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Neodymium isotopic variations in Northwest Pacific waters

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Abstract—Four vertical profiles of the concentration and isotopic composition of Nd in seawater were obtained in the western North Pacific. Two profiles from the Kuroshio Current regime showed congruently that although the Nd concentration increases gradually with depth, its isotopic composition varies significantly with depth depending upon the water mass occupying the water column. The high-salinity Kuroshio waters originating from the North Pacific Tropical Water (NPTW) carry the least radiogenic Nd ($\epsilon_{Nd} = -7.4$ to -8.7) to this region at ~250 m from the western margin continental shelves, most likely from the East China Sea. The Nd isotopic compositions in the North Pacific Intermediate Water (NPIW) that occurs at 600 to 1000 m in the subtropical region are fairly uniform at $\epsilon_{Nd} = -3.7$. The profile data from the ~38° to 40°N Kuroshio/Oyashio mixed water region off Sanriku of Honshu, Japan, also suggest that the newest NPIW with $\epsilon_{Nd} = -3.2$ is formed there by the mixing of various source waters, and the radiogenic component of Nd is derived mainly from the Oyashio waters.

In the Pacific Deep Water (PDW) below ~1000 m, the Nd isotopic composition is neither vertically nor horizontally homogeneous, suggesting that it serves as a useful tracer for sluggish deep water circulation as well. Two profiles from the Izu-Ogasawara Trench showed a minimum ϵ_{Nd} value at ~2000 m, suggesting that there exists a horizontal advective flow in the vicinity of Honshu, Japan. There is some evidence from other chemical properties to support this observation. The waters below 4000 m including those within the trench in the subtropical region have ϵ_{Nd} values of around -5, suggesting that the deep waters are fed from the south along the western boundary, ultimately from the Antarctic Bottom Water (AABW) in the South Pacific. This extends up to ~40°N along the Japanese Islands. In the subarctic region (>~42°N), the waters have more radiogenic Nd with $\epsilon_{Nd} > -4.0$ throughout the water column, presumably due to the supply of Nd by weathering in such igneous provinces as the Kuril-Kamchatska-Aleutian Island chain. The lateral inhomogeneity of the Nd isotopic composition in PDW suggests that there may be different circulation and mixing regimes in the North Pacific Basin. *Copyright © 2004 Elsevier Ltd*

1. INTRODUCTION

Variations in the Nd isotopic composition of seawater have been observed and discussed in terms of weathering, ocean circulation, and the scavenging residence time of Nd (Piepgras and Wasserburg, 1982, 1987; Spivack and Wasserburg, 1988; Jeandel, 1993; Bertram and Elderfield, 1993; Jeandel et al., 1995, 1998; Tachikawa et al., 1999; Amakawa et al., 2000; Lacan and Jeandel, 2001). The Nd isotopic composition is generally expressed as

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

where CHUR stands for Chondritic Uniform Reservoir and the present-day CHUR value is 0.512638 (Wasserburg et al., 1981). The $^{143}Nd/^{144}Nd$ isotopic ratio in various rocks varies depending upon the Sm/Nd ratio and age, because ^{147}Sm decays to ^{143}Nd with a half-life = 1.06×10^{11} years. The old continental crust has less radiogenic ϵ_{Nd} values of -10 to -36, whereas such mantle-derived igneous provinces as island arcs and midoceanic ridges have more radiogenic ϵ_{Nd} values ranging from ~0 to +10. Thus, weathering products carry various Nd isotopic signatures into the ocean. Unlike its concentration in seawater, the Nd isotopic ratio changes not by evaporation or particulate scavenging, but only by mixing of water masses or an external input of Nd with different isotopic ratios; therefore, it serves as a tracer of ocean circulation (Albarede and Goldstein, 1992; Rutberg et al., 2000).

We present new data on Nd concentration and its isotopic composition in the western North Pacific covering subtropical to subarctic regions, and compare them with two previously reported profiles in this region (Piepgras and Jacobsen, 1988). Based on the results, we attempted to clarify water mixing and circulation in this region of the Pacific.

2. STUDY AREA

Figure 1 shows the topography of the present study area in the western North Pacific. Its features include the existence of marginal seas with deep central basins (e.g., Japan and Okhotsk Seas) and the very deep trenches along the western rim of the northwest Pacific abyssal plain of ~6000 m (i.e., Kuril, Japan and Izu-Ogasawara Trenches).

The flow patterns in the upper layers of the subtropical and subarctic regions are governed largely by the Kuroshio and Oyashio Currents, respectively. During the past several decades, hydrographic investigations have been carried out intensively by Japanese scientists (see Stommel and Yoshida, 1972) and occasionally by U.S. and Russian workers (Talley, 1993;

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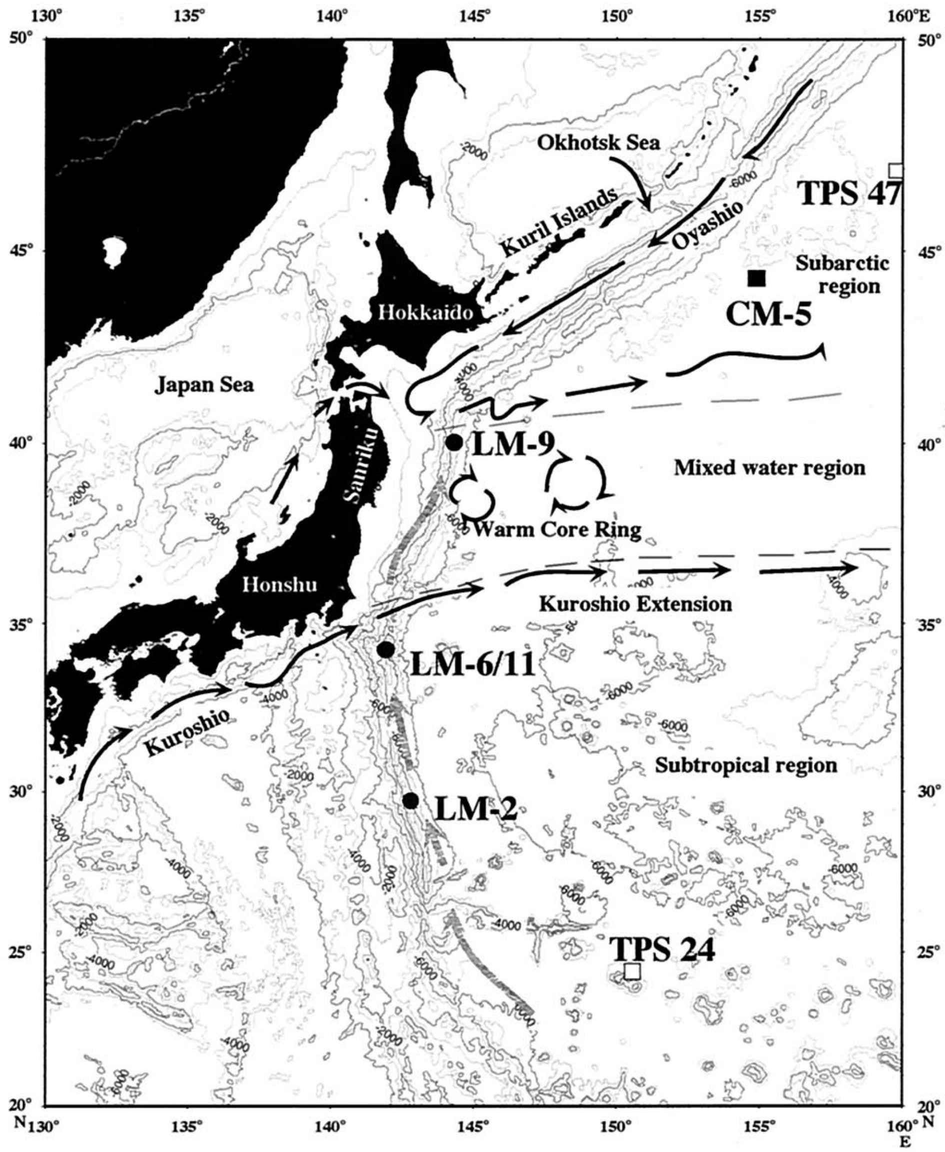

Fig. 1. Map showing topography and the surface current system (Kawai, 1972 and Talley et al., 1995) in the western North Pacific. The flow pattern of deep water along the Izu-Ogasawara Ridge and Honshu deduced in this work is shown by faint thick arrows. The sampling locations (LM-2, -6/11 and -9 (KH-94-3): filled circles; CM-5 (KH-98-3): filled square) are shown together with previously studied locations (TPS 24 and 47; open squares) by Piepgras and Jacobsen (1988).

Kawabe and Taira, 1995). The subtropical waters in the Kuroshio are characterized by high temperature ($>\sim 18°C$), high salinity, and low nutrient content, whereas the subarctic waters in the Oyashio are low in temperature ($<\sim 10°C$) and salinity and rich in dissolved oxygen and nutrients. Between the Kuroshio Extension and the Oyashio Front lies the Mixed Water Region (MWR; Talley et al., 1995) centered at around 39° to 40°N where several water masses originating in the Kuroshio, Oyashio and Tsugaru Warm Currents meet and mix in complex ways (Fig. 1). Warm core rings often appear there, changing their locations generally westward and disintegrating near the Japanese coast. This is an important area for the formation of the North Pacific Intermediate Water (NPIW) that prevails in almost the entire area of the North Pacific subtropical region (Talley, 1993; Talley et al., 1995) and hence, may serve as an important potential sink of anthropogenic carbon dioxide (Tsunogai et al., 1996). NPIW is generally defined by a salinity minimum at depths of 300 to 1000 m with a potential density surface of $\sigma_\theta = 26.8$. The more fresh and oxygenated Okhotsk Sea water appears to also contribute to NPIW (Reid, 1965). Hasunuma (1978) argued that the salinity minimum is not a convective core of a particular water mass like the Antarctic Intermediate Water (AAIW) and the Labrador Sea Water in the North Atlantic; rather, it is regarded as a boundary between subtropical and subarctic waters in the subtropical gyre. As pointed out by Talley et al. (1995), many questions remain regarding NPIW formation, including the relative importance of various possible sites in MWR, whether NPIW density is linked to the winter surface density of subarctic water, the importance of vertical and horizontal mixing, and how much NPIW is formed. Chemical tracers such as Nd isotopes may help answer some of these questions.

The Pacific Deep Water (PDW) lies beneath the NPIW. At depths greater than 2000 m, it has fairly uniform temperature and salinity (θ = 1.1–1.8°C, S = 34.6–34.7 psu). Although the possible flow patterns of PDW and AABW in the Pacific have been described (e.g., Kawabe and Taira, 1995), the origin and circulation of PDW particularly in the North Pacific are not well understood (see Schmitz, 1995, for example). Stommel and Arons (1960) predicted that the northward flow of the deep western boundary current coming straight up to the north from the South Pacific is expected to meet with its branch flowing anticlockwise along the rim of the North Pacific Basin somewhere near Japan. However, recent physical oceanographical model calculations (e.g., Ishizaki, 1994) do not agree with their prediction. This conflict cannot easily be resolved by field observations because the deep circulation is so sluggish that it can hardly be measured by direct physical means, such as CTD profiling, current meters and floats. Again, it may be best constrained by use of chemical tracers including Nd isotopes. This is the objective of the present study.

3. METHODS

Water samples were collected during two cruises on the R.V. Hakuho Maru: the Leo Minor (KH-94-3) expedition in September–October, 1994 and the Canis Minor (KH-98-3) expedition in July–August, 1998. The sampling locations are shown in Figure 1. For Nd isotope measurements, a twin 270l-PVC bottle sampler (Nichiyu-Giken Kogyo, N12-500) was used. Water samples were also collected for measurements of Nd concentration by using 12-L lever-action type Niskin bottles with Teflon-coated inside walls. The Niskin bottles were either mounted on 24-position Sea-Bird's 911plus CTD-rosette array for casts shallower than 6500 m or directly hung in series on a pure-titanium hydrowire together with digital reversing thermometers for the deeper casts. CTD measurements were conducted only down to 6500 m due to the tolerance limit of the pressure housing of the instrument. Near-bottom samples were obtained by monitoring the distance from the seafloor using an acoustic pinger.

Analyses of Nd concentration and its isotopic composition were based on unfiltered seawaters, except for some samples for Nd concentration analysis at CM-5 that were filtered through a 0.04 μm hollow-fiber (Millipore HF-400) cartridge. In a later section, we compare filtered CM-5 concentration data with other unfiltered data without correction, because as pointed out by Alibo and Nozaki (1999), the particulate fraction of trivalent REE is less than 5% in most cases.

The analytical procedure for Nd isotopic composition measurements was described by Amakawa et al. (2000). Nd in a large-volume (>20 L) seawater sample was coprecipitated with iron hydroxide and then purified by successive ion-exchange procedures. The purified Nd was loaded on a Re double filament in nitrate form and its isotopic ratio was measured using a Finnigan MAT 262 thermal ionization mass spectrometer. The measured $^{143}Nd/^{144}Nd$ ratios were normalized to $^{146}Nd/^{144}Nd = 0.7219$. During the measurements, the isotopic ratios were monitored against La Jolla and JNdi-1 standards, the latter provided by the Geological Survey of Japan. The values of the standards were 0.511856±0.000007 (2σ, n=9) and 0.512113±0.000007 (2σ, n=73), respectively, in good agreement with those reported by O'Nions et al. (1977) and Tanaka et al. (2000). Therefore, we did not apply any correction for instrumental bias.

The analytical procedure for REE concentration including Nd was described by Zhang and Nozaki (1996) and Alibo and Nozaki (1999). It follows the method developed by Shabani et al. (1990) including purification and concentration by solvent extraction and subsequent determination with an ICP mass spectrometer (Yokogawa Analytical Systems, PMS-2000). The precision based on replicate measurements was better than 4% at ~20 pmol/kg level and blank was less than 0.7 pmol/kg for Nd.

4. RESULTS

4.1. θ-S Diagram

Figure 2 shows the potential temperature-salinity diagrams at the four stations investigated in the study. The θ-S data clearly define the water masses in the Northwest Pacific, such as the North Pacific Tropical Water (NPTW), NPIW and PDW (Figs. 2a and b).

The data at LM-2 and LM-6/11 in the Kuroshio Current regime are similar to each other, showing salinity maxima at ~200 m attributable to NPTW entrainment (high salinity, high θ), which is believed to originate in the subtropical North Pacific (Kawai, 1972), and minima at 700 to 800 m corresponding to NPTW (low salinity, low θ). (Kawai, 1972). In contrast, the water column at CM-5 is characterized by low salinity due to dilution by fluvial discharge and precipitation, and low temperature due to winter cooling. The shallow water data at LM-9 in the MWR follow closely the trend of the Oyashio water. Below 2000 m, the θ-S diagrams at all stations depicted a characteristic trend defining PDW (high salinity, low θ; Fig. 2b), which underlies NPIW at LM-2 and LM-6/11.

The θ-S data at LM-2 and LM-6/11 and those at CM5 and LM-9 show good agreement with those at TPS 24 (a subtropical station; 24°N, 150°E) and TPS 47 (a subarctic station; 47°N, 161°E), respectively, as reported by Piepgras and Jacobsen (1988) (Fig. 1).

4.2. Nd Concentration and its Isotopic Composition

The Nd concentrations are given in Table 1 together with potential temperature, salinity and dissolved Si. Zhang et al. (1994) published the vertical profiles of REEs down to 3500 m at the same location as LM-2, based on analyses of samples collected by the R. V. Tansei Maru in August, 1993. The data are in good agreement with those obtained earlier. At CM-5, we filtered the water samples obtained from Niskin bottles by using a 0.04 μm hollow-fiber cartridge. Alibo and Nozaki (1999) determined the particulate fraction of REEs that can be removed by the filtration and found it to be 4.4 ± 2.7% (n=17) for Nd in ambient seawater near Japan. Exceptions are the anomalously high values of the REE concentrations that are often found near the bottom of slopes and troughs (Zhang and Nozaki, 1998). The particulate fractions for those samples are much larger due to contributions from resuspended sediment particles, although they can be removed by the filtration (Alibo and Nozaki, 1999).

The Nd isotopic compositions are given in Table 2. The bottom sample at LM-9 was collected from a depth of 7370 m. Therefore, it is likely that the Nd isotopic composition is affected by contributions from resuspended sediments, as deduced from the near-bottom high Nd concentrations (Table 1). The ϵ_{Nd} value for this sample is indeed the lowest at −6.0 among all the values for the deep waters obtained here. The Nd isotopic composition varies significantly from $\epsilon_{Nd} = -8.7$ to −2.8 depending upon the water mass and will be discussed in more detail below.

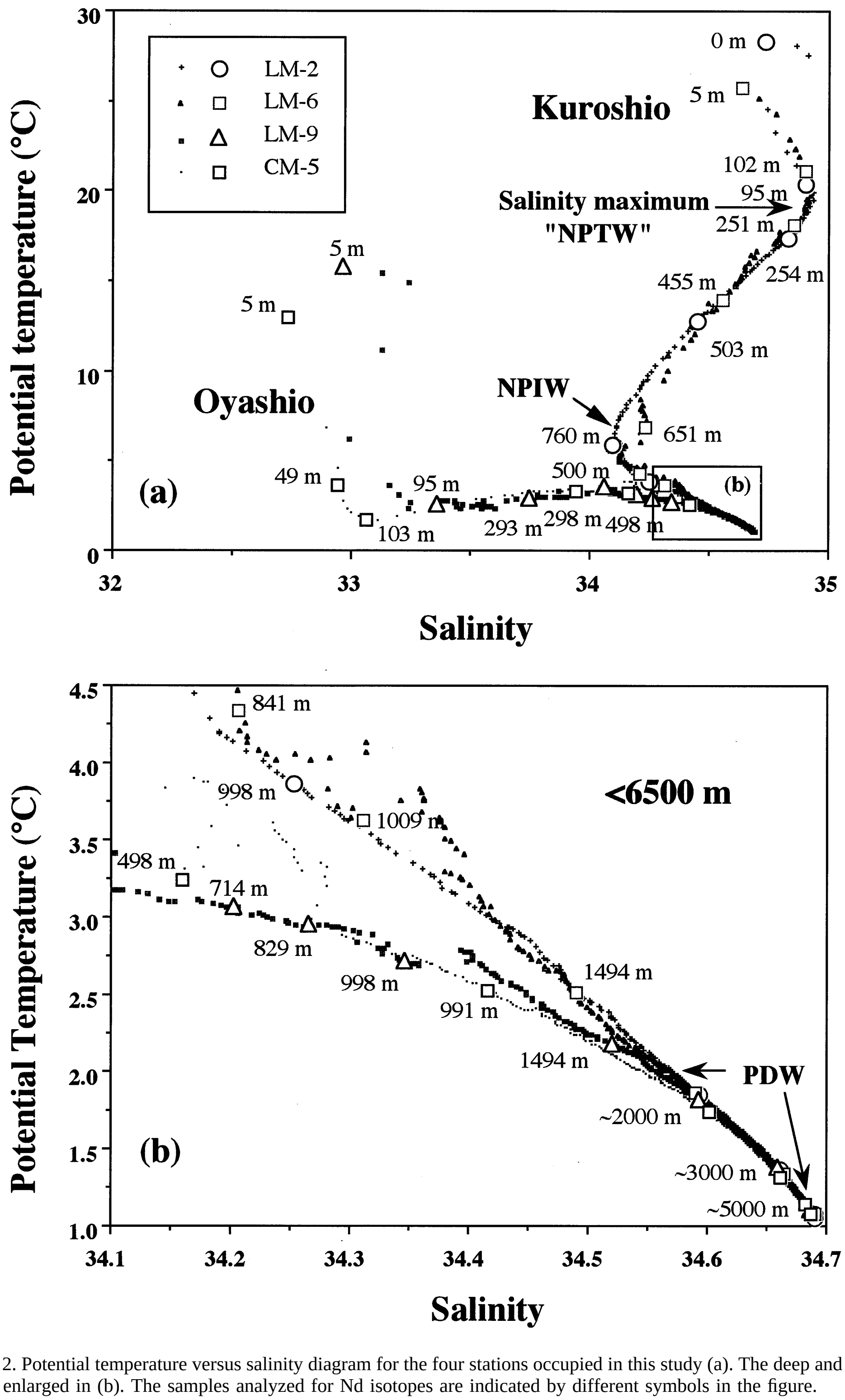

Fig. 2. Potential temperature versus salinity diagram for the four stations occupied in this study (a). The deep and bottom part is enlarged in (b). The samples analyzed for Nd isotopes are indicated by different symbols in the figure.

5. DISCUSSION

5.1. Nd Sources to the Surface Ocean

Since Nd in the ocean is ultimately derived from terrestrial sources, it seems possible to deduce source functions (i.e., eolian versus fluvial/coastal input), using data of the distribution and isotopic composition of Nd in surface waters. Amakawa et al. (2000) adopted this approach and concluded that remineralization of Nd from coastal and shelf sediments is more important than atmospheric input. Sholkovitz and Szym-

Table 1. Nd concentration in Northwest Pacific seawaters.

Depth m	Salinity psu	Pot. Temp. °C	SiO_2 μmol/kg	Nd pmol/kg
LM-2 (29°05′N, 142°51′E; Depth, 9738 m)				
0	34.733	28.274	2.6	5.9
499	34.451	12.784	17.1	6.7
1000	34.254	3.859	116.0	19.9
1500	34.492	2.506	154.2	23.6
2003	34.591	1.860	164.3	25.4
2502	34.640	1.539	158.7	28.8
3003	34.661	1.368	157.1	31.6
3501	34.674	1.242	156.3	34.5
4501	34.687	1.103	152.0	37.6
5998	34.691	1.050	146.9	39.1
7229	34.698	1.085	145.8	40.7
8727	34.699	1.102	150.8	38.9
9676	34.697	—	146.2	39.3
9721	34.693	—	146.4	39.9
9726	34.692	—	146.1	40.2
LM-6/11 (34°10′N, 142°00′E; Depth, 9194 m)				
0	34.640	25.728	0.0	6.0
199	34.907	19.076	3.4	5.6
398	34.645	15.312	12.8	7.5
505	34.448	12.158	20.3	8.9
794	34.171	4.668	84.7	18.2
994	34.303	3.682	117.6	21.0
1980	34.587	1.881	162.0	25.9
2467	34.636	1.545	156.3	30.9
2965	34.662	1.348	152.5	33.7
3997	34.683	1.149	159.4	38.9
4959	34.690	1.074	143.4	40.7
5957	34.692	1.050	140.3	39.0
7150	34.691	1.068	145.3	37.8
8156	34.689	1.096	145.5	37.3
8654	34.687	1.103	144.8	37.1
9095	34.687	1.075	144.7	37.5
9139	34.690	1.079	139.6	37.3
9159	34.690	1.076	139.4	38.1
LM-9 (40°26′N, 144°30′E; Depth, 7380 m)				
0	32.971	15.712	6.1	13.7
49	33.153	6.837	19.6	17.2
98	33.299	2.373	45.9	17.5
147	33.415	2.586	53.6	18.8
197	33.540	2.574	70.3	20.5
296	33.671	2.911	85.8	21.1
395	33.872	3.009	98.4	20.1
542	34.092	3.419	114.0	18.8
733	34.210	3.027	129.0	21.0
1002	34.354	2.695	148.8	23.0
1498	34.520	2.182	160.8	23.8
1998	34.593	1.817	160.9	24.6
2999	34.659	1.369	157.4	27.9
3496	34.671	1.259	154.4	30.5
3995	34.679	1.180	152.2	34.9
4994	34.687	1.094	148.3	37.6
5492	34.689	1.070	146.6	37.0
5992	34.690	1.055	144.5	37.7
6496	34.691	1.046	144.2	36.3
6877	34.692	1.082	145.8	39.7
7126	34.690	1.112	146.2	47.9
7315	34.689	1.064	146.3	70.2
7355	34.690	1.092	145.1	81.6
7375	34.685	1.060	145.9	85.7
CM-5 (44°00′N, 155°00′E; Depth, 5310 m)				
0	32.578	13.100	9.2	15.2
51	32.898	3.432	25.7	16.4
99	33.066	1.890	43.4	17.3
197	33.573	2.918	83.3	21.1
496	34.117	3.243	130.7	22.7
793	34.304	2.782	151.5	24.7
986	34.370	2.552	158.7	33.1

(Continued)

Table 1. (Continued)

Depth m	Salinity psu	Pot. Temp. °C	SiO_2 μmol/kg	Nd pmol/kg
1977	34.569	1.724	171.3	34.2
2959	34.628	1.308	165.1	39.1
3934	34.648	1.136	158.3	40.9
4421	34.651	1.097	158.1	42.0
4908	34.652	1.083	157.7	42.7
5288	34.663	1.081	158.1	42.9

czak (2000) and Lacan and Jeandel (2001) reached the same conclusion, independently. Nd concentration and isotopic composition obtained here are plotted against latitude (Fig. 3), together with those from the literature (Piepgras and Jacobsen, 1988; Amakawa et al., 2000). Nd concentration increases with latitude from ~3 pmol/kg in the tropical region to >15 pmol/kg at >45°N (Fig. 3a). There seems to be a significant difference in concentration between the subtropical and subarctic regions. This pattern is significantly different from that of the atmospherically derived ^{210}Pb that shows a maximum in the mid latitude centered around 30°N (Nozaki et al., 1976), and suggests that the eolian source is relatively minor for Nd.

The value of ϵ_{Nd} varies significantly from -5.6 to -0.1 in the surface waters (Fig. 3b). Greaves et al. (1994) indicated that the partial dissolution of aerosols, such as Saharan dust, into seawater may be an important source relative to riverine input. Thus, in the western North Pacific, Nd in seawater may be strongly affected by Asian dust input. However, the Nd isotopic composition in the surface waters obtained in this study does not support this idea. Asian dust originates from sedimentary deposit and soil in central Asia with generally unradiogenic Nd. Chinese loess has ϵ_{Nd} values of -10 to -11 (Liu et al., 1993, 1994; Jones et al., 1994). The ϵ_{Nd} values in the surface waters are far more radiogenic, and therefore it is unlikely that they are derived largely from atmospheric sources. This is consistent with the report of Lacan and Jeandel (2001), who rejected the eolian origin of Nd in surface water, based on the surface Nd isotopic data at 2°N, 0° and 12°S of the eastern equatorial Pacific.

Figure 3b shows a significant local variability in ϵ_{Nd} with a less radiogenic value of -5.6 near Japan and a radiogenic value of -0.1 near Kamchatska (TPS 47; Piepgras and Jacobsen, 1988). This suggests that Nd is strongly influenced by local sources. It is unclear if the less radiogenic value of -5.6 is due to fluvial input from Japan as Nd isotopic data are few in Japanese rivers. Goldstein and Jacobsen (1987) reported a high ϵ_{Nd} value of $+0.1$ for Mogami River in northern Japan. This value is expected because the Japanese Islands are of igneous origin. If it were typical of Japanese rivers, it would be difficult to explain the low ϵ_{Nd} near Japan. One possibility is that the low ϵ_{Nd} is derived from sediments along the Japanese coast, some of which may have less radiogenic Nd. The Nd isotopic composition of the near-bottom sample at LM-9 (Table 1), which may have been influenced by resuspended sediments, requires the underlying sediments in the Japan Trench to have ϵ_{Nd} values that are more negative than -6.0. If the shallow water sediments around Japan have such low ϵ_{Nd} values, sediment

Table 2. Nd isotopic ratios and ε_{Nd} values in Northwest Pacific seawaters.

Depth m	Salinity psu	Pot. Temp. °C	$^{143}Nd/^{144}Nd$	ε_{Nd}
LM-2 (29°05′N, 142°51′E; Depth, 9738 m)				
0	34.410	28.40	0.512414 ± 0.000019	−4.37 ± 0.36
95	34.969	20.59	0.512306 ± 0.000021	−6.47 ± 0.41
254	34.835	17.29	0.512261 ± 0.000027	−7.36 ± 0.53
503	34.439	12.67	0.512331 ± 0.000025	−5.99 ± 0.49
760	34.108	6.38	0.512384 ± 0.000029	−4.95 ± 0.56
998	34.270	3.85	0.512447 ± 0.000014	−3.72 ± 0.27
2021	34.593	1.84	0.512368 ± 0.000027	−5.26 ± 0.53
3032	34.661	1.36	0.512415 ± 0.000016	−4.34 ± 0.32
5060	34.689	1.08	0.512377 ± 0.000027	−5.09 ± 0.52
5972	34.677	1.07	0.512354 ± 0.000021	−5.14 ± 0.40
7024	34.707	1.06	0.512371 ± 0.000023	−5.22 ± 0.44
8003	34.684	1.07	0.512375 ± 0.000014	−5.14 ± 0.27
9017	34.701	1.08	0.512385 ± 0.000022	−4.93 ± 0.42
LM-6/11 (34°10′N, 142°00′E; Depth, 9194 m)				
5	34.559	25.50	0.512350 ± 0.000017	−5.63 ± 0.34
102	34.901	20.72	0.512245 ± 0.000020	−7.66 ± 0.38
251	34.874	18.18	0.512190 ± 0.000030	−8.74 ± 0.58
455	34.590	14.36	0.512315 ± 0.000028	−6.29 ± 0.54
651	34.208	7.41	0.512364 ± 0.000021	−5.34 ± 0.40
841	34.178	4.44	0.512453 ± 0.000016	−3.60 ± 0.31
1009	34.283	3.71	0.512442 ± 0.000015	−3.82 ± 0.30
1494	34.494	2.52	0.512386 ± 0.000014	−4.91 ± 0.27
1989	34.593	1.87	0.512331 ± 0.000019	−5.98 ± 0.37
3012	34.664	1.65	0.512423 ± 0.000013	−4.20 ± 0.25
4986	34.680	1.09	0.512386 ± 0.000011	−4.91 ± 0.22
6576	34.692	1.10	0.512370 ± 0.000014	−5.23 ± 0.26
9076	34.694	1.09	0.512399 ± 0.000016	−4.67 ± 0.31
LM-9 (40°26′N, 144°30′E; Depth, 7380 m)				
5	33.040	14.90	0.512432 ± 0.000021	−4.02 ± 0.41
95	33.427	3.24	0.512480 ± 0.000018	−3.07 ± 0.35
293	33.678	2.72	0.512471 ± 0.000013	−3.26 ± 0.26
500	34.062	3.52	0.512480 ± 0.000022	−3.08 ± 0.44
714	34.008	3.00	0.512458 ± 0.000029	−3.50 ± 0.56
829	34.277	2.92	0.512481 ± 0.000030	−3.06 ± 0.59
988	34.354	2.94	0.512407 ± 0.000026	−4.50 ± 0.51
1495	34.493	2.20	0.512419 ± 0.000019	−4.28 ± 0.37
2000	34.594	1.82	0.512410 ± 0.000017	−4.44 ± 0.33
3000	34.659	1.37	0.512397 ± 0.000013	−4.70 ± 0.26
4988	34.688	1.11	0.512348 ± 0.000012	−5.65 ± 0.23
7020	34.635	1.08	0.512364 ± 0.000020	−5.35 ± 0.39
7370	34.690	1.08	0.512330 ± 0.000010	−6.02 ± 0.20
CM-5 (44°00′N, 155°00′E, Depth, 5310 m)				
5	32.578	13.10	0.512494 ± 0.000013	−2.81 ± 0.25
49	33.144	4.08	0.512454 ± 0.000043	−3.60 ± 0.83
103	33.109	1.65	0.512517 ± 0.000021	−2.36 ± 0.40
298	33.939	3.38	0.512514 ± 0.000025	−2.42 ± 0.49
498	34.136	3.35	0.512490 ± 0.000035	−2.89 ± 0.68
991	34.384	2.52	0.512474 ± 0.000030	−3.19 ± 0.58
1978	34.571	1.71	0.512458 ± 0.000014	−3.51 ± 0.28
2974	34.628	1.30	0.512456 ± 0.000017	−3.54 ± 0.33
3977	34.648	1.13	0.512441 ± 0.000017	−3.84 ± 0.32
4999	34.653	1.08	0.512438 ± 0.000017	−3.90 ± 0.32

contact may imprint the Nd isotopic signal to the water. Similarly, the high ϵ_{Nd} of −0.1 must be derived from the Aleutian-Kamchatska-Kuril chain.

The above observations suggest that local REE contributions from sediments and source rocks influence REE abundances and Nd isotopic composition in the dissolved phase in this oceanic region. More detailed studies of the REE composition of sediments and fluvial inputs are needed to better constrain and quantify their contributions to the dissolved REE budget.

5.2. Vertical Variation of Nd Isotopic Composition in the Subtropical Region

The vertical profiles of Nd concentraion and its isotopic composition at LM-2 and LM-6/11 in the Kuroshio Current regime are shown in Figure 4, together with those of salinity and dissolved Si. A diagram of salinity versus ϵ_{Nd} values is also shown (Fig. 4g). Although the Nd concentration increases gradually with depth, there are significant differences in the profiles of nutrients and Nd. For example, dissolved Si is highly

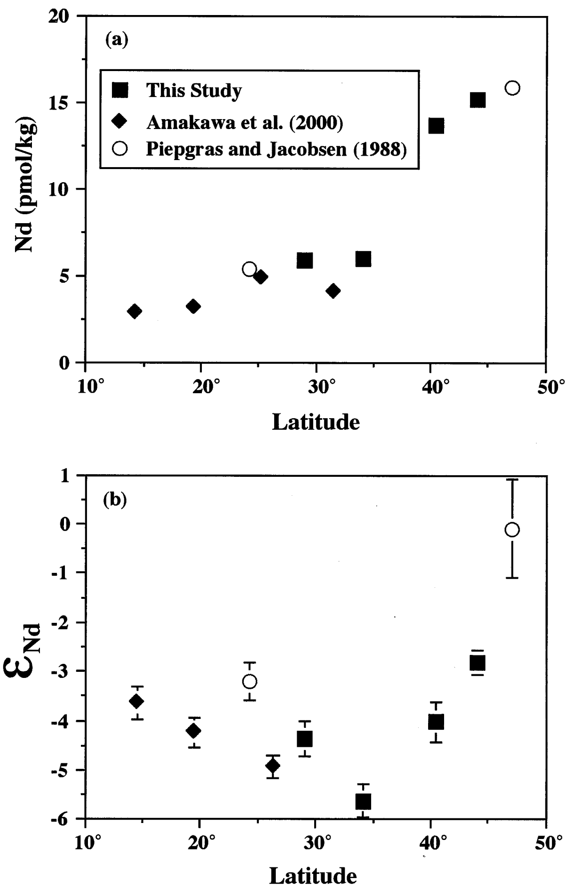

Fig. 3. Latitudinal distributions of Nd concentration (a) and its isotopic composition (b) in surface waters of the western North Pacific between 125°E and 161°E. Data from this study, Amakawa et al. (2000) and Piepgras and Jacobsen (1988) are plotted for comparison.

depleted in the surface water, whereas Nd has a finite concentration of ~6 pmol/kg. There is a maximum of dissolved Si at ~2000 m, whereas that of Nd either occurs at a much greater depth or is not seen (Figs. 4c and 4f). Despite the smooth "nutrient-like" profile of Nd, its isotopic composition varies significantly with depth. Although the two station locations are ~560 km apart, their vertical profiles are very similar (Figs. 4b and 4e). Clearly, the Nd isotopic signals are stratified according to various water masses occupying the water column. As suggested by previous studies (Piepgras and Wasserburg, 1987; Piepgras and Jacobsen, 1988; Jeandel, 1993), such variations in ϵ_{Nd} cannot be explained simply by such vertical processes as partial dissolution of eolian dust and solute-sinking particle exchange reactions.

The surface ϵ_{Nd} values of −4.4 and −5.7 obtained here are comparable to −4.9 in the Kuroshio Current around the Ryukyu Islands, southwest of Japan (Amakawa et al., 2000). There is a sharp ϵ_{Nd} minimum of −7.4 at LM-2 and −8.7 at LM-6/11 (Fig. 4) at depths around 250 m corresponding to the salinity maximum (>34.8 psu), which is attributable to entrainment of NPTW in the Kuroshio. The region for NPTW is clearly identified in the salinity vs. ϵ_{Nd} diagram (Fig. 4g).

The high-salinity NPTW is believed to be formed in subtropical North Pacific by excessive evaporation compared to precipitation, carried westward by the Northern Equatorial Current, and then incorporated into the Kuroshio off northern Philippines turning to the north (Kawai, 1972). It appears difficult to find such low ϵ_{Nd} values in the source water region. The shallow waters at the 24°N subtropical gyre station, TPS 24 (Piepgras and Jacobsen, 1988), have ϵ_{Nd} values that are more positive than −4.4 (Fig. 5), and the data for two surface waters east of the Philippines are −3.6 and −4.2 (Amakawa et al., 2000). This implies that the negative ϵ_{Nd} values are imprinted probably by sediment contact somewhere along the path of the Kuroshio. The best candidate is the East China Sea with a vast continental shelf since the Kuroshio can exchange various materials with the shelf waters along the western edge. In fact, Goldstein et al. (1984) reported the ϵ_{Nd} values of sedimentary loads from the Yangtze and Yellow Rivers, which were −10.7 and −10.9, and −12.6, respectively. Those sedimentary loads are believed to be the main sources for the sediments in the East China Sea. If the shelf sediments have $\epsilon_{Nd} = -11.4$, the average of three data by Goldstein et al. (1984), the fraction of Nd derived from the sediment source in the Kuroshio water can be calculated using a simple two-end-member mixing model. Assuming that the Kuroshio water and the original NPTW have ϵ_{Nd} values of −8 and −4, respectively, the continental shelf-derived Nd fraction is estimated to be 54% in the Kuroshio water. Although the exact mechanism of Nd release from the sediments is not well understood, there is no doubt that this contribution is significant.

With further increases in depth, ϵ_{Nd} increases, reaching a maximum of −3.7 at 998 m for LM-2 and -3.6 at 841 m for LM-6/11. Those depths agree with the salinity minimum and the potential density surface of $\sigma_\theta = \sim 26.8$ referred to as the North Pacific Intermediate Water (NPIW), which is also recognized in Figure 4g. In the TPS 24 station, the NPIW occurs at ~640 m and its ϵ_{Nd} value is −3.7 (Piepgras and Jacobsen, 1988). Thus, the NPIW has a uniform Nd isotopic composition at least in the western part of the North Pacific subtropical gyre. It is believed that the NPIW is formed in the mixed water region (MWR) between the Kuroshio Extension and the Oyashio front (Talley et al., 1995; Kawai, 1972), where complex mixing of waters from various sources occurs. Since our LM-9 station is located in the MWR, the Nd isotopic composition may shed light on the mechanism of NPIW formation. This aspect will be discussed later.

Below ~1000 m, potential temperature (θ) and salinity gradually change with depth, attaining near constant values of ~1.05 °C and ~34.69 psu below 4000 m. In the Pacific Deep Water (PDW), the Nd isotopic composition is not homogeneous, showing a minimum at ~2000 m and a maximum at ~3000 m in the two profiles (Figs. 4b, 4e and 4g); note that the range of ϵ_{Nd} values for PDW is much wider than those for NPTW and NPIW. This suggests that stratified advective flow of water exists in this region. Tsunogai et al. (1973) previously observed two discontinuities in the vertical profiles of chemical properties such as alkalinity and pH at 38.5°N, 145°E, approximately 110 km east of LM-2. The waters between 1700 and 3000 m are rich in dissolved oxygen and low in alkalinity and nutrients compared to those extrapolated smoothly from upper and deeper waters, suggesting that the waters are metabolically young and probably derived horizontally from the south. Thus,

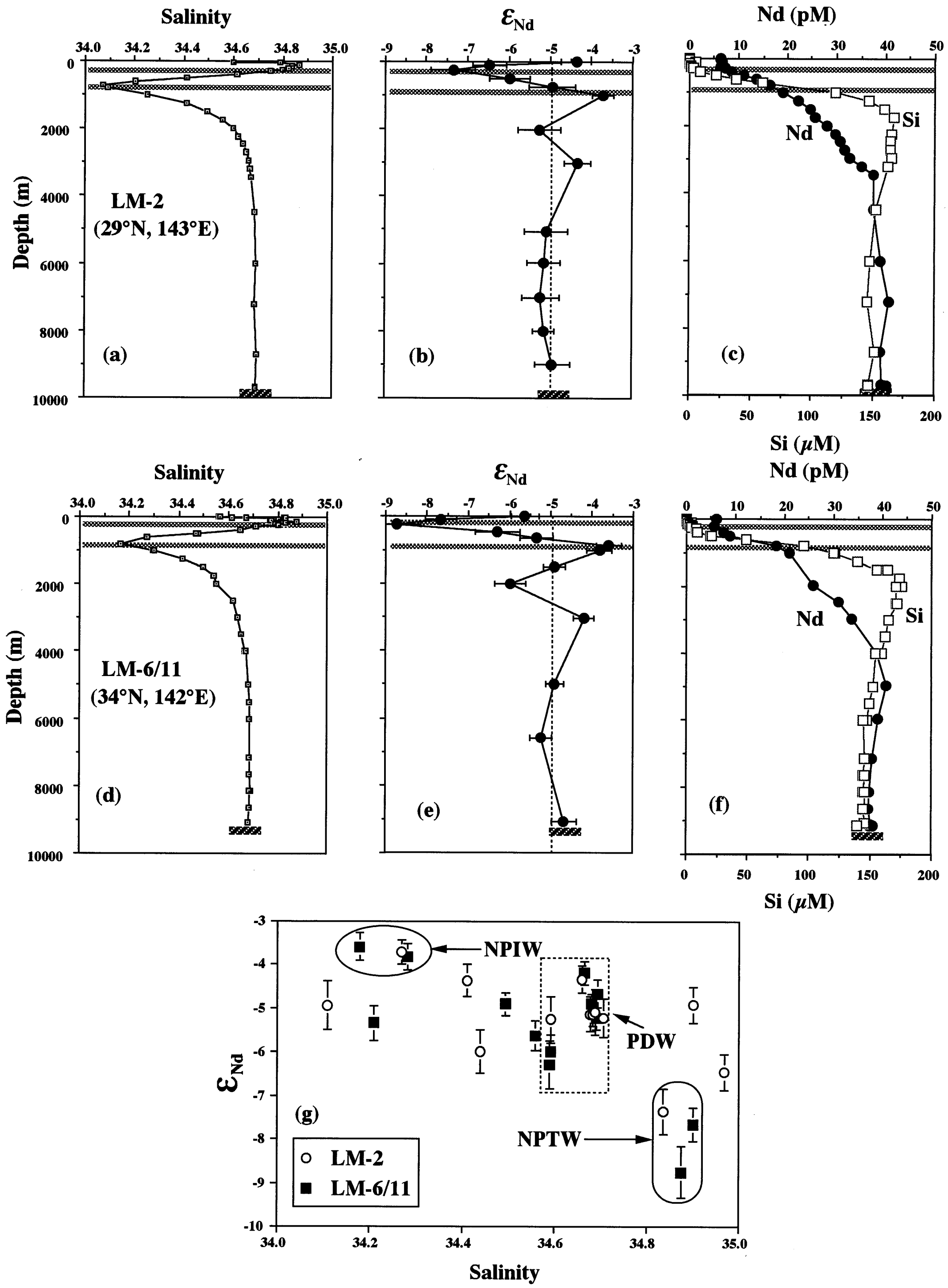

Fig. 4. Vertical profiles of salinity, Nd isotopic composition, Nd concentration and dissolved Si at LM-2 (a, b and c) and LM-6/11(d, e and f) in the Kuroshio Current regime. The Nd data of samples obtained by R.V. Tansei Maru at the same location as LM-2 are also plotted in (c). Note that Nd isotopic composition varies significantly, whereas Nd concentration shows a smooth and gradual increase with depth. The salinity-ϵ_{Nd} diagram of the above two stations is also shown in (g). NPIW and NPTW are identified in the diagram. The dot-dashed square indicates the region for PDW.

the Nd isotopic signatures in the deep water may be influenced by such lateral advection. Figure 5 indicates that the ϵ_{Nd} values of -3 to -4 between 1000 and 3000 m at TPS 24 (150°E) are systematically higher than those at LM-2 and LM-6/11, although the shapes of the profiles are somewhat similar to each other. Thus, it appears that the deep advective flow centered around 2000 m is limited to the regions close to the eastern topographic boundaries of the Japanese islands and the Izu-

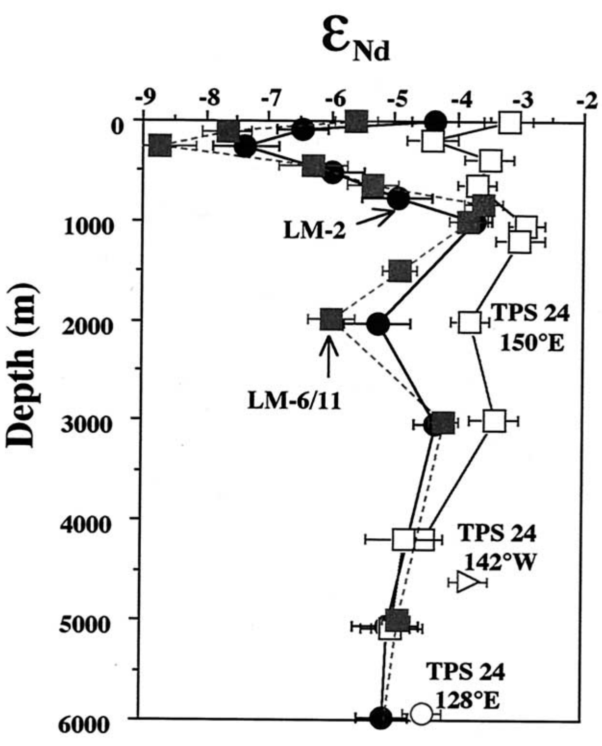

Fig. 5. Comparison of ϵ_{Nd} profiles obtained in this work with that at TPS 24 (Fig. 1; Piepgras and Jacobsen, 1988) in the subtropical region.

Ogasawara Ridge, and does not directly affect the central subtropical gyre.

The Pacific Deep Water (PDW) below ~4000 m is fed from the South Pacific along the western boundary by the northward flow ultimately originating from the Antarctic Bottom Water (AABW). The Nd isotopic composition (ϵ_{Nd}) is uniform at −5 for the PDW. This value is more radiogenic than those for the AABW in the South Pacific. Piepgras and Jacobsen (1988) obtained an ϵ_{Nd} value of −8.1 at 20°S, 160°E for the deep water from 4500 m, for example. The LM-2 and LM-6/11 stations are located in the deepest parts of the Izu-Ogasawara Trench and the data extend down to the bottom at depths greater than 9000 m. The waters inside the trench also have ϵ_{Nd} values of around −5, suggesting that the trench waters are well mixed and renewed rapidly by the PDW, as observed in an earlier study (Nozaki et al., 1998).

5.3. Radiogenic Nd in the Subarctic Region

The vertical profiles of Nd concentration and its isotopic composition at CM-5 (the subarctic region) and LM-9 (Kuroshio/Oyashio mixed water region) are shown in Figure 6, together with those of potential temperature, salinity and dissolved Si.

The vertical profile of the Nd isotopic composition at CM-5 (Fig. 6b) is simple compared to those in the subtropical Kuroshio region. The Nd isotopic ratios range only 2 in ϵ unit irrespective of salinity (Fig. 6g). The ϵ_{Nd} values of −2.8 at the surface and −3.6 at 49 m are more radiogenic than those of the Kuroshio surface waters, but are comparable to the surface ϵ_{Nd} of −3.2 at the subtropical TPS 24 (150°E) station (Piepgras and Jacobsen, 1988). Lacan and Jeandel (2001) also obtained values of −2.4 to −3.0 for the surface waters in the eastern equatorial Pacific. This agreement is probably fortuitous because the salinity and temperature characteristics are completely different among those oceanic sites. Piepgras and Jacobsen (1988) obtained a high ϵ_{Nd} value of −0.1 for surface water at the TPS47 (161°E) station approximately 560 km northeast of CM-5 (Fig. 7). The radiogenic Nd may have been derived from weathering sources such as volcanic rocks in the Kuril-Kamchatska and Aleutian Island chain, and can influence the surface waters of the western North Pacific by lateral mixing at various degrees depending on the location (Sholkovitz et al., 1999). Volcanic rocks in those islands have high ϵ_{Nd} values ranging from +6.7 to +10.1 (McCulloch et al., 1981; Zhuravlev et al., 1987). It appears that the surface waters in the entire North Pacific have generally more radiogenic Nd than those in the western North Pacific where the influence of Asian continent may be marked.

There is a temperature minimum of 1.65°C at 103 m at CM-5, which is typical of the Oyashio Current and the Okhotsk Sea, and ϵ_{Nd} is highest at −2.4. Again, the radiogenic ϵ_{Nd} values in the subsurface waters may be imprinted by local volcanic sources. The ϵ_{Nd} value decreases gradually with depth to −3.9 at 5000 m. Two explanations are proposed for the systematic decrease of ϵ_{Nd} with depth. One is that there are weak but nevertheless significant influences of the AABW component that increase toward the bottom. Another is that less radiogenic Nd is supplied from bottom sediments to the overlying water. Jones et al. (1994) reported ϵ_{Nd} values ranging from −5.2 to +3.8 for sediments in this region. If lower values are adopted for the site, the sediment release of Nd can reduce ϵ_{Nd} in the water column. More work is needed to resolve these issues.

The subarctic water ϵ_{Nd} values are invariably more radiogenic than −5, which is typical of subtropical deep waters (Fig. 4). This implies that ϵ_{Nd} is laterally inhomogeneous in the western North Pacific. Such inhomogeneity may arise from different circulation regimes of PDW, although we have no evidence to support it. Except for the surface samples, the ϵ_{Nd} data at CM-5 are in rough agreement with those at TPS 47 (Fig. 7).

5.4. Nd Profiles in the Kuroshio/Oyashio Mixed Water Region

Figure 6 (e, d and f) shows the vertical profiles at LM-9. This station is located in the MWR between the Kuroshio Extension and the Oyashio Front. The NPIW is formed in this region by complex mixing of various source waters originating from the Oyashio, Kuroshio and Tsugaru Currents. The ϵ_{Nd} value is somewhat low at −4 in the surface water presumably due to coastal influence, but is nearly constant at −3.2±0.2 in the subsurface waters down to 829 m from which the newest NPIW is formed (Talley, 1993). This value is slightly more radiogenic than the −3.7 in the NPIW in the subtropical stations, as discussed earlier. Thus, the Nd isotopic composition of the newest NPIW appears to be altered by vertical mixing with less radiogenic Nd in the waters above and below its trajectory. This is consistent with the observation of an increase in salinity

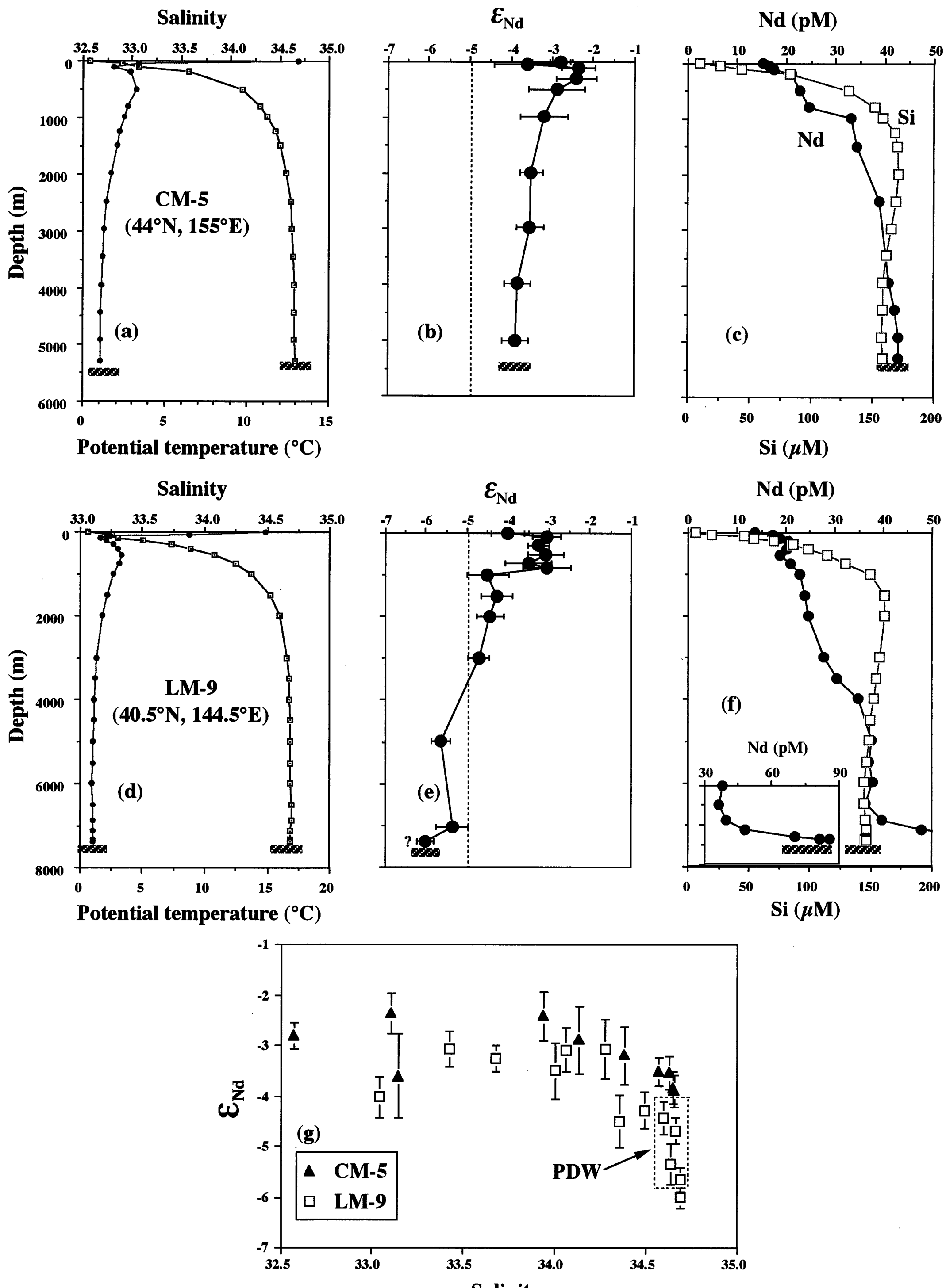

Fig. 6. Vertical profiles of potential temperature, salinity, Nd isotopic composition, Nd concentration and dissolved Si at the subarctic station, CM-5 (a, b and c), and the Kuroshio/Oyashio mixed water region station, LM-9 (d, e and f). The near-bottom high values in the inset in (f) are likely to be affected by resuspended sediments. The salinity-ϵ_{Nd} diagram of the above two stations is shown in (g). The region for NPTW in Figure 4g is not found in (g).

along the potential density surface of $\sigma_\theta = 26.8$ (see Talley, 1993). The ϵ_{Nd} values for the Kuroshio waters near Japan (see Table 1) range from -5.6 to -8.7, and are significantly less radiogenic than those for the newest NPIW. The Tsugaru Warm Water originates from the Japan Sea flowing eastward through the 130-m-deep Tsugaru Strait and contributing to NPIW. Although direct measurements of Nd isotopic composition are not available, it is likely to have an ϵ_{Nd} value of around -7.3,

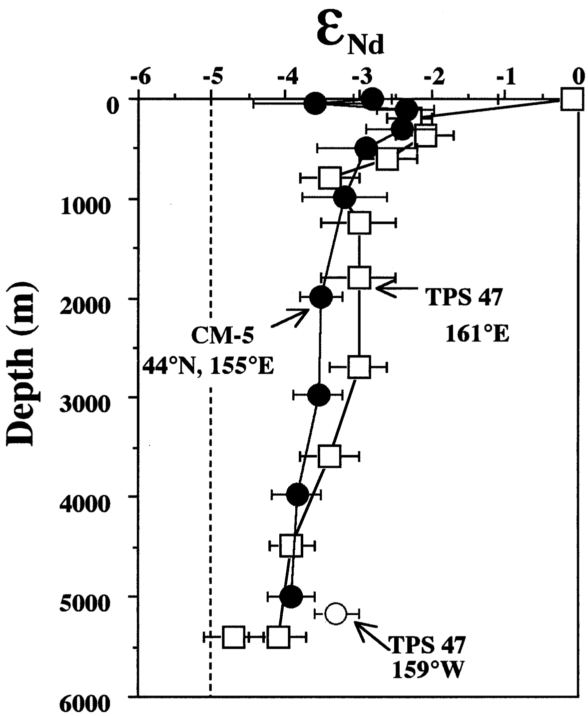

Fig. 7. Comparison of ϵ_{Nd} profiles obtained at CM-5 and TPS 47 (Piepgras and Jacobsen, 1988) in the subarctic region.

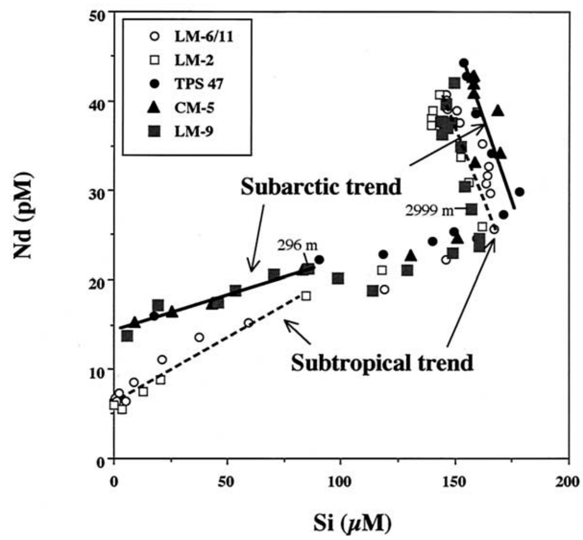

Fig. 8. Dissolved silica versus Nd diagram, showing separate trends for subtropical and subarctic stations. Note that the LM-9 data obtained at depths less than 1000 m are grouped in the subarctic trend, whereas those obtained at depths greater than 3000 m are close to the subtropical trend.

which is the mean value of our unpublished data for the Tsushima Current in the Japan Sea. Therefore, the radiogenic component of the NPIW must come from the Oyashio Current. The Oyashio water comprises the subarctic water and the Okhotsk Sea water (Fig. 1; Kawai, 1972). The subarctic waters with salinity <34.0 psu have ϵ_{Nd} values ranging from -0.1 to -3.6 with an average of -2.2 ± 1.0 (Table 1). Our unpublished value for the Okhotsk Sea water is -3.6, identical to that of NPIW; therefore, its contribution as a radiogenic component in the Nd isotopic composition of NPIW can be ruled out. This implies that the radiogenic Nd ultimately derived from such volcanic provinces as the Kuril-Kamchatska and Aleutian Islands is transported southwestward by the Oyashio Current to form the NPIW in the MWR immediately east of Japan. This radiogenic Nd isotopic signature may be useful as a tracer of vertical and horizontal mixing at the isopycnal surface of $\sigma_\theta =$ 26.8 for NPIW in the North Pacific subtropical gyre.

The ϵ_{Nd} value decreases sharply with depth from -3.1 at 829 m to -4.5 at 988 m. Between 988 m and 3000 m, ϵ_{Nd} values fall within a narrow range of -4.3 to -4.7, which are comparable to those in the subtropical LM-2 and LM-6/11 stations except for the values at ~2000 m, but are significantly lower than those of subarctic regions (Fig. 7). In the deep water, the two ϵ_{Nd} values of -5.7 at 4988 m and -5.4 at 7020 m are less radiogenic but roughly comparable to those in the subtropical stations, suggesting that the water is fed from the south. Figure 8 shows a diagram of Nd versus dissolved Si. The data can be separated into two trends for the subtropical and subarctic regimes. The LM-9 data obtained at depths less than ~300 m fall in the subarctic trend, whereas those obtained at depths greater than 3000 m fall in the subtropical trend. This is consistent with the arguments based on Nd isotopes given above.

6. SUMMARY AND CONCLUSION

The North Pacific is surrounded by geologically young volcanic provinces that can supply rare earth elements including Nd to the ocean; therefore, the waters in the basin generally have ϵ_{Nd} values ranging from ~0 to -4. The dissolved Nd isotopic compositions in the North Pacific are more radiogenic than $\epsilon_{Nd} = -8 \pm 2$ for the Indian Ocean and $\epsilon_{Nd} = -12 \pm 2.5$ for the Atlantic Ocean. There are at least two sources of less radiogenic Nd to the western North Pacific region. One is the Kuroshio Current that carries Nd with $\epsilon_{Nd} < -7$ at its salinity maximum (originating from NPTW), which is probably imprinted by sediment contact around the East China Sea continental shelf. The other is the northward flow of bottom water with $\epsilon_{Nd} < -5$ along the western boundary, which is influenced by the component of AABW from the South Pacific. There may be a third advective component at ~2000 m for less radiogenic Nd in the region close to Japan and the Izu-Ogasawara Ridge, although its source has yet to be identified. In contrast, the Oyashio Current carries radiogenic Nd of $\epsilon_{Nd} >$ -3 to the Kuroshio/Oyashio mixed water region to form NPIW that prevails in the large area of the subtropical region at depths 300 to 1000 m. These flow patterns produce significant spatial variability in the Nd isotopic composition in the western North Pacific that cannot be elucidated from the Nd concentration or some other oceanographic properties. Thus, the Nd isotopic composition serves as a novel tracer of ocean circulation and more intensive measurements are strongly recommended in the future.

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