

The distribution of radionuclides and some trace metals in the water columns of the Japan and Bonin trenches

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Abstract – Presented here is the first detailed geochemical data set on the U/Th series Th, Pa, Ac, and Pb isotopes and artificial fallout radionuclides (⁹⁰Sr, ¹³⁷Cs, and Pu isotopes), and some trace elements (V, Zn, Cd, Cu, Mn, and Ni) in two water columns of the Japan and Bonin trenches down to the bottom depths of 7585 m and 9750 m, respectively. Hydrographic properties such as temperature, salinity, dissolved oxygen, and nutrient content within the trench valley remain constant at the same levels as those in the bottom water of the Northwest Pacific basin (typically ~6000 m in depth). The radionuclide activities and most trace metal concentrations are also not *very* different from those in the overlying water at depths of around 5000–6000 m. This means that any chemical alteration which sea water undergoes during its residence within the trench was not obviously detected by the techniques used here. The suggestion follows that the trench water is rather freely communicating by isopycnal mixing with the bottom water overlying the Northwest Pacific abyssal plain. The trench waters contain high ^{239,240}Pu activities throughout, indicating that Pu is actively regenerating from rapidly sinking, large particles at the bottom interface, probably due to a change in the oxidation state. On the other hand, the vertical profiles of ²¹⁰Pb and ²³¹Pa show lower activities within the trench than those in the overlying deep waters, suggesting that the effect of boundary and bottom scavenging is significant in controlling their oceanic distributions. However, none of the trace metals studied here obviously follows the behaviour of the above nuclides. The ²²⁸Th data show scattering within the Bonin Trench that is largely ascribable to analytical errors. If, however we accept that the scatter of ²²⁸Th data is real and the variation is caused solely by decay of its parent ²²⁸Ra, we can set an upper limit of ~5 years for the renewal time of the trench water. © Elsevier, Paris

U/Th series nuclides / artificial radionuclides / scavenging / trace metals / renewal of trench water

Résumé – Répartition des nucléides radioactifs et de quelques métaux-traces dans les fosses du Japon et des îles Bonin. La première série détaillée de données géochimiques obtenues dans deux colonnes d'eau des fosses du Japon et des îles Bonin, respectivement jusqu'à 7585 m et 9750 m, est présentée ici. Elle porte sur la série uranium/thorium, sur les isotopes de thorium, protactinium, actinium et plomb et sur les retombées des nucléides radioactifs artificiels (strontium-90, césium-137 et plutonium) ainsi que sur quelques éléments à l'état de traces (vanadium, zinc, cadmium, cuivre, manganèse et nickel). Les caractéristiques hydrologiques telles que température, salinité, oxygène dissous et teneurs en nutriments prennent les mêmes valeurs aux mêmes profondeurs, dans les fosses comme dans l'eau de fond du bassin nord-occidental du Pacifique (typiquement, vers 6000 m). De même, les valeurs de l'activité des nucléides radioactifs et les concentrations

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de la plupart des traces métalliques ne sont pas très différentes dans la fosse et dans l'eau sus-jacente, vers 5000–6000 m. Si, au cours de son transit dans la fosse, l'eau a subi une modification chimique, celle-ci n'a pas été détectée par les techniques utilisées. Cela suggère que l'eau de la fosse communique assez librement par mélange isopycne avec l'eau de fond qui couvre la plaine abyssale dans le nord-ouest du Pacifique. Partout dans les eaux de la fosse l'activité du $^{239,240}\text{Pu}$ est élevée, ce qui indique que le plutonium est activement régénéré à partir des grosses particules qui sédimentent rapidement sur le fond, probablement par un changement de leur état d'oxydation. De plus les profils verticaux de ^{210}Pb et ^{231}Pa indiquent des activités plus faibles dans la fosse que dans les eaux profondes sus-jacentes, suggérant que l'effet de barrière et le balayage sur le fond sont significatifs dans la répartition de ces éléments dans l'océan. Aucun des éléments-traces étudiés ici ne se comporte comme les nucléides radioactifs. Dans la fosse des îles Bonin, les données de ^{228}Th montrent une dispersion qui est due essentiellement aux erreurs analytiques. Cependant, si la dispersion est réelle et résulte uniquement de la désintégration du ^{228}Ra , on peut fixer une limite supérieure d'environ cinq ans pour le temps de renouvellement de l'eau dans la fosse. © Elsevier, Paris

uranium – thorium / nucléide artificiel / balayage / métal-trace / temps de résidence

1. INTRODUCTION

The purpose of this paper is to describe the first detailed measurements of radionuclides and some chemical constituents in the water columns of the Japan and Bonin (also called "Izu-Ogasawara") trenches, together with their geochemical implications. Trenches, defined as narrow valley features of the ocean floor exceeding 7000 m in depth, are located along the plate boundaries where one plate is subducting under the other. Although trench regions occupy only ~1.2 % of the ocean floor, their near-bottom environments are attractive to marine scientists working in various disciplines as subjects of investigation. To date, however, hydrographic and geochemical investigations have been few. For example, during the Southern Cross Expedition of the R.V. Hakuho-Maru in 1968–69 [13], a hydrocast was made with Nansen bottles down to ~10000 m in the Tonga Trench, but all the reversing thermometers reaching depths greater than 8000 m were broken due to high water pressure. The GEOSECS expeditions between the 1970s and early 1980s occupied numerous stations in the world oceans, but samples were obtained at only two stations in the South Sandwich and Aleutian Trenches down to a maximum depth of 7800 m [45]. No other attempts have been reported so far, and therefore the oceanographic environment of the super-deep (e.g. > 8000 m) oceanic trenches remains virtually unexplored.

Fractional but plausible evidence available up to now suggests that active particle transport is taking place within the trench valley [32], and that relatively high biomass is sustained there [38]. The 1984 R.V. Hakuho-Maru Cruise (called the Antares Expedition) was

aimed at collecting geochemical data in the deepest parts of the Japan and Izu-Ogasawara trenches. The results indicate that the water within the trench is relatively uniform, with chemical characteristics which are not very different from those of overlying water in the adjacent abyssal plain around the depth of ~6000 m.

2. MATERIALS AND METHODS

2.1. Sampling

Two stations in the Japan and Izu-Ogasawara (also called Bonin) trenches were occupied by the Antares Expedition (KH 84-3 R.V. Hakuho-Maru Cruise) from August to September 1984. *Figure 1* shows the bottom topography and the station locations. The bottom depth is 9750 m for AN-1 and 7585 m for AN-4. A tapered steel wire (9 mm diameter at the top and 14 mm at the end of its 14 000 m length) was used for all large-volume water sampling and Niskin bottle casts deeper than 7000 m. Water temperature was measured using specially designed reversing glass thermometers available at that time for use in depths of up to 11 000 m. Samples for measurements of salinity, dissolved oxygen, pH, nutrients, and ^{210}Pb were obtained by standard 23 L Niskin bottles. For trace metals, either the Cal-Tech.-type [41] or the Tsubota-Kanamori-Murozumi beloze-type polyethylene bag samplers [16] were used. For radionuclide measurements, large-volume water samples were obtained by using a twin 270 L PVC-bottle sampler.

At the same time, suspended particulate matter and dissolved actinides adsorbable on MnO_2 -impregnated fiber

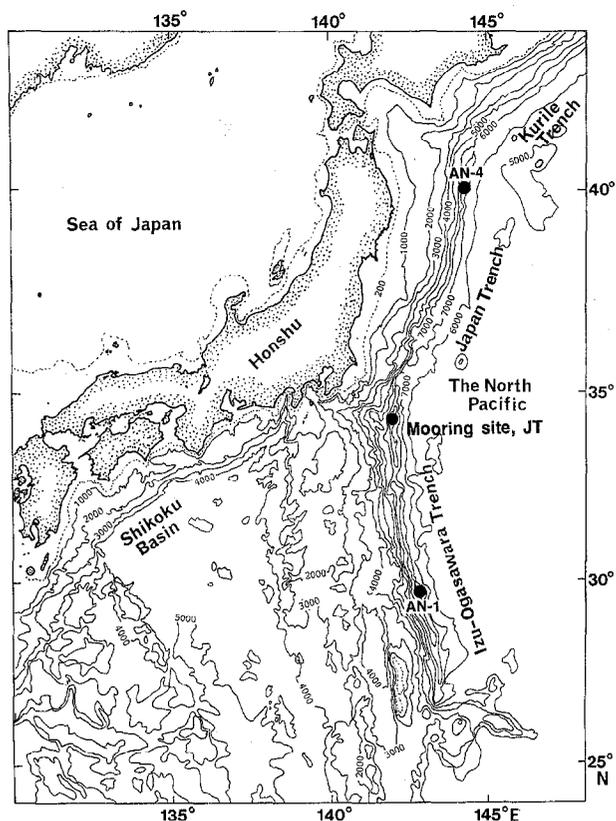


Figure 1. Sampling locations in the Japan and Izu-Ogasawara trenches during the Antares R.V. Hakuho-MarU Expedition. The JT site is the location where sediment-trap experiments have been carried out since 1986.

were also collected using battery-powered large-volume in situ pumps. Details of the sampling technique and analysis together with implications of Th isotope data are given in Nozaki et al. [31]. In addition, sediment-trap experiments using Honjo type time-series traps [12] have been carried out at JT site of the northern Izu-Ogasawara Trench (*figure 1*). Details of the experiments are given elsewhere [26].

2.2. Analysis

The analytical procedure for trace metals is described in detail by Shitashima [43]. Trace metals were concentrated using a chelex-100 column chromatography and determined by graphite furnace atomic absorption spectrometry. Since some deionized and distilled water was used for sampling, the results were suitably corrected for dilution effect based on salinity measurements.

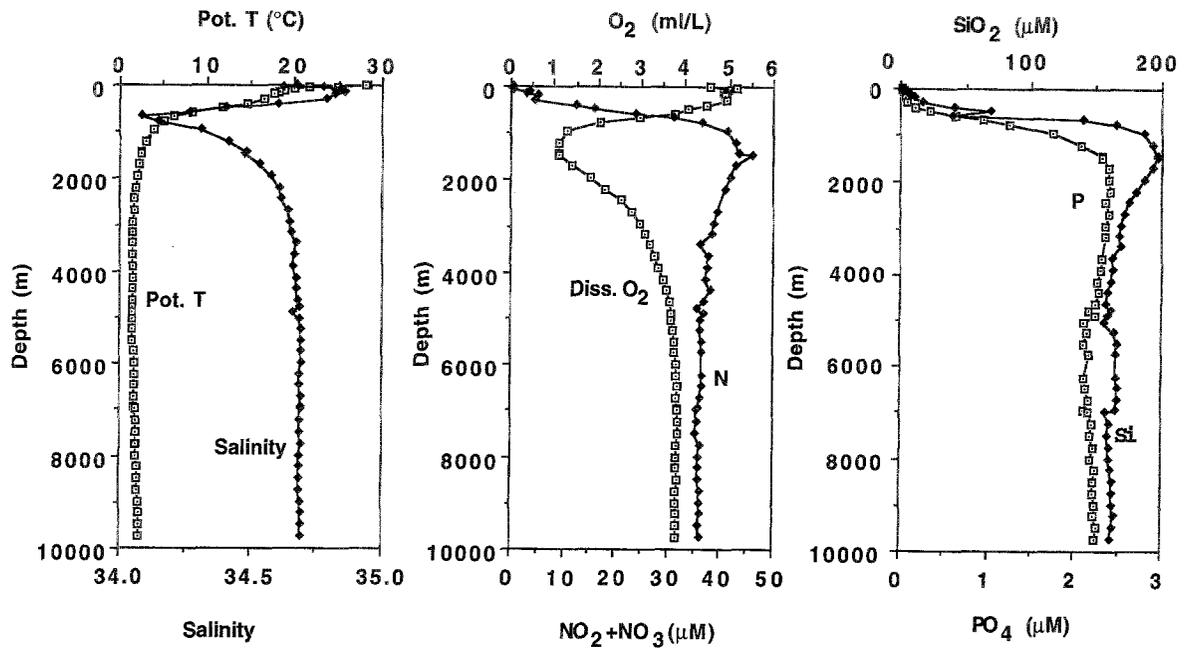
^{210}Pb was determined using ~ 3 L aliquots of unfiltered sea water obtained using Niskin bottles. The analysis was performed 2.5 years after the sampling, during which time ^{210}Po had reached equilibrium with ^{210}Pb . The procedure of Nozaki [25] which was followed includes coprecipitation of ^{210}Po with iron-hydroxide, isolation by electrodeposition onto a silver disc, and subsequent α -spectroscopy, using ^{208}Po as a yield monitor.

All the radionuclides except ^{210}Pb were determined using ~ 250 L water samples according to the method described in Nozaki [28]. The unfiltered water was acidified and degassed by purging nitrogen gas in order to extract CO_2 for radiocarbon measurement. Then appropriate amounts of ^{234}Th , ^{233}Pa , ^{242}Pu , and common Cs and Fe were added to the water and thoroughly mixed. The actinides and Pu in the solution were coprecipitated with iron hydroxide by neutralizing the water with ammonia. The hydroxide was removed from the water and redissolved in 8M HNO_3 solution. Th, Pa, Pu, and Ac isotopes were successively separated by the ion-exchange procedure and determined by α -spectrometry according to the methods described earlier ([30] for Th and Pa, [32] for Ac, [23] for Pu). The procedures for ^{137}Cs and ^{90}Sr in the remaining sea water, described in Nagaya and Nakamura [22], include coprecipitations of ^{137}Cs with ammonium molybdophosphate and ^{90}Sr with calcium carbonate, followed by chemical purification and β assay of the two nuclides.

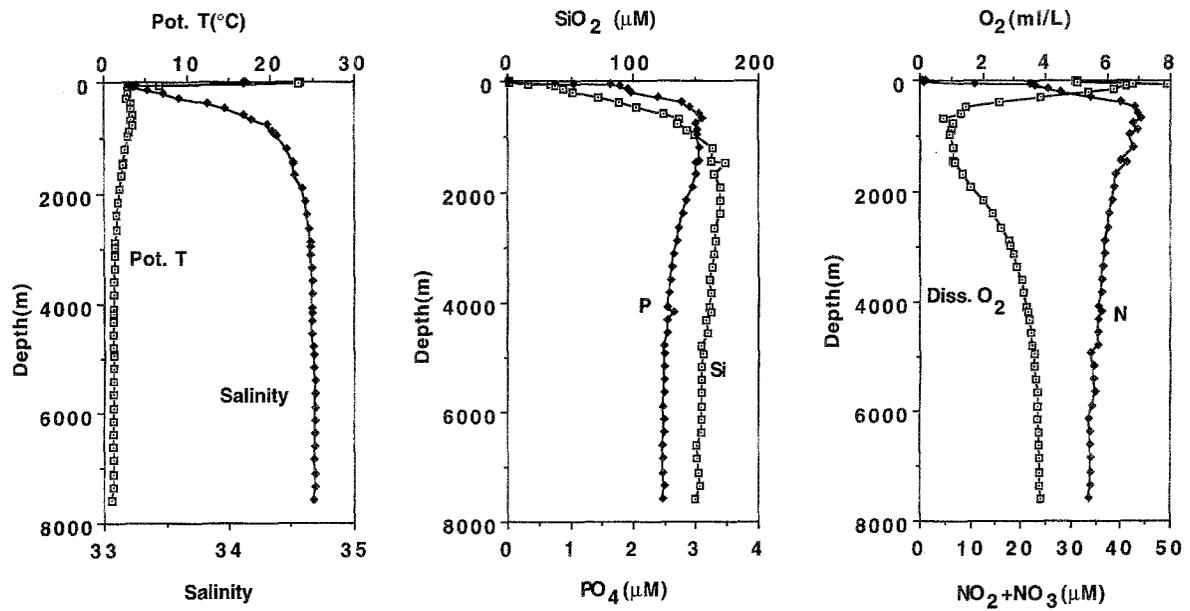
3. RESULTS

3.1. Hydrographic data

The vertical profiles of potential temperature, salinity, dissolved oxygen, and nutrients at the two locations are shown in *figure 2*. The AN-1 Bonin Trench station is located in the oligotrophic Kuroshio Current regime. A distinct salinity minimum, which is a characteristic of the North Pacific Intermediate Water, can be clearly seen centered around 700 m. The dissolved oxygen minimum, and nitrate and phosphate maxima, occur around 1250 m. The AN-4 Japan Trench station, on the other hand, is located in the Oyashio Current regime of relatively high productivity. The distinctively low salinity ($S = \sim 33.2$) and low temperature ($T = \sim 3^\circ\text{C}$) compared to the Kuroshio surface water ($S = 34.6\text{--}34.7$, $T = 18\text{--}20^\circ\text{C}$) appears at about 100 m depth (*figure 3*). The surface waters at the two stations have very low nutrient contents, but a notable difference is that the vertical gradient of the nutrients



(a)



(b)

Figure 2. Vertical profiles of temperature, salinity, dissolved oxygen, nitrate+nitrite, reactive phosphate, and silicate at stations AN-1 (a) and AN-4 (b).

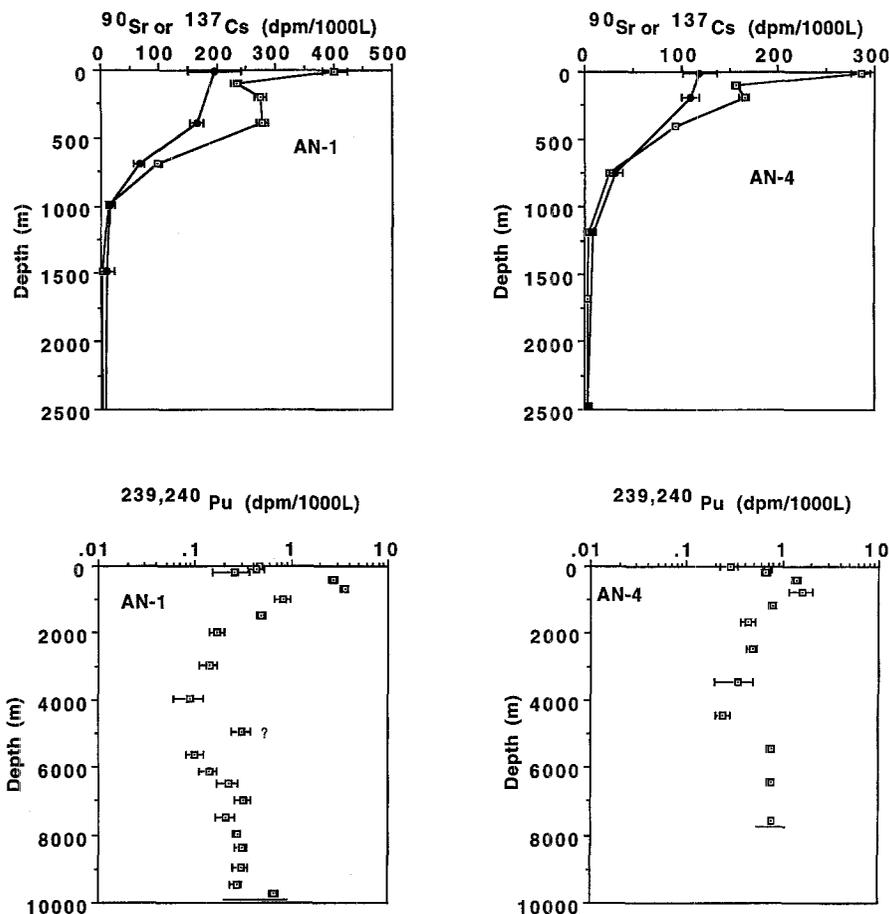


Figure 3. ^{90}Sr and ^{137}Cs in the upper water columns of stations AN-1 and AN-4 (a) and vertical profiles of $^{239,240}\text{Pu}$ at stations AN-1 and AN-4 (b).

across the seasonal thermocline is much higher at station AN-4 than at station AN-1.

In the deep waters between 5000 m and the bottom, all the hydrographic properties show very little vertical variation. There is no density stratification in the waters inside the two trenches.

3.2. Radionuclide data

Radionuclide data are given in *tables I-III* together with 1σ counting errors. ^{231}Pa , ^{227}Ac and ^{210}Pb were analyzed only at station AN-1 in the Bonin Trench. The analyses of the natural U/Th series nuclides were done in the ORI laboratory at the University of Tokyo. The artificial

nuclides, ^{90}Sr and ^{137}Cs , and Pu at Station AN-4 were analyzed at Nakaminato Branch of NIRS, and Pu at station AN-1 were measured at Kanazawa University. Since unfiltered water was used for the analysis, the data should be regarded as "acid-soluble" activities in the water. However, they probably represent total activities in the water because the Th data which indicated the largest particulate fraction among the nuclides showed good agreement with dissolved-plus-particulate Th determined independently by using large-volume in situ pumps at the two locations [35].

We are somewhat hesitant to present the non-radiogenic ^{232}Th data because of their large uncertainties. Since the contamination problem during handling of the large-

Table I. The activities of some actinides and artificial radionuclides in the water column of the Izu-Ogasawara Trench.

Depth (m)	Th-232 dpm/1000L	Th-230 dpm/1000L	Th-228 dpm/1000L	Pa-231* dpm/1000L	Ac-227* dpm/1000L	Sr-90* dpm/1000L	Cs-137* dpm/1000L	Pu-239,240 dpm/1000L
10	0.06 ± 0.03	0.10 ± 0.06	4.87 ± 0.34	ND	0.23 ± 0.06	195 ± 46	401 ± 22	0.47 ± 0.02
98	0.12 ± 0.04	0.21 ± 0.05	2.70 ± 0.18	0.19 ± 0.06	ND	ND	233 ± 9.3	0.43 ± 0.08
196	0.11 ± 0.04	0.23 ± 0.06	3.09 ± 0.22	0.18 ± 0.09	0.20 ± 0.05	ND	273 ± 9.3	0.43 ± 0.08
396	0.21 ± 0.05	0.51 ± 0.08	3.66 ± 0.26	0.28 ± 0.13	0.40 ± 0.04	164 ± 12	276 ± 9.8	2.72 ± 0.29
693	0.10 ± 0.03	0.32 ± 0.06	2.85 ± 0.21	0.41 ± 0.08	0.24 ± 0.03	66 ± 9	97 ± 6.0	3.58 ± 0.29
992	0.03 ± 0.02	0.35 ± 0.10	0.82 ± 0.19	0.39 ± 0.15	0.29 ± 0.05	16 ± 7	13 ± 2.9	0.82 ± 0.15
1488	0.05 ± 0.02	0.53 ± 0.09	0.53 ± 0.13	0.51 ± 0.10	0.32 ± 0.05	9 ± 13	3.1 ± 1.6	0.48 ± 0.05
1990	0.07 ± 0.03	0.55 ± 0.11	0.60 ± 0.14	0.40 ± 0.07	0.39 ± 0.05	ND	ND	0.17 ± 0.03
2985	0.09 ± 0.05	0.86 ± 0.17	0.59 ± 0.22	0.53 ± 0.13	0.44 ± 0.04	ND	2.9 ± 1.6	0.14 ± 0.03
3971	0.06 ± 0.03	1.18 ± 0.15	0.89 ± 0.17	0.58 ± 0.10	0.90 ± 0.07	6 ± 6	3.3 ± 2.0	0.09 ± 0.03
4964	0.10 ± 0.05	1.36 ± 0.16	1.62 ± 0.20	0.79 ± 0.11	1.30 ± 0.10	ND	2.0 ± 1.8	0.31 ± 0.07
5624	0.08 ± 0.04	1.47 ± 0.20	1.55 ± 0.24	0.45 ± 0.09	1.77 ± 0.12	ND	1.8 ± 2.0	0.10 ± 0.02
6125	0.11 ± 0.04	1.74 ± 0.18	2.38 ± 0.22	0.58 ± 0.15	1.38 ± 0.11	1 ± 8	1.8 ± 2.0	0.14 ± 0.03
6473	0.02 ± 0.02	1.08 ± 0.18	1.43 ± 0.26	0.40 ± 0.09	1.57 ± 0.11	ND	6.2 ± 1.8	0.22 ± 0.05
6971	0.05 ± 0.02	1.22 ± 0.13	1.97 ± 0.19	0.41 ± 0.10	1.54 ± 0.15	ND	0.0 ± 0.2	0.32 ± 0.06
7470	0.07 ± 0.04	1.60 ± 0.22	2.34 ± 0.32	0.23 ± 0.04	1.55 ± 0.17	ND	3.3 ± 1.6	0.21 ± 0.05
7968	0.04 ± 0.03	1.55 ± 0.17	2.41 ± 0.24	0.40 ± 0.09	1.50 ± 0.11	-2 ± 17	0.2 ± 1.8	0.27 ± 0.02
8381	0.06 ± 0.03	1.21 ± 0.16	1.84 ± 0.22	0.31 ± 0.09	1.69 ± 0.11	ND	1.1 ± 1.6	0.30 ± 0.04
8980	0.06 ± 0.03	1.34 ± 0.19	2.29 ± 0.30	0.52 ± 0.11	1.40 ± 0.10	5 ± 5	1.1 ± 1.8	0.30 ± 0.05
9470	0.04 ± 0.03	1.09 ± 0.23	1.66 ± 0.32	0.55 ± 0.12	ND	-2 ± 6	4.0 ± 3.3	0.27 ± 0.04
9738	0.12 ± 0.06	1.60 ± 0.24	2.59 ± 0.37	0.59 ± 0.20	1.36 ± 0.12	-3 ± 8	2.2 ± 1.6	0.67 ± 0.07

*ND : not determined.

Table II. ^{210}Pb in the water column of the Izu-Ogasawara Trench.

Depth (m)	Pb-210 dpm/1000L	Depth (m)	Pb-210 dpm/1000L
0	168 ± 8	5992	122 ± 6
483	142 ± 7	6234	166 ± 8
967	161 ± 8	7001	153 ± 8
1707	185 ± 9	7250	153 ± 8
1951	164 ± 8	7500	163 ± 8
2195	199 ± 10	7749	126 ± 6
2682	194 ± 10	7998	126 ± 6
2926	186 ± 9	8247	139 ± 7
3170	225 ± 11	8496	153 ± 8
3658	195 ± 10	8745	126 ± 6
4145	192 ± 10	8995	144 ± 7
4633	178 ± 9	9244	155 ± 8
5267	152 ± 8	9493	127 ± 6
5750	138 ± 7	9742	120 ± 6

volume sea water samples was not corrected, these values should be regarded as the upper limit. Nevertheless, the ^{232}Th data seem to show higher concentrations in the upper waters than in deep and bottom waters. The deep waters below 2000 m at station AN-4 have the ^{232}Th mean concentration of 0.03 ± 0.01 dpm 10^{-3}L , which is discernibly lower than the mean value of $0.06 \pm$

0.03 dpm 10^{-3}L at the AN-1 site. Since Nozaki et al. [35] have shown that 17–72 % of the total ^{232}Th is present in particulate forms at these locations, dissolved ^{232}Th may be significantly lower than the total given here. The ^{230}Th and ^{231}Pa activities are two or three orders of magnitude lower than those of parent ^{234}U and ^{235}U , respectively. They show systematic increase with depth down to ~6000 m, while the ^{228}Th concentrations become the lowest around 2000 m to 3000 m with higher concentrations in the surface and near-bottom waters. The mean activities in the trench waters (> 6000 m) are nearly the same for the AN-1 and AN-4 sites at 1.4 dpm 10^{-3}L for ^{230}Th and 2.2 dpm 10^{-3}L for ^{228}Th . The mean values inside the Bonin Trench are 0.4 dpm 10^{-3}L for ^{231}Pa , 1.5 dpm 10^{-3}L for ^{227}Ac , and 125 dpm 10^{-3}L for ^{210}Pb . Some Th and ^{227}Ac results were also reported previously together with their geochemical implications [34, 35].

The activities of ^{90}Sr ($t_{1/2} = 29$ y) and ^{137}Cs ($t_{1/2} = 30$ y) were found only in the upper water column. The deep waters below 1500 m contained insignificant amounts of these two nuclides, although ^{137}Cs often showed a positive sign of its presence in the deep water (table I). On the other hand, Pu was detected throughout the water column. The highest Pu activities were observed between

Table III. The activities of Th isotopes and artificial radionuclides in the water column of the Japan Trench.

Depth (m)	Th-232 dpm/1000L	Th-230 dpm/1000L	Th-228 dpm/1000L	Sr-90* dpm/1000L	Cs-137 dpm/1000L	Pu-239,240 dpm/1000L
10	0.03 ± 0.02	0.05 ± 0.04	1.88 ± 0.16	119 ± 17	286 ± 10	0.28 ± 0.06
98	0.06 ± 0.03	0.11 ± 0.05	3.21 ± 0.23	ND	157 ± 4.8	0.69 ± 0.08
196	0.05 ± 0.02	0.18 ± 0.06	4.57 ± 0.31	110 ± 9	166 ± 5.3	0.67 ± 0.07
400	0.05 ± 0.02	0.23 ± 0.05	2.22 ± 0.15	ND	95 ± 2.6	1.38 ± 0.14
750	0.04 ± 0.02	0.24 ± 0.06	1.31 ± 0.14	33 ± 6	26 ± 2.2	1.61 ± 0.44
1193	0.09 ± 0.03	0.38 ± 0.08	0.94 ± 0.14	9 ± 4	4.8 ± 1.5	0.78 ± 0.07
1686	0.07 ± 0.03	0.33 ± 0.07	0.69 ± 0.12	ND	2.4 ± 1.3	0.44 ± 0.07
2472	0.04 ± 0.02	0.47 ± 0.07	0.40 ± 0.08	3 ± 4	2.6 ± 1.8	0.48 ± 0.06
3460	0.04 ± 0.02	0.79 ± 0.12	0.47 ± 0.14	ND	2.6 ± 1.8	0.34 ± 0.15
4476	0.02 ± 0.02	1.19 ± 0.14	0.79 ± 0.15	5 ± 4	4.8 ± 2.2	0.24 ± 0.04
5470	0.03 ± 0.02	0.86 ± 0.15	1.10 ± 0.20	ND	2.0 ± 1.8	0.76 ± 0.07
6422	0.03 ± 0.02	1.39 ± 0.14	2.30 ± 0.19	11 ± 4	0.7 ± 1.8	0.75 ± 0.06
7565	0.02 ± 0.02	1.48 ± 0.15	2.44 ± 0.22	0 ± 2	0.0 ± 1.8	0.76 ± 0.04

*ND = not determined.

500 m and 750 m and the second highest values were found inside the trenches. The waters within the Japan and Izu-Ogasawara trenches contained Pu on average of 0.76 ± 0.05 dpm 10^{-3} L and 0.30 ± 0.06 dpm 10^{-3} L, respectively. We also note anomalously high values of Pu at 4964 m, and at the closest to the bottom in the Bonin Trench (*table I*). The former is not well understood at present. The near-bottom value was probably caused by resuspended sediment particles, as deduced from the high concentration of ^{232}Th (*table I*). Excluding this value, Pu is rather homogeneously distributed within the trenches.

3.3. Particulate data

The Th isotope results obtained for suspended particles were reported in Nozaki et al. [35]. Here, the ^{231}Pa data

together with ^{230}Th are given in *table IV*. Although the uncertainties of these measurements are large, the results show that the particulate ^{231}Pa is only less than 5 % of the total (*table I*) and seems to have a broad mid-depth maximum at around a depth of 3000 m.

The elemental and radiochemical data obtained for settling particulate matter collected by the sediment-trap experiments will be given elsewhere together with their geochemical implications. Here, we report only the mean activities of radionuclides in the settling particles collected at ~5000 m and 8800 m depths as necessary for the arguments (*table V*). Very high ^{210}Pb activities were observed in the particles due to scavenging in the water column. The activities of scavenged nuclides, ^{230}Th and ^{231}Pa also show significant increase with depth. Artificial

Table IV. ^{231}Pa and ^{230}Th in suspended particulate matter obtained by in situ filtration.

Station	Depth (m)	^{230}Th * dpm/1000L	^{231}Pa dpm/1000L	$^{230}\text{Th}/^{231}\text{Pa}$ activity ratio
<i>Bonin Trench</i>				
An-1	1100	0.054 ± 0.009	0.011 ± 0.004	4.9 ± 2.0
	2200	0.080 ± 0.037	0.025 ± 0.015	3.2 ± 2.4
	3300	0.187 ± 0.031	0.029 ± 0.008	6.4 ± 2.1
	4400	0.157 ± 0.031	0.017 ± 0.004	9.2 ± 2.8
	5500	0.257 ± 0.075	0.015 ± 0.002	17 ± 5
<i>Japan Trench</i>				
AN-4	1100	0.068 ± 0.017	0.010 ± 0.004	6.8 ± 3.2
	2200	0.073 ± 0.010	0.020 ± 0.004	3.7 ± 0.9
	3300	0.097 ± 0.023	0.028 ± 0.007	3.5 ± 1.2
	4400	0.188 ± 0.014	0.014 ± 0.002	13 ± 2
	5500	0.202 ± 0.024	0.013 ± 0.002	16 ± 3

*After Nozaki et al. (1987).

Table V. Nuclide activity and the activity ratio in sediment-trap particles at the JT site.

Trap depth	5000 m	8800 m
^{232}U (dpm/g)	1.24 ± 0.25	1.26 ± 0.27
^{232}Th (dpm/g)	1.12 ± 0.26	2.34 ± 0.61
^{230}Th (dpm/g)	8.9 ± 1.9	19.0 ± 3.8
^{228}Th (dpm/g)	12.8 ± 1.9	14.3 ± 2.9
^{231}Pa (dpm/g)	0.55 ± 0.11	1.46 ± 0.25
^{210}Pb (dpm/g)	970 ± 190	1550 ± 330
$^{239,240}\text{Pu}$ (dpm/g)	0.28 ± 0.03	0.36 ± 0.09
^{237}Cs (dpm/g)	-	2.41 ± 0.64
$(^{230}\text{Th}/^{231}\text{Pa})$	16	13
$(^{210}\text{Pb}/^{230}\text{Th})$	109	82
$(^{239,240}\text{Pu}/^{137}\text{Cs})$	-	0.15

radionuclides, Pu and ^{137}Cs were also detected in the samples, indicating that they are very rapidly transported to the deep sea in association with particulate matter.

3.4. Heavy metals

The concentrations of heavy metals for water samples from station AN-1 in the Bonin Trench were determined at Hiroshima University. Since the waters were not filtered, the data represent total acid-soluble concentrations. Vanadium shows some depletion in the surface water, whereas it is almost constant with depth in the deep water. The concentrations of Cd, Ni, and Zn increase from the surface to the depth of ~2000 m where they reach maxima, and then gradually decrease with depth. Surface concentrations for Zn and Cd are nearly zero, consistent with phosphate, nitrate, and reactive silica (*figure 2*). Nickel deviates from this, showing the definitely non-zero surface concentration of 2.8 nmol kg^{-1} . Copper shows rather monotonic increase with depth, having no mid-depth maximum. Details of these heavy metal profiles will be discussed later.

4. DISCUSSION AND CONCLUSION

4.1. Vertical profiles of anthropogenic nuclides

The vertical profiles ^{90}Sr and ^{137}Cs (*figure 3*) show the decreasing trends with depth, which are common features of bomb-produced soluble nuclides such as ^3H and ^{14}C . The total amount of ^{90}Sr and ^{137}Cs present in the water column is approximately two times higher at the AN-1

site than at the AN-4 site, this difference probably being due to the lateral redistribution of the nuclides along with the gyre circulation, as demonstrated for bomb-produced ^{14}C [6]. Although the distribution patterns of ^{90}Sr and ^{137}Cs in the water column are similar, the two nuclides show some difference in their oceanic behaviour. Sr exists in sea water as a soluble ionic form of Sr^{2+} . Therefore, ^{90}Sr has not been detected in marine particulate matter or sediments at a significant level, although it may be incorporated in biogenic tests such as carbonates and sulfates. In contrast, ^{137}Cs can be found in various lake and marine sediments. For example, Yang et al. [46] measured the ^{137}Cs and $^{239,240}\text{Pu}$ profiles in North Pacific deep-sea sediments to estimate the particle mixing rates due to bioturbation. More directly, ^{137}Cs is also found in settling particulate matter collected by sediment trap in the water column of the Bonin Trench (*table V*). The particle reactive nature is not expected from the ionic form of Cs^+ in sea water, but it seems likely that ^{137}Cs is incorporated into clay minerals by adsorption or through ionic exchange.

The unique behaviour of Pu isotopes in the marine environment has been demonstrated by previous workers [4, 11, 23]. Unlike ^{137}Cs and ^{90}Sr , the Pu concentration is detectable throughout the water column. In the North Pacific, the $^{239,240}\text{Pu}$ activity in the surface water is very low, increasing to a mid-depth maximum around 500 to 800 m and then decreasing with depth. The vertical profiles often show a secondary maximum near the bottom. Our Pu profiles in the upper water column (*figure 3*) are quite similar to those reported previously for the western North Pacific [4, 23], showing a maximum around 750 m at both locations. The low $^{239,240}\text{Pu}$ activities in the surface waters are probably due to intensive scavenging by particulate matter. This is more likely to occur if Pu exists in the forms of reduced state such as $\text{Pu}(\text{OH})_3^+$ and $\text{Pu}(\text{OH})_4^0$. The subsurface $^{239,240}\text{Pu}$ maximum may be explained by the vertical transport of Pu associated with settling particles and subsequent release of Pu from the particulate matter to the water. This mechanism may involve breakdown of large organic-rich particles bearing Pu into small particles and remineralization of Pu into the water. Since Pu(VI) appears to be a more stable form than Pu(IV) in sea water by forming plutonyl carbonate complex [17], a change in oxidation state of Pu along with degradation of particles may also be involved in the formation of subsurface Pu maximum.

Noteworthy in *figure 3* is that the waters inside the trench valley have higher activities of Pu (0.76 and 0.30

dpm 10^{-3} L, respectively for the Japan and Bonin trenches) than most waters above. Such was found in the Aleutian trench by Bowen et al. [4] who obtained the mean Pu value of 1.1 ± 0.2 dpm 10^{-3} L from 6000 m to 7200 m there during the 1973 GEOSECS expedition. The high activities of Pu in the trench waters probably occur by lateral transport and entry into the trench of Pu-rich bottom waters along isopycnal surface from the adjacent abyssal plain, where release of Pu is taking place due to oxidation of rapidly sinking, large organic-rich particles at the sediment-water interface [10]. The sediment-trap particles collected at ~8800 m of the JT site (*figure 1*) between September 1986 and February 1988 contained 0.2–0.5 dpm g^{-1} for $^{239,240}\text{Pu}$ (*table V*). Thus, the observed variation of Pu activity in the trench waters (more than a factor of 3) is in all likelihood dependent upon the vertical particulate flux of Pu which is carried by large particles such as marine snow and fecal pellets, and hence the surface ocean productivity. The difference in the Pu activity inside the trenches of the above three locations qualitatively agrees with the knowledge of phytoplankton productivity.

Although various implications are possible for the shallow Pu maximum as described above, it is likely to occur by vertical transport of slowly sinking, smaller-sized particles that escaped repackaging through biological mediation. The maximum value of 1.6 dpm 10^{-3} L at the AN-4 station is a factor of two to three lower than that at the AN-1 site (*figure 3*), and it is also low compared with those at the GEOSECS stations from the mid-latitudes of the North Pacific [4]. This may be ascribed to the intense removal of Pu from the surface water at the AN-4 site by large particles under the high-particle flux regime.

4.2. Depth profiles of Th isotopes

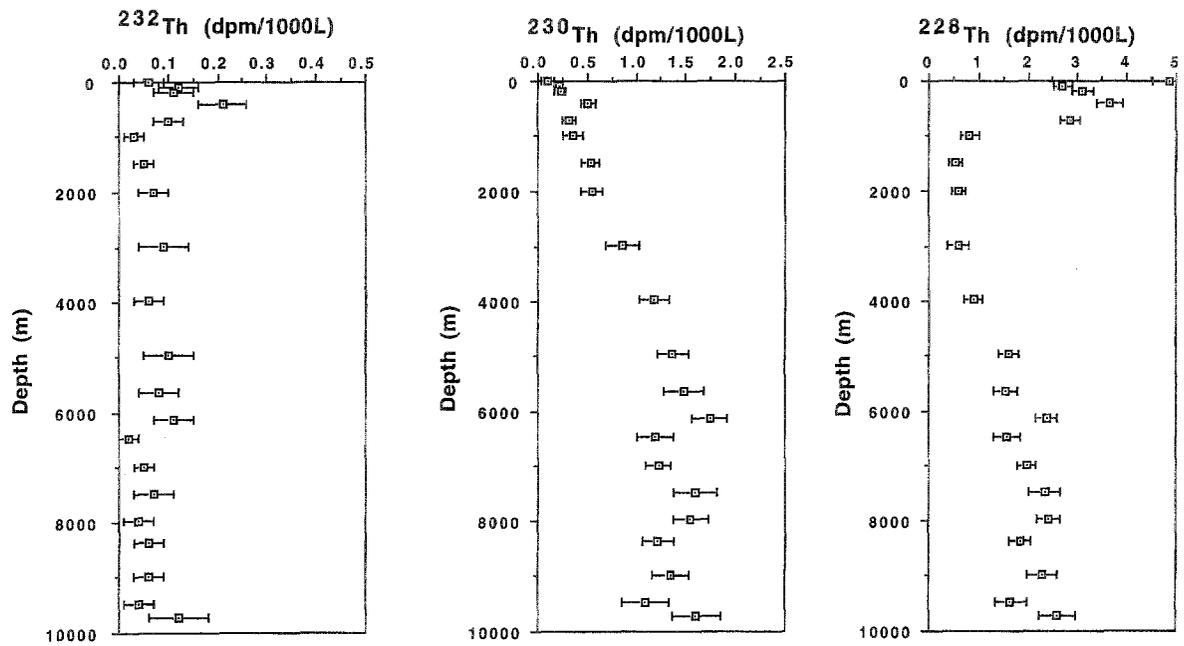
Because of its long half-life of 1.4×10^{10} years and non-radiogenic origin, ^{232}Th can be regarded as one of the reactive heavy metals whose concentrations in sea water are extremely low due to removal by particle scavenging. For the determination of ^{232}Th in sea water, more sensitive and precise methods than the α -spectrometry used here have recently been developed using thermal ionization mass spectrometry [8, 14]. Application of this method gave a variety of concentrations of ^{232}Th , depending on the locations where sea water samples were obtained. Roy-Barman et al. [39] obtained the lowest values around 16 pg kg^{-1} for dissolved ^{232}Th corresponding to ~ 0.005 dpm 10^{-3} L in the central North Pacific. In Sagami

Bay near Japan, Ida [15] reported dissolved ^{232}Th concentrations of ~ 200 pg kg^{-1} with a mean activity of ~ 0.06 dpm 10^{-3} L. Our acid-soluble ^{232}Th data in the upper water column are comparable with or higher than dissolved ^{232}Th in Sagami Bay. Somewhat higher ^{232}Th activity in shallow waters than in deep waters (*figures 4a, b*) suggests that ^{232}Th exhibits the scavenged type distribution like Al and Ce, showing decrease with depth [36]. However, this must be confirmed by thermal ionization mass spectrometry in the future.

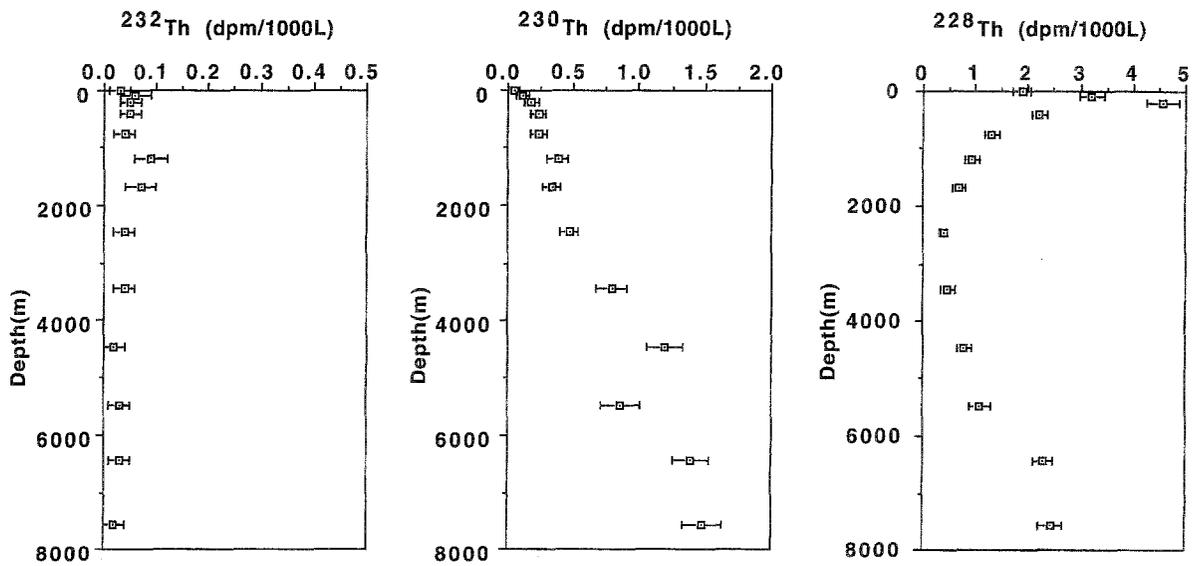
The profiles of ^{230}Th ($t_{1/2} = 75200$ y) and ^{228}Th ($t_{1/2} = 1.9$ y) down to ~ 6000 m (*figure 4*) show marked features which are similar to those in the North Pacific reported by Nozaki et al. [30, 33]. The monotonic increase of ^{230}Th concentration with depth is explained as a consequence of a constant ^{230}Th production by ^{234}U decay throughout the water column and the reversible scavenging of Th by particulate matter which settles downward [1, 33, 35]. Luo et al. [20] have shown that the ^{230}Th distribution in the water column is strongly affected by the surface ocean productivity. During the JGOFS EqPac study, the ^{230}Th concentration was systematically lower under the high productivity (and hence high-particle-flux) regime than in the low-particle-flux regime. Such a seasonal variation in the ^{230}Th profiles is consistent with the results obtained for the vertical flux of ^{230}Th by sediment-trap experiments. For example, Bacon et al. [3] found a marked seasonality in the ^{230}Th flux in the deep Sargasso Sea, corresponding to the variation in surface ocean productivity. Thus, somewhat lower concentrations of ^{230}Th , and possibly also ^{232}Th , for the same depth horizons at AN-4 compared to those at AN-1 may be attributable to the difference in the surface ocean productivity of the two locations.

Unlike ^{230}Th , the ^{228}Th profile, due to its relatively short half-life, is largely governed by its source distribution, namely decay of ^{228}Ra ($t_{1/2} = 5.7$ y). The ^{228}Ra typically shows higher concentrations in the surface waters due to lateral transport of excess ^{228}Ra (relative to ^{232}Th) from the shelf and coastal zones, and also in the bottom waters due to diffusion from sediment pore water.

Once inside the trench valley, the ^{230}Th and ^{228}Th activities do not follow the increasing trends with depth which continue from mid depths down to ~ 6000 m. They show some scatter, particularly in the Bonin Trench (*figure 4a*), but no systematic trend with depth. The question is whether this scatter of ^{230}Th and ^{228}Th data within the trench is significant. We examined the consistency of Th isotope data together with other nuclides (^{231}Pa , ^{210}Pb ,



(a)



(b)

Figure 4. Vertical profiles of Th isotopes in unfiltered water at stations AN-1 (a) and AN-4 (b).

^{227}Ac , and Pu isotopes) and chemical constituents (nutrients and trace metals) in terms of possible scavenging and water renewal processes. However, since there is no consistency among those radionuclides and other chemical properties, we regard this scatter of Th data as an experimental phenomenon rather than real.

4.3. Vertical profiles of ^{231}Pa , ^{227}Ac and ^{210}Pb

The data of ^{231}Pa ($t_{1/2} = 32800$ y), ^{227}Ac ($t_{1/2} = 21.7$ y), and ^{210}Pb ($t_{1/2} = 22.3$ y) at station AN-1 are plotted in figure 5. In general, the profiles of these nuclides from the surface to ~6000 m are similar to those in other locations of the western North Pacific [28]. The activity of ^{231}Pa is less than 1 % of the activity of its parent, ^{235}U , throughout the water column owing to scavenging by particulate matter. The increase of ^{231}Pa from the surface to the mid-depth concentration of ~ 0.5 dpm 10^{-3} L is similar to those in the western North Pacific [30]. Although a simple box model does not specify the mechanism of supplying to and removing from the ocean, it is useful to describe the overall mean residence times of elements [27]. For a soluble parent and reactive daughter pair, the mean residence time of the daughter (τ_d) is given by,

$$\tau_d = \left(\frac{R}{1-R} \right) \frac{1}{\lambda_d} \quad (1)$$

where R is the mean activity ratio of the daughter to the parent, and λ_d is the half-life of the daughter. Taking

0.4 dpm 10^{-3} L as the mean ^{231}Pa activity throughout the water column, the box-model scavenging residence time of ^{231}Pa is estimated as 180 years. The ^{231}Pa residence time is longer than that of ^{230}Th (~ 30 y [33]).

The profile of ^{210}Pb resembles that of ^{231}Pa , showing a mid-depth maximum around 3000 m. The ^{210}Pb activity in the deep waters is only half to one third of its precursor ^{226}Ra (~ 350 dpm 10^{-3} L), indicating removal of ^{210}Pb by particle scavenging. The estimated ^{210}Pb box-model residence time is relatively short at ~ 30 y, however. There are nearby GEOSECS stations and R/V Hakuho-Maru stations in the western North Pacific where the ^{210}Pb data are available. At most of the stations, the estimated ^{210}Pb residence times are around 100 years [36], decreasing from the center of the North Pacific gyre to the continental margins. In particular, the ^{210}Pb residence times in regions close to the boundary (e.g., Manus Basin and the Bering Sea) are extremely short. The present ^{210}Pb data suggests that a rapid transition in the deep-sea ^{210}Pb residence time occurs near the continental slope.

The activity of ^{227}Ac , a daughter nuclide of ^{231}Pa , is higher than its parent activities, particularly in the deep waters below 3000 m. This excess ^{227}Ac is presumably transported by eddy diffusion from the underlying sediments, very much as in the case of ^{228}Ra [21]. This is in agreement with previous findings in the deep water of the North Pacific [24] and in the interstitial water of a deep-sea sediment core [34].

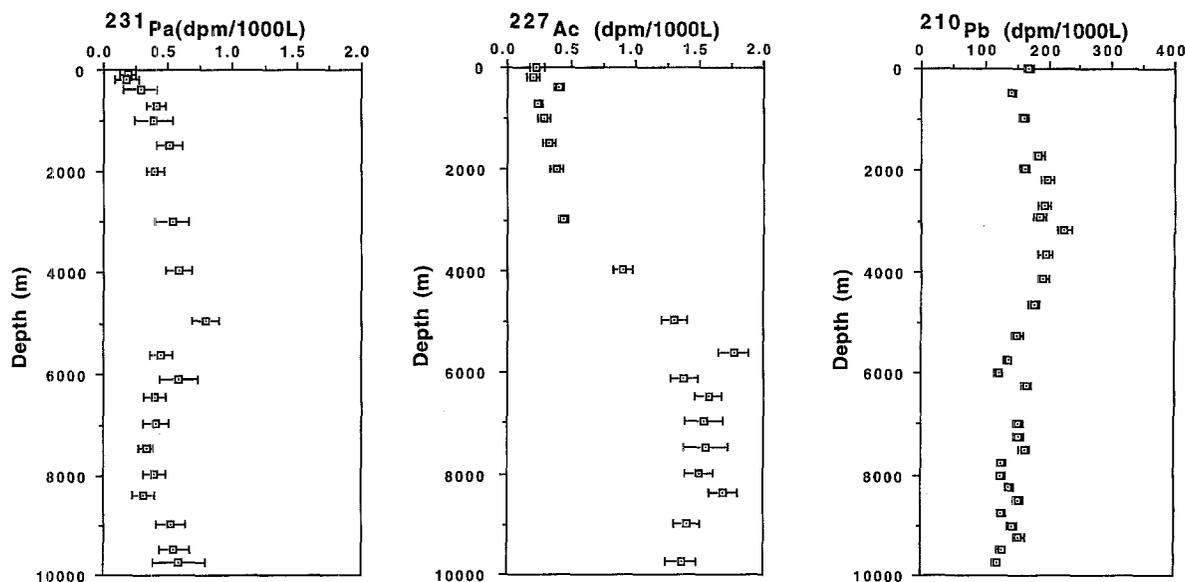


Figure 5. Vertical profiles of ^{231}Pa , ^{227}Ac , and ^{210}Pb in unfiltered water at station AN-1.

It is apparent in the three profiles shown in *figure 5* that there is no systematic trend in the activities at depths of below 6000 m. This is consistent with the distribution of Th isotopes. The uniformity of each nuclide in the trench water is presumably due to rapid vertical mixing of water within the trench.

4.4. Heavy metals

The vertical profiles of V, Zn, Cd, Cu, Mn, and Ni at station AN-1 are shown in *figure 6*. The profile of V, showing slightly lower concentrations in the upper part of the water column than in deep waters, obtained here agrees with that of Collier [9] for the eastern North Pacific. Cd, Ni, and Zn show the nutrient-type distributions as already demonstrated by Bruland [7], who has also shown that in the vertical profile, Cd resembles phosphate and nitrate, whereas Zn resembles reactive silica. The two patterns

cannot be distinguished from our data because Zn and Cd at the AN-1 site are very well correlated with each other ($r^2 = 0.96$).

Although the Cu concentration also increases with depth, the vertical pattern is discernibly different from those of Cd, Zn, and Ni. The copper shows a depth-profile of rather monotonic increase, with no mid-depth maximum. This deviation from the nutrient-type profile is attributed to the high affinity of Cu with respect to scavenging by particles [5]. Manganese also has a unique profile, showing a sub-surface maximum associated with the oxygen minimum layer (*figure 6*). This Mn profile is similar to that reported by Landing and Bruland [18] for the eastern North Pacific. Because the Mn data below 6000 m are scattered, it is difficult to draw a real trend from the data.

For all other heavy metals except for Mn, the vertical profiles are continuous down to the trench bottom, and there

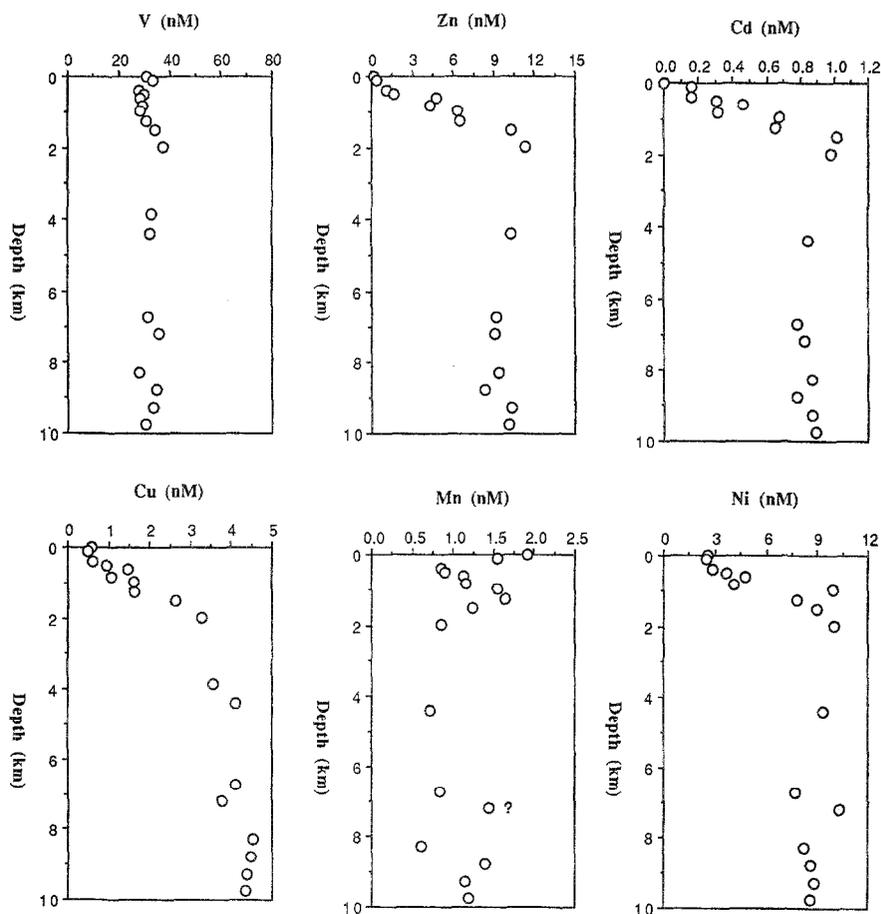


Figure 6. Vertical profiles of V, Zn, Cd, Cu, Mn, and Ni unfiltered water at the Bonin Trench station AN-1.

is no evidence that the water inside the trench is chemically different from the overlying water around 6000 m in depth. This situation in the trench is clearly different from that pertaining in anoxic basins such as the Cariaco Trench and in some Fjords like Saanich Inlet, where the density stratification limits the vertical exchange of water across the pycnocline, and thereby anoxic conditions develop in the deep waters.

4.5. Renewal of Bonin Trench water

One of the interesting questions about the trench environment is how rapidly the trench water is renewed. At first sight, the hydrographic properties (temperature, salinity, dissolved oxygen), the nutrients (*figure 2*), and trace metal concentrations (*figure 6*) in the trench waters are not significantly different from those in the bottom water overlying the western Pacific abyssal plain (~6000 m in depth), and their depth profiles are nearly constant. This implies that in situ alteration of the chemical composition of sea water is negligible within the trench, and suggests that renewal of the trench water with Northwest Pacific deep water must occur very rapidly. Some constraints on the renewal time may be obtained from the data of dissolved oxygen. In Pacific deep water near the bottom, the GEOSECS ^{14}C and dissolved oxygen data show a linear relationship with a slope corresponding to the oxygen utilization rate of $0.13 \mu\text{mol kg}^{-1}\text{y}^{-1}$ (*figure 7*). The con-

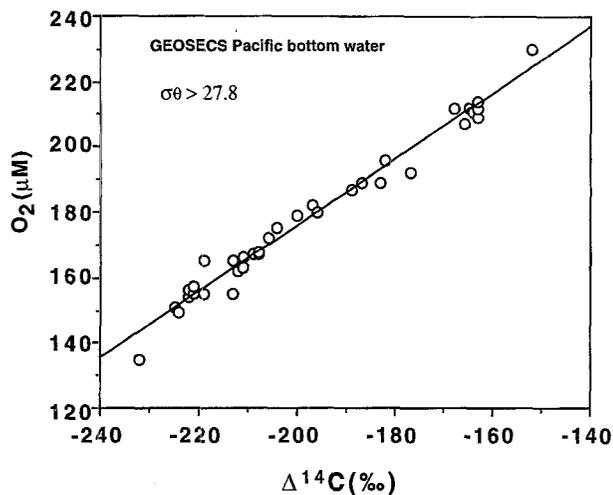


Figure 7. Correlation diagram of dissolved oxygen and $\Delta^{14}\text{C}$ in GEOSECS Pacific bottom waters (Ostlund et al., 1979). Converting the $\Delta^{14}\text{C}$ values to radiocarbon ages, an oxygen consumption rate of $0.13 \mu\text{mol kg}^{-1}\text{y}^{-1}$ is obtained from the slope of the regression line.

sumption of dissolved oxygen may take place largely at the sediment-water interface rather than in the water column. Dissolved oxygen inside the trench can only be supplied by replacement of water and it is then consumed along with the residence time of the water. The observed constancy of dissolved oxygen concentration below 6000 m at stations AN-1 and AN-4 may have an uncertainty of $\sim 5 \mu\text{mol kg}^{-1}$. Therefore, with the above oxygen utilization rate, the renewal time of water occupying the Japan and Izu-Ogasawara Trenches must be less than 40 years. The physical situations of the giant trenches in the North Pacific are clearly different from that of stratified basins like the Cariaco Trench.

Some radionuclide data are also useful in this context. ^{227}Ac and ^{228}Ra are known to be present in excess due to supply from the underlying sediments [21, 34]. The ^{227}Ac activity would be higher in the trench water than in the renewal water if additional input came from the walls of the trench. Nevertheless, it is almost vertically constant below 6000 m, and not very different from that of the bottom water overlying the western North Pacific abyssal plain [24]. This suggests that the renewal and vertical mixing of the trench water are rapid compared to the half-life of ^{227}Ac (21.7 y).

^{228}Ra with a half life of 5.7 years can provide better constraints. Although we have not measured ^{228}Ra directly, ^{228}Th may be used to deduce its parent ^{228}Ra activity. The reason for this comes from the box-model type relationship for the concordance of ($^{228}\text{Th}/^{228}\text{Ra}$) and ($^{230}\text{Th}/^{234}\text{U}$) ratios [35] given by,

$$\left(\frac{A_{\text{Ra}-228}}{A_{\text{Th}-228}}\right) = \left(\frac{\lambda_{\text{Th}-230}}{\lambda_{\text{Th}-228}}\right) \left(\frac{A_{\text{U}-234}}{A_{\text{Th}-230}}\right) + 1 \quad (2)$$

In deriving equation (2), the first-order scavenging rate constant is assumed to be the same for ^{228}Th and ^{230}Th . The measured ($^{234}\text{U}/^{230}\text{Th}$) for the deep waters is about 2000, and hence ($^{228}\text{Th}/^{228}\text{Ra}$) becomes 0.95, which is close to the equilibrium value.

The ^{228}Th data below 6000 m at station AN-1 are scattered somewhat from 1.4 to $2.6 \text{ dpm } 10^{-3} \text{ L}$. As discussed previously, this scatter may be ascribable to analytical errors, and the values are not significantly different from the mean of $2.0 \text{ dpm } 10^{-3} \text{ L}$ at 5624 and 6125 m, within the uncertainty of $\pm 30 \%$. This implies that the Bonin Trench water is replaced on a time scale much shorter than the half-life of ^{228}Ra (5.7 y). If we admit that the

variation within the trench is real and ascribable solely to decay of ^{228}Ra , then an upper limit of about five years is estimated as the time necessary for the trench water to be replaced by the overlying water. This is a conservative estimate, and the same conclusion may be applied to Japan Trench waters (see *figure 4b*).

Rapid refreshment of the trench waters may be related to the fact that these trenches are located in the western boundary with its prevailing deep currents and the Izu-Ogasawara Ridge and the Japanese Island Chain [44]. Although the deep water circulation below 6000 m has not as yet been thoroughly investigated, it is likely that the trench water may be flushed by following the prevailing deep current above the trench. However, our findings that the trench water has a uniform density and chemical characteristics similar to those around depths of 6000 m strongly suggest that the water communicates freely by isopycnal mixing with the bottom water overlying the abyssal plain of the western North Pacific.

4.6. Effect of Boundary and Bottom Scavenging

An effect of scavenging at the sediment-water interface was first proposed by Bacon et al. [2], based on the ^{210}Pb and ^{226}Ra disequilibrium distribution in the North Atlantic. Our ^{210}Pb profile data at station AN-1, which show a maximum at 3000–4000 m, then a decrease towards the abyssal plain depth (~6000 m), and constant in the trench water (*figure 5*), are consistent with boundary and bottom scavenging. ^{231}Pa shows similar features. In contrast, ^{230}Th increases almost linearly with depth down to 6000 m and remains constant at somewhat reduced activity than that at ~6000 m. The vertical profile of ^{230}Th is more strongly governed by adsorption/desorption equilibrium and the particle dynamics as described in Nozaki et al. [35], but, in the trench water, the rapid mixing tends to govern the vertical profile as modeled by Rutgers van der Loeff et al. [40] and Scholten et al. [42]. The particle associations of ^{210}Pb and ^{231}Pa are about 3–5 %, which are significantly lower than that of ^{230}Th (~20 %). Therefore, ^{210}Pb and ^{231}Pa behave differently from ^{230}Th in that they are more strongly influenced by boundary and bottom scavenging, resulting in different water column distributions.

It has been demonstrated that a large-scale fractionation between ^{231}Pa and ^{230}Th takes place in the pathways from the water column to the sedimentary sink [19, 47]. ^{230}Th tends to be removed vertically with its short residence time (~30 y), whereas ^{231}Pa is less particle-reactive than

^{230}Th , and hence is transported, within its oceanic residence time of ~150 years, to ocean margins where the nuclide is more intensively scavenged under the high-particle-flux regimes. As a result, the $^{230}\text{Th}/^{231}\text{Pa}$ activity ratios are higher in the oligotrophic central gyre and lower in the eutrophic ocean margins as compared to the value of 10.8 (the production ratio from U decay in sea water). The ^{231}Pa and ^{230}Th fractionation also takes place in the trench environment studied here. *Figure 8* shows the vertical profile of $^{230}\text{Th}/^{231}\text{Pa}$ activity ratio at station AN-1. The $^{230}\text{Th}/^{231}\text{Pa}$ ratio increases from ~1 in the surface water to ~3 in the trench water below 6000 m. These activity ratios are significantly lower than 10.8, and hence, the waters are enriched more in ^{231}Pa than ^{230}Th .

On the other hand, the $^{230}\text{Th}/^{231}\text{Pa}$ activity ratios of the suspended particles collected by in situ pumps (*table IV*) are a factor of 3–5 higher than those in sea water for the same depth horizon, and clearly show an increase with depth. Settling particles collected by sediment traps have the $^{230}\text{Th}/^{231}\text{Pa}$ activity ratios of 16 ± 5 at ~5000 m and

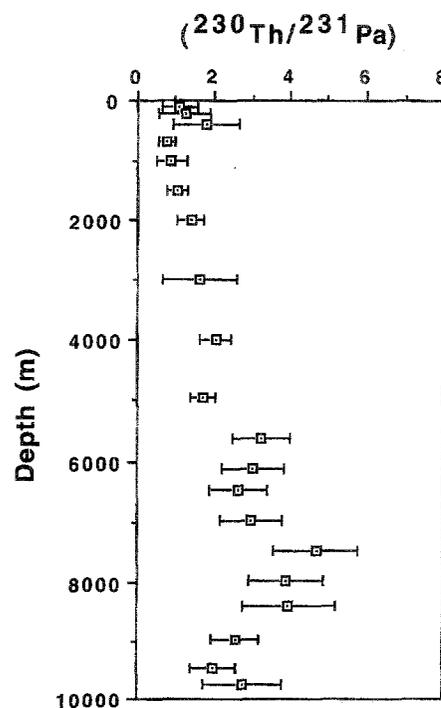


Figure 8. Depth profile of $^{230}\text{Th}/^{231}\text{Pa}$ activity ratio in unfiltered sea water at station AN-1.

13 ± 3 at 8800 m (*table V*), roughly in agreement with those of suspended particles for similar depths (*table IV*). Furthermore, the surface sediments collected at the trench bottom of the AN-1 site also gave the mean activities of 16 dpm g^{-1} for ^{230}Th and 1.1 dpm g^{-1} for ^{231}Pa and the $^{230}\text{Th}/^{231}\text{Pa}$ ratio of 15 ± 3 [32]. Although the measured vertical flux of ^{230}Th at JT site is, on average, some three times greater than that expected from production by ^{234}U decay in the water column, the fact that all the particles and sediments collected from depths greater than 5000 m have activity ratios greater than or equal to 10.8 implies that the sediments in the trench valley are not acting as an effective sink of ^{231}Pa over ^{230}Th . In contrast, suspended particles collected at depths shallower than ~3000 m are enriched in ^{231}Pa over ^{230}Th (*table IV*). This suggests that the shallow-depth and slope sediments may

be effectively accumulating ^{231}Pa over ^{230}Th in this western Pacific region.

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