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Isotopic Composition of Neodymium in Waters from the Drake Passage

Donald J. Piepgras and G. J. Wasserburg

Summary. The isotopic composition of neodymium has been determined in seawaters from the Drake Passage. The Antarctic Circumpolar Current, which controls interocean mixing, flows through this passage. The parameter $\epsilon_{Nd}(0)$, which is a function of the ratio of neodymium-143 to neodymium-144, is found to be uniform with depth at two stations with a value which is intermediate between the values for the Atlantic and the Pacific and indicates that the Antarctic Circumpolar Current consists of about 70 percent Atlantic water. Cold bottom water from a site in the south central Pacific has the neodymium isotopic signature of the waters in the Drake Passage. By using a box model to describe the exchange of water between the Southern Ocean and the ocean basins to the north together with the isotopic results, an upper limit of approximately 33 million cubic meters per second is calculated for the rate of exchange between the Pacific and the Southern Ocean. Concentrations of samarium and neodymium were also determined and found to increase approximately linearly with depth. These results suggest that neodymium may be a valuable tracer in oceanography and may be useful in paleo-oceanographic studies.

Studies of many oceanographic problems have been made with short-lived radionuclides such as ^{3}H, ^{14}C, ^{210}Pb, ^{226}Ra, and ^{230}Th. Because some of these nuclides have half-lives which are short relative to the time scales of the processes studied, they have been used, in conjunction with other hydrographic measurements, as tracers for studying oceanic circulation paths, mixing rates, and the chemical behavior and distribution of associated stable elements in seawater. One problem of interest to oceanographers is that of determining mixing rates in and between the oceans. Estimates of oceanic mixing rates are not well constrained, but ^{14}C studies (*1*) indicate that at least 1500 years are required for the exchange of deep water with the mixed layer. A longer time may be required for the exchange of deep waters between ocean basins. A minimum time of ~ 150 years to mix the world oceans is obtained by assuming that the Pacific Ocean is emptied by the flow through the Drake Passage and mixed with the Atlantic.

The isotopic compositions of elements connected to long-lived radioactive decay may also be useful as tracers for studying large-scale mixing in the oceans and chemical processes affecting their distribution in seawater if the elements have (i) isotopically distinct source regions and (ii) sufficiently short residence times in the water column compared to the time scale of interocean mixing. Analysis of deep-sea sediments showed that the lead, strontium, and neodymium which are supplied to the Atlantic Ocean from crustal rocks are isotopically distinct from those which are injected into the Pacific (*2–4*). The concentration of strontium in seawater [~ 8 parts per million (*5*)] is only a factor of 10 to 50 less than the average abundances in oceanic and continental crust. This indicates that Sr has a relatively long residence time (~ 10^{6} years) in seawater. As a consequence, $^{87}Sr/^{86}Sr$ is uniform in the different oceans (*6, 7*) even though there are several isotopically distinct sources of the element. Therefore, the Sr isotopic composition of seawater is not suitable as a large-scale oceanographic tracer but may be useful as a tracer in the immediate environment of Sr injection. The variations of $^{87}Sr/^{86}Sr$ observed in deep-sea sediments are then ascribed totally to the composition of the detrital material imparted at the source.

In contrast, the concentrations of Nd and Pb in deep waters are about 3×10^{-6} ppm (*4, 7, 8*, and this study) and 1×10^{-6} ppm (*9*), respectively. These are about 10^{-6} to 10^{-7} times the Nd and Pb concentrations in average oceanic and continental crustal rocks, indicating that these elements have very short residence times in seawater. In the case of Nd, the available data on rare earth elements in river water indicate that only a small proportion of the Nd weathered from crustal rocks enters rivers in solution (*5, 10*). However, Nd concentrations in rivers are 10 to 50 times those in seawater, which still implies a relatively short Nd residence time. Significant transport of rare earths to the oceans may be carried from the atmosphere by dust. As a result of the short residence times of Pb and Nd and their distinctive source characteristics, the oceans are not well mixed with respect to Nd and Pb isotopic compositions, indicating their usefulness as oceanographic tracers. In this article we discuss our results for Nd and its use in this way.

The abundance of ^{143}Nd in nature increases through geologic time due to the α-decay of ^{147}Sm (half-life $\tau_{1/2} = 1.08 \times 10^{11}$ years). Chemical fractionation of Sm and Nd during the formation of the continental crust relative to the depleted oceanic mantle results in differences in the isotopic composition of Nd in these lithic sources which reflect their age and Sm/Nd ratio (*11*). The change in ^{143}Nd is commonly related to the nonradiogenic isotope ^{144}Nd, and for convenience, because the differences are small, represented by $\epsilon_{Nd}(0) = [(^{143}Nd/^{144}Nd)_{measured}/(0.511847) - 1] \times 10^{4}$, where 0.511847 is the present-day average $^{143}Nd/^{144}Nd$ ratio in chondritic meteorites (*12*). These values reflect natural variations due only

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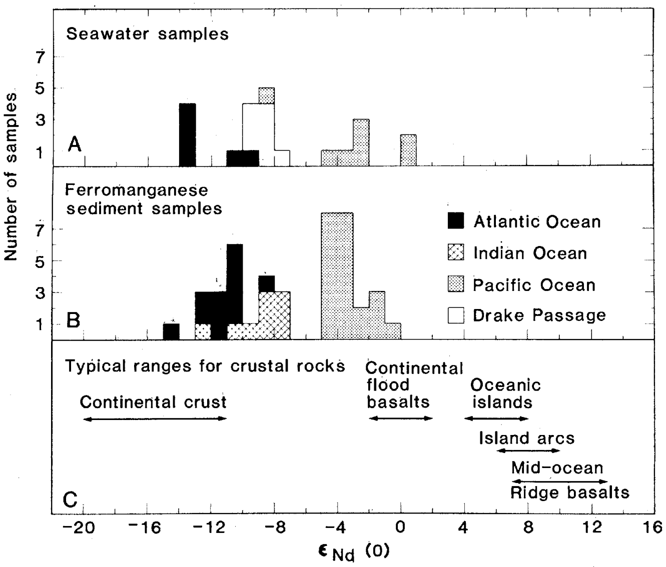

Fig. 1. Histograms of $\epsilon_{Nd}(0)$ values of (A) seawater and (B) ferromanganese sediments. Possible sources of rare earth elements in seawater and their typical $\epsilon_{Nd}(0)$ values are indicated in (C). Note the correspondence between the values of $\epsilon_{Nd}(0)$ in ferromanganese sediments and seawater from the respective oceans. Data for the Drake Passage are from this study. Atlantic seawater data are from Piepgras and Wasserburg (*7*). Pacific seawater data are from this study and Piepgras *et al.* (*4*). Ferromanganese sediment data are from Piepgras *et al.* (*4*), O'Nions *et al.* (*13*), Goldstein and O'Nions (*14*), and Elderfield *et al.* (*52*).

to the radioactive decay of ^{147}Sm and have been corrected for instrumental and natural mass fractionation. Typical ranges of $\epsilon_{Nd}(0)$ are shown in Fig. 1 for a variety of continental and oceanic crustal and mantle reservoirs which are potential sources of rare earth elements to the oceans. Neodymium isotopes measured in seawater would be blends of the Nd isotopic compositions from these reservoirs.

Recent studies have shown distinct differences in the ^{143}Nd/^{144}Nd ratio in seawater between the major ocean basins but only small differences within individual ocean basins (*4, 7*). This was inferred from measurements on ferromanganese sediments and direct measurements on seawater. O'Nions *et al.* (*13*) measured the Nd isotopic composition of ferromanganese sediments in the Pacific and Indian oceans. They observed variations between samples from these two oceans but concluded that this was due to a detrital component in their Indian Ocean sample and that the oceans were a relatively well-mixed reservoir for Nd with a ^{143}Nd/^{144}Nd ratio corresponding to $\epsilon_{Nd}(0) \approx -3$. Piepgras *et al.* (*4*) made isotopic measurements on ferromanganese sediments from the Pacific, Indian, and Atlantic oceans. They found differences in the Nd isotopic composition of the ferromanganese sediments and inferred that these differences were characteristic of the individual ocean basins. These results and data from O'Nions *et al.* (*13*) are shown in Fig. 1B. A recent study by Goldstein and O'Nions (*14*) of Nd isotopes in ferromanganese sediments obtained by a similar sampling of the oceans confirmed the results of Piepgras *et al.* (*4*). Direct Nd isotopic measurements in Pacific (*4*) and Atlantic (*7*) seawater (Fig. 1A) showed that the data on ferromanganese sediments for these oceans rather accurately reflected the Nd isotopic composition of modern seawater in the respective basins. There are no direct Nd isotopic measurements in Indian Ocean seawater, but a value for $\epsilon_{Nd}(0)$ can be inferred from ferromanganese sediment data. Estimated average $\epsilon_{Nd}(0)$ values for these oceans are: Pacific Ocean, -3; Indian Ocean, -8; and Atlantic Ocean, -12. Differences in $\epsilon_{Nd}(0)$ were also observed between surface water and deep water ($>$ 1000 meters) in the North Atlantic (*7*) and were attributed to the existence of different sources of Nd. On the average, however, $\epsilon_{Nd}(0)$ was found to be fairly uniform within individual ocean basins in comparison to the differences between basins. A comparison of the $\epsilon_{Nd}(0)$ values with those in possible sources of rare earth elements in seawater (Fig. 1C) shows that the Nd in seawater is derived predominantly from continental sources [$\epsilon_{Nd}(0) < 0$], with only small contributions from sources with oceanic mantle affinity [$\epsilon_{Nd}(0) \approx 10$]. The more negative $\epsilon_{Nd}(0)$ values observed in the Atlantic compared to the Pacific point to the dominance of an old continental source (having a low average Sm/Nd ratio) relative to young, mantle-derived materials for Nd injections into the Atlantic Ocean (see Table 1). This is in agreement with the fact that ~ 70 percent of the world continental drainage flows into the Atlantic.

Geological processes affecting Sm and Nd systematics have also resulted in chemical fractionation of U and Pb, causing differences in Pb isotopic compositions of various lithic sources. Thus there are several possible sources of Pb in seawater with distinctive isotopic compositions, and significant Pb isotopic variations in marine sediments have been observed. Chow and Patterson (*2*) found that Pb in Atlantic ferromanganese sediments was more radiogenic than that in Pacific sediments; this is consistent with an older continental source for the Atlantic Pb and is supported by the Nd results. Direct measurement of Pb isotopes in seawater is difficult because of contamination from industrial Pb during collection and handling of samples (*15*). There are no data for Pb isotopes in deep ocean waters. Data for Pb in seawater from coastal regions (*16*) and Pacific and Atlantic surface waters (*17*) reveal large variations in Pb isotopic composition which can be traced to anthropogenic input. The data base is insufficient for distinguishing Atlantic and Pacific characteristics, but recent anthropogenic effects have probably altered the natural distribution of Pb in surface seawater, making it difficult to compare these seawater measurements and data on deep-sea sediments.

Because of the difference in $\epsilon_{Nd}(0)$ between the Atlantic and Pacific oceans, it should be possible to use the Nd isotopic composition to monitor the exchange and mixing of water between these oceans. The Drake Passage is the only region where significant transport of water can take place between the Atlantic and Pacific basins, and it is presumed that some Pacific water is entrained into the Atlantic after entry through the Drake Passage, where there is a net eastward flow of water at ~ 130 sverdrups (1 Sv = 10^6 cubic meters per second) (*18*). We determined the isotopic composition of Nd in water flowing through the Drake Passage and compared it with results for the Atlantic and Pacific oceans. Because of the eastward transport through the Drake Passage, we expected to find Pacific signatures in the Nd isotopic composition of water flowing through this region. We will put forward some ideas derived from limited data on Nd isotopic variations which appear to be pertinent to problems of large-scale transport and mixing of the oceans. This represents an initial exploration of these problems with a new isotopic tracer. The data presented are a small supplement to the extensive and diverse observations made by oceanographers for many decades and can only be interpreted within the larger framework of oceanographic studies.

Sampling

Samples were collected during leg 11 of cruise 107 of the R.V. *Atlantis II* from vertical profiles of the water column at several stations in the Drake Passage and southeast Pacific. Water samples collected by K. Bruland from vertical profiles at two stations in the Central Pacific and material collected by Bruland from a sediment trap were analyzed in order to extend the data base and study the contribution of particles to the rare earth elements in seawater. Locations and depths for individual samples are shown in Table 1 and Fig. 2. Station 315 is south of the Antarctic polar front, which was located at about 58°S in the Drake Passage during this cruise (*19*). Station 327 is slightly to the north of the polar front.

Ten-liter samples were collected in Teflon-coated GO-FLO water samplers (*20*). They were transferred in a closed environment to cleaned polyethylene storage bottles and acidified with 25 milliliters of high-purity 10 normal HCl. Five-liter subsamples were analyzed for Sm and Nd concentrations and Nd isotopic composition. If only elemental concentrations were needed, the sample size could be reduced to 0.5 liter. The chemical separation of the rare earths and mass spectrometric techniques have been described (*4*, *7*). Samples collected by Bruland in the Central Pacific were filtered before analysis; the Drake Passage samples were analyzed without filtration. Data of Biscaye and Ettriem (*21*) for the North Atlantic show particle concentrations of 5 to 6 micrograms per liter averaged over the entire water column. Assuming an Nd concentration of 30 ppm in the particulate phase, about 200 picograms of Nd per liter of seawater would come from particles. As the total concentration of Nd in seawater is ten times larger than this, we assume it to be primarily in solution. Data for rare earths in pteropod tests in the South Atlantic (*22*) indicate much lower concentrations of Nd (< 5 ppm). The concentration of particles changes with depth, having a maximum at the surface and ocean bottoms which is four to five times the average (*21*). Therefore a substantial percentage of the Nd in near-surface waters could be adsorbed onto particles, but a correlation between particle distributions and Nd concentration changes with depth cannot be made at present. Although we have not compared filtered and unfiltered seawater samples from the same locations, Piepgras *et al.* (*4*) measured filtered and unfiltered samples from different locations in the Pacific and found similar concentrations; an unfiltered sample from the Galápagos Rise area had a slightly lower Nd concentration than filtered samples collected off California. Differences between filtered and unfiltered samples will need more attention and the chemical state of rare earth elements in seawater needs to be better defined, particularly with regard to the role of adsorption on sols and particles.

Results

Results of our Nd isotopic measurements on seawater samples from the Drake Passage and the Central Pacific are shown in Table 1 and Figs. 1 and 3. In addition to isotopic measurements, Sm and Nd concentrations were measured to ± 0.1 percent or better, and these results are shown in Table 1 and Fig. 4. These concentration data are also of importance in considering the origin, transport, and deposition of the rare earth elements.

The results for samples from station 315 show $\epsilon_{Nd}(0)$ values which are fairly uniform at all depths, ranging from −8.4 to −9.1 (Fig. 3A). Concentrations of Sm and Nd at station 315, however, increase with depth by a factor of ~ 2. Figure 4A shows that the Nd concentration increases approximately linearly with depth. The isotopic compositions are independent of concentration at this station. Station 327 is located north of the Antarctic polar front and two samples from this station have $\epsilon_{Nd}(0) = -9.2$ and −8.2. Measurements of the Nd concentration in three samples from station 327 show an approximately linear increase with depth (Fig. 4A) and a concentration gradient about a factor of 3 lower than that observed for station 315. Two samples from stations in the general vicinity of the Drake Passage were also analyzed (Fig. 3A). Station 261 is about 700 kilometers west of the coast of Chile in the Humboldt Plain and far north of the Drake Passage, and the sample has $\epsilon_{Nd}(0) = -7.9$. Station 292 is west of the Drake Passage in the Bellingshausen Plain within the Antarctic Circumpolar Current, and has $\epsilon_{Nd}(0) = -8.2$.

These data show that the Nd isotopic composition of seawater in the Drake Passage and in the deep waters of the Bellingshausen Plain is nearly uniform with depth and geographic location and that the concentration increases linearly with depth. The Antarctic polar front, which divides the transect along which the samples were taken, has no observable effect on the distribution of isotopic compositions of Nd in the Drake Passage. Further, the deep water in the Humboldt Plain northwest of the passage is indistinguishable in terms of Nd iso-

Table 1. Sample locations and results of Sm and Nd measurements in samples from the Drake Passage and the Pacific Ocean. Errors for concentration measurements are ~ 0.05 percent for Nd and ~ 0.1 percent for Sm. The $^{143}Nd/^{144}Nd$ ratio was determined in samples spiked with ^{150}Nd and normalized to $^{146}Nd/^{142}Nd = 0.636151$. Reported errors are 2 standard deviations from the mean.

Station	Depth (m)	Location	Nd (10^{-12} g/g)	Sm (10^{-12} g/g)	$\frac{^{147}Sm}{^{144}Nd}$	$\frac{^{143}Nd}{^{144}Nd}$	$\epsilon_{Nd}(0)$
315	50	61°01′10″S, 62°15′21″W	1.85	0.347	0.109	0.511381 ± 50	−9.1 ± 1.0
	800		2.30	0.432	0.113	0.511388 ± 39	−9.0 ± 0.7
	2000		3.25	0.610	0.113	0.511418 ± 38	−8.4 ± 0.7
	3600		4.21	0.800	0.115	0.511389 ± 21	−8.9 ± 0.4
327	50	56°27′55″S, 66°33′01″W	1.19				
	650		1.31			0.511375 ± 40	−9.2 ± 0.8
	1900		1.93			0.511429 ± 33	−8.2 ± 0.6
261	3900	47°47′33″S, 83°16′31″W	3.69	0.683	0.112	0.511444 ± 26	−7.9 ± 0.5
292	5050	60°54′02″S, 89°24′23″W	4.09			0.511430 ± 28	−8.2 ± 0.6
31	30	20°00′S, 159°59′W	0.411			0.511861 ± 39	0.3 ± 0.8
	2800		2.57			0.511617 ± 36	−4.5 ± 0.7
	4500		3.68			0.511435 ± 29	−8.1 ± 0.6
17	2000	14°41′N, 160°01′W				0.511848 ± 24*	0.0 ± 0.5
Sediment trap	580	36°44′42″N, 122°45′12″W	3.2 ppm†	0.60 ppm†	0.113		

*Unspiked, normalized to 146/142 as above. †The Sm and Nd were determined in an aliquot of the total sample; errors are ~ 5 percent.

topic composition from the waters in the Drake Passage.

Two profiles from the Central Pacific were analyzed (Table 1 and Figs. 3C and 4C). At station 31, $\epsilon_{Nd}(0)$ decreases with depth from 0.3 at 30 m to −8.1 at 4500 m. A sample from 2000 m at station 17 has $\epsilon_{Nd}(0) = 0.0$, compared to a value of −4.5 for a sample at 2800 m at station 31. These results show large differences in Nd isotopic composition between waters within the Central Pacific as well as between the Central Pacific and the Drake Passage. At least in the region of station 31, the Pacific now has a source of Nd near the sea surface which is more radiogenic than indicated by ferromanganese sediment data in this area (*4, 14*) or from seawater in the eastern Pacific (*7*). Comparison of $\epsilon_{Nd}(0)$ for the Central Pacific sample from 4500 m at station 31 [$\epsilon_{Nd}(0) = -8.1$] with values from the Drake Passage (Fig. 3A) shows them to be indistinguishable and indicates that this sample represents Antarctic Bottom Water underlying middle and deep Pacific water. This bottom water sample was collected with the hope of obtaining northward spreading Antarctic Bottom Water (*23*).

The Nd concentration profile at station 31 (Fig. 4C) shows an approximately linear increase with depth, as observed at other stations. The sample from 30 m has the lowest Nd concentration yet measured in seawater. In addition to the seawater samples, a sample of particulate matter from a sediment trap suspended at a depth of 580 m in the eastern North Pacific was analyzed for Sm and Nd and showed an Nd concentration of ~ 3 ppm and a $^{147}Sm/^{144}Nd$ ratio similar to that in seawater. This low concentration supports the argument that suspended material does not contribute significantly to the Nd content of unfiltered seawater.

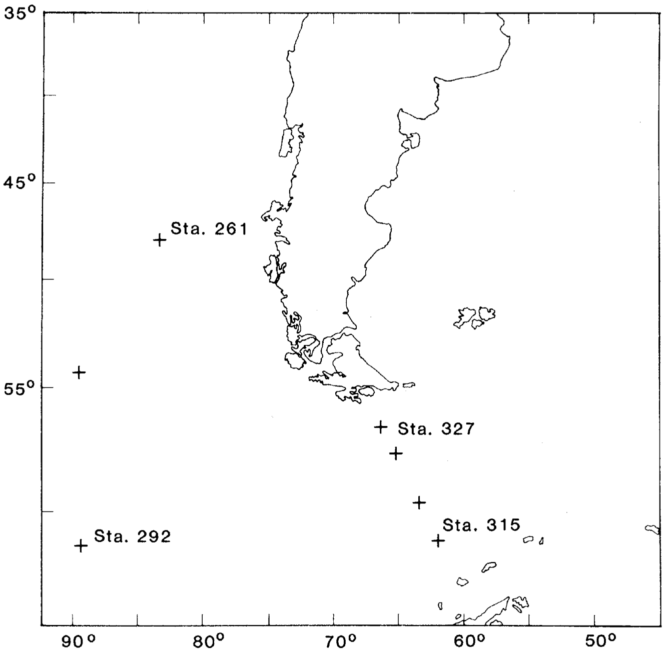

Fig. 2. Map showing locations of water sampling sites in the Drake Passage and southeast Pacific for Nd analysis. Stations for which Nd data were obtained in this study are labeled.

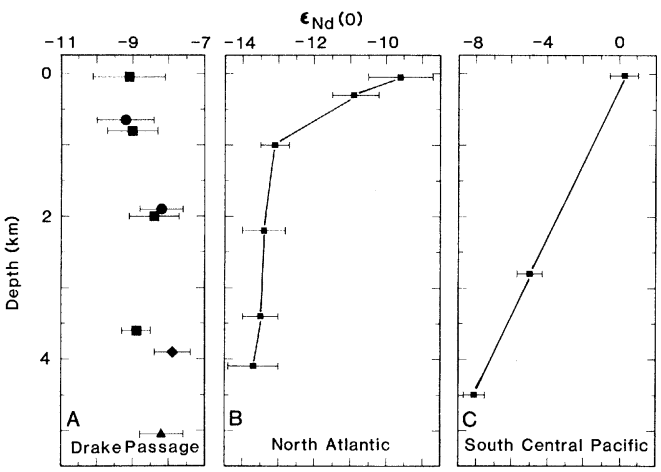

Fig. 3. Value of $\epsilon_{Nd}(0)$ as a function of water depth in (A) the Drake Passage, (B) the North Atlantic, and (C) the south central Pacific (station 31). North Atlantic data are from Piepgras and Wasserburg (*7*). Symbols in (A): (■) station 315; (●) station 327; (◆) station 261; (▲) station 292.

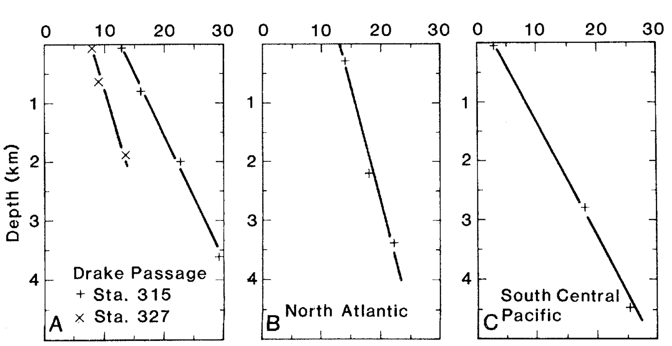

Fig. 4. Neodymium concentration as a function of depth in (A) the Drake Passage (stations 315 and 327), (B) the North Atlantic, and (C) station 31 in the south central Pacific. North Atlantic data are from Piepgras and Wasserburg (*7*). Errors on data points are negligible compared to the size of the symbols.

Discussion

The $\epsilon_{Nd}(0)$ values for the Drake Passage shown in Fig. 1 are close to −9. These values lie in the direction of more ancient continental sources than the Pacific and are far removed from young oceanic mantle-type sources, for which $\epsilon_{Nd}(0) \approx 10$. They are intermediate between North Atlantic and mid-depth Pacific values and closer to the Atlantic values, and they are similar to and possibly slightly lower than the values inferred for the Indian Ocean. From these data we conclude that the water flowing out of the Pacific sector of the Southern Ocean and into the Atlantic sector has Nd which is dominantly of Atlantic or Indian Ocean origin rather than dominantly of Pacific origin.

Bottom waters of the Bellingshausen Plain to the west of the Drake Passage and the Humboldt Plain west of central Chile far north of the Antarctic Convergence have the same $\epsilon_{Nd}(0)$ as the Drake Passage. These data show a coherence in Nd isotopic composition for the Southern Ocean and support its identification as a distinctive water body. In addition, a sample of cold bottom water from the Central Pacific has the same value of $\epsilon_{Nd}(0)$ as the Drake Passage, which identifies this bottom water with a northward spreading part of the Southern Ocean. These results are in agreement with observations by Reid and Lynn (*24*) and Reid (*25*), who showed that waters having physical oceanographic properties with Atlantic characteristics are preserved in the bottom waters of the Pacific Ocean. The oceanography of the Southern Ocean is reviewed in (*26*). More generally, our results support extensive observations of physical oceanographic properties which identify the Southern Ocean as a distinct water mass that is preserved in the bottom waters over extensive regions of the world ocean [see color plates in Dietrich *et al.* (*27*)].

If we assume that the water in the Drake Passage consists only of mixtures of Atlantic and Pacific seawater (ignoring fresh water) and that the Nd concentrations in these waters are equal, we can estimate the relative amounts of water contributed by these sources which would yield $\epsilon_{\mathrm{Nd}}(0) \approx -9$. Using the estimates of $\epsilon_{\mathrm{Nd}}(0)$ for the two oceans given earlier, we calculate that about 70 percent of the water flowing through the Drake Passage into the Atlantic Ocean is of Atlantic origin. Allowing for errors in our estimates of $\epsilon_{\mathrm{Nd}}(0)$ for the Atlantic and Pacific oceans, a lower limit of at least 50 percent Atlantic water is required to balance the Nd isotopic data for the Drake Passage. The Pacific contribution to the Nd budget appears to be smaller than or equal to that of the Atlantic.

This balance, however, is in disagreement with some views based on physical oceanographic observations which indicate that the Pacific should dominate the water properties of this region. The salinity maximum in the Drake Passage has been reported to be 34.725 per mil (*28, 29*), which we confirmed at our stations. If we use this salinity maximum for the Drake Passage and Montgomery's (*30*) values for the mean salinities of the Atlantic ($\approx$ 34.9 per mil) and the Pacific ($\approx$ 34.6 per mil), then at least 60 percent of the water in the Drake Passage should be of Pacific origin if only Pacific and Atlantic mixtures are considered. As there is a substantial contribution of fresh water to the Southern Ocean, it is not clear that Drake Passage water may be considered as a mixture of components having the mean salinity characteristics of the Atlantic and Pacific. Heat budget calculations by Hastenrath (*31*) indicate that there must be a net heat transfer from the Pacific to the Atlantic. Stommel (*32*), using data from Hastenrath (*31*), indicates that the South Pacific Ocean supplies heat dominantly by mass flow to the South Atlantic and that the transfer takes place through the Drake Passage [see figure 1 in (*32*)]. However, the available heat budget data are subject to large uncertainties and may not be a reliable indicator of the mass flow and hence the relative sources of water in the Drake Passage. We also considered the possibility that the Nd isotopic compositions in the Southern Ocean are controlled by contributions of dissolved Nd from Antarctica. Using an estimate for the runoff from Antarctica of 0.075 Sv (*33*) and an upper limit of 40 nanograms per liter for the Nd concentration in fresh water (*10*), we estimated the transport of dissolved Nd to the Southern Ocean from Antarctica as 3 grams per second. Gordon (*26*) estimates a rate of exchange of ~ 20 Sv between the Atlantic and Southern oceans. If deep water has an average Nd concentration of 3 ng/liter, the transport of dissolved Nd from the Atlantic to the Southern Ocean is ~ 63 g/sec. The value calculated for the Nd flux from Antarctica is only 5 percent of this, indicating that runoff from Antarctica is not an important source of Nd in the Southern Ocean. We conclude that, unless there is another source of rare earths in the Southern Ocean (possibly atmospheric dust), less than 50 percent of the water flowing through the Drake Passage is from the Pacific and the salt and heat budgets of the region should be reconsidered.

Figure 3 shows rather uniform values of $\epsilon_{\mathrm{Nd}}(0)$ with depth in the Drake Passage, in contrast to the data for the North Atlantic (*7*) and the south central Pacific. This indicates that the addition of Nd to Southern Ocean waters from other source regions (Pacific, Indian, and Atlantic oceans) must be slow relative to the residence time and vertical mixing rates of Nd in the Southern Ocean.

The changes in Nd isotopic composition with depth in the profiles from the North Atlantic (Fig. 3B) and south central Pacific (Fig. 3C) indicate that the profiles consist of water layers with different sources. The Atlantic data suggest a sill of water, possibly Mediterranean, underlain by North Atlantic deep water with some mixing. The Pacific data appear to represent mixtures of Southern Ocean water with what we infer to be more typical shallow or intermediate Pacific water. The limited data do not permit better identification of the sources of water in the profiles, but they indicate well-defined isotopic characteristics which may be used to characterize water masses and which are unaffected by changes in salinity due to evaporation or dilution with rainwater.

Box Model

We constructed a six-box model in order to characterize the mixing of the Southern, Atlantic, Indian, and Pacific oceans. Figure 5 depicts the model and the exchange paths. We divided the Southern Ocean into boxes 1, 2, and 3, adjacent to the Atlantic, Indian, and Pacific oceans, represented by boxes 1′, 2′, and 3′, respectively. This subdivision is to account for interactions with the other oceans. [Georgi (*28*) presented data for a series of transects of the Southern Ocean including the Drake Passage and regions south of Africa and New Zealand, which have boundaries between the Southern Ocean and major ocean basins to the north. He showed that salinity in the Southern Ocean decreases from a maximum south of Africa to a minimum in the Drake Passage and interpreted this as resulting from successive interactions with the Atlantic, Indian, and Pacific oceans.] Flow between the Southern Ocean boxes is constrained to be unidirectional and toward the east by the Antarctic Circumpolar Current. Flow between the Southern Ocean and the major oceans to the north is two-directional and constrained to be between adjacent primed and unprimed boxes only, as shown in Fig. 5. If we assume that there are no sources of Nd within the Southern Ocean, then for box 1 at steady state we have

$$0 = \epsilon_3 C_3 \dot{W}_{31} - \epsilon_1 C_1 \dot{W}_{12} + \epsilon_{1'} C_{1'} \dot{W}_{1'1} - \epsilon_1 C_1 \dot{W}_{11'} \qquad (1)$$

where the ϵ's are the Nd isotopic compositions, the *C*'s are Nd concentrations, and the $\dot{W}_{ij}$ are rates of volume transport of water from box *i* to box *j*. Taking the flow of the Antarctic Circumpolar Current to be uniform, we have $\dot{W}_{12} = \dot{W}_{23} = \dot{W}_{31}$. Letting $\dot{W}_{11'} = \dot{W}_{1'1}$ and taking the *C*'s to be approximately equal, we find that Eq. 1 reduces to

$$0 \approx (\epsilon_3 - \epsilon_1)\dot{W}_{12} - (\epsilon_1 - \epsilon_{1'})\dot{W}_{11'} \qquad (2)$$

Similarly, for boxes 2 and 3, respectively

$$0 \approx (\epsilon_1 - \epsilon_2)\dot{W}_{12} - (\epsilon_2 - \epsilon_{2'})\dot{W}_{22'} \qquad (3)$$

$$0 \approx (\epsilon_2 - \epsilon_3)\dot{W}_{12} - (\epsilon_3 - \epsilon_{3'})\dot{W}_{33'} \qquad (4)$$

With the available data base this box model is underdetermined, but we estimated $\epsilon_{\mathrm{Nd}}(0)$ for the undetermined Southern Ocean boxes in order to calculate an upper limit for the rate of exchange between the Southern Ocean and the Pacific ($\dot{W}_{33'}$). We may obtain bounds on the unknown ϵ values by assuming that $0 \le \dot{W}_{ii'}/\dot{W}_{12} \le 1$. This yields equations of the form $0 \le (\epsilon_i - \epsilon_j)/(\epsilon_j - \epsilon_{j'}) \le 1$. Taking $\epsilon_3 = -9$, $\epsilon_{3'} = -3$, and $\epsilon_{2'} = -8$, we get $-9.25 \ge \epsilon_2 \ge -10.5$. Using this result in Eq. 4 and taking $\dot{W}_{12} = 130$ Sv, we calculate an upper limit of $\dot{W}_{33'} \approx 33$ Sv for the flow rate between the Pacific and Southern oceans. From this we obtain a lower limit of ~ 800 years for the mean time for exchange between the Atlantic and Pacific. The most sensitive parameters are the values of $\epsilon_{Nd}(0)$ associated with the different segments of the Southern Ocean. If the difference between these values goes to zero, then mixing, as constrained by these approximations, cannot occur, and it would be necessary to consider concentration differences as well.

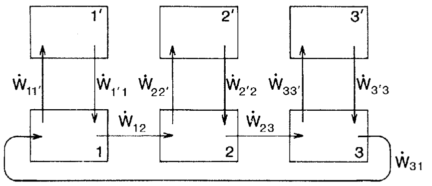

Fig. 5. Box model for mixing in the Southern Ocean. The Southern Ocean is divided into three boxes (1, 2, and 3) adjacent to the Atlantic (1′), Indian (2′), and Pacific (3′) oceans, respectively.

Transport of Neodymium in the Water Column

In contrast to the differences in $\epsilon_{Nd}(0)$ in the water column at different locations, Nd concentration profiles all exhibit approximately linear increases with depth. Samples where the Sm concentration was measured yielded a constant $^{147}Sm/^{144}Nd$ of ~ 0.11 at all levels. This is similar to $^{147}Sm/^{144}Nd$ in continental crustal rocks but much lower than the ratio (0.1967) in a chondritic uniform reservoir (*34*), indicating that the same processes control the vertical transport of Sm and Nd in the water column at the different sampling locations in the oceans.

Potential temperature versus salinity (*T-S*) diagrams for stations 315 and 327 in the Drake Passage and station 31 in the Pacific are shown in Fig. 6. The diagrams for stations 315 and 327 indicate that four components of water having distinctive temperature and salinity characteristics are involved in mixing. In general, these *T-S* relations indicate that lateral as well as vertical transport must play an important role. The points in Fig. 6 representing the sampling for Nd do not lie on straight-line segments in the *T-S* diagram. The samples from the three lower depths at station 315 appear to be on a linear segment, while those from the other stations lie on divergent trends. As pointed out by Craig (*35*), colinearity on the *T-S* diagram is a necessary condition for treating element distribution in terms of a two-component mixing model. However, $\epsilon_{Nd}(0)$ is uniform at stations 315 and 327, indicating only one component of Nd in the water column in the Drake Passage. From the *T-S* diagram for station 31 we infer that there are at least four components of water involved in mixing at this location, whereas the $\epsilon_{Nd}(0)$ data indicate mixing of two components of Nd, one with $\epsilon_{Nd}(0) \approx -8$ and the other with $\epsilon_{Nd}(0) \approx 0$. The different *T-S* relations at these stations are also in contrast with the linear Nd concentration profiles at all stations analyzed. These observations indicate that the distribution and transport of Nd in seawater are not dependent on the factors that directly influence local temperature and salinity variations.

Any explanation for the observed distribution of Nd in the water column must be considered in the context of the numerous studies of other trace element distributions in the oceans. We therefore outline some key observations related to the problem of trace element transport. A number of stable elements (Zn, Ni, Cd, and Ba) increase with depth in a complex fashion (*36–39*). For example, Ba increases to a maximum at about 2 km and then remains relatively constant or decreases toward the bottom (*38, 39*). These elements are found to correlate with nutrient profiles in the oceans, indicating that their distribution is controlled by uptake into planktonic organisms in surface waters with subsequent re-solution in deep waters. The sources of the individual elements and their transport have been subjects of study for three decades. A key to some of the transport processes that may occur in the marine environment has come from study of abundances of the decay products of U and Th (*40*). These decay series are important because some of the nuclides are at extremely low concentrations due to their removal from solution. For example, from the near absence of ^{232}Th in seawater and the presence of the daughter ^{228}Th ($\tau_{1/2} = 1.9$ years) it is inferred that the more soluble intermediate daughter ^{228}Ra ($\tau_{1/2} = 5.7$ years) is transported from coastal waters into the surface ocean waters to provide the source of ^{228}Th with a lateral transport time scale of ~ 20 years. Dissolved U has a relatively high and uniform concentration in seawater (~ 3 μg/liter) (*41*), but one of its daughters, ^{230}Th ($\tau_{1/2} = 7.7 \times 10^4$ years), has a very low concentration, well below secular equilibrium (~ 10^{-3}) (*42*). Recent experiments (*43*) showed the dissolved ^{230}Th to increase regularly with depth in an approximately linear manner. These data can be explained by constant production of ^{230}Th throughout the water column from ^{238}U decay [^{238}U ($\tau_{1/2} = 4.5 \times 10^9$ years) $\rightarrow$ ^{234}Th ($\tau_{1/2} = 24$ days) $\rightarrow$ ^{234}U ($\tau_{1/2} = 2.4 \times 10^5$ years) $\rightarrow$ ^{230}Th ($\tau_{1/2} = 7.7 \times 10^4$ years) $\rightarrow$ ^{226}Ra ($\tau_{1/2} = 1.6 \times 10^3$ years)] with subsequent adsorption of ^{230}Th on particulate matter, which settles to the bottom (*40, 42*). For some nuclides from this decay series such as ^{210}Pb there are direct observations of the proportions associated with particles (~ 10 percent) and in solution (~ 90 percent) (*44*). In addition to scavenging from solution and particle settling, there is evidence for element transport from the sediments into the water column over at least a few hundred meters from the bottom. This is demonstrated by the presence of excess ^{222}Rn ($\tau_{1/2} = 3.8$ days) (*45*) as well as ^{228}Ra and ^{228}Th (from decay of dissolved ^{228}Ra) (*46*) in the lower levels of the deep sea. There is in general no direct connection between the processes controlling these short-lived nuclides and the more abundant stable elements; however, Chan *et al.* (*39*) showed an excellent correlation between Ra (~ 6×10^{-14} grams per kilogram) and Ba (~ 10^{-5} grams per kilogram) in regions above a bottom layer for basins in widely different areas.

These observations indicate that the concentration and isotopic composition of Nd may depend on scavenging by particles, re-solution from particles, and re-solution from sediments at the sea bottom. The regular change of Nd concentration with depth [similar to that of Cu (*36*) and ^{230}Th (*43*)] cannot be due simply to scavenging as there is no steady source such as that for ^{230}Th. Removal of Nd from near-surface waters by settling particles with continuous re-solution during settling coupled with vertical transport or circulation might provide a net effective local source. Some workers have formally used an arbitrary constant source function for elements such as Ba (which has a long residence time) in the water column in order to explain the concentration as a function of depth. The source term presumably results from dissolution of particles ($CaCO_3$) containing Ba, a mechanism suggested by Turekian *et al.* (*22*). We do not find an a priori basis for assuming a uniformly distributed source for Nd (which has a short residence time) in the water column. In addition, if the time scale for removal is short [20 years, as indicated from Th data (*43*)], this would result in rapid removal of Sm and Nd during the history of a water mass.

Theoretical analyses of element distribution in ocean water due to these processes are primarily based on transport models that involve diffusion (with eddy diffusion parameter $\kappa \sim 1$ square centimeter per second and upward advective velocity $w \sim 10^{-5}$ cm/sec). These phenomenological transport models are based on the view that there is an upward component of velocity in the ocean from the cold deep waters that balances the downward diffusion of heat from the warm surface layers (*47*). This approach has led to a set of abyssal recipes (*48*). Craig has discussed this class of models (*35*) as well as the problem of scavenging for the case of Cu (*49*).

The usual form of the one-dimensional equations at steady state for θ (either temperature or salinity) is

$$0 = \kappa \frac{\partial^2 \theta}{\partial z^2} - w \frac{\partial \theta}{\partial z} \qquad (5)$$

For the concentration of species i in solution (C_i) the equation becomes

$$0 = \kappa \frac{\partial^2 C_i}{\partial z^2} - w \frac{\partial C_i}{\partial z} - (\lambda_i + \psi_i)C_i + P_i \qquad (6)$$

Here z is the height upward from the ocean bottom, P_i is the local generic production rate of species i, and $\lambda_i + \psi_i$ are the decay constant plus adsorption rate. These transport equations, which require layers at the upper and lower boundaries to satisfy continuity, have been used extensively as a basis for discussing observational data. By fitting either temperature or salinity to Eq. 5 over regions where temperature and salinity are linearly correlated, the parameter (κ/w) is determined, and by using the known concentration of species i the source term P_i can be explicitly calculated (*35*) (it is usually taken to be a constant).

In explicitly considering transport with sequestering on particles that are raining out, we derive the one-dimensional equation for species i

$$\frac{\partial C_i}{\partial \tau} + \frac{\partial m_p C_{pi}}{\partial \tau} = \kappa \frac{\partial^2 C_i}{\partial z^2} - w \frac{\partial C_i}{\partial z} + \frac{\partial m_p C_{pi} v}{\partial z} - \lambda_i(C_i + m_p C_{pi}) + P_i \qquad (7)$$

where C_{pi} is the concentration of i on particulate matter, m_p is the mass of particles per gram of seawater, and v is their characteristic settling velocity in the rising advective stream. We have neglected particle diffusion, but if it were included C_i in the first term on the right should be $C_i + m_p C_{pi}$. For ^{234}Th, which is dominantly associated with particles, $C_i \rightarrow 0$ (*50*).

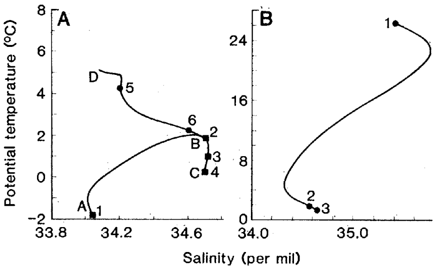

Fig. 6. Potential temperature versus salinity diagrams for stations 315, 327, and 31. (A) Drake Passage. Station 315 follows points ABC. Boxes labeled 1 to 4 correspond to Nd samples at 50, 800, 2000, and 3600 m, respectively. Station 327 follows points DB, and circles labeled 5 and 6 correspond to Nd samples at 650 and 1900 m, respectively. The *T-S* data are for 50-m intervals from CTD casts. (B) South central Pacific, station 31. Points 1 to 3 correspond to Nd samples at 30, 2800, and 4500 m, respectively. The shape of this curve is inferred from eight *T-S* measurements made at this station.

If there is quasi-equilibrium partitioning between the adsorbed and dissolved species of the form $C_{pi}/C_i = \alpha$ (a constant), the settling velocity v is constant, there is no true in situ source, and the species are stable, then the steady-state equation becomes

$$0 = \kappa \frac{\partial^2 C_i}{\partial z^2} - w \frac{\partial C_i}{\partial z} + \alpha v \frac{\partial m_p C_i}{\partial z} = \frac{\partial}{\partial z}\left[\kappa \frac{\partial C_i}{\partial z} - (w - \alpha v m_p)\, C_i\right] \qquad (8)$$

The expression in brackets is the downward net flux, which is constant at steady state.

Comparison of this result with Eq. 6 shows that the coefficient of $\partial C_i/\partial z$ in Eq. 8 is $-w + \alpha v m_p$ rather than $-w$. If $\alpha v m_p$ is comparable to w, then the advective velocity terms are not the same for species whose transport involves particle settling and the transport equations for salinity and temperature. In such cases Eqs. 5 and 8 may be decoupled. If we use a Stokes velocity of $8 \times 10^{-5}\, D^2$ cm/sec for particles of diameter D micrometers and take $\alpha m_p \sim 10^{-1}$ (9 percent adsorbed on particles), then for 3-μm particles we get $\alpha v m_p \sim 7 \times 10^{-5}$ cm/sec. The advective velocity in Eq. 5 is usually taken as $w \sim 10^{-5}$ cm/sec. It follows that if only a few percent is adsorbed in equilibrium on grains, the settling term may govern the transport. The characteristic length then becomes $\kappa/\langle(\alpha v m_p - w)\rangle$ ($\langle\ \rangle$ indicates the average value). For attenuation by a factor of e for a depth of 4 km and $\kappa \sim 1$ cm^2/sec we obtain $\langle(\alpha v m_p - w)\rangle \sim 3 \times 10^{-6}$ cm/sec. For $(\alpha v m_p - w)C_i$ = constant, the concentration is a linear function of depth, and the distribution of particles is hyperbolic. According to this model (Eq. 8), the Nd data require that the net downward flux be less than $(\alpha v m_p - w)C_i$. If equilibration is not achieved, then C_{pi}/C_i is not constant and the falling grain may continue to sequester material from solution. In this case $d(m_p C_{pi}) = -\psi_i m_p C_i dz/v$ if the particulate mass is constant. Substituting this in Eq. 7 at steady state gives a result analogous to Eq. 6 and does not yield an advective term that competes with w. The Nd isotopic and concentration data are not yet sufficient to test these models or to establish whether linear mixing requires linear behavior between T and S. It appears that, in order to understand the distribution of Nd and related elements, a detailed comparison of experimental data, taking into account the vertical distribution of particles, will be needed to assess the validity of the class of models described by Eq. 7.

If Nd and other rare earth elements were injected from bottom sediments, they would be distributed over a scale of only a few hundred meters from the sea floor on an appropriate short time scale, as indicated by the transport coefficients from Rn and Ra studies (*45, 46*). The time for Nd injected from the bottom to reach the surface must be ~ 1000 years. The isotopic differences observed in some profiles indicate preservation of intrinsic differences in Nd. In contrast, the regular behavior of the Nd concentration suggests continuous mixing or exchange between surface and bottom waters on a short time scale. A complex transport mechanism involving precipitation of Nd from solution, scavenging by particles, and re-solution of bottom materials is suggested above. Layering of mid-depth water masses is known to occur in the ocean and must play an important role. A large contribution from the sediments would imply that the observed Nd in solution was not directly related to the dissolved Nd from the continents but resulted from a dissolution-precipitation-saltation process. Such considerations could alter the model of simple uniform ocean basins that we proposed on the basis of the isotopic composition data.

Conclusions

Water flowing through the Drake Passage has an average Nd isotopic composition corresponding to $\epsilon_{Nd}(0) \approx -9$. The $\epsilon_{Nd}(0)$ values reported here indicate that one-half to two-thirds of the Nd in these waters is of Atlantic origin and at most one-half is of Pacific origin. These sources of the water mass do not agree with those based on the estimated salt budget for the Drake Passage or on heat transport considerations. From the Nd data reported here, the Antarctic Circumpolar Current is dominated by Atlantic water, which it spreads to other ocean basins and recirculates back into the Atlantic. These observations are in qualitative agreement with those of Reid and Lynn (*24*) and Reid (*25*), who inferred the presence of Atlantic water in the Indian and Pacific oceans from physical properties of the water and concluded that the Antarctic Circumpolar Current spreads the Atlantic water to other ocean basins, but made no quantitative estimates of the amount of Atlantic water in the Circumpolar Current.

Concentrations of Sm and Nd in the water column show an approximately linear increase with depth in the Drake Passage and at other locations. This is similar to results for Cu (*36*). The concentration gradient requires some means of transport and replenishment of these elements in the water column on a short time scale. This transport appears to be governed by settling and re-solution of particles. If replenishment of ND in the bottom waters is due to re-solution of the underlying sediments, the Nd isotopic composition must reflect materials previously deposited rather than recent continental drainage. The concentration data are difficult to explain by the simple model used to discuss the isotopic data. Further advances will require an understanding of transport characteristics of rare earth elements in the ocean.

An upper limit of 33 Sv for the rate of exchange between the Pacific and Southern oceans has been calculated on the basis of a box model describing Nd transfer between the oceans. At present, the model is underdetermined, but with more data for the Nd isotopic composition of Southern Ocean waters, it may be possible to put further constraints on the rates of exchange between the Southern Ocean and ocean basins to the north.

Insofar as the differences observed in $\epsilon_{Nd}(0)$ between the Atlantic and the Pacific reflect the rate of water exchange between these oceans, then this must be controlled by the flow through the Drake Passage. We may imagine that as the passage was opened in late Oligocene time (*51*), the degree of isotopic difference decreased. If the Nd isotopic composition of seawater is preserved in sediments over geologic time, it may be possible to study the degree of isolation of earlier oceans under different arrangements of the continents. This has considerable interest in terms of determining the disposition of earlier ocean basins and may possibly be used to trace the flow of major paleo-ocean currents. In the case of the Southern Ocean, we believe that the dominant source must be North Atlantic deep water. The changes of such flow over the past few million years may be of use in understanding climate change.

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