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Neodymium isotopic composition and concentration in the Southwest Pacific Ocean

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We report two vertical profiles of the concentration and isotopic composition of neodymium (Nd) in seawater samples collected at 20°S and 30°S along a longitude of 170°W in the Southwest Pacific Ocean. At depths below 500 m, Nd isotopic composition values are less radiogenic than those obtained from the subtropical North Pacific. The minimum ε_{Nd} values of –8.0 and –6.9 at depths of 800 to 1000 m correspond to Antarctic Intermediate Water (AAIW). The differences in ε_{Nd} values between upper deep water (2000–3000 m) and lower deep water (>4000 m) seem to be associated with Upper and Lower Circumpolar Deep Waters (UCDW and LCDW, respectively). The radiogenic Nd supply to AAIW from the Society Islands is estimated to be 30 ± 22 tons Nd/yr, which is consistent with the estimates from previous studies conducted in the same location and the oceanic region close to the Hawaiian Islands. Our study demonstrates the uniqueness of Nd isotopic composition distributions in the Southwest Pacific Ocean. These results may be valuable for tracing present and past ocean circulation in this region.

Keywords: Southwest Pacific, Nd isotopic composition, Antarctic Intermediate Water (AAIW), Upper and Lower Circumpolar Deep Waters (UCDW and LCDW), GEOTRACES

INTRODUCTION

The Atlantic, Indian, and Pacific Oceans are connected through the Southern Ocean by the thermohaline circulation (THC) (Broecker and Peng, 1982). In the Southern Ocean, the Antarctic Circumpolar Current (ACC) flows eastward around the Antarctic. Circumpolar Deep Water (CDW), which is the most voluminous water mass that contributes to the ACC in the southern Pacific and Indian Oceans. The CDW is not formed at the surface; instead, CDW is a mixture of various water masses including Antarctic Deep Water (AADW) and North Atlantic Deep Water (NADW) (Emery, 2002; Talley *et al*., 2003). In the Antarctic Polar Front zone, the water mass called Antarctic Intermediate Water (AAIW) that has a low temperature and salinity is formed through the process of subduction (Talley, 1996). The AAIW flows to the southern part of each ocean and progresses northward, and CDW flows to the Pacific and Indian Oceans (Schmitz, 1996). The flow pathways and fluxes of AAIW and CDW could be affected by changes in THC, which is strongly associated with changes in the ocean and climate (e.g., Rutberg *et al*., 2000; Pahnke *et al*., 2008). Understanding the circulation patterns of the AAIW and CDW from the past through the present, therefore, may provide valuable information on THC and climate changes. The South Pacific, South Atlantic, and southern Indian Oceans may be key oceanic regions to monitor and record such changes because of their proximity to the regions where AAIW and CDW form.

The isotopic composition of Nd is generally expressed as $\varepsilon_{Nd} = [(^{143}Nd/^{144}Nd)_{Sample}/(^{143}Nd/^{144}Nd)_{CHUR} - 1] \times 10^4$, where CHUR stands for Chondritic Uniform Reservoir and represents a present day average earth value. The accepted CHUR value $(^{143}Nd/^{144}Nd)_{CHUR}$ is 0.512638 (Jacobsen and Wasserburg, 1980). The $^{143}Nd/^{144}Nd$ isotopic ratio in rock varies because of the radioactive decay of ^{147}Sm to ^{143}Nd (half-life = 1.06×10^{11} yr). The typical ranges of ε_{Nd} in old continental rock and mantle-derived igneous rock are –40 to –10 and 0 to +10, respectively (Jeandel *et al*., 2007). The Nd contained in rock is supplied to seawater by processes such as river input, remobilization from sediment deposited on the ocean margins, and dust input. The isotopic signals from these

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inputs are imprinted to seawater with little isotopic fractionation from the original sources (Frank, 2002; Lacan and Jeandel, 2005). Therefore, the seawater Nd isotopic composition varies primarily because of the range of ε_{Nd} values in the Nd source. In the ocean, the Nd isotopic composition is not fractionated by biological activity or particulate scavenging, and the composition is largely controlled by mixing of water masses or external fluxes of Nd with different isotopic compositions (Amakawa *et al*., 2004b). The short residence time of Nd (300–500 yr) (Tachikawa *et al*., 2003; Amakawa *et al*., 2004a; Arsouze *et al*., 2009) relative to ocean mixing (1500 yr) (Broecker and Peng, 1982) keeps the Nd isotopic composition from completely homogenizing, and its isotopic composition remains variable. The Nd isotopic composition of seawater can thus be considered a "quasi-conservative" tracer for water mass circulation (Arsouze *et al*., 2010). Neodymium data have been widely applied in studies of present and past oceanic circulation (e.g., Piepgras and Wasserburg, 1987; Jeandel, 1993; Rutberg *et al*., 2000; Amakawa *et al*., 2004a, b; van de Flierdt *et al*., 2004; Piotrowski *et al*., 2005; Pahnke *et al*., 2008). Some of these studies were carried out to reveal ocean-climate changes during glacial and interglacial periods through a focus on specific water masses such as NADW (Rutberg *et al*., 2000; Piotrowski *et al*., 2005) and AAIW (Pahnke *et al*., 2008). The discussion in these studies is based heavily on knowledge of ocean circulation patterns revealed by seawater Nd isotopic distributions in the present ocean (Piepgras and Wasserburg, 1987; Jeandel, 1993; Goldstein and Hemming, 2003). Therefore, available data on seawater Nd isotopic compositions are gradually increasing in response to the demands for paleoceanographic research, as well as for chemical oceanographic research (Lacan *et al*., 2012). However, few data are available for some regions of the ocean, particularly for oceans south of 30°S (Carter *et al*., 2012; Lacan *et al*., 2012; Stichel *et al*., 2012).

In the South Pacific, Piepgras and Wasserburg (1982) reported the first Nd isotopic depth profile for a western oceanic station. Although the profile has only three data points (30 m, 2800 m, and 4500 m), the range in values is about 8 epsilon units (ε_{Nd} of –8.1 to +0.3), which is larger than that observed for profiles in the North Pacific (less than 5 epsilon units for each profile) (Piepgras and Jacobsen, 1988; Amakawa *et al*., 2004b, 2009; Zimmermann *et al*., 2009). Although Lacan and Jeandel (2001) presented one profile for the equatorial South Pacific and found that AAIW shows a characteristic ε_{Nd} value of –8.0 ± 0.3, they did not report data obtained at depths below 1000 m. Therefore, neither of the previous studies presented a detailed full-depth profile of the Nd isotopic composition in the South Pacific. Recently, Carter *et al*. (2012) reported six depth profiles in the eastern Pacific sector of the Southern Ocean, which demonstrated homogeneity of the CDW (ε_{Nd} = –8.7 ± 0.1). However, a detailed full-depth profile of Nd isotopic composition in the central to western South Pacific has not been reported to date. Models of the Nd isotopic composition distribution in the ocean have been successful at producing results comparable with observations (Arsouze *et al*., 2007, 2009, 2010; Jones *et al*., 2008; Rempfer *et al*., 2011), but few studies have focused on the Nd isotopic distribution in the South Pacific. These facts are surprising because the Nd isotopic composition of the South Pacific may yield clearer information on AAIW or CDW because of the South Pacific's less complicated water mass structure compared with the South Atlantic (Schmitz, 1996; Stramma and England, 1999) where there is a strong influence of NADW that originates from the North Atlantic. Profile studies in the North Pacific have detected no present deep-water formation in that region (Piepgras and Jacobsen, 1988; Amakawa *et al*., 2004b, 2009).

The aims of this study are 1) to obtain the first full-depth profiles of Nd isotopic composition from the Southwest Pacific, 2) to compare these profiles with hydrographic parameters, and 3) to clarify characteristics of the Nd isotopic distributions in these areas. We find a difference in Nd isotopic composition between the upper deep layer (2000–3000 m) and the lower deep layer (below 4000 m), which correspond approximately with that between the upper and lower Circumpolar Deep Water (UCDW and LCDW, respectively). In addition, we discuss an observed shift to radiogenic AAIW in terms of external Nd sources. The South Pacific Nd isotopic datasets presented in this study can be used as a valuable reference for paleoceanography and model simulations.

Regional Hydrography of the Study Area

The sample locations for this study and several previous studies are shown in Fig. 1a along with oceanic circulation patterns in the South Pacific Ocean (Figs. 1b to 1d). The South Equatorial Current (SEC) and the East Australian Current (EAC) are located between the surface and intermediate depths (800–1000 m) (Fig. 1b) (Sokolov and Rintoul, 2000). Off the east coast of Australia, the SEC flows westward on the surface and then, turns into the EAC that flows southward along the coast. Between 30 and 40°S, the EAC leaves the coastal zone and flows eastward (Sokolov and Rintoul, 2000). At intermediate depths, the water column is comprised of the AAIW. Here, a portion of the AAIW flows further eastward to around 90°W and then, turns westward until it reaches the coast of Papua New Guinea (Fig. 1b). The other part of the AAIW flows along a path similar to that of the SEC/EAC along the east coast of Australia, and the water mass circulates between 20 and 40°S (Sokolov

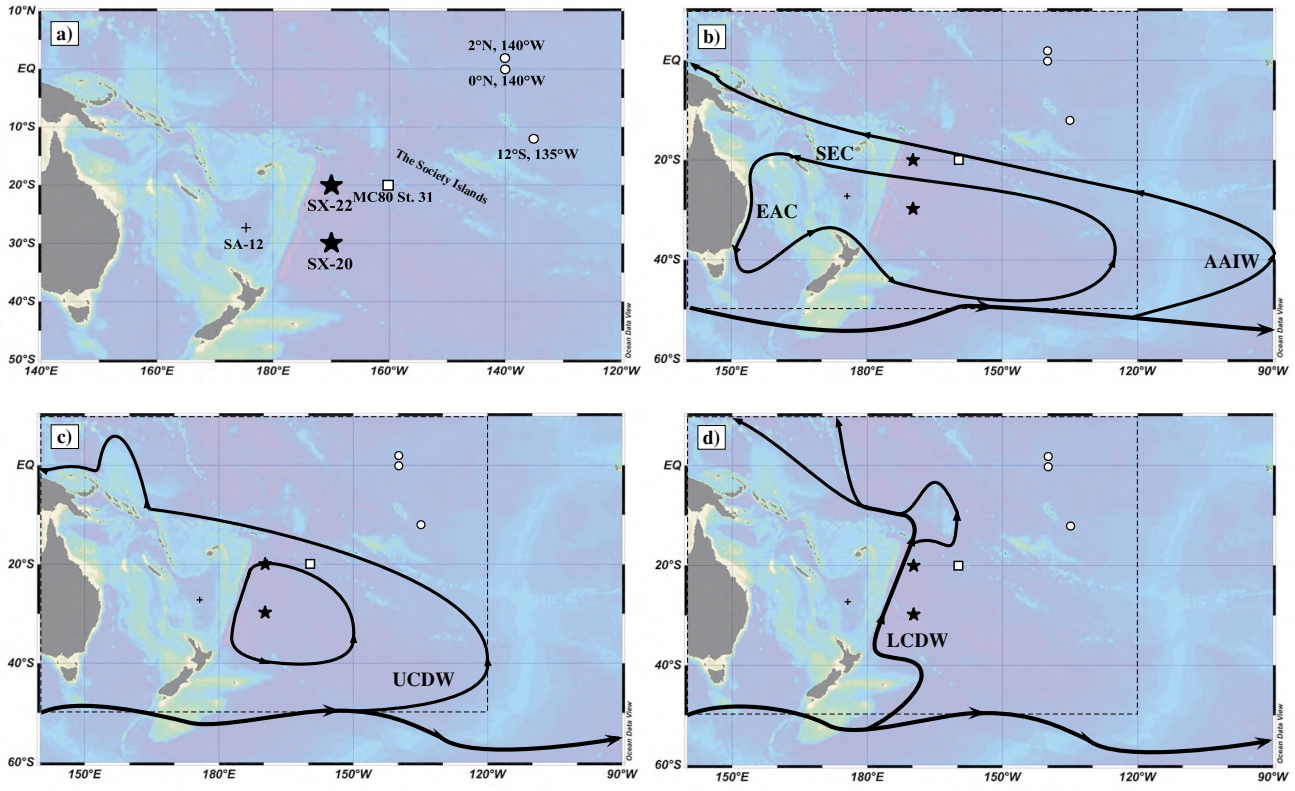

Fig. 1. Sample location map (a) and ocean circulation patterns in the South Pacific (b)–(d) (Kawabe and Fujio, 2010). (a) The sample locations in this study (SX-20 and SX-22; stars) are shown along with the locations studied by Piepgras and Wasserburg (1982) (an open square) and Lacan and Jeandel (2001) (open circles). The sampling location with neodymium (Nd) concentrations only (SA-12; cross) is also shown (Zhang and Nozaki, 1996). (b) Circulation patterns in the surface to intermediate depth (~800 m), (c) upper deep depth (2000 to 3000 m), and (d) lower deep depth to bottom (below 3000 m). The dashed line indicates the area depicted in (a).

and Rintoul, 2000; Kawabe and Fujio, 2010). The upper deep layer (2000–3000 m) in this region is dominated by UCDW that originates in the Southern Ocean. The UCDW, which is characterized by an oxygen minimum derived from source waters in the eastern South Pacific and western Indian Oceans (Callahan, 1972), flows eastward then, turns northwestward at around 120°W, eventually reaching the equator (Fig. 1c). The main water mass in the lower deep layer (3000 m to bottom) is LCDW that originates from the NADW in the Southern Ocean by mixing with other water masses such as Weddell Sea Deep Water (van Aken *et al*., 2004). Between 160°E to 170°W, LCDW meanders northward to the equator (Fig. 1d).

Samples and Methods

Samples were collected during the "Southern Cross—II (SX)" Expedition (KH-04-5) from the R.V. Hakuhou-Maru from January to February 2005 at two stations, SX-20 (30°S, 170°W) and SX-22 (20°S, 170°W), that were located within the Southwest Pacific Basin (Fig. 1a). This expedition was carried out as a precedent to the GEOTRACES program in Japan. The GEOTRACES program is an international effort to study the marine biogeochemical cycles of trace elements and their isotopes (van de Flierdt *et al*., 2012).

Seawater samples for Nd concentration measurements were collected using acid-cleaned 12-L Niskin-X bottles with external springs and Teflon-coated interior walls. The bottles were mounted on a 36-position conductivity, temperature, and depth (CTD) profiler (Sea-Bird 911 plus, Sea-Bird Electronics Inc.) and Carousel Water Sampler (Sea-Bird 32, Sea-Bird Electronics Inc.), and suspended from a titanium armored cable. After sampling, the seawater was filtered through Stera disk membrane filters (Kurabo Industries Ltd.) with 0.2 μm pores and acidified to pH < 2 with ultrapure 6M HCl (Tamapure-AA-100). For Nd isotopic measurements, seawater samples were collected using an acoustically triggered large-volume polyvinyl chloride (PVC) water sampler (N12-1000, Nichiyu-Giken Kogyo Co. Ltd.) equipped with four 250-L bottles.

Table 1. Nd concentration in South Pacific seawaters

Depth (m)	Salinity	Pot. Temp. (θ) (°C)	Pot. Dens. (σ_θ)	SiO_2 (μmol/kg)	Nd* (pmol/kg)
SX-20 (January 26–27, 2005; 30°00′ S 170°00′ W; Depth, 5394 m)					
0	35.696	23.700	24.26	—	2.50
11	35.697	23.735	24.25	—	4.64
30	35.697	23.731	24.25	—	2.60
49	35.697	23.694	24.26	—	2.21
100	35.642	18.175	25.72	—	2.53
149	35.565	16.827	25.99	—	3.41
200	35.477	16.034	26.11	—	3.51
398	34.753	10.714	26.63	3.2	5.10
598	34.443	7.395	26.92	6.0	6.62
796	34.359	6.196	27.02	11.0	6.77
993	34.340	4.969	27.16	25.4	7.70
1485	34.525	2.778	27.53	78.5	9.78
1978	34.626	2.120	27.66	114.1	11.9
2468	34.652	1.820	27.71	126.3	13.4
2959	34.674	1.547	27.75	131.9	14.3
3447	34.716	1.266	27.80	113.2	18.0
4473	34.712	0.663	27.84	121.7	23.0
4909	34.710	0.613	27.84	123.7	24.3
5390	34.709	0.604	27.84	124.4	25.4
SX-22 (January 31, 2005; 20°00′ S, 170°00′ W; Depth, 5328 m)					
11	35.134	28.533	22.32	—	2.56
99	35.519	24.439	23.90	—	4.71
199	35.644	21.078	24.96	—	4.11
398	34.998	13.294	26.33	2.4	3.66
595	34.414	7.288	26.92	8.4	6.09
793	34.371	5.288	27.14	26.9	6.74
992	34.433	3.864	27.35	54.5	9.16
1486	34.590	2.487	27.60	101.4	10.0
1978	34.633	2.069	27.67	119.0	14.9
2961	34.672	1.518	27.75	135.4	19.1
3939	34.720	0.920	27.83	115.0	26.9
4913	34.710	0.619	27.84	123.0	25.5
5287	34.709	0.610	27.84	123.7	23.1

**Uncertainties in concentration (2σ) are 3%.*

Neodymium concentrations were determined by the isotope dilution (ID) method (Amakawa *et al*., 2009). A ^{145}Nd enriched spike was added to seawater samples (100–500 mL). Iron was also added to the samples followed by the addition of ammonia for iron co-precipitation. Before the addition of ammonia, seawater samples were shaken multiple times for more than three days to ensure that samples and spikes were equilibrated. Sample Nd was then purified through an anion-exchange column. The concentration of Nd in the procedure blank was 8 pg, which corresponded to less than 7% of the sample Nd levels. Concentration data (Table 1) were corrected for the procedure blank. Neodymium concentrations were determined using an inductively coupled plasma mass spectrometer (ICP-MS) (HP 4500, Hewlett Packard Co.) in the Atmosphere and Ocean Research Institute at the University of Tokyo. The long-term reproducibility of the procedure was 3% (2σ) (Amakawa *et al*., 2009).

Analyses of Nd isotopic composition were performed on unfiltered water samples. The presence of particles should not affect the measured seawater Nd isotopic composition because the particle fraction of Nd does not exceed 5% of the total Nd at the open ocean with low particle concentrations (Bertram and Elderfield, 1993; Alibo and Nozaki, 1999; Lacan *et al*., 2012).

The overall procedure for Nd isotopic measurements was described in Amakawa *et al*. (2009). Briefly, Nd in 20–40 L of seawater was co-precipitated with iron hydroxide and then purified through successive ion-exchange columns and a reverse phase chromatographic column (Ln resin column, LN-C50-A, Eichrom Technologies Inc.). The procedure blank for Nd isotopic composition analysis was negligible (120 pg or less than 1% of the Nd content of samples). The purified Nd was loaded on a rhenium double filament assembly and its isotopic composition was measured as Nd^+ with a thermal ionization mass spectrometer (Finnigan MAT262, Thermo Fisher Scientific Inc.) that was located in the Atmosphere and Ocean Research Institute at the University of Tokyo. The measured $^{143}Nd/^{144}Nd$ ratios were normalized to $^{146}Nd/^{144}Nd = 0.7219$. The $^{143}Nd/^{144}Nd$ ratio of the JNdi-1 standard from the Geological Survey of Japan was measured at 0.512105 ± 0.000013 (2σ, $n = 40$), which is in close agreement with the reported value of 0.512115 ± 0.000006 for JNdi-1 (Tanaka *et al*., 2000).

The reliability of our data was verified by the GEOTRACES intercalibration program for Nd isotopes and concentrations (van de Flierdt *et al*., 2012).

RESULTS

Measurements of potential temperature and salinity collected by the CTD profiler are plotted in Fig. 2. Concentrations and isotopic data for seawater Nd, combined with hydrographic data and nutrient data (SiO_2), are presented in Tables 1 and 2. Depth profiles of the SX stations are shown in Fig. 3.

θ-S diagrams

The similar θ-S curves collected at stations SX-20 and SX-22 indicate that there is a close hydrographic correspondence at these stations. South Pacific Subtropical Water (θ of 18–20°C, salinity of ~35.7) occupies the near-surface regions of both stations (Zhang and Nozaki, 1996). At mid depths (800–1000 m), AAIW (θ of 5–6°C, salinity of 34.3–34.5) occupies the water column (Fig. 2a) (Tsuchiya and Talley, 1996; Lacan and Jeandel, 2001). Circumpolar Deep Water from the Southern Ocean underlies AAIW. As shown in Fig. 2b, the CDW is further divided into two sections: the upper section (i.e., UCDW)

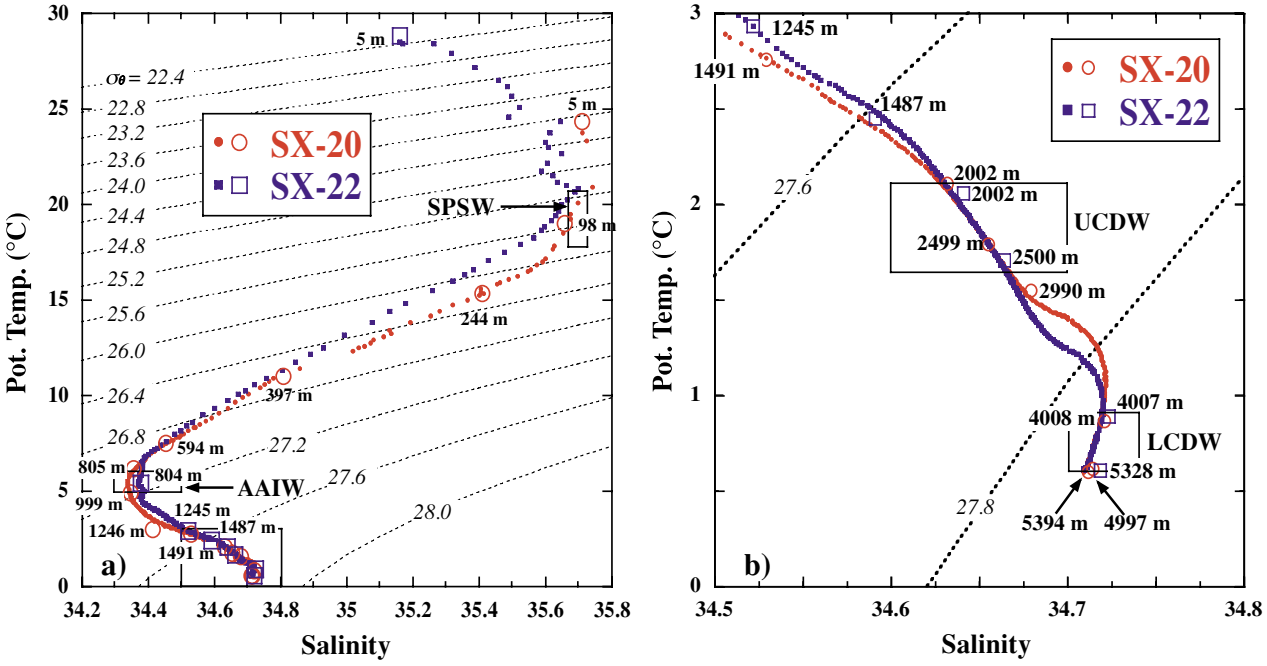

Fig. 2. (a) θ (potential temperature) - S (salinity) diagram plots for stations SX-20 and SX-22. Data from the conductivity, temperature, and depth (CTD) profiler are shown with small symbols. Samples for neodymium (Nd) isotopic composition determinations (large-volume samples) are shown with the large symbols marked by depth. The dotted lines show potential density curves (σ_θ = 22.4–28.0). Boxes indicate the compositions of South Pacific Subtropical Water (SPSW) and Antarctic Intermediate Water (AAIW). (b) Bottom-deep part of (a). Boxes indicate the compositions of Upper and Lower Circumpolar Deep Waters (UCDW and LCDW, respectively).

(θ of 1.6–2.1°C, salinity of 34.6–34.7) that is characterized by low oxygen and high nutrient concentrations and the lower section (i.e., LCDW) (θ of 0.6–0.9°C, salinity of >34.7) that is characterized by high salinity (Orsi *et al.*, 1995, 1999; Moffat *et al.*, 2009; Horikawa *et al.*, 2011). In the subpolar region of the South Pacific, Antarctic Bottom Water (AABW) that originated in the Weddell and Ross Seas is found. However, this water mass does not extend north of 60°S in the Pacific because of the topographic blocking by the Pacific-Antarctic ridge (Orsi *et al.*, 1999; Horikawa *et al.*, 2011). Consequently, LCDW dominates the lower deep and bottom portions of the South Pacific (Horikawa *et al.*, 2011).

Nd concentrations and isotopic composition

Neodymium concentration profiles at the SX stations are shown in Fig. 3. Both SX stations show similar profiles and concentration ranges (at SX-20 Nd concentration is 2.5–25.4 pmol/kg, at SX-22 Nd concentration is 2.6–26.9 pmol/kg) with low values at the surface and high values at greater depths. Similar results were reported by a previous study at a neighboring station (SA-12) (Zhang and Nozaki, 1996).

Depth profiles of the Nd isotopic composition at stations SX-20 and SX-22 are also presented in Fig. 3. The SX-20 and SX-22 depth profiles are characterized by maxima at the surface (ε_{Nd} = –1.9 at SX-20 and ε_{Nd} = –2.3 at SX-22) and gradual decrease to depths around 1000 m where AAIW occupies the water column. However, over the same depth interval, station SX-22 has a profile with more radiogenic ε_{Nd} values (e.g., ε_{Nd} of –6.9 ± 0.3 at 804 m) than those observed at station SX-20 (e.g., ε_{Nd} –8.4 ± 0.3 at 805 m and –8.3 ± 0.3 at 999 m). At depths below 1000 m, the ε_{Nd} values of station SX-20 exhibit an increasing trend to depths around 2500 m where UCDW is found. Then, the ε_{Nd} values sharply decrease to approximately –9 at depths >2500 m where LCDW is the major water mass. For station SX-22, although data for depths between 3000 and 4000 m are not available, a large difference between depths above and below 3000 m is also observed, which may correspond to differences between UCDW and LCDW. Both profiles show constant values (ε_{Nd} of –9.2 to –8.6) at depths below 3000 m.

Discussion

Nd concentrations and isotopic composition distributions in the Southwest Pacific

Figure 4 shows Nd concentration profiles at the SX stations together with data from adjacent stations includ-

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

Depth (m)	Salinity	Pot. Temp. (θ) (°C)	Pot. Dens. (σ_θ)	$^{143}Nd/^{144}Nd$[c]			ε_{Nd}[d]		
SX-20 (January 25–26, 2005; 30°00′ S 170°00′ W; Depth, 5394 m)									
5[a]	35.71	24.3	24.09	0.512543	±	0.000005	–1.9	±	0.3
98	35.657	18.97	25.53	0.512477	±	0.000015	–3.1	±	0.3
244	35.409	15.30[b]	26.22	0.512429	±	0.000011	–4.1	±	0.3
397	34.811	10.99	26.63	0.512337	±	0.000012	–5.9	±	0.3
594	34.454	7.50	26.92	0.512315	±	0.000010	–6.3	±	0.3
805	34.357	6.16	27.02	0.512206	±	0.000013	–8.4	±	0.3
999	34.347	4.91	27.17	0.512215	±	0.000015	–8.3	±	0.3
1246	34.414	2.97	27.42	0.512246	±	0.000016	–7.6	±	0.3
1491	34.529	2.76	27.53	0.512308	±	0.000018	–6.4	±	0.3
2002	34.632	2.11	27.67	0.512305	±	0.000016	–6.5	±	0.3
2499	34.655	1.79	27.71	0.512399	±	0.000011	–4.7	±	0.3
2990	34.679	1.55	27.75	0.512176	±	0.000023	–9.0	±	0.4
4008	34.721	0.87	27.83	0.512168	±	0.000013	–9.2	±	0.3
4501	—	0.87	—	0.512190	±	0.000016	–8.7	±	0.3
4997	34.714	0.62	27.84	0.512184	±	0.000016	–8.9	±	0.3
5394	34.712	0.60	27.84	0.512199	±	0.000013	–8.6	±	0.3
SX-22 (January 30–31, 2005; 20°00′ S, 170°00′ W; Depth, 5328 m)									
5[a]	35.16	28.8	22.25	0.512519	±	0.000004	–2.3	±	0.3
804	34.376	5.40	27.13	0.512287	±	0.000013	–6.9	±	0.3
1245	34.522	2.93	27.51	0.512288	±	0.000015	–6.8	±	0.3
1487	34.591	2.45	27.61	0.512355	±	0.000012	–5.5	±	0.3
2002	34.641	2.06	27.68	0.512401	±	0.000016	–4.6	±	0.3
2500	34.664	1.71	27.73	0.512386	±	0.000012	–4.9	±	0.3
4007	34.723	0.89	27.83	0.512179	±	0.000015	–9.0	±	0.3
5328	34.718	0.61	27.84	0.512195	±	0.000011	–8.6	±	0.3

[a]*Data from Tazoe et al. (2013).*
[b]*Deduced from CTD data.*
[c]*Errors are $2\sigma_m$ (internal errors).*
[d]*Errors are based on 2 sigma standard errors of repeated JNdi-1 runs (see text) except for SX-22 2990 m datum (calculated from the internal error).*

ing the Sagittarius Expedition (SA)-12 (27°16′ S, 175°25′ E) from Zhang and Nozaki (1996) and the Mar. Chem. (MC) 80 St. 31 (20°00′ S, 159°59′ W) from Piepgras and Wasserburg (1982). The Nd concentration vertical profile is controlled by sources of Nd and various processes in the water column including scavenging (Byrne and Kim, 1990; Bertram and Elderfield, 1993; Zhang and Nozaki, 1996; Alibo and Nozaki, 1999; Nozaki and Alibo, 2003). At depths below 1500 m, the Nd concentrations (12–27 pmol/kg) are lower than those found in the subtropical North Pacific (30–57 pmol/kg at the Bootes Expedition (BO)-3 (30°N, 160°W) and BO-5 (20°N, 175°W)) (Amakawa *et al.*, 2009), which is consistent with previous studies that found that water aging (the age of North Pacific deep water is greater than that of the South Pacific) increases Nd concentrations similar to SiO_2 (Jeandel, 1993; Goldstein and Hemming, 2003). Data collected previously at station MC80 (20°S) covered only three depths (30 m, 2800 m, and 4500 m), and the results show a profile similar to that of SX-22 at the same latitude. At depths above 3000 m, station SA-12 in the South Fiji Basin (Zhang and Nozaki, 1996) also exhibit a profile similar to SX-22. The station SX-20 has lower Nd concentrations than those of the other stations at depths between 2000 and 4000 m (Fig. 4), which is presumably associated with strong Nd scavenging at this station. Neodymium concentrations at SX-22 are observed to decrease with depth below 4000 m, which likely result from active boundary scavenging at the sediment-water interface (Nozaki and Alibo, 2003).

The Nd isotopic profiles of the SX stations and previous findings from the southwestern and equatorial Pacific (Piepgras and Wasserburg, 1982; Lacan and Jeandel, 2001) are plotted in Fig. 5a. The range of ε_{Nd} values found for the SX stations (–9.0 to –1.9) is similar to that of previously studied stations (–8.1 to +0.3). Lacan and Jeandel

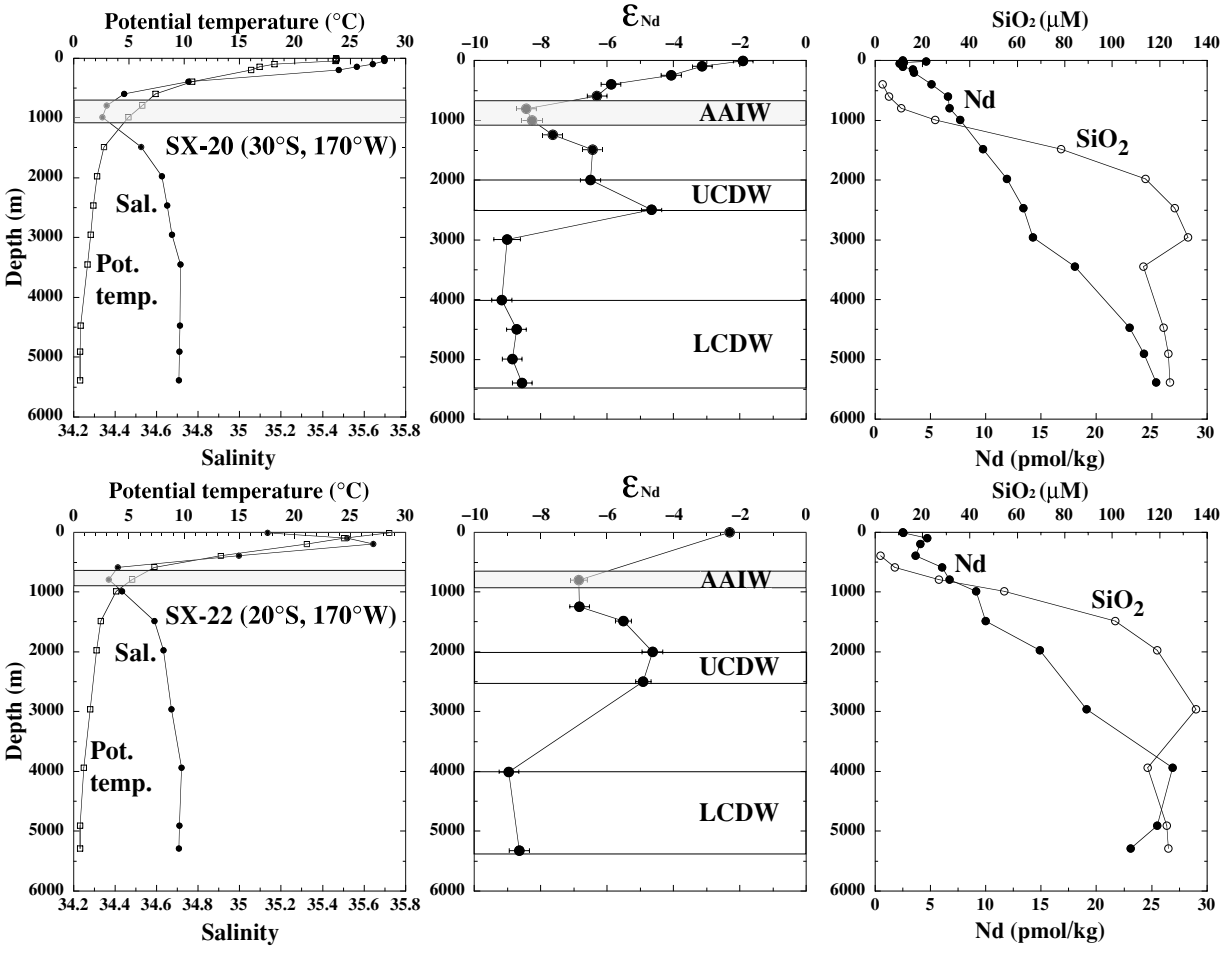

Fig. 3. Depth profiles of salinity, potential temperature, Nd isotopic composition, Nd concentrations, and SiO_2 concentrations at stations SX-20 and SX-22. Uncertainties in Nd concentrations are about 3%.

(2001) reported an ε_{Nd} value of –8.0 ± 0.3 for AAIW based on a datum collected at one South Pacific station from 835 m (12°S, 135°W), and this value is similar to measurements obtained at station SX-20 from 805 m (–8.4 ± 0.3) and 999 m (–8.3 ± 0.3). In the Pacific, AAIW originates in the southeastern Polar Front zone (Talley, 1996). Between the southeastern oceanic region and the station at 12°S (Fig. 1b), there is almost no land area to supply external Nd in amounts that could modify the Nd isotopic signal of AAIW. Therefore, the ε_{Nd} value of AAIW at the 12°S station most likely retains that derived from the formation region. The similar ε_{Nd} value of AAIW at station SX-20 may imply that the Nd isotopic signal of the original AAIW is also preserved at this station, even if the flow paths of AAIW at those stations are somewhat different. As discussed in Subsection "Nd concentrations and isotopic composition", the station SX-22 has more radiogenic values at the depth interval of 804 m (–6.9 ± 0.3) than values from comparable depths at the other station (–8.4 ± 0.3 at 805 m). This indicates that there is a radiogenic Nd supply to AAIW along its flow path to station SX-22.

Piepgras and Wasserburg (1982) reported two data points for depths between 2500 and 5000 m (–4.5 ± 0.7 for 2800 m and –8.1 ± 0.6 for 4500 m), and those values are consistent with the profile observed at the SX-20 station that shows changes at depths from about 3000 to 4000 m. As mentioned above, such changes are also observed between the depths above 3000 m and below 4000 m for station SX-22 (–4.9 ± 0.3 for 2500 m and –9.0 ± 0.3 for 4007 m). As shown in Fig. 2b, UCDW and LCDW occupy, respectively, the upper (2000–3000 m) and lower (>4000 m) portions of the deep depths at the SX stations. The range of ε_{Nd} values for UCDW and LCDW are –6.5 to –4.6 and –9.2 to –8.6, respectively, which is indicative of a clear difference between those water masses. This observation, which is consistent with SiO_2 profiles that show lower values at depths between 3000 and 4000 m than those at shallower and deeper depths (Fig. 3), has not been previously reported in the North Pacific (Fig.

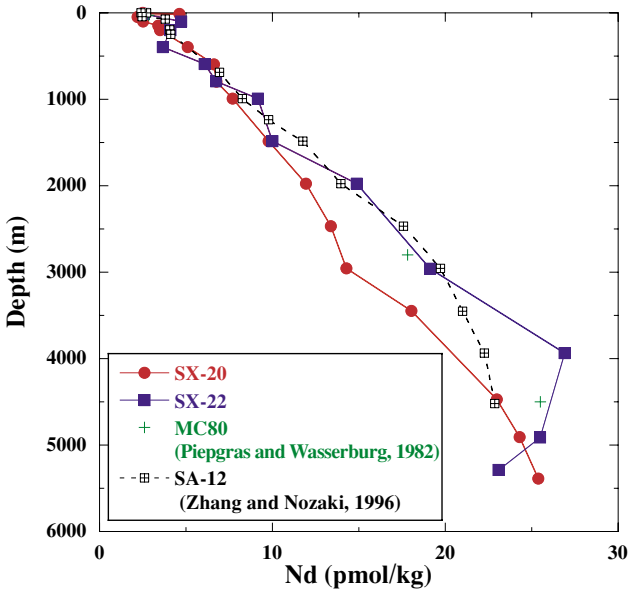

Fig. 4. Depth profiles of Nd concentrations in the South Pacific. The data from Piepgras and Wasserburg (1982) and Zhang and Nozaki (1996) are also plotted.

5b). It is interesting to note that one seawater sample from the SX-20 station (2990 m) with hydrographic parameters close to UCDW had an $\varepsilon_{Nd} = -9.0 \pm 0.4$, which is indistinguishable from the value for LCDW (Fig. 3). We propose that a local Nd input with an unradiogenic signal ($\varepsilon_{Nd} < -9$) could be the reason for the observed ε_{Nd} value because such a signal would shift the Nd isotopic composition of the water mass without changing the hydrographic parameters (Lacan and Jeandel, 2005).

The Nd isotopic data of the SX stations are plotted along with two subtropical North Pacific datasets (station BO-5 at 20°00′ N, 175°00′ W and station 7 at 24°15′ N, 170°20′ E) in Fig. 5b. The data from BO-5 and station 7 are from Amakawa *et al.* (2009) and Zimmermann *et al.* (2009), respectively. Except for depths above 500 m, the subtropical North Pacific profiles are more radiogenic than those of the SX stations. However, the differences in ε_{Nd} values are not constant throughout the depths. Large differences exist at depths around 1000 m and below 3000 m, and smaller differences are observed at depths between 1500 and 2500 m. The upper deep part (2000–3000 m) of the South Pacific is under a stronger influence of return flow to the Southern Ocean from the North Pacific than the lower deep part (>4000 m) (Schmitz, 1996; Horikawa *et al.*, 2011). Hence, this flow shows a more radiogenic value than the water masses in the South Pacific, which may increase the ε_{Nd} of the UCDW and further result in the close ε_{Nd} values of the North and South Pacific stations at depths between 1500 and 2500 m.

Comparison of Nd isotopic profiles from the Southern Ocean with adjacent oceanic regions

Figure 5b shows that the profiles in the South Pacific are quite different from those in the North Pacific. Here, we compare the profiles with those in the eastern Pacific and Atlantic sectors of the Southern Ocean in addition to adjacent oceanic regions to clarify the characteristics of the water masses observed in the Southwest Pacific (AAIW, UCDW, and LCDW).

In Fig. 5c, the data from the SX stations are plotted together with data from the Southern Ocean north of 65°S and its adjacent oceanic regions in the South Atlantic (Jeandel, 1993; Carter *et al.*, 2012; Stichel *et al.*, 2012). Station 022 (60°32′ S, 108°18′ W) located between the sub-Antarctic front and polar front in the eastern Pacific sector of the Southern Ocean has a homogeneous profile ($\varepsilon_{Nd} = -8.8$ to -8.4) at depths occupied by CDW (1504 to 2503 m), which is a typical feature in the Southern Ocean (Piepgras and Wasserburg, 1982; Carter *et al.*, 2012; Stichel *et al.*, 2012). This implies that the ε_{Nd} difference between UCDW and LCDW at the SX stations was not observed at St. 022. Carter *et al.* (2012) claimed that rapid horizontal advection results in the homogeneity of the CDW Nd isotopic signal in the Pacific sector of the Southern Ocean. The different ε_{Nd} values between UCDW and LCDW observed at the SX stations suggest that such a mechanism does not effectively operate to homogenize the Nd isotopic signal of UCDW and LCDW at those stations, presumably because of the long distance from the polar region.

Profiles collected at five stations in the Atlantic sector of the Southern Ocean (the South Atlantic Ventilation Experiment Expedition (SAVE) 271, Stations 101/102, and 104) and the South Atlantic (SAVE217 and SAVE302) are shown in Fig. 5c (Jeandel, 1993; Stichel *et al.*, 2012). At the same water depths, most of the ε_{Nd} values are less radiogenic than those of the SX stations. This matches the general trend observed for the ε_{Nd} values of the Pacific, Atlantic, and Southern Oceans (Lacan *et al.*, 2012). Four of the five profiles show no clear change at depths below 2500 m, which is different from the profiles observed at the SX stations. This can be attributed to efficient vertical and horizontal homogenization of the CDW Nd isotopic composition in the Southern Ocean as was pointed out by Stichel *et al.* (2012). A clear change at depths below 2500 m is only found in the profile from SAVE302 (33°15′ S, 41°47′ W). At 3156 m, the ε_{Nd} value at SAVE302 is –11.4, while at 3937 m the ε_{Nd} value is –7.8 in the Southwest Atlantic (Jeandel, 1993). Such a change was not observed for the Southeast Atlantic station, SAVE217 (30°S, 1°25′ W). The study by Jeandel (1993) assigned NADW to the water mass present at the 3156 m depth and LCDW to the water mass at lower depths. The difference between NADW and LCDW may

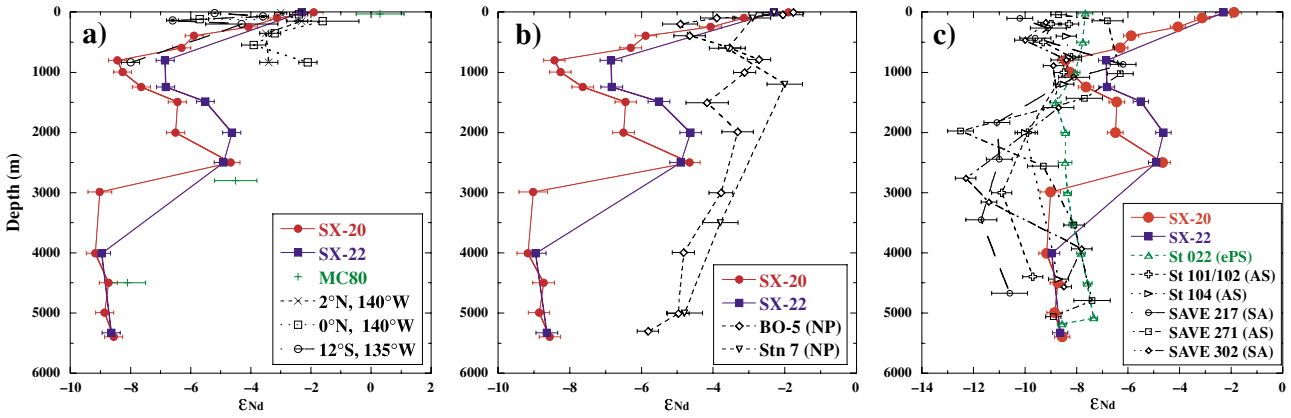

Fig. 5. (a) Depth profiles of Nd isotopic composition in the South Pacific. The data at MC80 and 140°–135°W are from Piepgras and Wasserburg (1982) and Lacan and Jeandel (2001), respectively. Note that our data are consistent with previous studies for depths around 1000 m (corresponding to AAIW) and 3000 m (corresponding to UCDW). (b) Comparison of Nd isotopic profiles between the SX stations and those of the subtropical North Pacific ("NP"). The data at BO-5 (20°N, 175°W) and St. 7 (24°15′N, 170°20′E) are from Amakawa et al. (2009) and Zimmermann et al. (2009), respectively. (c) Comparison of Nd isotopic profiles between the SX stations and those of the eastern Pacific sector of the Southern Ocean ("ePS"), Atlantic sector of the Southern Ocean ("AS"), and South Atlantic ("SA"). The data at St. 022 (60°32′S, 108°18′W), St. 101 (42°21′S, 8°59′E), St. 102 (44°40′S, 7°6′E), St. 104 (47°40′S, 4°16′E), SAVE 217 (30°S, 1°25′W), SAVE 271 (49°39′S, 33°23′W), and SAVE 302 (33°15′S, 41°47′W), are from Carter et al. (2012), Stichel et al. (2012), and Jeandel (1993).

be similar to that between UCDW and LCDW observed for the SX stations. The difference in deep depth Nd isotopic profiles observed between the Southeast (SAVE217) and the Southwest (SAVE302) Atlantic can result from different deep ocean circulation patterns between those oceanic regions as was suggested by Stramma and England (1999). Their study showed that the major southward transport in the South Atlantic is mainly due to the Deep Western Boundary Current (DWBC), and partly due to the eastern boundary current. This implies that the Southwest Atlantic is under a stronger influence of NADW than the Southeast Atlantic. As we discussed in the previous section, there is a possible strong influence of North Pacific Deep Water (NPDW) at the depths occupied by UCDW for the SX stations. In the oceanic regions that show clear ε_{Nd} differences between UCDW and LCDW, such differences may be caused by the strong influence of water masses from the north that have distinctive ε_{Nd} values from the water mass in the Southern Ocean. For example, in the South Pacific the NPDW has a ε_{Nd} = –3.9 (Amakawa *et al.*, 2009), and in the South Atlantic the DWBC associated with NADW has a ε_{Nd} = –13.5 (Piepgras and Wasserburg, 1987). Both of these values differ from values in the Southern Ocean (ε_{Nd} = –9 to –8).

The difference in ε_{Nd} values between UCDW and LCDW found in the present South Pacific may have important implications for interpreting Nd isotopic compositions of paleo-proxy samples such as fish teeth or ferromanganese crusts. Horikawa *et al.* (2011) reported ε_{Nd} data for core-top fossil fish teeth collected at depths around 3000 m in the South Pacific, which could be the boundary depth between the UCDW and LCDW as mentioned above. The implications based on past ε_{Nd} data of fossil fish teeth collected at those depths would be misleading without accurate water depth information on the time of deposition. We suggest that special care should be taken when interpreting ε_{Nd} data of paleo-proxy samples collected at those depths in the South Pacific.

Figure 5c shows a very interesting Nd isotopic feature at depths around 1000 m where AAIW occupies the water column of the SX stations. Except for three stations, SX-22, SAVE217 (30°S, 1°25′ W), and SAVE 271 (49°39′ S, 33°23′ W), the ε_{Nd} profiles converge to a value of –9 to –8 at this depth. Although AAIW is found at depths of 400 m at Station 104, the ε_{Nd} value (–8.4 ± 0.4) (Stichel *et al.*, 2012) is indistinguishable from that observed at depths around 1000 m at SX-20 (–8.3 ± 0.3 at 999 m). Stichel *et al.* (2012) reported that the average ε_{Nd} value of AAIW in the Atlantic sector of the Southern Ocean is –8.1 ± 0.2, which is similar to the AAIW value found at the SX-20 station. This fact is very important because the AAIW in the South Pacific and South Atlantic are considered to have different origins. The former is thought to originate from surface waters in the southeastern Pacific, and the latter is thought to originate from surface waters in the northern Drake Passage and the Falkland Current loop (Talley, 1996). The similar ε_{Nd} values of AAIW at some stations in the South Pacific, Southern Ocean, and South Atlantic suggest that the ε_{Nd} values of

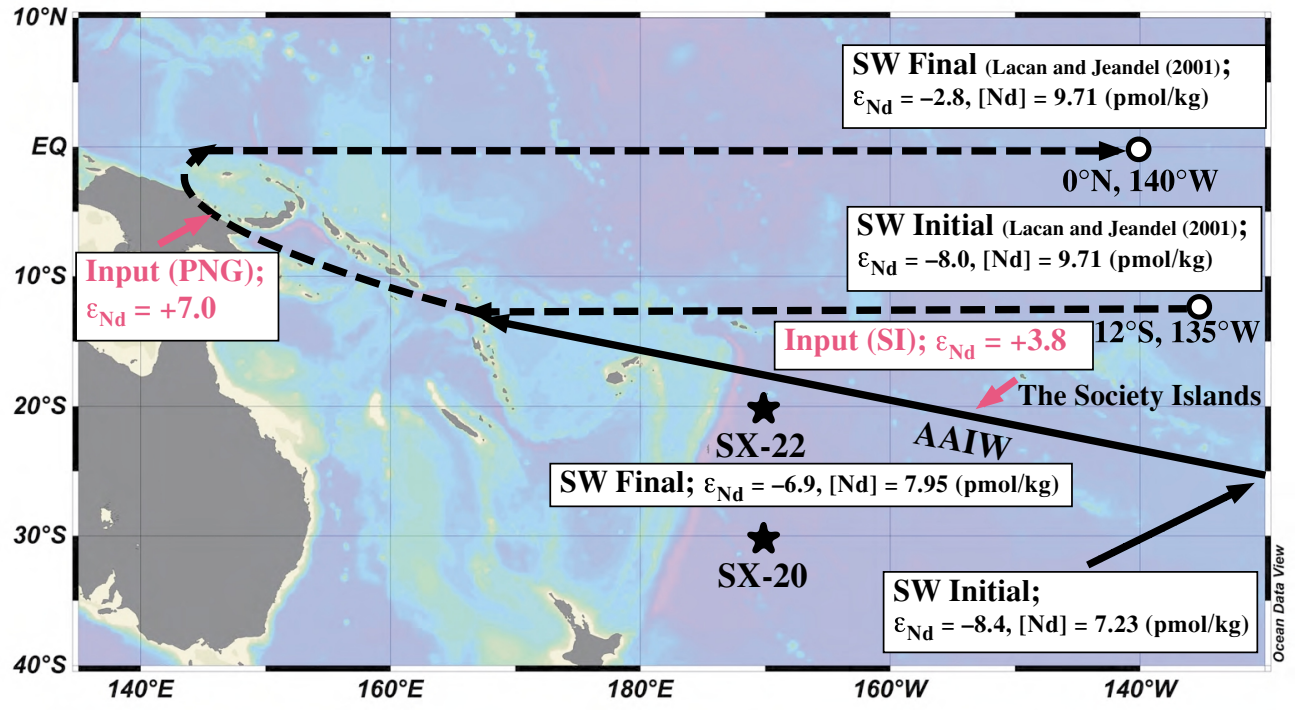

Fig. 6. Radiogenic Nd supply to the AAIW from islands in the South Pacific. The trajectory of AAIW (solid line) is the same as that shown in Fig. 1b. The terms SI and PNG stand for the Society Islands and Papua New Guinea, respectively. The dashed line shows the trajectory of the AAIW used for estimating the Nd flux from the PNG islands by Lacan and Jeandel (2005). We use the AAIW values at station SX-20 as the initial values to calculate the Nd flux from PNG as detailed in Table 3.

the original surface waters may be close to each other. Actually, the ε_{Nd} values for shallow water at depths less than 200 m at the northern Drake Passage ($\varepsilon_{Nd} = -8.1 \pm 0.4$) (Stichel *et al.*, 2012) and the southeastern Pacific sector of Southern Ocean ($\varepsilon_{Nd} = -7.7 \pm 0.3$) (Carter *et al.*, 2012) are close to each other. On the other hand, Rickli *et al.* (2009) and Stichel *et al.* (2012) have shown that there is a difference in ε_{Nd} values of AAIW in the South Atlantic and Atlantic Sector of the Southern Ocean. According to Rickli *et al.* (2009), the ε_{Nd} values of AAIW from the eastern Atlantic north of 25°S range from –11.5 to –11.1, which is less radiogenic than the above AAIW values (ε_{Nd} = –9 to –8). However, the reason for the discrepancy remains unexplained. Stichel *et al.* (2012) discussed the difference in AAIW values between their eastern South Atlantic stations (stations 101/102 and 104) and those reported by Jeandel (1993) (SAVE217 and SAVE271) (Fig. 5c). In that study the AAIW occupying the surface of SAVE271 (147 m) showed a slightly radiogenic ε_{Nd} value (=–6.8), and Stichel and co-authors suggested that a radiogenic Nd supply from volcanic rocks forming the South Sandwich Islands may shift the ε_{Nd} to more radiogenic values. However, they were unable to present a clear explanation for the radiogenic ε_{Nd} value at SAVE217 (ε_{Nd} = –6.2 at 869 m) located in the Southeast Atlantic. As the ε_{Nd} values of AAIW at stations 101/102 and 104 located near SAVE217 range from –8.4 to –8.2 (Stichel *et al.*, 2012), we think that there is likely a local radiogenic Nd source for the SAVE217 station that modifies the AAIW value.

The AAIW at station SX-20 ($\varepsilon_{Nd} = -8.3 \pm 0.3$) and the 12°S station ($\varepsilon_{Nd} = -8.0 \pm 0.3$) have similar values to the shallow seawater of St. 022 (503 m, $\sigma_\theta = 27.4$, $\varepsilon_{Nd} = -7.8 \pm 0.2$), which is located close to the formation region of AAIW in the Pacific. These data also support the conservative ε_{Nd} feature of AAIW in the South Pacific (see Subsection "Nd concentrations and isotopic composition distributions in the Southwest Pacific"), which does not seem to be the case for station SX-22 that shows a more radiogenic ε_{Nd} value for AAIW. Consequently, a radiogenic Nd supply from local sources is likely present during the pathway to station SX-22, which will be discussed in next section.

"Boundary Exchange" Nd flux for the AAIW

As shown in Fig. 3, the Nd concentration profile normally displays lower values at the surface and higher values at greater depths, indicating a residence time similar to nutrients such as SiO_2 (~10^4 yr). Conversely, the Nd isotope distribution in different oceans (Lacan *et al.*, 2012)

Table 3. Nd fluxes to AAIW from the Society Islands and Papua New Guinea estimated under the boundary exchange processes

Pathway	Initial		Final		ε_{Nd} of source[c]	Flow magnitude[d]	$F^{Addition}$	$F^{Removal}$
	ε_{Nd}[a]	[Nd][b] (pmol/kg)	ε_{Nd}[a]	[Nd][b] (pmol/kg)		(10^6 m^3/s)	(tons Nd/y)	(tons Nd/y)
East of 135°W to SX-22	–8.4	7.23	–6.9	7.95	+3.8	6.5 ± 1.5	30 ± 22	9 ± 4
SX-22 to 2°N	–6.9	7.95	–2.8	9.71	+7	3 ± 1	45 ± 24	21 ± 8

[a]*The initial and final Nd isotopic compositions for "East of 135°W (SX-20) to SX-22" are calculated from the samples collected at depths between 800 and 1000 m. The data for "2°N" are from Lacan and Jeandel (2005). The uncertainties of initial and final Nd isotopic compositions are 0.3 in epsilon units.*
[b]*The initial and final Nd concentrations are calculated from the samples collected at depths between 790 and 1000 m. The data for "2°N" are from Lacan and Jeandel (2005). The uncertainties of initial and final Nd concentrations are 3%.*
[c]*The source ε_{Nd} values for "East of 135°W to SX-22" and "SX-22 to 2°N" are from Cordier et al. (2005) and Lacan and Jeandel (2005), respectively.*
[d]*The flow magnitudes are from Kawabe and Fujio (2010) and Sloyan and Rintoul (2001) for "East of 135°W to SX-22", and Lacan and Jeandel (2005) for "SX-22 to 2°N".*

implies that the residence time of Nd is shorter than the ocean mixing time (~10^3 yr) (Broecker and Peng, 1982). This decoupled distribution of Nd concentrations and isotopic composition in the ocean has been referred as the "Nd paradox" (Goldstein and Hemming, 2003). This contradiction led Lacan and Jeandel (2005) to propose a concept called "Boundary Exchange" (BE) (Arsouze *et al.*, 2007), whereby an exchange of elements at a continental margin could modify the ε_{Nd} value of a water mass with or without changes in Nd concentrations.

A clear shift is observed in the ε_{Nd} values of AAIW between stations SX-20 and SX-22 (Fig. 5). According to the BE theory, there should be a radiogenic Nd flux along the AAIW flow path from the formation area of AAIW to station SX-22. We think that a possible radiogenic Nd source may be located close to station SX-22 because the ε_{Nd} value of AAIW at the 12°S station (Lacan and Jeandel, 2005) shows the same value as that at SX-20 (Fig. 1a). This is considered to be the same as the original value of AAIW in the South Pacific (see Subsection "Comparison of Nd isotopic profiles from the Southern Ocean with adjacent oceanic regions"). If the radiogenic Nd source is located to the east of 135°W, then the 12°S station would be expected to show a higher radiogenic value. However, this is not the case. Consequently, the Nd source is likely located west of 135°W. One candidate for the Nd source is the volcanic rocks of the Society Islands including Tahiti (Fig. 1a). Amakawa *et al.* (2009) has shown that volcanic islands like the Hawaiian Islands could be sources for seawater Nd through the BE process. Using the concepts of BE, we estimate the radiogenic Nd flux from the Society Islands.

According to Lacan and Jeandel (2005), the exchange flux from the Society Islands ($F^{Addition}$) and removal flux from the surface current ($F^{Removal}$) are defined as follows:

$$F^{Addition} = F_W \times [Nd]^{Initial} \times (\varepsilon^{Final} - \varepsilon^{Initial})/(\varepsilon^{Addition} - \varepsilon^{Final}) \quad (1)$$

$$F^{Removal} = F_W \times \{[Nd]^{Final} \times (\varepsilon^{Final} - \varepsilon^{Addition}) - [Nd]^{Initial} \times (\varepsilon^{Initial} - \varepsilon^{Addition})\}/(\varepsilon^{Addition} - \varepsilon^{Final}) \quad (2)$$

where F_W is the flow magnitude (m^3/s) of AAIW, and $[Nd]^{Final}$ and $[Nd]^{Initial}$ stand for Nd concentrations of the final water mass measurement (AAIW at SX-22) and the initial measurement (AAIW at SX-20), respectively, assuming that the parameters of AAIW to the east of 135°W are identical with those at the SX-20 station. The parameters ε^{Final}, $\varepsilon^{Initial}$, and $\varepsilon^{Addition}$ signify the final and initial values of ε_{Nd} and the flux from the Society Islands, respectively.

The flow magnitude of AAIW in the South Pacific is estimated to be 6.5×10^6 m^3/s (Sloyan and Rintoul, 2001; Kawabe and Fujio, 2010). We employ ε_{Nd} = +3.8 as a radiogenic source value, which is based on the average value of rocks from the Society Islands (Cordier *et al.*, 2005). The parameters used for the calculations presented above are shown in Fig. 6 and Table 3.

Table 3 shows the results of calculations for Nd flux from the Society Islands. In the same oceanic region, Lacan and Jeandel (2005) estimated the Papua New Guinea radiogenic Nd flux to AAIW and obtained a $F^{Addition}$ value = 70 ± 21 metric tons Nd/yr, which is of the same order of magnitude as our estimate ($F^{Addition}$ = 30 ± 22 tons Nd/yr). The latter flux is comparable to, but lower than, the radiogenic Nd flux from the Hawaiian Islands to the surface current ($F^{Addition}$ = 66–130 tons Nd/yr) that was estimated by the same method (Amakawa *et al.*, 2009).

We estimate the Nd flux from Papua New Guinea us-

ing our SX-22 AAIW values as the initial parameters and the equator values of Lacan and Jeandel (2005) as the final parameters. As shown in Table 3, the calculated $F^{\text{Addition}} = 45 \pm 24$ tons Nd/yr is somewhat smaller than the estimate by Lacan and Jeandel (2005). Lacan and Jeandel (2005) may slightly overestimate the flux by attributing too much flux to Papua New Guinea and neglecting fluxes from other volcanic islands in the region, such as the Society Islands.

A study on the distribution of the Nd isotopic composition in the oceans using an ocean circulation model indicates that a radiogenic Nd input to the deep Pacific Ocean is needed to reproduce the reported Nd isotopic distribution in the area (Jones *et al*., 2008). We think that an accurate estimation of radiogenic Nd flux from volcanic islands in the Pacific may provide a clue to the constrained Nd flux to the intermediate and deep ocean depths.

Summary and Conclusions

We have reported the first full-depth profiles of Nd isotopic composition from the Southwest Pacific Ocean, along with Nd concentrations. We find two characteristic features in the Nd isotopic profiles: a minimum ε_{Nd} value at depths around 1000 m and a difference in ε_{Nd} values between depths above 3000 m and depths below 4000 m. The former observation is likely due to the influence of AAIW, which shows a uniform isotopic signal in the South Pacific. The latter finding seems to correspond to a difference in ε_{Nd} values between the upper and lower CDW. We think that these characteristic features will be helpful for studies of past and present ocean circulation in the South Pacific.

Based on BE theory, we have estimated the radiogenic Nd flux from the Society Islands to AAIW to be 30 ± 22 tons Nd/yr. Although this value is smaller than a previous estimate for the radiogenic Nd flux from Papua New Guinea, it is on the same order of magnitude. This suggests that small islands in the South Pacific could be important Nd sources for water masses in the South Pacific.

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