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Nd concentration and isotopic composition distributions in surface waters of Northwest Pacific Ocean and its adjacent seas

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Nd isotopic composition and concentration distributions were determined in surface waters of the Northwest Pacific Ocean and its adjacent seas, i.e., the Okhotsk and Japan Seas. The Okhotsk and Japan Seas samples showed higher Nd concentrations (19~31 pmol/kg) than the Northwest Pacific Ocean samples, suggesting a large amount of Nd input from the land areas surrounding the Okhotsk and Japan Seas. Although the Nd isotopic composition data were limited for those regions, each oceanic area showed a distinctive value. The lone sample from the Okhotsk Sea had radiogenic Nd (ε_{Nd} = –3.6) that was comparable to samples from the Pacific Ocean, indicating a large amount of radiogenic Nd supply from the Kuril Islands. On the other hand, the Japan Sea samples had the most highly unradiogenic Nd (ε_{Nd} = –8.9~–7.2). The southernmost sample had the most negative value, due to the influence of the Tsushima Warm Current (TWC) having a low ε_{Nd} value. The latitudinal distributions of Nd isotopic composition and concentration in the surface waters of the Northwest Pacific Ocean showed marked variations. Nd concentration was low at low latitudes (3~8 pmol/kg) and was increased with increasing latitude (16~20 pmol/kg). On the other hand, Nd isotopic composition showed a mid-latitude minimum (ε_{Nd} = ~–6) and high ε_{Nd} values at low and high latitudes (ε_{Nd} = –2~–1). The distribution of the ε_{Nd} values seemed to be controlled by three currents: Kuroshio Extension, Oyashio Current and North Equatorial Current (NEC). The Kuroshio Extension, which showed a low ε_{Nd} value, seemed to play an important role in transporting continentally derived Nd to the central Pacific. The Oyashio Current and NEC are supplied with large amounts of radiogenic Nd from the Kuril and Aleutian Islands, and the Hawaiian Islands, respectively.

Based on the Nd isotopic composition and concentration distributions, we calculated the radiogenic Nd flux to the Pacific Ocean surface waters and estimated the Nd residence time. Our calculation suggested that the radiogenic Nd flux required to account for the Nd isotopic composition is higher than 70% of the unradiogenic Nd flux including Nd remobilization from coastal and shelf sediments and atmospheric input. It was also revealed that a large Nd remobilization flux is necessary to attain a global Nd residence time of ~400 y as estimated in previous studies.

Keywords: Nd isotopic composition, Nd concentration, surface water, North west Pacific Ocean, current

INTRODUCTION

Nd isotopic composition, one of the most versatile isotopic tracers, is utilized in such diverse fields as marine chemistry, cosmochemistry, and petrology. The Nd isotopic composition is frequently used for identifying water masses (Piepgras and Wasserburg, 1982, 1987; Spivack and Wasserburg, 1988; Jeandel, 1993; Bertram and Elderfield, 1993; Jeandel *et al*., 1995, 1998; Tachikawa *et al*., 1999; Lacan and Jeandel, 2001; Amakawa *et al*., 2004), because, compared with the rare earth elements (REE) concentration of seawater, it is less susceptible to evaporation or scavenging and more faithfully keeps its original signal.

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The Nd isotopic composition is generally expressed as:

$$\varepsilon_{Nd} = \left(\frac{\left(^{143}Nd/^{144}Nd\right)_{measured}}{\left(^{143}Nd/^{144}Nd\right)_{CHUR}} - 1 \right) \times 10^4 \qquad (1)$$

where CHUR stands for Chondritic Uniform Reservoir, the present-day CHUR value being 0.512638 (Wasserburg *et al*., 1981). The Nd isotopic compositions ($^{143}Nd/^{144}Nd$) of terrestrial rocks vary because of their initial Sm/Nd ratios and their formation or metamorphic ages. If the Nd isotopic growth curve of a rock were identical with CHUR, the ε value would be 0. A rock with time-integrated $(Sm/Nd)_N$ (normalized value of the CHUR Sm/Nd ratio) lower than 1 would give a negative ε value and

vice versa. Typical rocks for the former are continental crustal rocks with $\varepsilon_{Nd} = -30 \sim -10$ and those for the latter are mantle-derived rocks, such as Mid Ocean Ridge Basalts (MORB), Island Arc Basalts (IAB) and Ocean Island Basalts (OIB), with $\varepsilon_{Nd} = 0 \sim +10$. The Nd in the rocks is transported to the ocean by various processes and mixed there. The relative contribution of continental crustal materials and mantle-derived materials is revealed by the determination of Nd isotopic composition.

Based on the Nd isotopic compositions of surface seawaters in the East Indian Ocean, Amakawa *et al*. (2000) pointed out the predominance of fluvial and coastal input over eolian input of dissolved Nd in the surface water. They further estimated the Nd residence time for the entire ocean. Lacan and Jeandel (2001) reported the Nd isotopic compositions of waters sampled from several Equatorial Pacific stations and suggested that radiogenic Nd is supplied from the Papua New Guinea slope. Recently, Amakawa *et al*. (2004) found that the North Pacific Intermediate Water (NPIW) and the North Pacific Tropical Water (NPTW) collected at stations close to the Japanese Islands show distinctive isotopic signals. They also pointed out that the Kuroshio and Oyashio Currents show distinctive Nd isotopic compositions. However, detailed circulation patterns including those for the surface currents in the Northwest Pacific are yet to be elucidated.

The objectives of this study are to determine the Nd concentration and isotopic composition distributions in surface waters collected from the Northwest Pacific Ocean and its adjacent seas, i.e., the Okhotsk and Japan Seas, and to elucidate the factors controlling such distributions. We also discuss the Nd budget in the Pacific Ocean.

METHODS

Unfiltered surface seawaters were collected along the tracks of the Canis Minor Expedition (KH-98-3, July 15, 1998 to August 14, 1998) and the Bootes Expedition (KH-00-3, June 20, 2000 to July 27, 2000) on R.V. Hakuho-Maru. For Nd isotopic analysis, approximately 300 L of seawater was collected and transferred by means of a built-in pump system from an inlet located at the ship bottom (~5 m below sea level) to a plastic container on the ship deck.

The procedure for the chemical separation of Nd was identical with that described in Amakawa *et al.* (2004). Nd isotopic composition was determined with a thermal ionization mass spectrometer (Finnigan MAT262). Nd isotopes were measured as Nd^+ using a Re double filament assembly. The measured Nd isotopic ratios were normalized to $^{146}Nd/^{144}Nd = 0.7219$. During the Nd isotopic measurements, the isotopic ratios measured for the standards, i.e., La Jolla and JNdi-1 (supplied by the Geological Survey of Japan), were 0.511856 ± 0.000007 (2σ, $n = 9$) and 0.512112 ± 0.000006 (2σ, $n = 29$), respectively, and were in good agreement with those reported by O'Nions *et al*. (1977) and Tanaka *et al*. (2000). As the contribution of blank to each sample was less than 1%, no blank correction was performed.

For Nd concentration measurement, surface seawaters were also collected *via* the inlet at the ship bottom. Immediately after collection, the seawaters were filtered on board through a 0.04 μm hollow-fiber membrane (Millipore HF-400) in a built-in clean room of the vessel.

The filtered samples were transferred into pre-cleaned 5-L plastic cubic containers and then acidified to pH < 1.5 with ultra-pure HCl (TAMAPURE AA-100). The water samples were brought to the Ocean Research Institute, University of Tokyo. The concentrations of REE, including Nd, were measured with an ICP mass spectrometer (YOKOGAWA PMS-2000) according to the procedures described in Zhang and Nozaki (1996) and Alibo and Nozaki (1999).

RESULTS

Hydrography and sampling locations

The hydrographic data for the occupied stations are given in Tables 1 and 2. We focused on the stations for Nd isotopic analysis.

Along the west coast of the Japanese Islands, a warm high-salinity current called "Kuroshio" and a cold low-salinity current called "Oyashio" flow northward and southward, respectively (Fig. 1). The Kuroshio Current is a western boundary current similar to the Gulf Stream

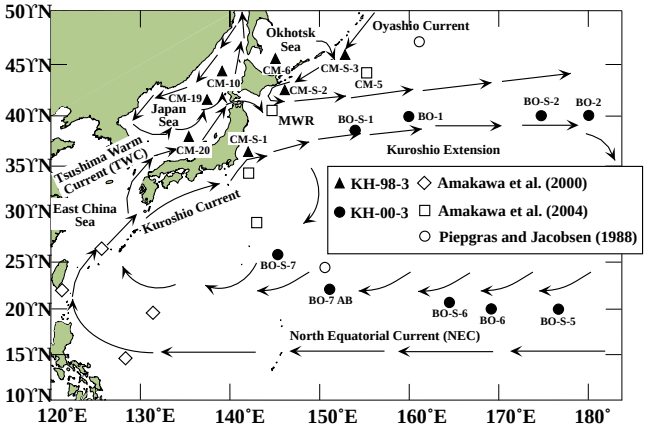

Fig. 1. Map showing sampling stations for Nd isotopic composition measurement (closed triangles and circles) and surface current patterns in the Northwest Pacific Ocean and its adjacent seas. The previously studied locations (open diamonds, squares and circles) for Nd isotopic composition measurement (see Fig. 2) are also shown.

in the Atlantic, and plays an important role in transporting heat from low to high latitudes.

The origin of the Kuroshio Current is traced back to the North Equatorial Current (NEC) that flows westward at 10°N~15°N. Five sampling stations (BO-S-5, BO-6, BO-S-6 and BO-7 (duplicate)) located around 20°N are north of the NEC pathway. Their locations should be regarded as within the Subtropical Gyre, as their temperature (T > 25°C) and salinity (S > 34.9) are similar to those of previously studied locations within the Subtropical Gyre, i.e., TPS 24 (Piepgras and Jacobsen, 1988) and LM2 (Amakawa *et al.*, 2004). Station BO-S-7 is also within the Subtropical Gyre.

The NEC makes a turn near the Philippine Islands, where the Kuroshio is considered to originate (Nitani, 1972). The Kuroshio Current flows at the east boundary of the East China Sea along the Ryukyu Islands, and separates into two at the east of the Kyushu Island. One flows further along the east coast of the Honshu Island, the main part of the Japanese Islands, and the other flows into the Japan Sea through the Tsushima Strait, and is called the Tsushima Warm Current (TWC, Lie *et al.*, 1998). The former meets with the Oyashio Current at the Mixed Water Region (MWR) centered around ~40°N, which is famous for its good fisheries industry (station CM-S-2 is located within the MWR). Subsequently, it flows to the central North Pacific, which is called the Kuroshio Extension. The four samples collected around ~40°N (BO-S-1, BO-1, BO-S-2 and BO-2) were in between the Kuroshio Extension and the Oyashio Current circulated along the Subarctic Gyre. The latter two samples showed lower salinity and temperature than the former two samples, probably because of the relatively large contribution of the Oyashio Current compared to that of the Kuroshio Extension in the latter two samples. The water sampled from stations CM-S-3 and CM-5 showed very low salinity (S = 32.6, 32.9), indicating the regime of the Oyashio Current.

As mentioned above, TWC flows into the Japan Sea through the Tsushima Strait. Lie and Cho (1994) reported that water in the Tsushima Strait has very low salinity (S < 32.5) in summer, and pointed out the influence of fresh water from the southern coast of Korea and the Changjiang River. The water sampled from the southernmost station, CM-20, in the summer of 1998 showed the lowest salinity (S = 32.9) among the samples

Table 1. Nd isotopic compositions in surface waters (KH-98-3 and KH-00-3)

Sample code	Location		Region	Salinity	Temp.	$^{143}Nd/^{144}Nd$	ε_{Nd}	Nd#
	Latitude	Longitude		(psu)	(°C)			(pmol/kg)
KH-98-3								
CM-S-1	36°13′ N	141°55′ E	Pacific Ocean	34.34	24.0	0.512351 ± 0.000011	−5.6 ± 0.2	(13.0)
CM-S-2	42°23′ N	145°50′ E	Pacific Ocean	32.63	14.3	0.512481 ± 0.000012	−3.1 ± 0.2	(16.1)
CM-S-3	45°43′ N	152°42′ E	Pacific Ocean	32.94	8.8	0.512527 ± 0.000011	−2.2 ± 0.2	(15.1)
CM-5*	44°01′ N	155°00′ E	Pacific Ocean	32.58	13.1	0.512494 ± 0.000013	−2.8 ± 0.3	15.2
CM-6	45°25′ N	145°04′ E	Okhotsk Sea	32.38	12.6	0.512455 ± 0.000010	−3.6 ± 0.2	26.5
CM-10	44°10′ N	139°00′ E	Japan Sea	33.83	18.4	0.512258 ± 0.000009	−7.4 ± 0.2	29.4
CM-19	41°21′ N	137°20′ E	Japan Sea	33.70	22.4	0.512269 ± 0.000008	−7.2 ± 0.2	23.5
CM-20	37°44′ N	135°14′ E	Japan Sea	32.90	26.4	0.512184 ± 0.000011	−8.9 ± 0.2	31.4
KH-00-3								
BO-S-1	38°26′ N	153°47′ E	Pacific Ocean	34.15	19.6	0.512384 ± 0.000015	−5.0 ± 0.3	(6.7)
BO-1	39°57′ N	159°57′ E	Pacific Ocean	34.27	14.5	0.512406 ± 0.000012	−4.5 ± 0.2	—
BO-S-2	40°00′ N	174°34′ E	Pacific Ocean	33.85	12.5	0.512436 ± 0.000011	−3.9 ± 0.2	(8.9)
BO-2	40°00′ N	180°00′ E	Pacific Ocean	34.00	12.7	0.512428 ± 0.000014	−4.1 ± 0.3	(8.3)
BO-S-5	20°00′ N	176°26′ E	Pacific Ocean	35.25	28.4	0.512556 ± 0.000015	−1.6 ± 0.3	(6.3)
BO-6	20°00′ N	169°00′ E	Pacific Ocean	35.05	28.9	0.512553 ± 0.000018	−1.7 ± 0.3	(6.8)
BO-S-6	20°34′ N	163°57′ E	Pacific Ocean	35.30	28.6	0.512555 ± 0.000013	−1.6 ± 0.3	(6.2)
BO-7 A	22°00′ N	151°00′ E	Pacific Ocean	35.05	29.3	0.512498 ± 0.000014	−2.7 ± 0.3	(5.8)
BO-7 B	22°00′ N	151°00′ E	Pacific Ocean	35.05	29.3	0.512509 ± 0.000029	−2.5 ± 0.6	(5.8)
BO-S-7	26°10′ N	145°03′ E	Pacific Ocean	34.90	28.0	0.512435 ± 0.000015	−4.0 ± 0.3	(6.0)

Errors are $2\sigma_m$.

**Amakawa et al. (2004).*

#The data are from Table 2 (CM samples) and Hongo et al. (submitted, BO samples). Note that the Nd concentration data are not reported for all the stations. The values in parentheses are data from the nearest stations where Nd concentration data are available (within 250 km, see Table 2 and Fig. 3).

Table 2. Dissolved Nd concentration (pmol/kg) in surface waters (KH-98-3)

Sample code	Location		Region	Salinity	Temp.	Nd
	Latitude	Longitude		(psu)	(°C)	
KH-98-3						
CM SSW-1	37°06′ N	142°28′ E	Pacific Ocean	34.05	22.3	13.0
CM SSW-3	42°22′ N	145°50′ E	Pacific Ocean	32.65	13.5	16.1
CM SSW-4	43°22′ N	151°20′ E	Pacific Ocean	32.49	11.6	19.0
CM SSW-5	43°39′ N	152°58′ E	Pacific Ocean	32.57	11.4	15.3
CM-5*	44°01′ N	155°00′ E	Pacific Ocean	32.58	13.1	15.2
CM SSW-6	46°09′ N	152°05′ E	Pacific Ocean	32.70	10.5	15.1
CM SSW-17	40°28′ N	144°30′ E	Pacific Ocean	33.07	21.9	14.0
CM SSW-7	45°44′ N	146°14′ E	Okhotsk Sea	32.32	12.8	22.1
CM-6	45°25′ N	145°04′ E	Okhotsk Sea	32.38	12.6	26.5
CM SSW-8	44°45′ N	145°10′ E	Okhotsk Sea	32.70	13.2	26.6
CM SSW-9	44°31′ N	145°00′ E	Okhotsk Sea	32.49	15.5	28.5
CM SSW-10	45°38′ N	141°38′ E	Soya Strait	33.74	16.7	24.3
CM-10	44°10′ N	139°00′ E	Japan Sea	33.83	18.4	29.4
CM SSW-12	43°16′ N	139°31′ E	Japan Sea	33.79	23.1	25.6
CM-18	42°43′ N	138°13′ E	Japan Sea	33.94	22.6	24.5
CM-19	41°21′ N	137°20′ E	Japan Sea	33.70	22.4	23.5
CM SSW-14	39°29′ N	135°03′ E	Japan Sea	33.67	23.6	23.1
CM-20	37°44′ N	135°14′ E	Japan Sea	32.90	26.4	31.4
CM SSW-15	40°17′ N	138°46′ E	Japan Sea	33.68	25.3	21.9
CM SSW-16	41°20′ N	140°18′ E	Tsugaru Strait	33.51	24.4	19.0

**Amakawa et al. (2004).*

collected from the Japan Sea including those for Nd concentration determination. Considering both salinity and location, station CM-20 was much more strongly influenced by TWC than stations CM-10 and CM-19.

The Okhotsk Sea has been intensively studied in terms of the formation mechanism of NPIW (Talley, 1993). Based on the Nd isotopic composition, Amakawa *et al*. (2004) discussed the formation mechanism of NPIW. The surface water of the Okhotsk Sea actively exchanges with the Pacific seawater. By contrast, there is little water exchange between the Okhotsk Sea and the Japan Sea (Preller and Hogan, 1998). The low salinity of the water sampled at station CM-6 (S = 32.4) is indicative of the large input of fresh water.

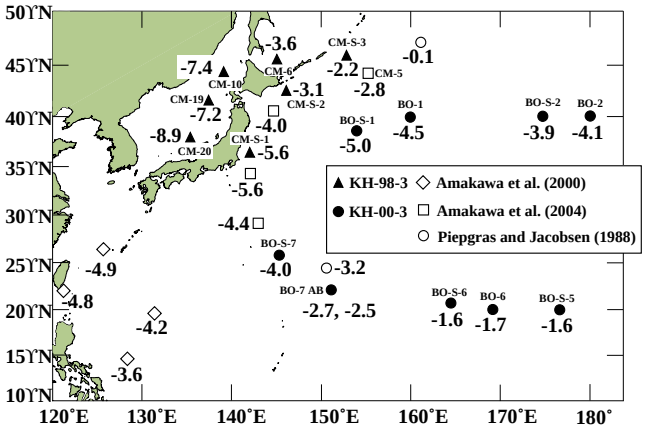

Fig. 2. Distribution of ε_{Nd} values in surface waters.

Nd isotopic composition and concentration

The Nd isotopic composition and concentration data are given in Tables 1 and 2, respectively. Not all stations have both Nd isotopic composition and concentration data because the water samples for those two determinations were separately collected. To this end, for the stations with no Nd concentration data, the Nd concentrations from the nearest stations (within ~250 km) are shown as surrogates in Table 1.

The Nd concentrations reflect those of dissolved Nd, being free from particulate matter due to the filtration. On the other hand, the unfiltered samples were used for determining Nd isotopic compositions. In hemipelagic and open oceans, particulate Nd is less than 5% of total Nd (Alibo and Nozaki, 1999). We believe that the Nd isotopic compositions of the samples from the Pacific Ocean are almost identical with the "dissolved" ones, whereas

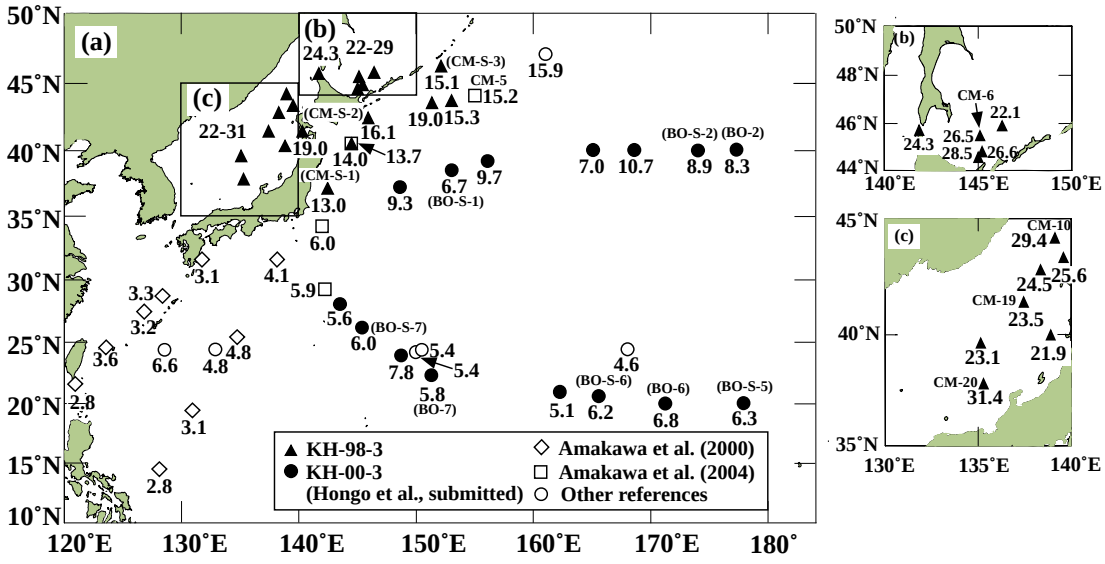

Fig. 3. (a) Nd concentration distribution (pmol/kg) in surface waters. The Okhotsk Sea and the Japan Sea are enlarged in (b) and (c), respectively. The stations where Nd isotopic compositions were analyzed are indicated. The stations in parentheses indicate the nearest stations where Nd isotopic composition data are available (see Table 1).

particulate Nd may significantly contribute to the Nd isotopic compositions of the samples from such semi-closed basins as the Okhotsk and Japan Seas. We surmise that the Nd isotopic data of those regions reflect both dissolved and particulate Nd.

To check the reproducibility of the Nd isotopic composition determination, we collected two samples at station BO-7 (A and B). The results showed good agreement with each other within $2\sigma_m$ error (see Table 1).

The Nd isotopic composition distribution is shown in Fig. 2 together with previously reported data (Piepgras and Jacobsen, 1988; Amakawa *et al*., 2000, 2004). The Nd isotopic compositions differed among the oceanic regions. The Japan Sea samples had the most highly unradiogenic Nd (ε_{Nd} = –8.9 to –7.2). The lone sample from the Okhotsk Sea had relatively less radiogenic Nd than a sample from the Pacific Ocean close to the Kuril Islands (ε_{Nd} = –2.2). It should be noted that the Northwest Pacific Ocean samples showed a very wide range of ε_{Nd} values (–5.6~–0.1). At the latitude around 20°N and above 40°N, most of the samples showed ε_{Nd} values higher than –3, whereas the samples collected from the latitude between 25°N and 40°N showed ε_{Nd} values lower than –4. Within those latitudes, a systematic change in ε_{Nd} values was observed in both our data and the previously reported ones. This feature will be discussed later. Although the number of data was limited, some heterogeneity in ε_{Nd} value was observed for the Japan Sea samples.

The Nd concentration distribution is shown in Fig. 3, together with the reported and unpublished data of the Northwest Pacific Ocean near the Japanese Islands (Piepgras and Jacobsen, 1988; Greaves *et al*., 1999; Amakawa *et al*., 2000, 2004; Hongo *et al*., submitted). Good agreement was observed between our data and the previously reported data at one location, i.e., 40°30′ N, 144°30′ E. It was revealed that the Okhotsk and Japan Seas showed almost the same range of Nd concentrations. In the Pacific Ocean, the data showed a systematic increase from south (3~8 pmol/kg) to north (16~20 pmol/kg).

DISCUSSION

Nd concentration and isotopic composition distributions in semi-closed oceanic basins, the Okhotsk and Japan Seas

The Okhotsk and Japan Seas are semi-closed oceanic basins surrounding the Japanese Islands. The Nd concentration and isotopic composition distributions in the surface waters from semi-closed oceanic basins have been reported by several researchers.

Andersson *et al*. (1992) reported very high Nd concentrations (>30 pmol/kg) and low ε_{Nd} values (<–14) for surface waters collected from the inner regions of the Baltic Sea that was surrounded by very old rocks having ε_{Nd} < –20. Henry *et al*. (1994) reported Nd concentrations and isotopic compositions of surface waters from the Mediterranean Sea. Combined with the data of Spivack and Wasserburg (1988), they discussed surface water transport in the Mediterranean Sea. More recently, Amakawa *et al*. (2000) reported Nd concentrations and isotopic compositions of surface waters from the Sulu and

South China Seas. Although those oceanic areas did not show distinctive Nd concentrations compared with the open ocean, their Nd isotopic compositions showed characteristic values. The Sulu Sea sample showed an ε_{Nd} of –1.3, which was indicative of a supply of radiogenic Nd from surrounding young volcanic rocks. On the other hand, the South China Sea samples showed ε_{Nd} values ranging from –9.5 to –6.4, suggesting a greater contribution of continentally derived Nd than that of the Sulu Sea. Taken together, the above observations suggest that the Nd isotopic compositions of surface waters in semi-closed basins are strongly influenced by the surrounding rock type.

The Okhotsk and Japan Seas surface water samples showed relatively high Nd concentrations (>20 pmol/kg) compared with the Northwest Pacific Ocean samples. As for the Japan Sea, the high Nd concentrations suggest the strong influence of fluvial/coastal inputs because the surface waters have very high ^{228}Ra concentrations (76~164 dpm/L; Nozaki *et al*., 1998) that indicate a large supply from coastal areas. The low salinity of Okhotsk Sea samples (S = 32.3~32.7) suggests a large riverine input from land areas, which may result in the high Nd concentrations of those samples.

The difference in Nd isotopic composition among those seas is worth noting, even if the isotopic composition may reflect both dissolved and particulate Nd due to the absence of filtration. The lone Okhotsk sample shows much more radiogenic Nd than the Japan Sea samples, indicating the strong influence of the volcanic islands, i.e., the Kuril Islands (ε_{Nd} = +6.7~+10.1, McCulloch and Perfit, 1981; Zhuravlev *et al*., 1987; Amakawa *et al*., 2004). The radiogenic Nd isotopic composition of the Pacific Ocean surface water near the Kuril Islands (CM-S-3, ε_{Nd} = –2.2) supports this idea.

Two Japan Sea samples collected from stations located at latitudes higher than 40°N, CM-10 and CM-19, showed almost identical ε_{Nd} values, whereas one sample collected from a station located at a latitude of ~38°N, CM-20, showed a slightly lower value (ε_{Nd} = –8.9). The latter sample is located more closely to the TWC pathway than the former two samples. As shown in Fig. 1, one branch of the Kuroshio Current flows along the west of the Kyushu Island, the southern part of the Japanese Islands, and enters the Japan Sea through the Tsushima Strait. TWC may be supplied with unradiogenic Nd during its passage through the Strait, because the Korean Peninsula is composed of very old rocks with low ε_{Nd} values (Lan *et al*., 1995). Furthermore, in summer, TWC has very low salinity due to the supply of fresh water from the Changjiang River (Lie and Cho, 1994), which may have a low ε_{Nd} value. These facts suggest that TWC may have supplied Nd with low ε_{Nd} value to water sampled at station CM-20.

It should be noted that although the surface waters at stations CM-19 and CM-20 showed different ε_{Nd} values, the deep waters below 500 m at both stations showed almost the same ε_{Nd} values of approximately –7 (Amakawa *et al*., 1999). Furthermore, it has been reported that the ^{226}Ra concentrations in waters below 500 m collected at various stations in the Japan Sea are very uniform (Harada and Tsunogai, 1986), indicating an active mixing of the deep waters of the Japan Sea. Although the number of data is limited, most of the Japan Sea water samples would show a uniform ε_{Nd} value of approximately –7. We think that the low ε_{Nd} value observed at station CM-20 is not ubiquitous in the Japan Sea.

Assuming that the ε_{Nd} value of the entire Japan Sea water is –7 and explaining it with a simple two-component mixing model, i.e., the continental source and the mantle source, the fraction of the continentally derived Nd can be calculated. We employ ε_{Nd} = –11.2 (Liu *et al*., 1994) as the continental end member and ε_{Nd} = +0.1, the only datum of a Japanese river that flows into the Japan Sea (Goldstein and Jacobsen, 1987), as the mantle source. The calculated fraction of the continental source is approximately 63%, which is only nearly twice as large as the fraction of the mantle source (37%). Amakawa *et al*. (2000) found that the ε_{Nd} values (less than –11) of surface waters from the Gulf of Bengal indicated the continental source fraction to be almost 100%. Considering the location of the Japan Sea, which is in the vicinity of the Asian continent, the fraction of the continental source for the Japan Sea seems to be small. The influence of Japanese coastal sediments with a radiogenic ε_{Nd} signal may play an important role in supplying Nd to the Japan Sea. However, to well elucidate the contribution of the Japanese coastal sediments to the Nd budget in the Japan Sea, we need more data on the Nd isotopic compositions of Japanese rivers and the coastal sediments around the Japan Sea. It is also necessary to determine the contribution of particulate matter to the Nd isotopic composition in the Japan Sea.

Nd isotopic composition distribution in Northwest Pacific Ocean surface water

^{228}Ra, a naturally occurring nuclide (half-life, 5.76 y), is regarded as a useful tracer for monitoring the lateral transport of elements from the continental shelf to the open ocean. The main sources of ^{228}Ra are estuarine, coastal and shelf sediments, where ^{228}Ra is produced by the α decay of ^{232}Th. The supplied ^{228}Ra spreads over the ocean surface waters by lateral mixing and decays with transport, resulting in high concentrations in the coastal region and low concentrations in the central oceanic region. Nozaki *et al*. (1998) compiled the distribution of ^{228}Ra in the Northern Hemisphere. ^{228}Ra data in the central Pacific Ocean is limited, partly because of its very

low concentration. In such regions, other chemical tracers are required to well elucidate the mechanism of the lateral transport of elements from the coastal regions. Nd isotopic composition is one candidate alternative for ^{228}Ra, because the minimum surface water volume required for the determination of Nd isotopic composition (~50 L) is fivefold lower than that for ^{228}Ra.

There are several pathways for supplying Nd to the ocean, such as fluvial/coastal, eolian and hydrothermal inputs. In ocean surface waters, as hydrothermal input is negligible, the first two inputs are perceived to mainly control Nd concentration and isotopic composition. Previous studies have revealed that Nd remobilization from coastal and shelf sediments is a much more important Nd source than atmospheric input (Amakawa *et al*., 2000, 2004; Sholkovitz and Szymczak, 2000; Lacan and Jeandel, 2001). Amakawa *et al*. (2004) studied Nd concentration and isotopic composition distributions in the surface waters of the Northwest Pacific Ocean near the Japanese Islands and concluded that the eolian input is relatively minor in the surface water. However, as the stations employed in the study of Amakawa *et al*. (2004) were located only in the vicinity of the Japanese Islands, it should be verified whether that conclusion could be extended to the central North Pacific region.

We plot our data together with reported and unpublished North Pacific data between 125°E and 180°E against latitude (Fig. 4). For comparison, the Nd isotopic data from the other North Pacific regions are also shown (Lacan and Jeandel, 2001). Both Nd concentration and isotopic composition showed trends similar to those of the oceanic region near the Japanese Islands presented recently by Amakawa *et al*. (2004). Although our Nd concentration data show a slight scatter (Fig. 4(a)), they show a common trend with data from the other studies, i.e., an increase with increasing latitude. As suggested by Amakawa *et al*. (2004), this increase with increasing latitude indicates a small influence of atmospheric input, because this trend is quite different from that of ^{210}Pb distribution having a mid-latitude maximum (Nozaki *et al*., 1976), which is a good indicator of atmospheric flux.

It should be noted that the "V-shaped" distribution of Nd isotopic composition (Fig. 4(b)), i.e., a mid-latitude minimum with gradual increases toward both high and low latitudes, is clearly distinguishable, even if samples from the central Pacific are included. This implies that the observed mid-latitude minimum is a feature common to both the Northwest and the Northcentral Pacific. On the other hand, the data of Amakawa *et al*. (2000) and Lacan and Jeandel (2001) did not follow the "V-shaped" distribution. This may be due to differences in the sample location and the surface current pattern. The samples collected between 128°E and 132°E were close to the continent and subject to unradiogenic Nd from the continental margins, whereas those collected from 140°W were within the South Equatorial Current and were not susceptible to the surface currents in the North Pacific, such as NEC.

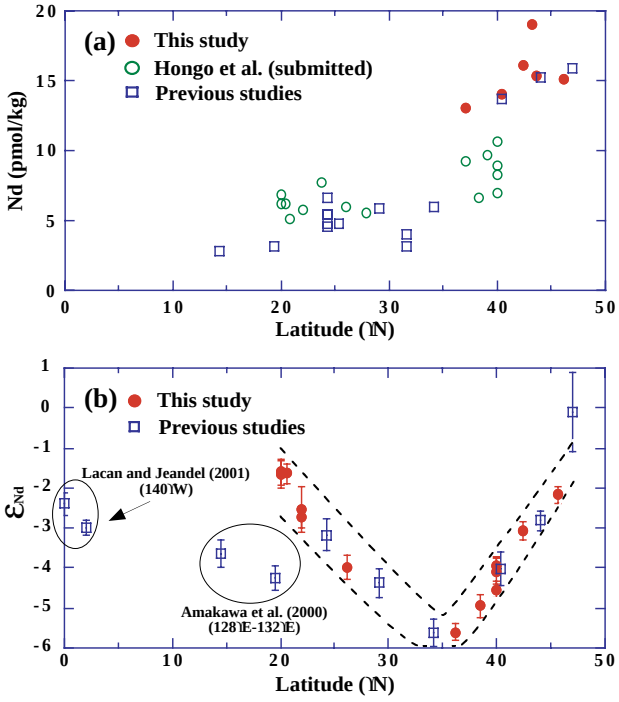

Fig. 4. Latitudinal distributions of Nd concentration (a) and its isotopic composition (b) in surface waters of the Northwest Pacific Ocean between 125°E and 180°E. The reported data ((a) Piepgras and Jacobsen, 1988; Greaves et al., 1999; Amakawa et al., 2000, 2004, and (b) Piepgras and Jacobsen, 1988; Amakawa et al., 2000, 2004) and unpublished data ((a) Hongo et al., submitted) from the same area are also plotted. The Nd isotopic data from other North Pacific regions (Lacan and Jeandel, 2001) are also shown in (b).

Figure 4(b) reveals that there are at least three water masses or currents showing distinctive Nd isotopic compositions: two radiogenic at high and low latitudes and one unradiogenic at mid latitude. This seems to correspond to the surface current patterns in the North Pacific (Fig. 1), i.e., the predominance of the westward current (NEC) and eastward currents (Kuroshio Extension and part of the Oyashio Current circulating along the Subarctic Gyre) in the Northwest Pacific. The Kuroshio Extension, which flows eastward at a latitude of around 35°N~40°N, shows ε_{Nd} values ranging from –5.6 to –3.9. While flowing eastward, the Kuroshio Extension mixes with the circulating Oyashio Current that shows a higher ε_{Nd} value ($\varepsilon_{Nd} > -3$) than the Kuroshio Extension. Water sampled from stations BO-S-2 and BO-2 showed slightly higher ε_{Nd} values (–3.9 and –4.1) than those sampled from sta-

tions BO-S-1 and BO-1 (–5.0 and –4.5) at the same latitude (Fig. 2), indicating the larger contribution of the Oyashio Current than the Kuroshio Extension in the former samples. This is compatible with the salinity data (see Subsection "Hydrography and sampling locations").

The radiogenic Nd signal of the Oyashio Current is due to such radiogenic sources as the Kuril and Aleutian Islands (Amakawa *et al.*, 2004). As suggested by Amakawa *et al.* (2004), the ultimate source of unradiogenic Nd in the Kuroshio Extension is a continental margin, i.e., the East China Sea.

As shown in Fig. 1, NEC flows westward at 10°N to 15°N, and changes its direction to the north near the Philippine Islands. The gradual decrease of ε_{Nd} values from 20°N to 30°N as observed in Fig. 4(b) suggests the decrease of the influence of NEC (a radiogenic Nd source) toward the north. The gradual decrease in salinity of the samples from 20°N to 30°N (~20°N, S > 35; ~26°N, S = ~34.9; ~29°N, S = ~34.7 (LM2, Amakawa *et al.*, 2004)) supports this idea. We deduce that NEC would show ε_{Nd} values that are higher than or comparable to those of the samples from 20°N (ε_{Nd} = –1.7~–1.6), because the water samples collected from the North Pacific Gyre were supplied with unradiogenic Nd from the Kuroshio or Kuroshio Extension, resulting in the lower ε_{Nd} values than those of NEC.

NEC flows into the East China Sea where it attains unradiogenic Nd isotopic signals. It flows further eastward to become the Kuroshio Extension (Fig. 1). These surface current patterns may have transported the continentally derived Nd to the central Pacific. On the other hand, the radiogenic Nd signals of the samples collected at a latitude around ~20°N suggest that the Hawaiian Islands, which are composed of mantle-derived volcanic rocks (ε_{Nd} = +3~+8, Mukhopadhyay *et al.*, 2003), may be the main source of radiogenic Nd for NEC. The contribution of volcanic island rocks, such as those from the Hawaiian and Kuril Islands, will be discussed later.

Nd budget in the Pacific Ocean

In the above section, we pointed out the importance of radiogenic Nd flux from volcanic islands in the Pacific Ocean. Goldstein and Jacobsen (1987) reported the dissolved Nd load in rivers from various areas and discussed the Nd budget in seawater. Jeandel *et al.* (1998) proposed that coastal areas could be the important sources of additional Nd to the oceans. Amakawa *et al.* (2000) pointed out that the estimation of Nd flux to the ocean by Goldstein and Jacobsen (1987) would result in an overestimation of the residence time because of the non-incorporation of Nd remobilization from coastal and shelf areas. We cannot neglect the remobilization of Nd in our discussion of the global Nd budget in the ocean.

Based on Nd concentration and isotopic composition, we discuss herein the Nd budget in the Pacific Ocean surface waters and estimate the radiogenic Nd flux, i.e., the Nd supplied from volcanic islands, such as the Kuril and Hawaiian Islands.

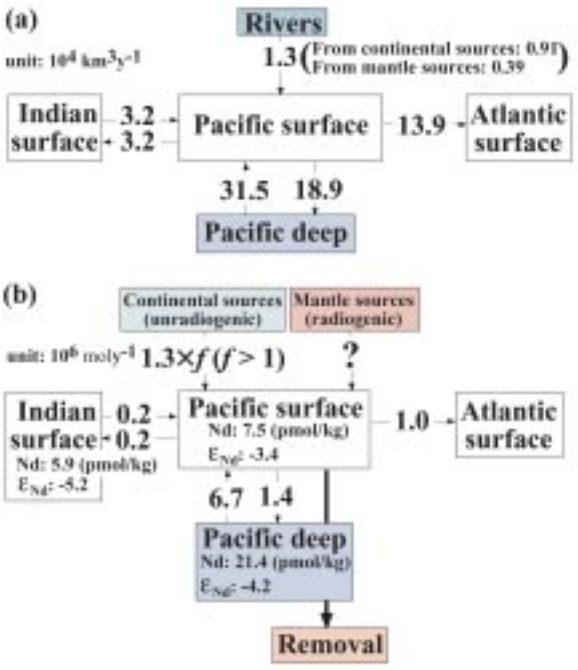

Fig. 5. Schematic diagrams of the box model used for calculating radiogenic Nd flux and Nd residence time in the Pacific Ocean surface water. Water mass transport is shown in (a). Water fluxes are from Bertram and Elderfield (1993). The Nd fluxes shown in (b) were calculated from the parameters listed in Table 3.

We employed the box model presented by Bertram and Elderfield (1993), which was originally proposed by Keir (1988) (Fig. 5), to estimate radiogenic Nd flux to the Pacific Ocean surface waters. The Pacific Ocean was separated into two reservoirs, namely, surface (above 100 m) and deep (below 100 m).

A schematic diagram of water mass transport is shown in Fig. 5(a). The water input and output, as well as the water discharge from various land areas are shown in Table 3 (Goldstein and Jacobsen, 1987; Bertram and Elderfield, 1993), together with the average Nd concentration and isotopic composition data of the water masses and river waters. Evaporation and precipitation at the surface were assumed to be equal.

For convenience, we set two major Nd land sources, a radiogenic source and an unradiogenic source. The former corresponds to the flux from the continental areas in Table 3, and the latter, to the flux from volcanic islands that are made of mantle-derived rocks, such as the Hawaiian and Kuril Islands. The total flux from the continental ar-

Table 3. Parameters used for Nd budget calculation

	Water input (10^4 km^3/y)*	Water output (10^4 km^3/y)*	Nd (pmol/kg)	ε_{Nd}	Reference#
Pacific surface	—	—	7.5 ± 3.9	−3.4 ± 1.6	[1], [2], [3], [4]
Pacific deep	31.5	18.9	21.4 ± 10.9	−4.2 ± 1.5	[1], [3], [5]
Indian surface	3.2	3.2	5.9 ± 1.7	−5.2 ± 1.4	[6]
Atlantic surface	—	13.9	—	—	
Water discharge from various land areas					
Asia (continent)	4.2×10^{-1}	—	165	−10.1	[7]
North America	2.5×10^{-1}	—	163	−3.3	[7]
South America	1.3×10^{-1}	—	166	−3.0	[7]
Australia	1.1×10^{-2}	—	104	−2.0	[7]

**Water fluxes from other oceans and water discharges from various land areas are from Bertram and Elderfield (1993) and Goldstein and Jacobsen (1987), respectively.*
#References: [1] Zhang and Nozaki (1996); [2] Nozaki (2001); [3] Amakawa et al. (2004); [4] Hongo et al. (submitted); [5] Piepgras and Jacobsen (1988); [6] Amakawa et al. (2000); [7] Goldstein and Jacobsen (1987).

eas is 1.3×10^6 (mol/y) (see Fig. 5(b)), which was calculated from the data shown in Table 3 (water discharges from various land areas).

The total riverine flux shown in Table 3, however, is not necessarily equal to the flux from the continental areas. As mentioned above, Nd remobilization from coastal and shelf areas may be an important source of Nd in the ocean. Furthermore, we cannot completely neglect the contribution of atmospheric flux from the continental areas to the ocean surface. Here, we introduce a parameter, f, which incorporates the contributions of Nd remobilization from coastal and shelf areas and atmospheric flux (Fig. 5(b)). f is defined as follows:

$$f = \frac{F^{CT}}{F^{C}} \tag{2}$$

where F^C and F^{CT} (mol/y) are, respectively, Nd input flux from rivers in the continental areas and real total Nd input flux from the continental areas including Nd remobilization from coastal and shelf areas and the atmosphere, etc. If f is 1, the flux from the continent will be equal to the continental riverine flux; therefore, due to the Nd remobilization, etc., f is usually higher than 1.

Under a steady state, if f and the average of the Nd isotopic composition of the mantle sources are given, the flux from the mantle sources is calculated from a balanced equation of the Nd isotopic composition in the Pacific Ocean surface. The isotopic balanced equation for inputs of the Pacific Ocean surface is given by:

$$\varepsilon_{Nd}^{PS} = \frac{\sum \varepsilon_{Nd}^{W} \bullet F_I^{W} + \sum \varepsilon_{Nd}^{C} \bullet F^{CT} + F^{M} \bullet \varepsilon_{Nd}^{M}}{\sum F_I^{W} + \sum F^{CT} + F^{M}} \tag{3}$$

where $\varepsilon_{Nd}{}^{PS}$, $\varepsilon_{Nd}{}^{W}$, $\varepsilon_{Nd}{}^{C}$ and $\varepsilon_{Nd}{}^{M}$ are ε_{Nd} values of the Pacific Ocean surface, the input from the Indian Ocean and the Pacific Ocean deep (the exchanged water masses), and the river inputs from continental areas (unradiogenic sources) and mantle sources (radiogenic sources), respectively. On the other hand, $F_I{}^{W}$ and F^M (mol/y) are the Nd input flux from other oceans and mantle sources, respectively. Combining the above equations, we obtain:

$$\varepsilon_{Nd}^{PS} = \frac{\sum \varepsilon_{Nd}^{W} \bullet F_I^{W} + \sum \varepsilon_{Nd}^{C} \bullet f \bullet F^{C} + F^{M} \bullet \varepsilon_{Nd}^{M}}{\sum F_I^{W} + \sum f \bullet F^{C} + F^{M}}. \tag{4}$$

Solving this equation, F^M is expressed as follows:

$$F^{M} = \left\{ \varepsilon_{Nd}^{PS} \bullet \left(\sum F_I^{W} + \sum f \bullet F^{C} \right) - \sum \varepsilon_{Nd}^{W} \bullet F_I^{W} - \sum \varepsilon_{Nd}^{C} \bullet f \bullet F^{C} \right\} \Big/ \left(\varepsilon_{Nd}^{M} - \varepsilon_{Nd}^{PS} \right). \tag{5}$$

Furthermore, the Nd removal flux from the Pacific Ocean surface (F^R) is calculated using the obtained mantle flux from a balanced equation of Nd concentration. F^R is given as follows:

$$F^{R} = \sum F_I^{W} - \sum F_O^{W} + \sum f \bullet F^{C} + F^{M} \tag{6}$$

where $F_O{}^{W}$ is the Nd output flux from the Pacific Ocean surface to the other oceans. This process consequently leads to the estimation of Nd residence time in the Pacific Ocean surface by dividing the Nd inventory in the Pacific Ocean surface by F^R. The calculated results are shown in Table 4. We tentatively set the ε_{Nd} values of the mantle sources at 0 and +2. There are scarce river

Table 4. Nd budget in Pacific Ocean surface

f	ε_{Nd} value of mantle sources	Calculated radiogenic Nd flux (F^M, 10^6 mol/y)	Radiogenic Nd flux /Unradiogenic Nd flux	Nd removal from Pacific Ocean surface (F^R, 10^6 mol/y)	Nd residence time in Pacific Ocean surface (y)
1	0	3.12	2.34	8.7	14.2
1	2	1.96	1.47	7.5	16.4
2	0	4.46	1.67	11.4	10.8
2	2	2.80	1.05	9.7	12.7
5	0	8.49	1.27	19.4	6.4
5	2	5.32	0.80	16.2	7.6
10	0	15.2	1.14	32.8	3.8
10	2	9.52	0.71	27.1	4.6

data showing ε_{Nd} values higher than +2 (Goldstein and Jacobsen, 1987); therefore, we think our choice of ε_{Nd} values is reasonable. Calculation errors due to uncertainties of Nd concentrations and isotopic compositions (see Table 3) are approximately 70% (data not shown).

As shown in Table 4, the radiogenic flux/unradiogenic flux ratio (F^M/fF^C) is decreased as f is increased. Furthermore, even at the highest f value, the mantle source flux is required for at least 70% of the unradiogenic flux.

Although the calculated values contain large uncertainties, it is revealed that a high f value is required for the high Nd removal flux from the ocean surface, and hence, the short residence time. Amakawa *et al.* (2000) estimated Nd residence time in the ocean surface to be 1.5 to 2.6 y. We need high f values to match our calculated values with their estimates. Nd remobilization from the coastal area as suggested by previous studies is one of the candidates for the additional flux (Jeandel *et al.*, 1998; Amakawa *et al.*, 2000, 2004; Sholkovitz and Szymczak, 2000; Lacan and Jeandel, 2001).

If we assume that the shortest obtained residence time of 3.8 y can be applied to the global oceans, the Nd residence time in the entire ocean can be estimated. Following Amakawa *et al.* (2000), the total dissolved Nd in the ocean surface (100 m in thickness) is 2.5×10^8 moles for an average concentration of 7 pmol/kg. The Nd flux to the ocean surface is calculated to be 6.6×10^7 (mol/y). Assuming a global average concentration of 20 pmol/kg (Nozaki, 2001), the Nd inventory in the entire ocean is 2.8×10^{10} moles. The Nd residence time is calculated by dividing the inventory by the flux to the surface ocean, i.e., 420 y, which somehow shows good agreement with the previous estimates by Tachikawa *et al.* (1999, ~400 y), Tachikawa *et al.* (2003, ~500 y) and Amakawa *et al.* (2000, 260~440 y). We conclude that both the large Nd remobilization from coastal and shelf areas and the mantle-derived fluxes are required for realizing a reasonable Nd residence time in the ocean.

SUMMARY AND CONCLUSION

The Okhotsk and Japan Seas surrounding the Japanese Islands show higher surface Nd concentrations than the Pacific Ocean. This may be due to the large Nd input from the land area close to the seas. However, the Nd isotopic compositions show highly contrasting values: a high ε_{Nd} value in the Okhotsk Sea and low ε_{Nd} values in the Japan Sea. The radiogenic ε_{Nd} value in the Okhotsk Sea apparently indicates the large Nd supply from the Kuril Islands that also have high ε_{Nd} values. There are at least two unradiogenic Nd sources for the Japan Sea: the fluvial/coastal sediments of the Asian continent and TWC. We think that the lowest ε_{Nd} value measured for the sample from the southernmost station, CM-20, is mainly influenced by TWC that may be supplied with unradiogenic Nd from the Korean Peninsular or the East China Sea. However, the contribution of particulate matter to Nd isotopic composition is yet to be elucidated for these semi-closed oceanic basins.

As for the North Pacific surface waters, the Nd concentration shows a gradual increase with increasing latitude. By contrast, the Nd isotopic composition distribution shows a more complicated feature that is not found in Nd concentrations. There are at least three surface waters with distinctive Nd isotopic compositions. One water with unradiogenic Nd signal prevails at the mid latitude and two waters with radiogenic Nd signals prevail at high and low latitudes. The mid-latitude minimum is ascribable to the lateral Nd transport by the Kuroshio Extension. The radiogenic ε_{Nd} values at high and low latitudes are due to the influence of the Oyashio Current and NEC, respectively, which are supplied with large amounts of radiogenic Nd from the volcanic islands. The lateral Nd transport of the three currents results in the latitudinal ε_{Nd} variation in the North Pacific. Again, this feature is not found in the Nd concentration distribution.

Based on our data and those of previous studies, we

estimated radiogenic Nd flux and Nd residence time in the Pacific Ocean surface waters. The calculations indicate that a large Nd remobilization from coastal and shelf areas is required to match our results with the previous estimation of Nd residence time in the surface waters. Using the shortest calculated residence time at the surface, we tentatively estimated Nd residence time in the global ocean and obtained a value of ~400 y, which is almost identical with previous estimates (Tachikawa *et al*., 1999, 2003; Amakawa *et al*., 2000).

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