrations is larger in regions close to the margins than in regions far away from the continents. The results in Figures 5e and 5f imply that input from shallow boundary sources, riverine supply, and release from dust might not be of primary importance notably in the Southern Ocean and the Pacific, which supports our case made in section 3.1. Unfortunately, the low simulated Nd concentrations compared to observations complicate the estimation of the quantitative importance of internal Nd sources (i.e., subsurface Nd fluxes) on a global scale. Improving the ability of models to simulate surface Nd concentrations in agreement with observations is beyond the scope of this study. Nevertheless, it has become apparent from our discussion that such improvement cannot simply be achieved by increasing the shallow coastal Nd input. A recent study [*Akagi*, 2013] proposed that incorporation of Nd into diatoms and subsequent diatom dissolution in deep water (thus irreversible intake and release) is the dominant mechanism in explaining the nonzero surface seawater REE (including Nd) concentrations as well as enrichment of REE at depth. While a close link of Nd with silicic cycling (or other nutrient elements) is not surprising, as discussed in section 3.2, our modeling above clearly shows that nonzero surface Nd concentrations and first-order surface Nd concentration distribution can be produced using a reversible scavenging model. Moreover, given that reversible scavenging of Nd is required to successfully simulate both Nd concentrations and isotope compositions of the deep water profiles [e.g., *Bertram and Elderfield*, 1993; *Siddall et al.*, 2008; *Arsouze et al.*, 2009; *Oka et al.*, 2009; *Rempfer et al.*, 2011], it appears that future modeling studies should aim at resolving the surface concentration issue by focusing on the fluxes related to vertical supply and reversible scavenging parameters of Nd in the upper ocean.

## 4. Conclusions

[21] The knowledge on Nd cycling in surface waters is important for interpreting oceanic Nd isotope records in terms of oceanic environmental changes, such as ocean circulation versus local shallow weathering supply [e.g., *Rempfer et al.*, 2012b]. Our study implies that open ocean surface Nd concentrations are influenced by processes other than shallow coastal/dust input and subsequent surface current mixing and scavenging. We propose that vertical supply of Nd from subsurface waters to shallower depths, itself limited by upper ocean stratification, may represent an important contribution to the Nd budget of open ocean surface waters. This is supported by (1) the systematic differences of global distribution of Nd concentrations and $^{228}$Ra activities, (2) the positive correlation between Nd concentrations and the potential density anomaly at 200 m water depth (i.e., the base of euphotic layer), (3) the estimation of the magnitude of required Nd input flux to maintain the observed surface ocean Nd concentrations, and (4) results from a three-dimensional modeling experiment. Future field and modeling studies focusing on the scavenging of Nd and stratification-related Nd flux in the surface layer are thus of high priority in order to precisely understand the surface water Nd cycle as well as to better constrain the use of Nd isotopes as a (paleo)circulation proxy.

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