DISTRIBUTION OF RADIOCARBON IN THE SOUTHWESTERN NORTH PACIFIC

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ABSTRACT. Radiocarbon measurements in seawater samples taken at six stations in the southwestern North Pacific visited during the International Atomic Energy Agency (IAEA) 1997 Pacific Ocean Expedition were carried out at the accelerator mass spectrometry (AMS) facility of the Japan Atomic Energy Research Institute (JAERI). Three stations were located close to GEOSECS stations, and three were in the vicinity of Bikini and Enewetak Atolls, which may be influenced by former nuclear weapons testing. Compared with the GEOSECS data (1973), our results show an increase of ^{14}C in intermediate waters. Furthermore, it is estimated that bomb-produced ^{14}C inventories in the water column have increased by more than 20% during the last 24 years. However, vertical profiles of $\Delta^{14}C$ at the stations near Bikini and Enewetak Atolls show a similar general trend to those found in other stations.

INTRODUCTION

In 1995, the International Atomic Energy Agency's Marine Environment Laboratory (IAEA-MEL), began a five-year project "Research on Worldwide Marine Radioactivity". In the framework of the project, the "IAEA '97 Pacific Ocean Expedition" was carried out in the southwestern North Pacific. The objective of this expedition was to develop an understanding of the present open ocean distribution of radionuclides for comparison with data sets obtained in national and international surveys of radionuclides at sites used for radioactive waste dumping or nuclear bomb tests.

The sampling work to analyze radionuclides in the southwestern North Pacific was carried out at 20 stations (Figure 1), including five GEOSECS stations (Stations 1, 2, 2B, 3, and 4) and seven stations close to Bikini and Enewetak atolls (Stations 5B, 6, 6B, 6C, 7, 7B, and 7D) (Povinec et al. 1998). Seawater samples for ^{14}C measurements were collected at 10 stations (see Figure 1).

^{14}C measurements at six stations (see Figure 1) were carried out at the AMS facility of the Japan Atomic Energy Research Institute's Marine Research Laboratory (JAERI-MRL). Three stations, Station 1, 2B and 4 were located close to the GEOSECS stations 224, 225, and 227, respectively. Three others, Stations 5, 8, and 9 were around the Bikini and Enewetak atolls. According to GEOSECS radionuclides data, these stations are influenced by close-in fallout from nuclear testing (Bowen et al. 1980). ^{14}C measurements of seawater samples collected at four additional stations were carried out at the University of Arizona and these data are given elsewhere (Jull et al. 1998). In this paper, we present the results of ^{14}C analysis of seawater samples taken in the southwestern North Pacific, and discuss temporal changes of vertical profiles over the last 24 years (1973–1997) at the GEOSECS stations. Further, we discuss the effect of close-in fallout from nuclear weapons testing at the stations around Bikini and Enewetak Atolls.

METHODS

Sample Preparation

Seawater was collected in 500–1000 mL glass bottles with high-quality stoppers and 100–200 µl of saturated $HgCl_2$ solution was added as poison on board. The DIC in seawater samples was cryogenically extracted as CO_2 gas from the samples by adding 4 mL of 100% H_3PO_4 and using pure N_2 gas

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in the vacuum system (Aramaki et al. 2000). The CO_2 gas was reduced to graphite with pure H_2 gas over an Fe catalyst at 650 °C (Kitagawa et al. 1993; Aramaki et al. 2000). Graphite was pressed into targets for AMS-^{14}C measurements.

^{14}C Measurement

The $^{14}C/^{12}C$ ratios of the sample graphite were measured by a Tandetron AMS (HVEE, model 4130-AMS) at JAERI-MRL (Aramaki et al. 2000), using a NIST oxalic acid ^{14}C standard (HoxII, SRM-4990C). The value of 0.7459 times the $^{14}C/^{12}C$ ratio of HOxII normalized to $\delta^{13}C_{PBD} = -25‰$ is equal to that of 0.95 times the $^{14}C/^{12}C$ ratio of HOxI, SRM-4990 normalized to $\delta^{13}C_{PBD} = -19‰$ (Stuiver 1983).

The ^{14}C concentration is expressed by R and Δ^{14}C values. The R value is given by the following equation (Stuiver and Polach 1977):

$$R = A_{SN} / 0.7459A_{ON} \times e^{\lambda(y-1950)}, \quad (1)$$

where A_{SN} and A_{ON} are equal to the $^{14}C/^{12}C$ ratios of a sample and HOxII normalized to $\delta^{13}C_{PBD} = -25‰$, λ is equal to 1/8267 yr^{-1} and y is equal to a measurement year. The Δ^{14}C values are given by the following equation using the calculated value for R,

$$\Delta^{14}C = (R-1) \times 1000\ (‰). \quad (2)$$

The precision of our AMS measurement is typically less than ±5‰. The accuracy of our ^{14}C measurements has been examined using IAEA intercomparison standards and HoxII standard material produced with our sample preparation system (Aramaki et al. 2000). The background level is estimated to be 0.2% fraction modern. The $\delta^{13}C_{PBD}$ measurements of a sample and the standard used to normalize are made by analyzing sub-samples of the CO_2 gas generated during graphite production, using a triple collector mass spectrometer, Finnigan, DELTA^{plus} with a precision of ±0.05‰.

RESULTS AND DISCUSSION

Radiocarbon in the Southwestern North Pacific

Figure 2 shows a vertical profile of Δ^{14}C values at Station 4 and at GEOSECS Station 227, both at the same location (25.0°N, 170.5°E). Below 2000 m water depth where no effects of nuclear weapons testing were observed, both Δ^{14}C profiles are in good agreement with each other within analytical error. Therefore, our Δ^{14}C data measured by AMS can be directly compared with the GEOSECS data obtained by β-counting.

Figure 3 and Table 1 show vertical profiles of Δ^{14}C values at six stations in the southwestern North Pacific. Generally, the profiles are typical for the North Pacific, similar to those of WOCE (Key et al. 1996). Each profile shows an excess in the surface water, decreasing sharply with depth down to 1000 m, and reaches a minimum at 2000–3000 m depth, increasing gradually to the bottom. Although in the early 1970s in the North Pacific, a maximum Δ^{14}C value was observed in the mixed layer, especially in the air-sea interface (Ostlund and Stuiver 1980), that of the Δ^{14}C value in the late 1990s was observed at 100–200 m depth, below the mixed layer. It indicates that ^{14}C from nuclear bomb tests was transferred below the mixed layer into seawater by mixing with and/or diffusing into intermediate waters over the last 24 years (1973–1997).

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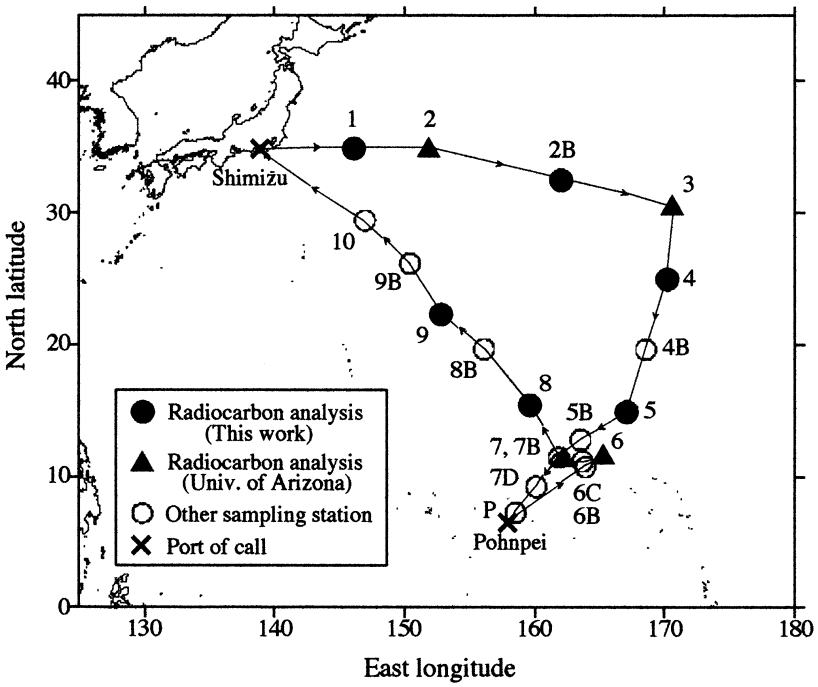

Figure 1 Route and sampling stations for the IAEA '97 Pacific Ocean Expedition

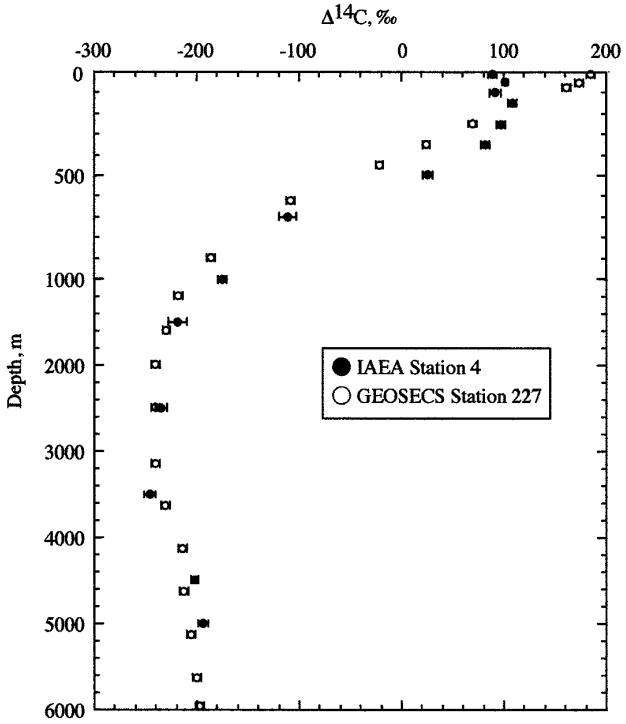

Figure 2 Comparison of $\Delta^{14}C$ vertical profiles between IAEA and GEOSECS stations. Closed and open circles denote IAEA and GEOSECS stations, respectively.

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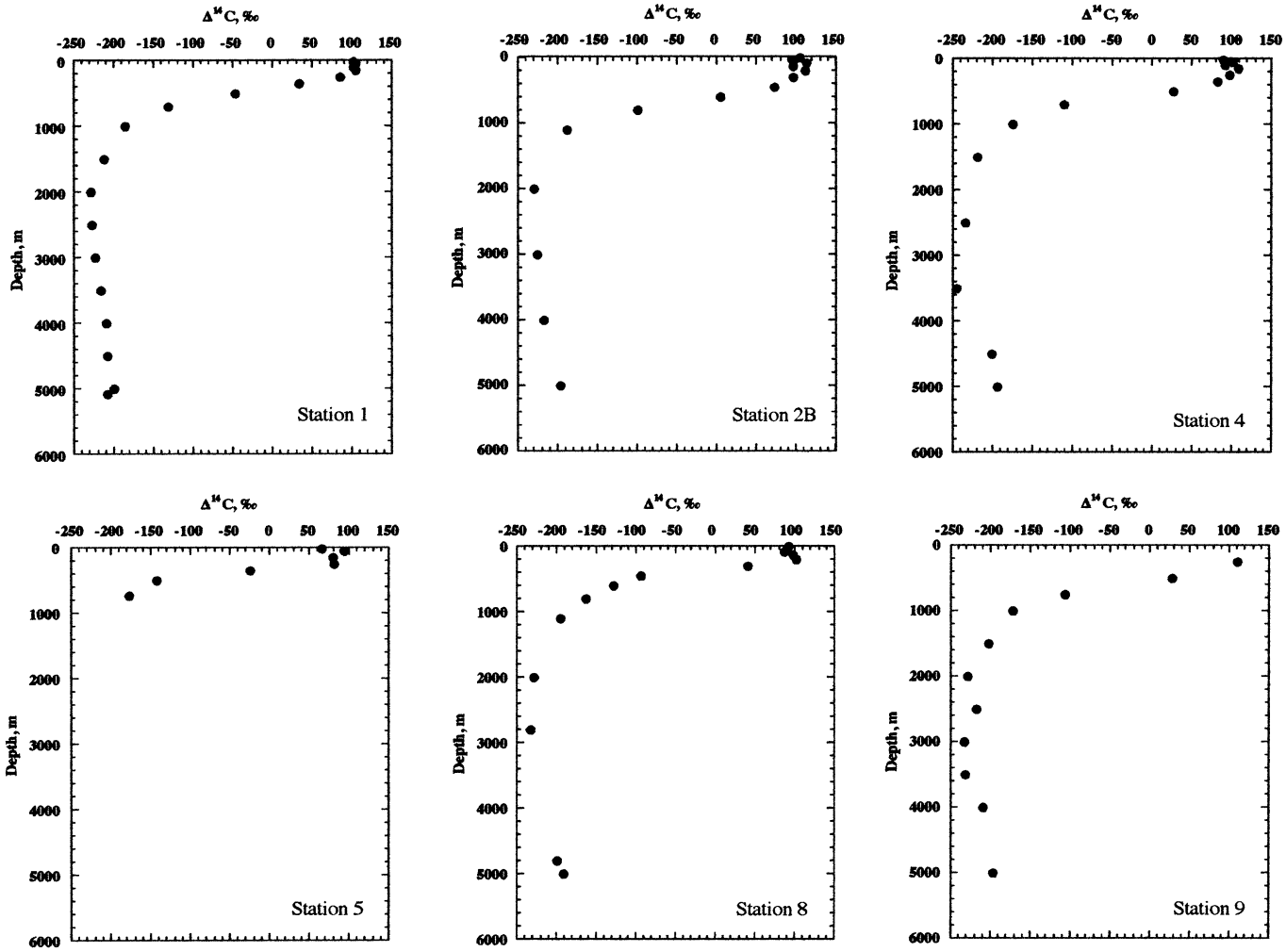

Figure 3 Vertical profiles of $\Delta^{14}C$ at six sampling stations visited during the IAEA '97 Pacific Ocean Expedition

Vertical profiles of $\Delta^{14}C$ at the stations around Bikini and Enewetak Atolls (Stations 5, 8, and 9) show a general trend similar to other stations (Stations 1, 2B, and 4). No effects of close-in fallout from nuclear weapons testing were found for radiocarbon on both of 1970s (GEOSECS) and this observation. For other radionuclides such as ^{90}Sr, ^{137}Cs, and $^{239,240}Pu$, however, this area was influenced by close-in fallout at 1970s (Bowen et al. 1980). The distribution of other radionuclides in the expedition is discussed elsewhere (Livingston et al. 2000).

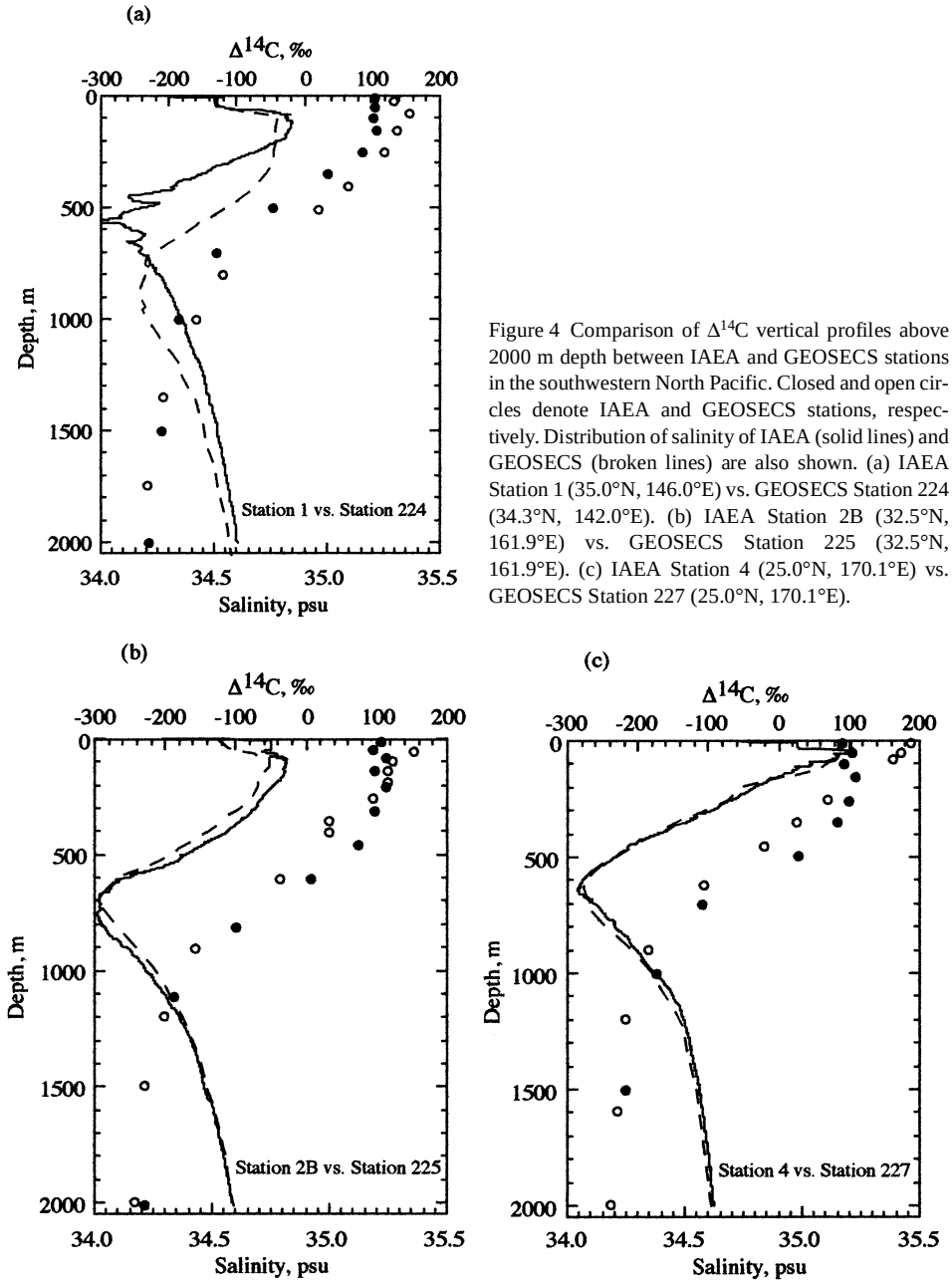

Figure 4 Comparison of $\Delta^{14}C$ vertical profiles above 2000 m depth between IAEA and GEOSECS stations in the southwestern North Pacific. Closed and open circles denote IAEA and GEOSECS stations, respectively. Distribution of salinity of IAEA (solid lines) and GEOSECS (broken lines) are also shown. (a) IAEA Station 1 (35.0°N, 146.0°E) vs. GEOSECS Station 224 (34.3°N, 142.0°E). (b) IAEA Station 2B (32.5°N, 161.9°E) vs. GEOSECS Station 225 (32.5°N, 161.9°E). (c) IAEA Station 4 (25.0°N, 170.1°E) vs. GEOSECS Station 227 (25.0°N, 170.1°E).

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Comparison with GEOSECS Data

Figure 4 compares our ^{14}C and salinity data obtained for water above 2000 m depth with those from GEOSECS (Ostlund and Stuiver 1980) at three locations, 34–35°N, 142–146°E (Station 1 and GEOSECS Station 224), 32.5°N, 161.9°E (Station 2B and GEOSECS Station 225), and 25.0°N, 170.1°E (Station 4 and GEOSECS Station 227). At Stations 2B and 4, our $\Delta^{14}C$ values in the surface layer (< 200 m) are lower than the GEOSECS values (by up to 50–80‰), but higher (by up to about 60‰) in the subsurface layer (200–1000 m). On the other hand, at Station 1, the $\Delta^{14}C$ values are lower (by up to 50‰) in the layer above 1500 m. Since other recent $\Delta^{14}C$ measurements in the subtropical western North Pacific indicate an increase of $\Delta^{14}C$ in intermediate waters (Gamo et al. 1987; Watanabe et al. 1999; Kumamoto et al. 2000), the results from Stations 2B and 4 are consistent with these results. However, at Station 1 and GEOSECS Station 224, a difference was found in the vertical profiles of salinity, which show stronger effects of the influence of the Kuroshio Extension water (high salinity and high$\Delta^{14}C$ content) at the GEOSECS station than at our Station 1. Further, it would seem that the Oyashio Current (low salinity and low $\Delta^{14}C$ content) has an influence on water at around 500 m depth at Station 1. This is because Station 1 lies about 500 km to the east of GEOSECS Station 224. The decrease of $\Delta^{14}C$ values, shown in Figure 4a, may be due to the difference in the water masses. Thus it is difficult to compare directly the data from Station 1 with those of GEOSECS Station 224.

Bomb-Produced ^{14}C Inventories

Based on observed $\Delta^{14}C$, silicate and ^{3}H data from GEOSECS (1973–1974), Broecker et al. (1995) found a good relationship between $\Delta^{14}C$ and silicate contents in deep water where no effects of nuclear weapons testing were observed. They proposed a method for reconstructing the vertical profile of the natural $\Delta^{14}C$ value ($\Delta^{14}C_{nat}$) as follows:

$$\Delta^{14}C_{nat} = -70 - SiO_2, \tag{3}$$

where the unit for SiO_2 is μmole kg^{-1} and that for $\Delta^{14}C_{nat}$ is ‰. Furthermore, the value for surface water is derived from that adopted by Broecker et al. (1985), –50‰ (for latitudes between 45°N and 45°S). The values for four selected densities (26.2 σ_ϑ, 26.4 σ_ϑ, 26.6 σ_ϑ, and 26.8 σ_ϑ for the Pacific) are calculated using the above equation, and the depth of ^{3}H-free (less than 0.1TU) water is taken as the lower limit of the penetration depth of bomb-produced ^{14}C. Figure 5 provides a plot of $\Delta^{14}C$ versus silicate at below 27.2 σ_ϑ depth at the time of the IAEA expedition. It is assumed that seawater below 27.2 σ_ϑ is Pacific deep water where no effects of bomb testing can be observed. The uncertainty of the above equation is estimated as ± 10‰ for GEOSECS (Broecker et al. 1995), however our results are applied with an uncertainty of ± 20‰. The data used to estimate the vertical profiles of $\Delta^{14}C_{nat}$ value for our 6 stations are given in Table 1. Using the vertical profiles of the observed $\Delta^{14}C$ ($\Delta^{14}C_{obs}$) and $\Delta^{14}C_{nat}$, the bomb-produced ^{14}C ($^{14}C_{bomb}$) inventories at each station were estimated to investigate the spatial distribution of $^{14}C_{bomb}$. The $^{14}C_{bomb}$ content is expressed as follows:

$$^{14}C_{bomb} = (\Delta^{14}C_{obs} - \Delta^{14}C_{nat}) \times \alpha \times \sum CO_2, \tag{4}$$

where α is the ratio of $^{14}C/^{12}C$ at the $\Delta^{14}C$ = 0‰, 1.176×10^{-12}, $\sum CO_2$ is a total carbonate content in mole kg^{-1}, at the time of "observe" and "natural", both measurements of $\sum CO_2$ at the same depth are regarded as an approximately similar value. Therefore, we can calculate the bomb-produced ^{14}C inventory (I_{bomb}) to integrate $^{14}C_{bomb}$ from surface to depth (z m) of ^{3}H-free (less than 0.1TU) by the following equation:

$$I_{bomb} = \int_0^z {}^{14}C_{bomb}\, dz \times \rho \times N_A, \tag{5}$$

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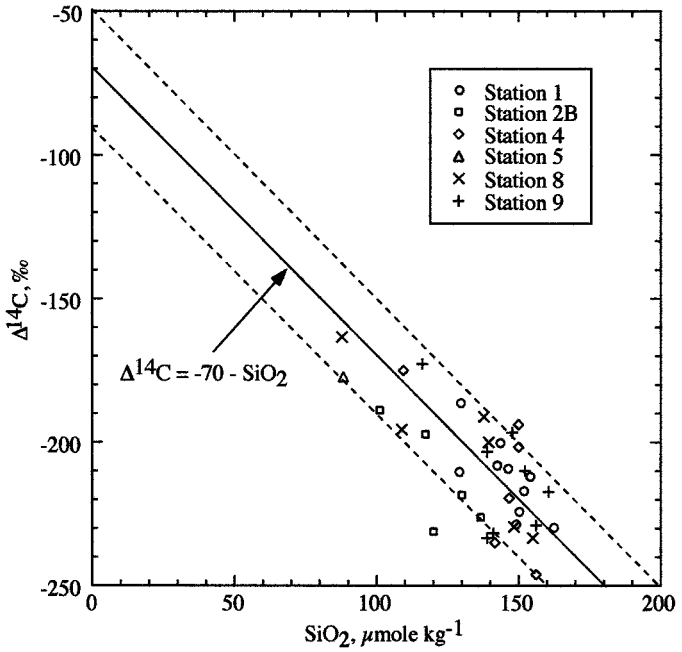

Figure 5 $\Delta^{14}C$ vs. dissolved SiO_2 at the below density 27.2 σ_ϑ depth during the IAEA '97 Pacific Ocean expedition. Also shown are scales (solid and broken lines) for natural $\Delta^{14}C$ where $\Delta^{14}C$ (± 20 ‰) = −70–SiO_2.

where ρ and N_A are the density of seawater in kg m^{-3}, and Avogadro number (= 6.02×10^{23}), respectively. The ^3H data used in the above equations were obtained during the IAEA expedition (Povinec and Togawa 1999) and decay corrected to the day of collection of the seawater sample. However, previous data on $\sum CO_2$ observed in the southwestern North Pacific were used here (Ostlund and Stuiver 1980; Tsunogai et al. 1993), as $\sum CO_2$ was not measured during the IAEA expedition.

The estimated I_{bomb} values are given in Table 2, and those of GEOSECS (Stations 224, 225, and 227) recalculated by the above equations (3) to (5) are also given to investigate the temporal change of I_{bomb} over the last 24 years (1973–1997). Here we show, respectively, I_{bomb} in the total water column, I_{bomb} above 26.2 σ_ϑ as surface water, I_{bomb} between 26.2 and 26.8 σ_ϑ as intermediate waters, and I_{bomb} below 26.8 σ_ϑ as deep water. Concerning I_{bomb} in the total water column, there is a clear distinction between the subtropical area (north of 20°N) and the tropical area (south of 20°N), which is consistent with previous studies (Broecker et al. 1985; Broecker et al. 1995; Watanabe et al. 1999). However, comparing our results (1997) with those from GEOSECS (1973), I_{bomb} in the total water column had increased by 27% for 32.5°N, 161.9°E (Stations 2B and GEOSECS Station 225), and by 21% for 25.0°N, 170.1°E (Station 4 and GEOSECS Station 227) over the last 24 years, respectively. The increase is related to that in the layer between 26.2 and 26.8 σ_ϑ. This result indicates that bomb-produced ^{14}C is transported southwards from intermediate waters in a more northern area. The water mass of density between 26.2 and 26.8 σ_ϑ in the western North Pacific, named North Pacific Intermediate Water (NPIW), is thought to be formed in the area between subarctic and subtropical regions in the western North Pacific and to flow southward (Talley 1988). Furthermore, there are reports that the excess atmospheric anthropogenic carbon (CO_2) dissolve into the NPIW at the northern North Pacific (Tsunogai et al. 1993, 1995). Our results support these reports. However, more measurements of ^{14}C at GEOSECS stations are required to confirm this observation.

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Table 1 Data for the stations occupied during the IAEA 1997 Pacific Ocean expedition

Depth (m)	Potential temp (°C)	Salinity (psu)	Density (σ_ϑ)	SiO_2 (μmole kg^{-1})	Observed $\Delta^{14}C$ (‰)
Station I 35.0°N, 146.0°E 23 Oct. 1997					
9.4	23.1336	34.512	23.5268	2.2	102.1 ± 2.7
49.2	23.1502	34.531	23.5362	2.2	103.6 ± 3.3
100.1	19.3916	34.825	24.7878	3.7	101.6 ± 3.1
150.8	18.0977	34.834	25.1220	4.3	104.9 ± 2.9
250.3	15.4629	34.687	25.6292	12.9	84.9 ± 2.3
350.0	12.0961	34.450	26.1442	26.9	33.8 ± 4.8
500.2	7.8304	34.193	26.6653	53.2	–47.7 + 4.6
700.1	4.7895	34.168	27.0397	94.7	–131.2 ± 2.5
1000.4	3.3299	34.353	27.3379	129.4	–186.0 ± 3.0
1499.8	2.2886	34.513	27.5583	153.7	–211.9 ± 5.4
2000.2	1.7935	34.601	27.6682	162.5	–229.5 ± 3.9
2502.2	1.4950	34.647	27.7269	148.7	–228.1 ± 9.1
3001.2	1.3227	34.668	27.7566	150.3	–224.1 ± 3.7
3501.0	1.2211	34.680	27.7728	151.9	–216.6 ± 5.1
4001.5	1.1375	34.688	27.7851	128.8	–209.7 ± 4.1
4500.0	1.0902	34.692	27.7921	146.0	–208.7 ± 4.1
5000.0	1.0584	34.695	27.7965	143.5	–199.9 + 4.9
5088.2	1.0554	34.695	27.7969	142.3	–207.6 ± 5.9
Station 2B 32.5°N, 161.9°E 28 Oct. 1997					
8.7	23.1825	34.749	23.6923	1.6	104.5 ± 3.9
40.0	23.1828	34.751	23.6935	1.3	93.9 ± 4.5
79.3	19.2756	34.798	24.7971	2.7	112.0 ± 6.3
130.3	17.5203	34.804	25.2398	3.7	95.7 ± 5.2
201.1	16.3669	34.756	25.4766	4.7	111.3 ± 5.0
303.0	15.2293	34.684	25.6782	6.0	95.6 ± 4.0
453.5	12.6032	34.468	26.0596	17.1	72.1 ± 3.9
600.8	8.6574	34.131	26.4921	33.5	4.6 ± 4.4
803.3	5.1633	34.031	26.8883	75.7	–100.1 ± 4.6
1104.2	3.5471	34.296	27.2713	101.3	–188.8 ± 4.6
2000.8	1.9040	34.584	27.6456	120.0	–231.1 ± 5.6
3001.9	1.3676	34.663	27.7492	136.7	–226.0 ± 4.3
3996.9	1.1680	34.684	27.7803	129.8	–218.1 ± 3.9
5000.2	1.0642	34.694	27.7954	117.3	–197.4 ± 6.0
Station 4 25.0°N, 170.1°E 31 Oct. 1997					
10.1	27.0171	34.979	22.6986	1.7	88.4 ± 4.2
48.8	26.7438	35.207	22.9576	1.6	101.8 ± 3.0
99.5	20.0206	35.042	24.7894	2.3	91.7 ± 6.0
150.0	17.9702	34.912	25.2130	3.4	108.2 ± 4.2
254.9	15.8375	34.713	25.5644	6.4	97.2 ± 4.4
350.0	13.4339	34.508	25.9246	9.9	82.0 ± 4.3
495.9	9.6711	34.207	26.3886	25.7	25.8 ± 5.0
698.7	5.3658	34.110	26.9274	79.8	–111.2 ± 8.3
1001.7	3.6722	34.370	27.3182	109.4	–174.8 ± 4.1
1499.7	2.5323	34.563	27.5774	146.8	–219.2 ± 9.1
2499.1	1.5320	34.652	27.7285	141.8	–234.9 ± 5.2
3500.9	1.2501	34.679	27.7705	156.3	–246.2 ± 5.3
4497.5	1.0840	34.693	27.7926	150.0	–210.5 ± 3.6
5000.0	1.0134	34.698	27.8019	150.0	–194.1 ± 5.0
Station 5 15.0°N, 166.0°E 3 Nov. 1997					
10.4	28.5048	34.396	21.7775	1.7	65.8 ± 3.1
48.6	28.5032	34.397	21.7788	1.7	93.1 ± 2.2
150.4	21.4257	35.048	24.4152	2.6	79.6 ± 5.3

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Table 1 Data for the stations occupied during the IAEA 1997 Pacific Ocean expedition *(Continued)*

Depth (m)	Potential temp (°C)	Salinity (psu)	Density (σ_ϑ)	SiO_2 (μmole kg^{-1})	Observed $\Delta^{14}C$ (‰)
247.8	13.2639	34.441	25.9073	11.7	80.5 ± 3.9
351.5	9.1741	34.243	26.4974	36.3	–24.5 ± 3.6
498.6	7.2111	34.428	26.9393	59.5	–143.1 ± 10.5
735.6	5.2443	34.505	27.2542	88.1	–177.2 ± 3.9
Station 8 15.5°N, 159.5°E 14 Nov. 1997					
8.6	28.5193	34.218	21.6383	4.0	91.5 ± 2.3
39.6	28.5031	34.296	21.7025	2.8	90.7 ± 3.6
79.0	26.6899	34.852	22.7069	2.2	86.1 ± 2.4
128.8	23.4007	35.071	23.8732	3.2	97.9 ± 2.6
199.7	18.2700	34.930	25.1529	7.0	100.8 ± 2.7
299.9	11.0473	34.313	26.2327	23.0	40.4 ± 4.1
451.9	8.0756	34.430	26.8151	51.0	–94.4 ± 2.7
600.2	6.7710	34.467	27.0305	67.0	–129.7 ± 2.3
800.8	5.4914	34.499	27.2204	87.8	–163.5 ± 1.9
1101.9	4.1449	34.546	27.4102	108.8	–195.4 ± 3.0
2000.4	2.1037	34.629	27.6659	148.4	–229.3 ± 3.6
2801.0	1.5062	34.668	27.7434	155.1	–233.5 ± 2.7
4799.1	1.0292	34.701	27.8032	139.7	–200.2 ± 5.5
5000.0	1.0164	34.702	27.8047	137.9	–217.4 ± 2.6
Station 9 22.4°N, 152.7°E 16 Nov. 1997					
248.4	17.0480	34.835	25.3771	7.0	109.5 ± 3.0
500.7	10.3047	34.257	26.3197	29.4	27.8 ± 2.6
748.9	5.2792	34.157	26.9749	79.6	–106.2 ± 4.1
1001.6	3.9858	34.397	27.3079	116.4	–172.6 ± 2.2
1500.1	2.6477	34.547	27.5546	138.9	–203.3 ± 4.4
2000.4	1.9180	34.609	27.6651	156.3	–229.1 ± 5.3
2499.7	1.5475	34.650	27.7259	160.7	–217.4 ± 2.6
2997.5	1.3639	34.668	27.7538	138.7	–233.2 ± 3.3
3499.6	1.2397	34.680	27.7723	141.0	–231.4 ± 7.4
3998.3	1.1551	34.688	27.7843	152.5	–210.0 ± 8.0
5001.1	1.0498	34.697	27.7989	147.7	–196.8 ± 2.3

Table 2 Bomb-produced ^{14}C inventories (I_{bomb}) in the southwestern North Pacific in 1997 (IAEA) and 1973 (GEOSECS)

				Bomb ^{14}C inventory, I_{bomb} ($\times 10^9$ atom cm^{-2})			
Area	Station	Latitude (°N)	Longitude (°E)	Total	Above 26.2 σ_ϑ	26.2–26.8 σ_ϑ	Below 26.8 σ_ϑ
North of 20°N							
	1	35.0	146.0	13.4	9.1	2.9	1.4
	2B	32.5	161.9	17.7	12.5	4.2	1.0
	4	25.0	170.1	13.9	10.0	3.0	0.9
	9	22.4	152.7	15.7	12.5	2.9	0.3
South of 20°N							
	5	15.0	166.0	8.4	6.7	1.7	0.0
	8	15.5	159.5	9.5	6.9	1.9	0.7
GEOSECS							
	224	34.3	142.0	14.7	13.3	1.3	0.1
	225	32.5	161.9	13.9	10.5	2.5	0.9
	227	25.0	170.1	11.5	9.9	1.5	0.1

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CONCLUSIONS

After measuring ^{14}C in seawater samples taken in the southwestern North Pacific in 1997, we can draw the following conclusions:

1. Generally the vertical profiles of ^{14}C were typical for the North Pacific, similar to those of WOCE. No effect of close-in fallout from bomb testing was found at the stations around Bikini and Enewetak Atolls.
2. Compared with the GEOSECS ^{14}C data (1973), our results showed an increase of ^{14}C in intermediate waters. After estimating the bomb-produced ^{14}C inventory, the value had increased by more than 20% over the last 24 years, which may have been caused by an increase in the layer between 26.2 and 26.8 σ_ϑ.

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