

## Neodymium isotopes distribution and transport in the central North Pacific deep water

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Radiogenic Neodymium (Nd) isotopes in the central North Pacific deep water have been generally interpreted as reflecting Pacific arcs input according to previous studies, while little is known about the specific path and mode of radiogenic Nd from these arcs to deep water. In this study, Nd isotopic data from surface scrapings of 17 Fe-Mn crusts from the central North Pacific deep water (around 20°N) have been reported. Based on these data together with recently published data on dissolved Nd isotopes in seawater for this region, we propose that radiogenic Nd isotopes in North Pacific Intermediate Water (NPIW 300–800 m) from the western Pacific margin is an important source for the central North Pacific deep water, while the surface water of this area has little influence on dominating deep water Nd isotopes. Such a view emphasizes the role of vertical mixing and advection of the marginal ocean currents in balancing Nd isotopes in central North Pacific deep water, and helps to understand oceanic Nd cycling. Moreover, the values of  $\epsilon_{Nd}$  from surface scrapings of Fe-Mn crusts are relatively homogenous, with an average value of about  $-3.4$ , which is higher than that of modern seawater ( $\sim -3.9$ ). This difference over short time scales reflects the quick evolution of Nd isotopes in central North Pacific deep water.

**central north Pacific deep water, Nd isotopes, Fe-Mn crusts, intermediate water, Nd budget**

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Oceanic Neodymium (Nd) isotopic studies (including authigenic ferromanganese deposits), which began in the late 1970s [1], have now become a powerful method to reconstruct modern and past ocean circulation. Compared with other tracers of paleo-oceanic currents (such as C isotopes and Cd/Ca of foraminifera), Nd isotopes are less affected by factors not directly related to ocean currents (e.g. biological activity) [2], and have thus attracted growing attention from the paleoceanographic community.

Of the global oceans the North Pacific contains the most radiogenic Nd isotopes [1]. Based on the study of three North Pacific deep water ferromanganese crusts, Ling et al. [3] discovered the discrepancy of Nd isotopic compositions at different depths, and concluded such discrepancies can be traced back to at least 25 Ma. In fact, there were already

many Atlantic Nd isotopic data on seawater and ferromanganese crusts, while data on the Pacific were still very scarce, and sample depth information was often missing in the early studies [1,4]. In general, the high  $\epsilon_{Nd}$  value of the Pacific can be interpreted as weathering input from the widely distributed active volcanic arcs around the Pacific, most of which have high  $\epsilon_{Nd}$  (e.g. [1]). However, the transport path and mode of radiogenic Nd in the North Pacific deep water are still not well understood. Consequently, the interpretations of past Nd isotopic variations in this area were not very clear. Given the trend of more and more high-resolution Nd isotope studies for tracing the paleo-ocean circulation (e.g. glacial-interglacial) in recent years [5–9], it becomes more important to understand the radiogenic sources of the North Pacific deep water, which will help to reconstruct the North Pacific glacial-interglacial or even millennium-scale circulation changes.

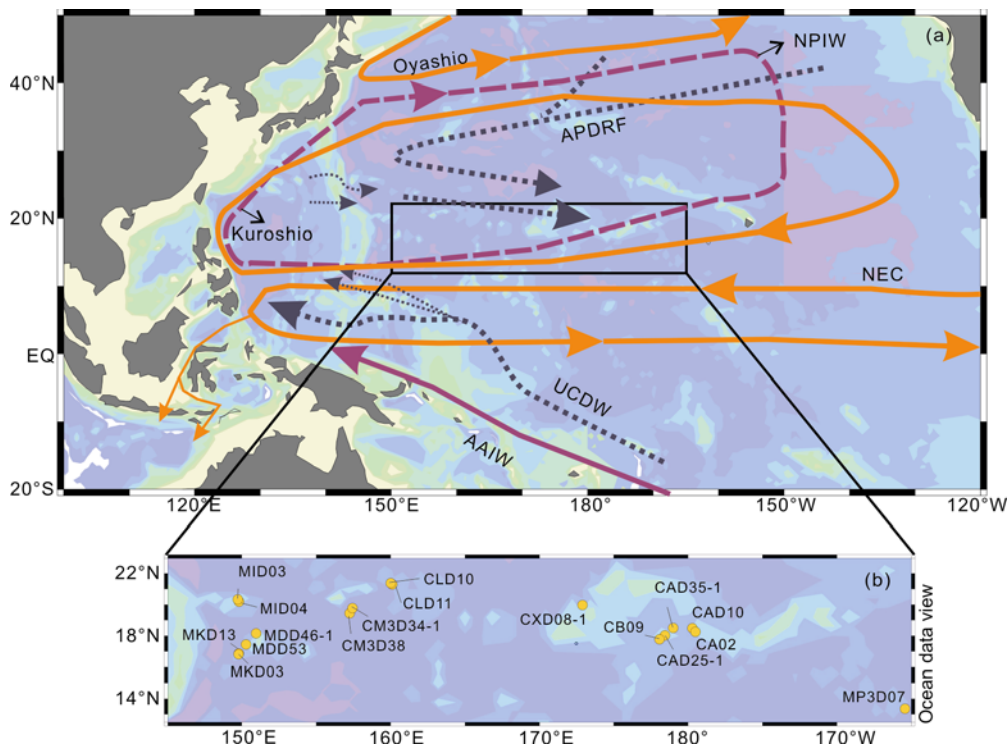
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To further understand the radiogenic Nd sources in the deep central North Pacific, we report ferromanganese crust Nd isotope data with different water depths near 20°N in this study. Hydrothermal Nd has an insignificant effect on seawater [10,11]. Thus the Nd isotopes in seawater are mainly controlled by continental inputs (including the island-arc weathering). When terrigenous materials are transported by rivers and dust to the ocean surface, they can release part of their Nd to the deep water by advection and diffusion [12], so that seawater can acquire a corresponding Nd isotopic composition. Meanwhile Nd can be absorbed onto and desorbed from particles in the seawater, and then be introduced into the deep water during particle sinking [13,14]. The shelf sediment-seawater Nd isotope exchange can also transfer Nd to the deep ocean [15–17]. Based on these and the modern ocean current studies and data of this study, we demonstrate that the advective transport of Nd by North Pacific Intermediate Water is an important source of radiogenic Nd. Furthermore, we also find the quick evolution of Nd isotope composition of the North Pacific deep water in a short time scale.

## 1 Modern oceanography in the North Pacific

The North Pacific surface currents are schematically shown in Figure 1(a) (the orange solid line and purple dotted line), with the Kuroshio Current being part of this system. On the

other hand, in the Subarctic Pacific (north of 40°N), Oyashio is characterized by counterclockwise circulation. High salinity North Pacific Tropical Water (NPTW,  $\theta=18\text{--}25^\circ\text{C}$ ,  $S=34.8\text{--}35.3$ ,  $\sigma_\theta=23.0\text{--}25.0$ , depth 100–200 m) is widely distributed in the North Pacific subsurface depth [25]. Although in the modern North Pacific there are no deep water mass formations, the intermediate water can still have good ventilation [26]. While the intermediate water mainly originates from the Sea of Okhotsk [18–20], there is also sporadic formation in the Gulf of Alaska [27]. North Pacific Intermediate Water (NPIW,  $\theta=1.4\text{--}6.4^\circ\text{C}$ ,  $S=33.3\text{--}33.4$ ,  $\sigma_\theta=26.6\text{--}27.4$ ) is about 300–800 m deep and circulates clockwise [18–20]. After the major currents at about 40°N flow eastward to about 150°W, they turn southward to about 20°N, and then there is some return flow to the west. At about 3500 m, the deep ocean currents, mainly from the Upper Circumpolar Deep Water (UCDW), are transported along the Southwest Pacific from the Southern Ocean. Near the equator, the deep ocean currents mainly export to the Philippine Sea, and ultimately form part of the Pacific Deep Water (PDW) (the major path is shown as the dark blue dotted line in Figure 1(a) [21–23]). Water masses deeper than 3500 m are the northward transport of the Lower Circumpolar Deep Water (LCDW) which is characterized by high salinity and low silica, and is the remnant of the North Atlantic Deep Water (NADW) [24]. The North Pacific deep return flow mainly concentrates on the eastern Pacific basins, whereas the upwelling is mostly located east of 20°W



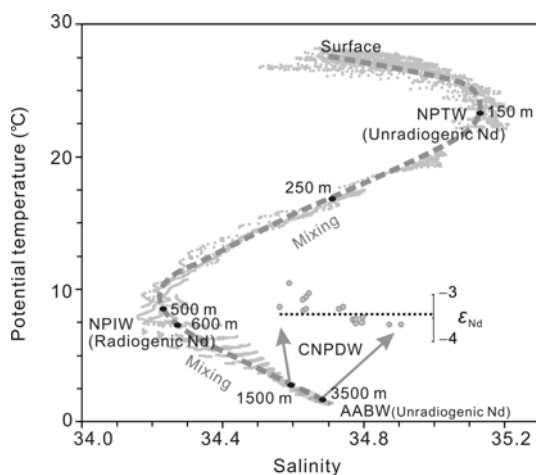
**Figure 1** (a) A schematic map showing the major currents of the North Pacific: Surface (<200 m, orange solid line); intermediate (200–1500 m, dashed purple line), and deep (deep blue dotted line). The black box indicates the area of this study [18–24]. APDRF, Subarctic Pacific return flow; NEC, North Equatorial current; UCDW, Upper Circumpolar Deep Water; AAIW, Antarctic Intermediate Water; (b) sample location map.

and north of 40°N in the Pacific [24]. More details about present-day North Pacific circulation can be found in [24].

The study area is located in the central North Pacific near 20°N (Figure 1). Based on the North Pacific circulation background discussed above, the water mass structure of the study area is illustrated by the traditional Temperature-Salinity diagram (Figure 2). According to Figure 2, we can see that NPTW (about 150 m) is characterized by high salinity (about 35.1), while NPIW (500–600 m) is characterized by lower salinity in this area. In contrast, the central North Pacific Deep Water (CNPDW, below 1500 m) has a very narrow range of temperature and salinity, which is formed by the Antarctic Bottom Water (AABW) mixed with a small amount of NPIW. It should be noted that, due to partial mixing with the surrounding water masses during transport, the salinity of NPTW in this area is slightly decreased, while the temperature and salinity of NPIW are slightly increased.

## 2 Materials and methods

Ferromanganese crusts in this study were all collected from the central North Pacific seamounts (Figure 1(b)) during the cruises organized by the State Oceanic Administration of China. For Nd isotope analysis, about 10 mg of each sample was gently scraped from the crust surface using a blade. The depth of these crusts can be seen in Table 1. Neodymium isotope measurement was carried out at the State Key Laboratory for Mineral Deposits Research at Nanjing University, using LN resin and HCl leaching for the chemical separation of Nd. Neodymium measurements were performed with a Finnigan Triton thermal ionization mass spectrometer (TIMS).  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios were normalized to



**Figure 2** The temperature-salinity distribution of seawater in the study area (data source: World Ocean Atlas 2005. [http://odv.awi.de/en/data/ocean/world\\_ocean\\_atlas\\_2005/](http://odv.awi.de/en/data/ocean/world_ocean_atlas_2005/)). The circles represent  $\epsilon_{\text{Nd}}$  distribution of the crust surface samples. As shown by the gray arrows, the depth of samples increases from 1500 to 3500 m (left to right).

**Table 1** Nd isotopic compositions for the crust surface samples<sup>a)</sup>

Sample	Depth (m)	$^{143}\text{Nd}/^{144}\text{Nd}$ (surface)	$\epsilon_{\text{Nd}}$
CA02	3285	0.512452 ± 4	-3.6
duplicate		0.512453 ± 2	-3.6
CB09	3474	0.512452 ± 2	-3.6
duplicate		0.512454 ± 4	-3.6
CM3D34-1	2748	0.512454 ± 1	-3.6
duplicate		0.512460 ± 2	-3.5
CLD10	2850	0.512454 ± 27	-3.6
MDD53	2700	0.512457 ± 2	-3.5
MP3D07	2860	0.512460 ± 5	-3.5
MKD13	1530	0.512471 ± 2	-3.2
CAD35-1	2000	0.512486 ± 1	-3.0
MID03	1680	0.512497 ± 3	-2.8
MKD03	1900	0.512479 ± 1	-3.1
CXD08-1	1910	0.512467 ± 2	-3.3
MID04	1950	0.512469 ± 3	-3.3
CAD10	2480	0.512469 ± 1	-3.3
CAD25-1	2750	0.512453 ± 2	-3.6
CLD11	2750	0.512458 ± 1	-3.5
CM3D38	2548	0.512471 ± 1	-3.2
MDD46-1	1950	0.512482 ± 3	-3.0

a) Longitudes and latitudes of these samples are available upon request.

$^{146}\text{Nd}/^{144}\text{Nd}=0.7219$  to correct for the instrumental mass bias during measuring. Repeated analyses of the Nd standard JNdi-1 yielded  $0.512121 \pm 2$  ( $2\sigma$  external standard deviation,  $n=15$ ).

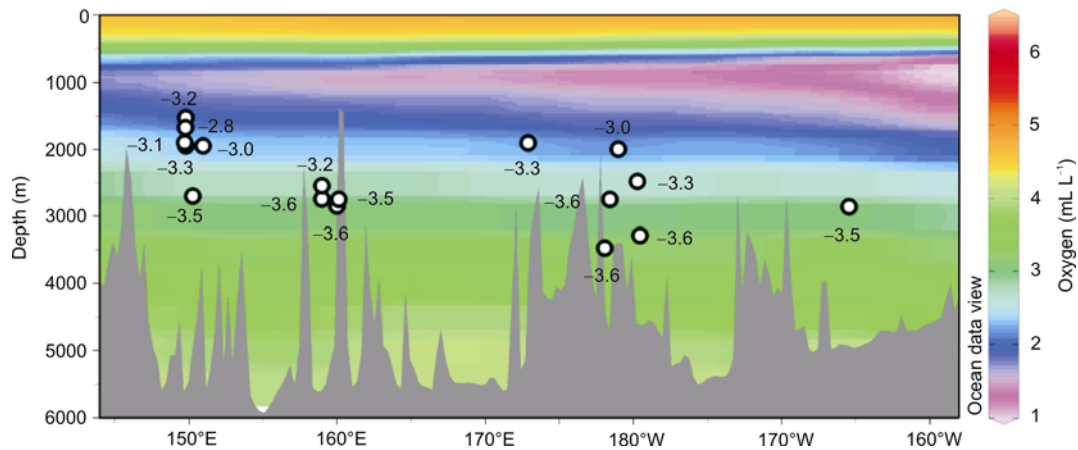
## 3 Results and discussion

### 3.1 Results for Nd isotopic compositions of surface samples

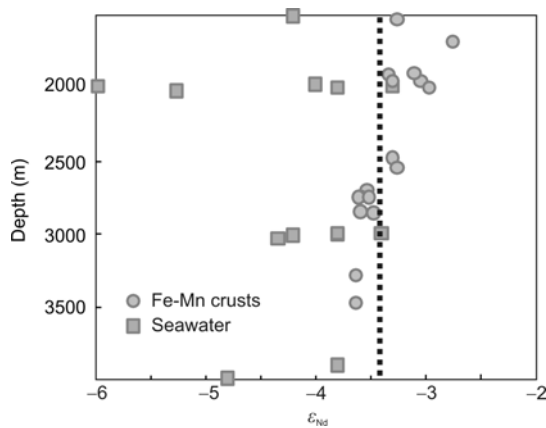
The Nd isotopic compositions of these samples are listed in Table 1. In general, the ferromanganese samples should represent the average Nd isotopic composition of ambient seawater over a certain period of time. For the surface samples, the Nd isotopes of the central North Pacific deep water were integrated through the late Quaternary. It can be seen from Figure 2 that the  $\epsilon_{\text{Nd}}$  values are relatively uniform in late Quaternary central North Pacific Deep Water, with a maximum value of -2.8, a minimum value of -3.6, and an average of about -3.4. Nevertheless, Nd isotopes still became slightly more unradiogenic with increasing depth (Figure 3). In the upper part of deep water (e.g. < 2500 m), greater variations of  $\epsilon_{\text{Nd}}$  at a similar depth are found than in the lower part of deep water (Figures 3 and 4). However, there is almost no horizontal  $\epsilon_{\text{Nd}}$  gradient.

### 3.2 Distribution and transport of radiogenic Nd to deep water

As shown in Figure 3, an obvious feature of the distribution



**Figure 3**  $\epsilon_{\text{Nd}}$  distribution of the crust surface samples along the 20°N profile. It is noted Nd isotopes show almost no horizontal gradient. The filled color represents the contents of  $\text{O}_2$  ( $\text{mL L}^{-1}$ ). One can easily see that the southern component water is characterized by higher  $\text{O}_2$  content at the bottom (near 170°E). The figure was generated using Ocean data view. Data source for oxygen content: World Ocean Atlas (2009) (odv.awi.de).



**Figure 4** A comparison of  $\epsilon_{\text{Nd}}$  between modern seawater [28,29] and crust surface samples.

of deep water  $\epsilon_{\text{Nd}}$  values is their homogeneity at a similar depth. For example, at about 3000 m, two samples (MDD53, MP3D07) which are separated by a distance of nearly 5000 km at about 165°W and 150°E, show  $\epsilon_{\text{Nd}}$  values of  $-3.5$  to  $-3.6$ , clearly more radiogenic than in the Southern Ocean ( $\epsilon_{\text{Nd}} < -5$ ). The issue is where the sources of the radiogenic Nd are, and how the central North Pacific deep water, which has an average  $\epsilon_{\text{Nd}}$  of about  $-3.5$  but comes from the Southern Ocean ( $\epsilon_{\text{Nd}} < -5$ ), acquires its radiogenic signature.

Previous views about the potential sources and input mechanisms of radiogenic Nd isotopes for the central North Pacific deep water include: (a) physical convection-diffusion taking surface Nd to the deep water [12]; (b) vertical sinking of particles through adsorption and desorption drawing the surface Nd (<200 m) down into the deep sea [30]; (c) dissolution of highly radiogenic volcanic ash and glass in the seawater adding extra radiogenic Nd [31]; and (d) Nd being released from the continental shelf and seafloor sediments into the deep Pacific water [32].

To simulate Nd isotope distribution in the seawater, Jones et al. [12] set Nd isotopic compositions of the surface ocean as boundary conditions, and assumed Nd was a fully conservative element. For the North Atlantic, the simulation agrees well with the observed distribution of Nd isotopes, but for the North Pacific deep water the simulated  $\epsilon_{\text{Nd}}$  values are lower than the observed values. Therefore, the Nd isotopes from the surface ocean through diffusion and convection are not enough to account for the deep radiogenic feature. Siddall et al. [30] used Nd isotopes and concentration at the ocean surface as boundary conditions, in combination with the reversible scavenging model, to explain the global seawater Nd isotopes and their concentrations. The simulation results for the North Pacific deep water are much closer to the observed values compared with the results of Jones et al. [12]. This indicates that reversible scavenging is important in taking the upper North Pacific radiogenic Nd into deep water, but there are still some unresolved issues: (a) Siddall et al. [30] did not effectively simulate either the observed widespread un-radiogenic signature in the North Pacific subsurface water [28] ( $\sim 100\text{--}300$  m) or the gradually increasing value of  $\epsilon_{\text{Nd}}$  downwards (to about 1000 m). This indicates that Nd from the North Pacific surface waters alone is unable to generate the observed distribution of radiogenic Nd in deep water. (b) Most of the North Pacific radiogenic signature of Nd (i.e.  $\epsilon_{\text{Nd}}$  higher than  $-4$ ) is somewhat underestimated in the modeling. To explain such discrepancy, Siddall et al. [30] point out that since the modeled surface boundary conditions are too simple, some of the radiogenic sources with high  $\epsilon_{\text{Nd}}$  are not included in the model simulation (e.g. near the Hawaiian islands). However, even if some surface areas with high  $\epsilon_{\text{Nd}}$  have not been considered in the boundary conditions [30], these local surface regions are still unlikely to contribute much to the deep radiogenic Nd isotopes. For example, the Hawaii Island chain, which could provide much radiogenic Nd to the surface

water, has almost no effect on deep water Nd isotopes [28]. Thus there is no reliable evidence to support an argument that the central North Pacific surface water provides enough radiogenic Nd to the deep water.

It is still difficult to estimate the dissolved flux of highly radiogenic volcanic ash and glass in the seawater. Previous studies have shown that most of the leachable components of volcanic ash are released into the seawater during the initial stages of dissolution, and then dissolution rate decreases rapidly. Consequently, the dissolution of ash and volcanic glass should take place mainly on the ocean surface [33]. As mentioned above, the ocean surface is unlikely to fully explain the North Pacific deep water radiogenic Nd isotope distribution. Thus we consider that dissolution of ash and volcanic glass in seawater is not an important source of deep water radiogenic Nd.

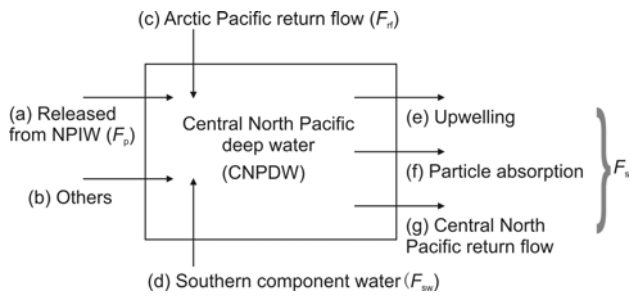
One possible explanation is that the radiogenic Nd of the central North Pacific deep water comes from re-dissolution of deep shelf sediments (for example, 0–3000 m [32]), and is advected by deep currents to the central North Pacific deep water which is mixed with un-radiogenic southern component water. In this case, the radiogenic Nd is characterized by pervasive input from the continental shelf. For example, Arsouze et al. [32] assume that ocean sediment at a depth of less than 3000 m would export Nd to the ocean. However, there are still questions remaining about this model which assumes the dissolution of deep sea sediments as the most important source of radiogenic Nd for the central North Pacific deep water: (a) the role of shelf sediment at more than 1000 m is not yet understood, i.e. it is still unclear whether this zone is the source or sink for marine Nd, or has no noticeable influence on the seawater. Based on the Nd isotopes at the station near Hawaii, Amakawa et al. [28] found that although the ocean surface may be influenced by the Nd exchange of volcanic rocks in Hawaii, the deep water does not seem to be affected. (b) Arsouze et al. [32] used a biogeochemical ocean circulation model to simulate the Nd distribution in seawater. They used a pre-set shelf sediment flux as the boundary condition and found that the shelf sediments might account for 90% of the Nd input. Although the Nd isotopic compositions can be reproduced well, the concentrations of Nd are not satisfactorily simulated.

We suggest that the radiogenic NPIW [34] may be an important source for deep water Nd isotope distribution. With a reversible scavenging model [30], the Nd isotope distribution of the central North Pacific deep water can be readily explained as follows: NPIW transfers its Nd from the Northwest Pacific to the central North Pacific, and Nd is then brought down to the deep water through reversible adsorption-desorption of particulates. The Nd isotopic compositions of NPIW are not uniform ( $\epsilon_{\text{Nd}} = -5.3$  to  $-3.1$ , with an average of  $-3.6$  [28]), due to the mixing of many different water masses with different Nd isotopic compositions [34], such as the Kuroshio ( $-3.9$  to  $-5.6$ ), Oyashio ( $-1.6$  to  $-2.7$ ) and the Tsugaru warm currents [19,35], dur-

ing the formation process of the NPIW in the Northwest Pacific. However, the Nd isotopes of NPIW become more uniform during the advection process, which is consistent with the observation that the Nd isotopes of surface samples of crusts in the lower deeper water (e.g. <2500 m) are more uniform than in the upper deep water. Although it is still difficult to accurately calculate the flux of Nd carried by NPIW from the Northwest marginal Pacific, the following first order estimate shows that such flux is considerable: The estimated flux of newly formed NPIW is about  $6 \times 10^6$  m<sup>3</sup>/s in the mixed water region near Japan (Talley et al. [19]). If all the newly formed NPIW transport into the central North Pacific, with an Nd concentration of  $\sim 20$  pmol/ka [28], the Nd flux carried by NPIW would then be  $5.6 \times 10^8$  g/a, which is about twice the current global riverine flux ( $2.6 \times 10^8$  g (Nd)/a [32]). Therefore, we suggest that NPIW is an important source for Nd in deep water.

We can further calculate the Nd isotope composition of the central North Pacific deep water through a simple box model approach. As shown in Figure 5, if we consider only the contribution of Subarctic Pacific (north of 40°N) return flow, southern component water, and NPIW, without considering the contribution from surface and deep-sea volcanic ash dissolution, then at steady state, the Nd isotopic composition can be calculated as  $\epsilon_{\text{CNPDW}} = (F_{\text{sw}} \times \epsilon_{\text{sw}} + F_{\text{rf}} \times \epsilon_{\text{rf}} + F_{\text{NPIW}} \times \epsilon_{\text{NPIW}}) / (F_{\text{sw}} + F_{\text{rf}} + F_{\text{NPIW}})$ . The  $\epsilon_{\text{Nd}}$  value obtained is  $-4.1$ , which is close to the observed  $\epsilon_{\text{Nd}}$  average value ( $\sim -4.0$ ) of North Pacific deep water [12]. This further quantitatively supports the assumption that other radiogenic sources of Nd appear to be unimportant, while the Subarctic Pacific return flow and NPIW are sufficient to explain the radiogenic Nd isotope content for central North Pacific deep water.

Physical mixing can introduce some Nd into the deep water. However, according to the temperature-salinity diagram (Figure 2), it is clear the central North Pacific deep water mainly inherits the characteristics of the southern component water, whereas NPIW has little effect on modifying its temperature and salinity. Specifically, the temperature and salinity of central North Pacific deep water are very close to the AABW but far from the NPIW along the mixing line. Because the southern component water enters this area with an  $\epsilon_{\text{Nd}}$  value of about  $-5$  [28], while the  $\epsilon_{\text{Nd}}$  of the crust surface samples vary from  $-3$  to  $-4$ , with a  $-3.4$  average and a slightly increase in  $\epsilon_{\text{Nd}}$  values downwards (Figure 4), the deep water should have received significant contributions of radiogenic Nd from other sources (NPIW). Therefore, Nd in the North Pacific deep water cannot be conservative. In fact, reversible scavenging of dissolved Nd may be the main mechanism that removes Nd from seawater (e.g. [13,14,37]). This is demonstrated by the fact that the concentration of seawater Nd increases with depth. For example, in the North Pacific, Nd concentrations increase from  $\sim 5$  pmol/kg in the surface water to  $\sim 30$  pmol/kg in deep water. This concentration profile supports the theory



**Figure 5** Illustration of the Nd budget (south of 40°N) in central North Pacific deep water (1500–3500 m). (a) Nd released from the sinking particles of the NPIW should contain the surface ocean flux. Such a surface flux is categorized in (b). Here only the Nd flux from the NPIW source area is taken into account.  $F_p = 5.6 \times 10^8$  g/a (see calculation in the text); Nd isotopic composition:  $\epsilon_p = -3.6$  [28]. (b) Dissolution of dust, volcanic ash, and input from central North Pacific surface water are included. (c) Subarctic North Pacific deep water return flux is about  $4 \times 10^6$  m<sup>3</sup>/s [24]. Nd concentration was set to be the same as the central North Pacific deep water ( $\sim 30$  pmol/kg [28]). Thus the Nd flux  $F_{tr}$  is approximately  $5.6 \times 10^8$  g/a. Nd isotopic composition ( $\epsilon_{tr}$ ) of crust surface samples of this area is  $\sim -2.1$  [36], which may be higher than that of the present ocean levels. Based on data from the seawater of the subarctic Pacific station [28], we choose  $\epsilon_{tr}$  as  $-3.0$ . (d) The modified southern component water flux to the central North Pacific is about  $8 \times 10^6$  m<sup>3</sup>/s [36]. Our southern component water of interest is actually UCDW ( $< 3500$  m), which flows along the western Pacific [24]. Therefore, we set the concentration of Nd in the southern component water to be the same as the central North Pacific deep water ( $\sim 30$  pmol/kg [28]). Then the Nd flux  $F_{tr}$  is about  $11 \times 10^8$  g/a. Nd isotopic composition  $\epsilon_{sw}$  is set to be  $-5.0$  (seawater:  $-4$  to  $-5$  [28], crust surface samples (4800 m) are about  $-4.6$  [3]). (e), (f) and (g) They are considered as the sinks of the North Pacific deep water Nd. In steady state, we can suppose that their Nd isotopic compositions are the same as the central North Pacific deep water ( $-4.0$ , [12]), and that the total output flux is equal to the total input ( $F_s = F_{tr} + F_{bw} + F_p$ ).

that marine Nd is strongly linked to biogeochemical cycles [14]. Particles formed in the surface water would sink and be re-mineralized in the deep water, during which most of the adsorbed Nd would be released to or exchanged within the deep water. Oka et al. [14] studied the distribution of rare earth elements in the North Pacific and proposed that only when these elements (including Nd) have a reversible adsorption and desorption behavior on the particles, can their concentration profiles be correctly reproduced by simulation. In summary, we suggest that Nd from NPIW is introduced into deep water mainly through reversible scavenging.

Another question that needs to be further discussed is why the Nd isotope signal of NPIW remains stable during its transport in the North Pacific and is not affected by Nd addition from the overlying water. Since the surface water is mainly controlled by riverine and dust input, the flux from the marginal to oligotrophic open ocean surface would be largely limited because a large part of Nd would be absorbed by strong biological activity in the shelf area. The downward flux from the North Pacific gyre surface water would then be small. Moreover, dust directly supplying the central North Pacific (mainly from western China and central Asia [38,39]) has very un-radiogenic Nd ( $\epsilon_{Nd}$  around

$-10$  [39]), and obviously cannot be the source for the radiogenic Nd in deep water. Our calculations above show that Nd from NPIW could be twice that of the global riverine input. Therefore, as a rapid ventilating water mass, NPIW [26] could carry enough Nd from its source region in the Northwest Pacific near Japan to the central North Pacific without being significantly modified by Nd desorbed from the particles sinking from the surface.

### 3.3 Implications for marine Nd cycling

The above discussion shows that NPIW is a major source that transports Nd to the deep central North Pacific. Together with the Subarctic Pacific return flow, it could account for the Nd isotopic distribution in the central North Pacific deep water. This improves our previous understanding about the path and transport of Nd in the central North Pacific, where the traditional view is that central North Pacific surface waters may significantly influence the Nd isotopes of deep water [30]. NPIW forms in the western North Pacific margin, and its own Nd sources may include rivers, dust and shelf sediment-water exchange. Accordingly, it may not be necessary to introduce a widely distributed deep sediment-seawater exchange mechanism [32] for the central North Pacific deep water Nd. However, as noted above, the amount of Nd carried by NPIW has to be further constrained quantitatively, and attention should be paid to such an input mechanism in future studies. We suggest that further modeling of Nd distribution in North Pacific Deep Water could try to incorporate Nd isotopes of NPIW as the boundary condition.

### 3.4 Implications of the difference in Nd isotopes between crust surfaces and modern/Cenozoic records

As shown in Figure 3, the  $\epsilon_{Nd}$  of crust surface samples slowly increases as depth reduces from 3500 to 1500 m, which is consistent with the present pattern of the vertical distribution of Nd isotopes in the North Pacific. The overall variation of  $\epsilon_{Nd}$  values is  $\sim 0.6$  units, however, much less variation ( $\sim 0.3$  units) is found near 180° at below 2000 m. However, Ling et al. [40] reported larger than 0.6 units difference in Nd isotope composition between crusts CB12 ( $\sim 2400$  m) and CJ01 ( $\sim 3100$  m) in the same area before the Quaternary. Our recently obtained Nd isotopic records from two crust profiles (unpublished data) near 150°E (MKD13, MDD53, see Figure 1(b)) also indicate that  $\epsilon_{Nd}$  differences could be larger than 1 unit in the early Pliocene deep water. This suggests that vertical homogenization of Nd isotopes may have been strengthened in the central North Pacific deep water in the near past.

It is also noteworthy that Nd isotopes of the crust surface samples (average value of  $-3.4$  in this study) are apparently more radiogenic than those for the present ambient seawater ( $-3.9$  [28]). Nd residence time in the deep sea is about several

hundreds of years [41], so the modern deep water Nd isotopes only reflect the integration of a few centuries. In contrast, the ferromanganese crusts grow very slowly, usually only 1–3 mm/Ma in the North Pacific deep water (e.g. [42]). Although the composition of ferromanganese crusts could well reflect the influence of orbital-scale changes in ocean currents and climate (see the *in situ* high resolution studies of Han et al. on ferromanganese nodules [43] and crusts [44]), the data obtained in this study represent an integration of Nd isotopes over a relatively longer time (but still less than 1 Ma, because the sampling depth is less than 1 mm) [31]. Therefore, apart from the previously discovered long-term Nd isotopic changes on a million year scale [3,40], it is very likely that Nd isotopes also vary over a very short time scale (within the last 1 Ma) in the central North Pacific. Because the samples may well contain an integration of several glacial-interglacial cycles [31], one possible reason for the discrepancy between seawater and crust surfaces is that the North Pacific deep water in glacial stages is more radiogenic than the present interglacial times. It is also possible that Nd isotopic variations in the North Pacific deep water may not be cyclic, so the exact cause of such inconsistency remains to be clearly understood. Up to now, no Nd isotope data have been published over orbital or millennium time scales in the studied area, which certainly deserves future research.

#### 4 Conclusions

In this study, the Nd isotope compositions of crusts surface samples from the central North Pacific deep water have been reported. In combination with published Nd isotope data of modern seawater, we draw the following conclusions:

(1) The North Pacific Intermediate Water (NPIW) formed in the Northwest Pacific carries radiogenic Nd from the source area to the central North Pacific. This is probably an important way for the central North Pacific deep water to acquire their radiogenic Nd isotopes, while the central North Pacific surface water may have little influence on the deep water Nd isotopes.

(2) The marginal Nd cycling and input to the deep water may be important in understanding the oceanic Nd budget, which deserves more attention in future studies, especially in modeling studies.

(3) The crust surface samples have more radiogenic Nd isotopes than ambient modern seawater, indicating that Nd isotopic compositions are not stable over relatively short time scales in the central North Pacific.

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