

RADIOCARBON IN THE WATER COLUMN OF THE SOUTHWESTERN NORTH PACIFIC OCEAN—24 YEARS AFTER GEOSECS

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ABSTRACT. In the framework of the Worldwide Marine Radioactivity Studies (WOMARS) project, water profile samples for radiocarbon measurements were taken during the IAEA'97 cruise at 10 stations in the southwestern North Pacific Ocean. While ¹⁴C concentrations were rapidly decreasing from the surface ($\Delta^{14}\text{C}$ about 100‰) down to about 800 m at all visited stations ($\Delta^{14}\text{C}$ about –200‰), the concentrations below 1000 m were almost constant. Some stations were in proximity to the GEOSECS stations sampled in 1973; thus, ¹⁴C profiles could be compared after a 24-yr interval. Generally, ¹⁴C concentrations had decreased in surface waters (by 50–80‰) and increased (by about the same amount) in intermediate waters when compared with GEOSECS data. In deep waters (below 1000 m), the observed ¹⁴C concentrations were similar to GEOSECS values. The bomb-produced ¹⁴C inventory had increased by more than 20% over the 24 yr from 1973 to 1997 and was estimated to be about $(32 \pm 5) 10^{12}$ atom m⁻², with an annual ¹⁴C flux of $(1.3 \pm 0.3) 10^{12}$ atom m⁻² yr⁻¹. The results suggest that bomb-produced ¹⁴C has been advected northwards by the Kuroshio Current and the Kuroshio Extension and stored in the intermediate layer as North Pacific Intermediate Water.

INTRODUCTION

From 1995 to 2002, the International Atomic Energy Agency's Marine Environment Laboratory (IAEA-MEL), Monaco, in collaboration with 35 institutes, carried out a research project entitled Worldwide Marine Radioactivity Studies (WOMARS). The main objectives were the following:

- To identify the major sources of anthropogenic radionuclides in the world's oceans and seas;
- To develop present knowledge of concentrations and distributions of the key radionuclides—³H, ¹⁴C, ⁹⁰Sr, ¹²⁹I, ¹³⁷Cs, and Pu isotopes—in water and sediment of the world's oceans and seas;
- To study the changes in radionuclide concentrations in the oceans with time using good quality historical data (e.g. from the Geochemical Ocean Sections Programme [GEOSECS] of the mid-1970s) and new data sets collected recently.

In the framework of the WOMARS project, in November 1997, IAEA-MEL organized a research cruise (IAEA'97 Pacific Ocean Expedition) to sample seawater, sediment, and biota in the southwestern North Pacific Ocean. The objectives of this expedition were to develop an understanding of the present open ocean distribution of radionuclides for comparison with data sets obtained in an earlier (GEOSECS) and a more recent (World Ocean Circulation Experiment [WOCE]) international survey of radionuclides in the same region and, specifically, to trace global fallout radionuclides and radionuclides produced during nuclear weapons tests (local fallout) carried out at the Marshall Islands (Povinec and Togawa 1999; Povinec et al. 2003).

Background Oceanography

Specific circulation patterns in the southwestern North Pacific Ocean make radionuclide tracers particularly useful. In the equatorial region, the North Equatorial Current (NEC) is the dominant surface current which flows westward at the southern boundary of the survey area (around Pohnpei),

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and on reaching the island of Honshu, merges into the Kuroshio Current. The Kuroshio Current leaves the Japanese coast at between 36°N and 39°N and meets the Oyashio Current (subarctic gyre). The Kuroshio Extension bifurcates at around 155°E into the main current and a northern branch (Kawabe and Taira 1998).

The dominant water masses in the North Pacific are the North Pacific Tropical Waters (NPTW), the North Pacific SubTropical Mode Waters (NPSTMW), and the North Pacific Intermediate Waters (NPIW). NPTW are composed of saline surface water produced in the central region of the North Pacific, while NPSTMW and NPIW are generated in the western North Pacific. NPSTMW are defined as a voluminous water mass in an isothermal layer around 16.5 °C and a layer of potential vorticity minimum $< 2 \times 10^{-12} \text{cm}^{-1} \text{s}^{-1}$, which is formed by deep vertical convection in winter just south of the Kuroshio Current and the Kuroshio Extension (Suga et al. 1989). NPIW exist in the intermediate minimum salinity layer at isopycnal 26.8 σ_θ (potential density) and at isopycnal 27.28 σ_θ , respectively (Reid 1965). NPIW are formed in the region where subtropical waters mix with subarctic waters and then extend to the subtropical gyre (Talley 1993). The deep waters in the investigated area are composed of North Pacific Deep Waters (NPDW).

¹⁴C in the Water Column of the Southwestern North Pacific

Radiocarbon enters the ocean through air-sea CO₂ gas exchange, with an equilibration time of 7 to 10 yr. It mainly exists as dissolved inorganic carbonic acid in seawater. Due to its radioactive decay, dissolved inorganic ¹⁴C (DI¹⁴C) in the ocean decreases from the time it enters the ocean, allowing DI¹⁴C/¹²C ratios in seawater to be used to calculate the “age” of seawater (Tsunogai 1981). In the 1950s and 1960s, atmospheric testing of nuclear weapons released large amounts of ¹⁴C into the atmosphere, approximately doubling the pre-bomb levels (Levin et al. 1985). Subsequently, surface water in the ocean containing ¹⁴C originating from these tests gradually spread deeper into the ocean. Thus, the ¹⁴C of bomb origin has provided a means to study air-sea gas exchange and trace intermediate and deep water circulation in the oceans over the last few decades (e.g. Broecker et al. 1985). Broecker et al. (1995) found a relationship between ¹⁴C concentration and silicate contents in deep water where no effects of bomb tests were observed, and proposed a method for separation of natural and bomb components of observed vertical $\Delta^{14}\text{C}$ profiles. Their method is widely used for estimating the bomb-¹⁴C inventory in the ocean (Peng et al. 1998; Lebourcier et al. 1999; Watanabe et al. 1999).

In this paper, we present and discuss ¹⁴C data obtained for the water column of the southwestern North Pacific Ocean sampled during the IAEA'97 Pacific Ocean Expedition. Some of the northern stations were located close to GEOSECS stations, and preliminary results have been published by Aramaki et al. (2001). In the current paper, we present the complete results and discuss the behavior of bomb ¹⁴C in the southwestern North Pacific, including the bomb-produced ¹⁴C inventory, over the 24-yr interval between 1973 and 1997.

METHODS

Sampling

In 1997 (21 October–20 November), seawater profile samples for ¹⁴C measurements were collected at 10 stations in the southwestern North Pacific during the “IAEA'97 Pacific Ocean Expedition” on board the research vessel “Bosei Maru” (Tokai University, Japan) (Figure 1). Four IAEA stations (IAEA-2, 2B, 3, and 4) were close to GEOSECS stations (GEOSECS-223, 225, 226, and 227) sampled in 1973. IAEA Station 1 was located between GEOSECS Stations 224 and 223. IAEA Stations

6 and 7 were located close to the Bikini and Enewetak Atolls, respectively. IAEA Stations 5, 8, and 9, located north of the Marshall Islands, were chosen in order to trace radionuclides released as local fallout from the nuclear weapons tests.

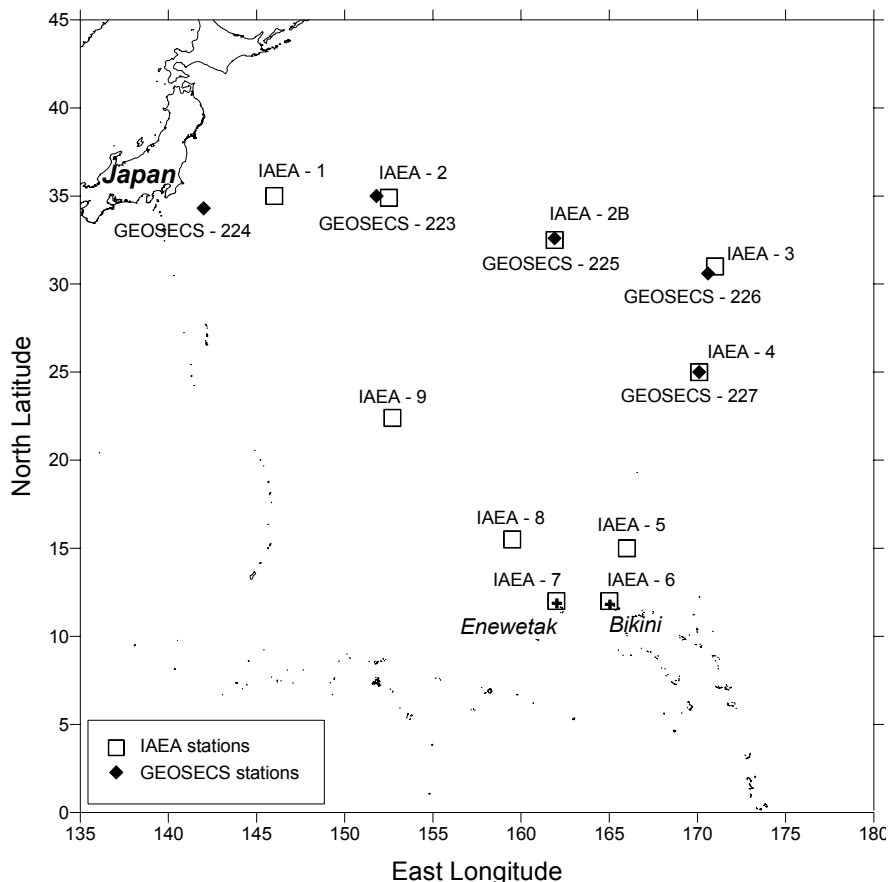


Figure 1 Sampling stations for ¹⁴C measurements of the IAEA '97 Pacific Ocean Expedition and GEOSECS stations (1973)

Seawater samples for ¹⁴C measurements were collected at each station using a CTD/RMS system equipped with Niskin bottles. Collected samples were stored in 500-mL glass bottles with hermetic stoppers mixed with 100 μL of saturated HgCl₂ solution to poison the sample.

Sample Preparation and ¹⁴C Measurements

¹⁴C measurements of samples taken at 6 stations (1, 2B, 4, 5, 8, and 9) were carried out at the Japan Atomic Energy Research Institute's Marine Research Laboratory in Mutsu, Japan (JAERI-MRL), and measurements of samples from the other 4 stations (2, 3, 6, and 7) were carried out at the University of Arizona's Accelerator Mass Spectrometry (AMS) facility in Tucson, Arizona. The seawater samples were prepared for the measurement of the ¹⁴C/¹²C or ¹⁴C/¹³C ratios in dissolved inorganic carbon (DIC) by both JAERI-MRL and the University of Arizona, using each laboratory's specific methods.

In the JAERI-MRL measurements, the DIC in the sample was extracted as CO₂ gas by adding 4 mL of 100% H₃PO₄, stripped using pure N₂ gas, and cryogenically collected in a vacuum (Aramaki et

al. 2000). The CO₂ gas was reduced to graphite with pure H₂ gas over an Fe catalyst at 650 °C (Kitagawa et al. 1993; Aramaki et al. 2000). Graphite was pressed into targets for AMS ¹⁴C measurements. The stable isotopic composition of sub-samples of the CO₂ gas was measured in a Finnigan mass spectrometer, DELTA^{plus}. The ratio of ¹⁴C to ¹²C was measured by a Tandem AMS (HVVEE, model 4130-AMS) at JAERI-MRL. The ¹⁴C activity in seawater samples has been expressed as Δ¹⁴C (Stuiver 1980; Aramaki et al. 2001).

In the University of Arizona measurements, the DIC in seawater was extracted by acidification of the sample to pH 3 and the CO₂ released was cryogenically extracted in a flow of high-purity O₂ gas. The CO₂ gas was reduced to graphite over an Fe catalyst, following the procedure described by Donahue et al. (1990a). Graphite was pressed into targets for AMS ¹⁴C measurements. The stable isotopic composition of sub-samples of CO₂ gas was measured in an Optima mass spectrometer. The ratio of ¹⁴C to ¹³C was measured at the NSF Accelerator Facility for Radioisotope Analysis at the University of Arizona. The ¹⁴C activity in seawater samples is expressed by the Δ¹⁴C value (Stuiver 1980; Donahue et al. 1990b).

Intercomparison measurements were carried out on seawater samples collected at Station 1 which showed good agreement between both laboratories (Liong et al., these proceedings).

RESULTS AND DISCUSSION

¹⁴C Vertical Profiles

¹⁴C vertical profile measurements are presented in Figure 2 and Table 1 (only data from the University of Arizona [Stations 2, 4, 6, and 7] are given in the table, as the JAERI-MRI data were published by Aramaki et al. 2001). All profiles have similar curves, although differences can be observed at surface and sub-surface water depths. Generally, the Δ¹⁴C values below 1000 m are the same within uncertainties, except for Station 7 which shows the lowest observed Δ¹⁴C values (at the surface and 1000–2500 m water depths) in contrast to the other radionuclides measured (Povinec et al. 2003). We do not understand at present the reason for this offset, since there are not available comparable data from GEOSECS and WOCE. Surface concentrations (in top 150 m) differ for different stations, and as shown in Figure 3, there is still a visible latitudinal effect, although it probably differs from the original global fallout input. A similar trend was observed for other radionuclides as well (Povinec et al. 2003).

Comparison with WOCE and GEOSECS Data

A very comprehensive Δ¹⁴C data set for the Pacific Ocean has been published recently by the WOCE collaboration (Key et al. 2002). Although the WOCE stations in the southwestern Pacific (sampled in 1993) differ from the WOMARS stations, a comparison presented in Figure 4 for stations in similar latitudinal belts (IAEA Stations 1, 2B, and 3 versus WOCE Stations 78 and 80) shows that WOMARS data are comparable to WOCE data.

A comparison of our Δ¹⁴C profiles with GEOSECS results (Ostlund and Stuiver 1980) are presented in Figure 5 (only stations situated in the northwest Pacific could be compared, as there are no GEOSECS data for stations in the southwest). At Stations 2B, 3, and 4, our Δ¹⁴C 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 sub-surface layer (200–800 m). As other recent ¹⁴C measurements in the subtropical western North Pacific also indicate an increase of Δ¹⁴C values in intermediate waters (Gamo et al. 1987; Watanabe et al. 1999; Aramaki 2002; Key et al. 2002), our results are consistent with their observations.

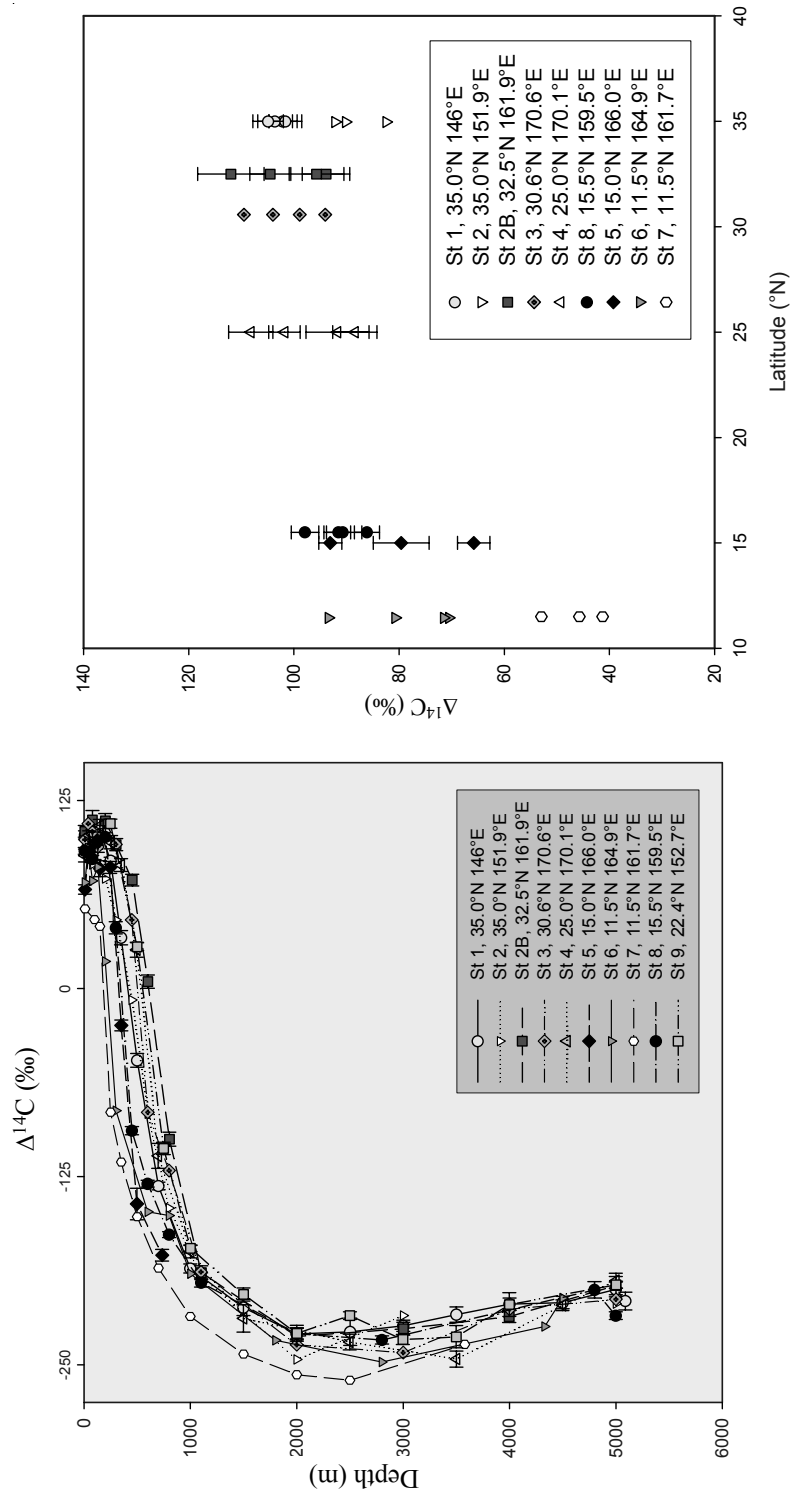


Figure 2. Vertical $\Delta^{14}\text{C}$ profiles observed at IAEA'97 stations

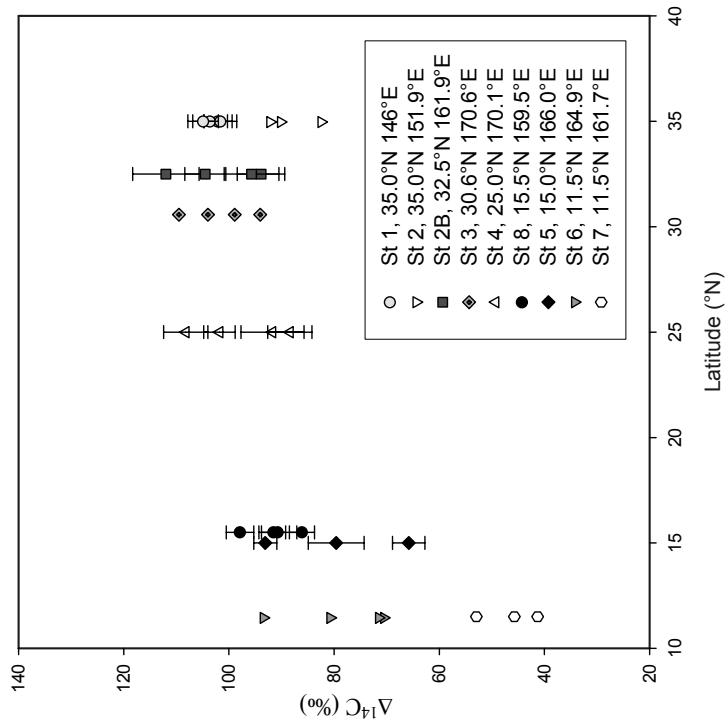


Figure 3. $\Delta^{14}\text{C}$ in surface water (top 1.50 m) as a function of latitude

Table 1 $\Delta^{14}\text{C}$ data for IAEA'97 Stations 2, 3, 6, and 7.

Location	Depth (m)	$\Delta^{14}\text{C}$ (‰)	Location	Depth (m)	$\Delta^{14}\text{C}$ (‰)
Station 2			Station 6		
34.97°N, 151.89°E	10	92.2 ± 5.1	11.45°N, 164.88°E	10	70.5 ± 4.1
	40	90.2 ± 5.0		40	93.4 ± 5.2
	80	82.5 ± 4.2		80	71.5 ± 4.2
	130	—		130	80.7 ± 4.6
	200	73.0 ± 4.1		200	18.1 ± 2.9
	300	45.7 ± 2.9		300	-81.1 ± -4.4
	450	-7.6 ± -3.0		450	—
	600	-83.3 ± -4.3		600	-148.3 ± -7.5
	800	-145.9 ± -7.5		800	-150.8 ± -8.0
	1100	-191.2 ± -10.0		1000	-189.6 ± -9.7
	2000	-246.5 ± -12.0		1800	-233.7 ± -12.1
3000	-217.3 ± -11.0	2800	-248.2 ± -1.2		
4000	—	4330	-224.5 ± -11.5		
5000	-209.4 ± -10.6	4500	-205.9 ± -10.7		
Station 3			Station 7		
30.57°N, 170.62°E	10	98.9 ± 5.5	11.5°N, 161.74°E	10	53.0 ± 3.4
	40	109.5 ± 5.8		100	45.7 ± 2.9
	80	104.0 ± 5.7		150	41.3 ± 2.9
	130	94.0 ± 5.3		250	-82.2 ± -4.8
	200	—		350	-115.3 ± -6.6
	300	95.8 ± 5.3		500	-151.7 ± -8.1
	450	45.6 ± 2.8		700	-185.8 ± -9.6
	600	-82.2 ± -4.3		1000	-217.9 ± -11.1
	800	-121.0 ± -6.4		1500	-242.9 ± -12.1
	1100	-188.3 ± -9.8		2000	-256.6 ± -13.0
	2000	-236.7 ± -12.0		2500	-260.3 ± -13.0
3000	-241.9 ± -12.1	3580	-236.5 ± -1.2		
4000	-213.1 ± -11.3	—	—		
5000	-206.5 ± -10.5	—	—		

On the other hand, at Stations 1 and 2, the $\Delta^{14}\text{C}$ values are lower (by up to 50‰) in the layer above 1500 m. At these stations, the differences between our and GEOSECS observations are found in vertical salinity profiles as well, and this may be caused by the influence of the saline Kuroshio Extension waters in the sampling region (Aramaki et al. 2001). Furthermore, Station 1 lies about 500 km to the east of GEOSECS Station 224 (see Figure 1). Thus, the differences in $\Delta^{14}\text{C}$ vertical profiles at these stations are due to the presence of different water masses at the time of the 2 sampling missions, and it is difficult to directly compare data from Stations 1 and 2 with data from GEOSECS Stations 224 and 223, respectively. In Figure 6, $\Delta^{14}\text{C}$ values at Stations 1 and 2 are plotted against density (σ_θ), and compared with GEOSECS Stations 224 and 223, respectively. Our $\Delta^{14}\text{C}$ values above 25.5 σ_θ (250 m at Station 1 and 170 m at Station 2) are lower than the GEOSECS values, but higher in the intermediate water layer with 25.5–27.0 σ_θ (27.0 σ_θ = 680 m at Station 1 and 27.0 σ_θ = 700 m at Station 2). Stations 2B, 3, and 4 show a similar tendency, confirming that at least at Station 2, water masses have changed since the GEOSECS mission.

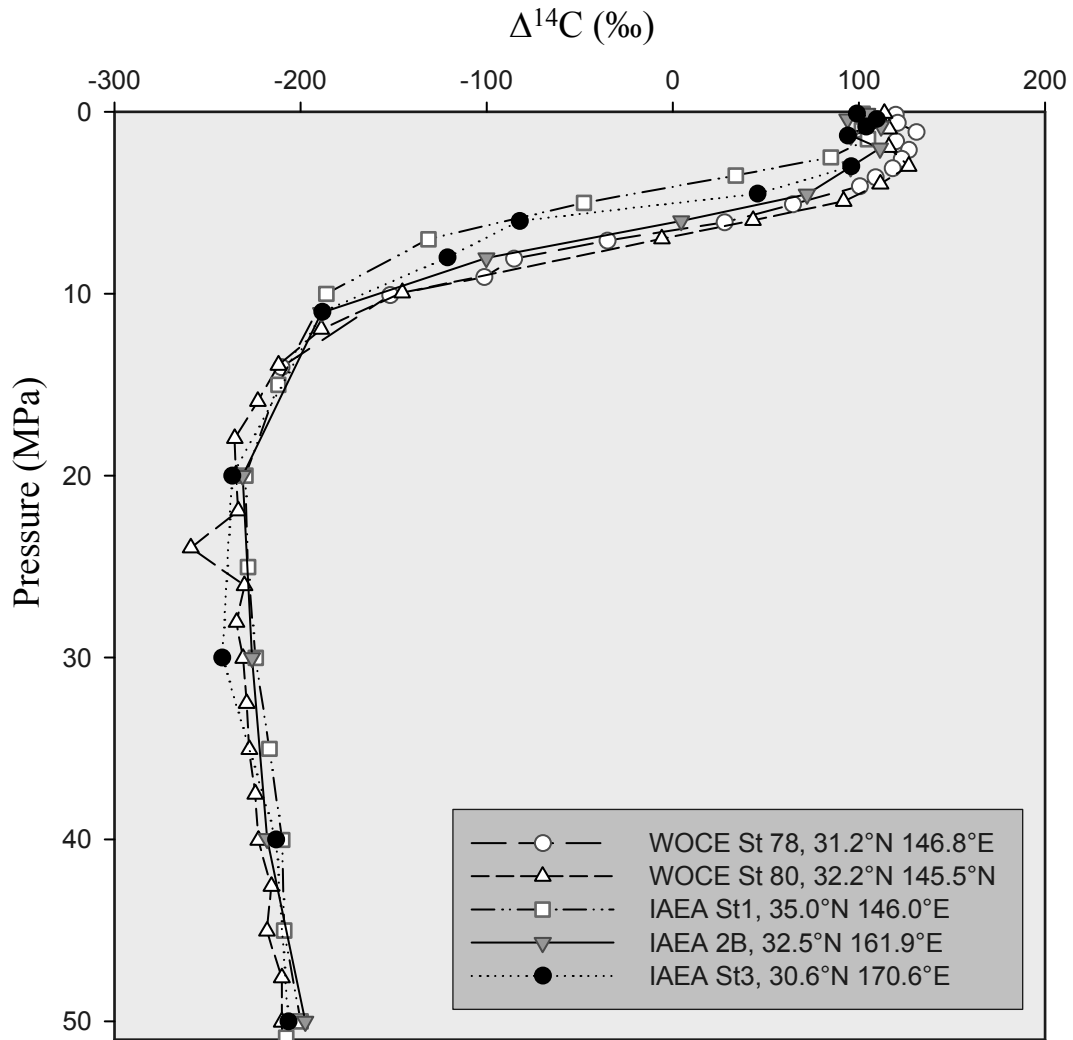


Figure 4 Comparison of vertical $\Delta^{14}\text{C}$ profiles observed at IAEA'97 and WOCE (1993) stations

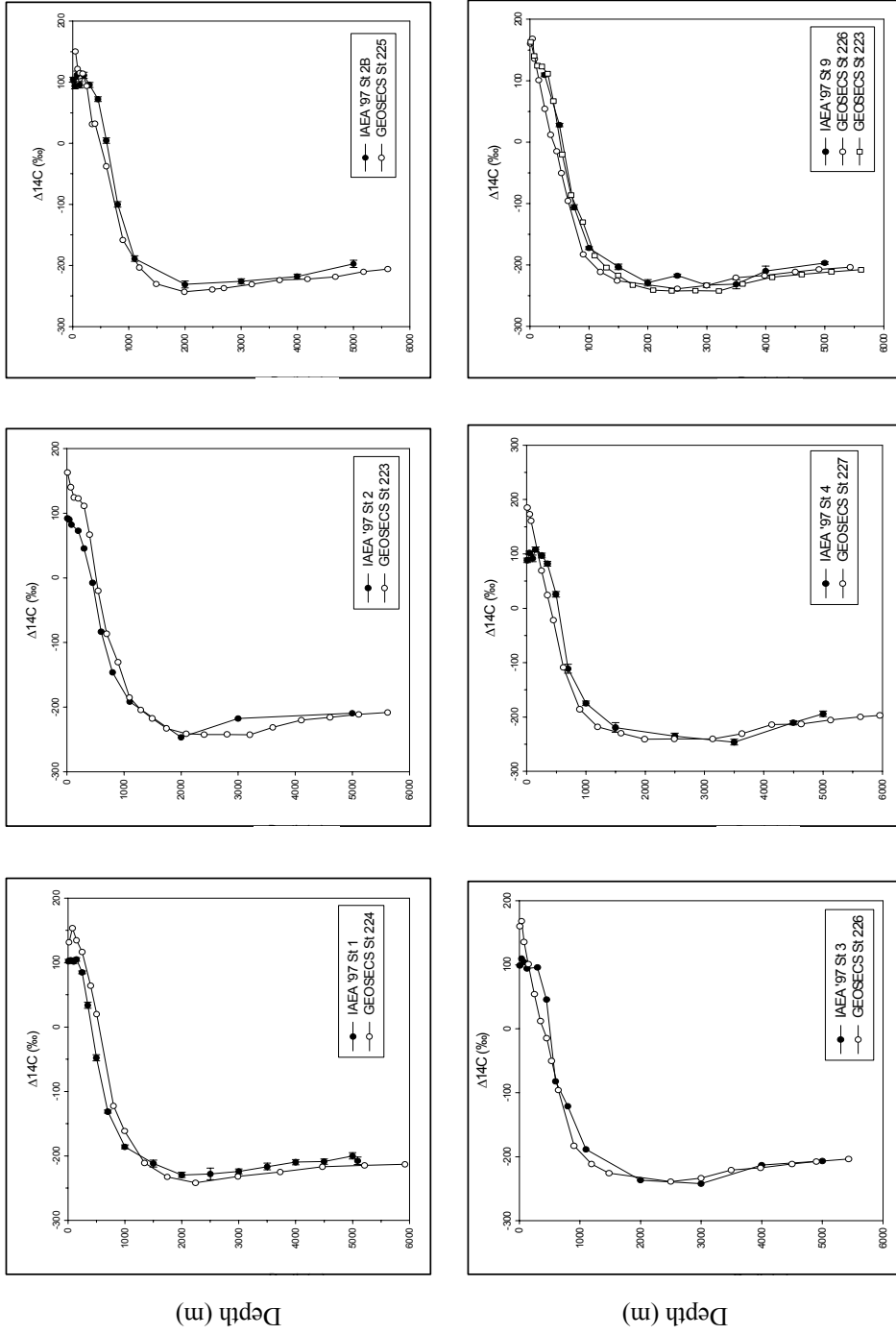


Figure 5 Comparison of IAEA 97 and GEOSECS vertical $\Delta^{14}\text{C}$ profiles

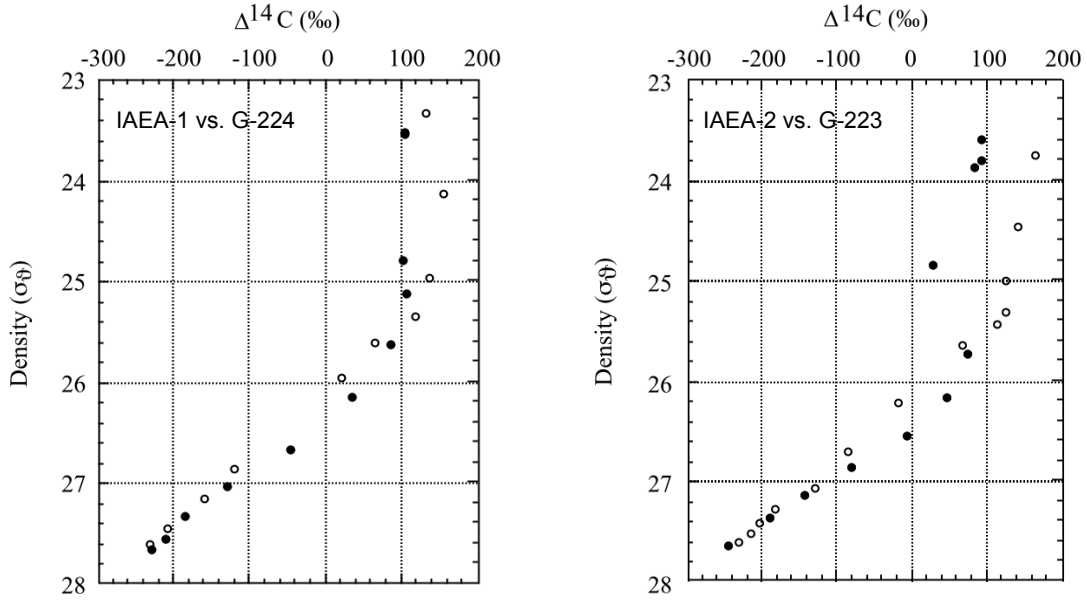


Figure 6 Comparison of $\Delta^{14}\text{C}$ vertical profiles against density (σ_θ) between IAEA (dots) and GEOSECS (open circles) stations.

Bomb ¹⁴C Behavior in the Southwestern North Pacific

Two sets of vertical profiles (this work and GEOSECS) are used to analyze the behavior of bomb ¹⁴C in the southwestern North Pacific over the 24-yr period between 1973 and 1997. The bomb ¹⁴C content (¹⁴C_{bomb}) is expressed as

$$^{14}\text{C}_{\text{bomb}} = (\Delta^{14}\text{C}_{\text{obs}} - \Delta^{14}\text{C}_{\text{nat}}) \times \alpha \times \Sigma\text{CO}_2 \tag{1}$$

where $\alpha = 1.176 \times 10^{-12}$ is the ratio of ¹⁴C/¹²C at $\Delta^{14}\text{C} = 0\text{‰}$, and ΣCO_2 is the total carbonate content in mole kg^{-1} , at the time of “observation” and “natural.” Both ΣCO_2 at the same depth are regarded as approximately similar values. Therefore, we can calculate the bomb ¹⁴C inventory (I_{bomb}), integrating ¹⁴C_{bomb} from the surface to the lower limit of the bomb ¹⁴C penetration depth (z , m) by the following equation:

$$\Delta I_{\text{bomb}} = \int_0^Z {}^{14}\text{C}_{\text{bomb}} dz \times \rho \times N_A \tag{2}$$

where ρ and N_A are the density of seawater in kg m^{-3} and Avogadro number ($= 6.02 \times 10^{23}$), respectively. The variability (ΔI_{bomb}) of I_{bomb} in each water column during the last 24 yr is expressed as follows:

$$\Delta I_{\text{bomb}} = I_{1997} - I_{1973} \tag{3}$$

By substituting equations (2) at 1973 and 1997 for equation (3), we can obtain the following equation:

$$\Delta I_{\text{bomb}} = \int_0^Z {}^{14}\text{C}_{1997-1973} dz \times \rho \times N_A \tag{4}$$

where ${}^{14}\text{C}_{1997-1973}$ is defined as

$${}^{14}\text{C}_{1997-1973} = (\Delta^{14}\text{C}_{1997} - \Delta^{14}\text{C}_{1973}) \times \alpha \times \Sigma\text{CO}_2 \quad (5).$$

The calculated ΔI_{bomb} inventories at Stations 2B, 3, and 4 are given in Table 2, where we assume the following: (i) surface water is seawater above $25.5 \sigma_\theta$ of which the average depth is 200 m; (ii) intermediate water is a water mass of $25.5\text{--}27.0 \sigma_\theta$; and (iii) deep water is seawater below $27.0 \sigma_\theta$ (830 m), as defined by Reid (1965), see also Figure 6. At Stations 1 and 2, ΔI_{bomb} cannot be calculated because of the presence of different water masses at the time of the 2 expeditions (this work and GEOSECS). An average value of total of ΔI_{bomb} was $(31.9 \pm 5.0) \times 10^{12}$ atom m^{-2} . The annual flux of bomb ${}^{14}\text{C}$ in the southwestern North Pacific from 1973 to 1997 is $(1.3 \pm 0.3) 10^{12}$ atom $\text{m}^{-2}\text{yr}^{-1}$, and is consistent with a previous study in the western North Pacific (Watanabe et al. 1999). The amount of bomb ${}^{14}\text{C}$ in surface water decreased by $(9.4 \pm 3.0) 10^{12}$ atom m^{-2} (0.4×10^{12} atom $\text{m}^{-2}\text{yr}^{-1}$); conversely, the amount of bomb ${}^{14}\text{C}$ in intermediate water increased by $(41.3 \pm 3.4) 10^{12}$ atom m^{-2} (1.7×10^{12} atom $\text{m}^{-2}\text{yr}^{-1}$). On the other hand, in deep water, no variability of I_{bomb} over the 24-yr period was found, suggesting that no bomb ${}^{14}\text{C}$ had been transported into deep water up to the time of sampling in 1997.

Table 2 The variability of the bomb ${}^{14}\text{C}$ inventory (ΔI_{bomb}) in the water column, 1973–1997.

Station	Location	ΔI_{bomb} ($\times 10^{12}$ atom m^{-2})			
		Total	above $25.5 \sigma_\theta$	$25.5\text{--}27.0 \sigma_\theta$	below $27.0 \sigma_\theta$
IAEA-2B G-225	32.5°N 161.9°E	37.5	−7.3	44.9	0
IAEA-3 G-226	30.6°N 170.6°E	30.1	−8.0	38.1	0
IAEA-4 G-227	25.0°N 170.1°E	28.2	−12.8	40.9	0
Average:		31.9 ± 5.0	$−9.4 \pm 3.0$	41.3 ± 3.4	0
Annual flux:		1.3×10^{12} atom/ m^2 yr			

The increase of bomb ${}^{14}\text{C}$ in the water column can be considered to be equal to the increase in the intermediate layer. Atmospheric $\Delta^{14}\text{C}$ increased substantially during the 1960s, and then decreased from the late 1960s to the present (Levin et al. 1994). The main input of atmospheric bomb ${}^{14}\text{C}$ was absorbed into surface water over the whole Pacific Ocean between the 1960s and the 1970s. At the same time, excess bomb ${}^{14}\text{C}$ in surface water was transported northward by advection by the Kuroshio Current and/or Kuroshio Extension Current. Bomb ${}^{14}\text{C}$ absorbed into surface water in subarctic areas was carried to the intermediate layer by strong vertical mixing. The excess bomb ${}^{14}\text{C}$ in the intermediate layer in subarctic areas was transported to the subtropical area by North Pacific Intermediate Water (NPIW) (Mahadevan 2001; Aramaki 2002).

CONCLUSIONS

In the framework of IAEA-MEL's project on Worldwide Marine Radioactivity Studies (WOM-ARS), water profile samples were taken during the IAEA'97 Pacific Ocean cruise at 10 stations in the southwestern North Pacific Ocean. Some stations were in proximity to the GEOSECS stations sampled in 1973; thus, ${}^{14}\text{C}$ profiles could be compared over the 24-yr interval. The following conclusions were drawn from the present work:

- ¹⁴C concentrations had decreased in surface waters (by 50–80‰) and increased (by about the same amount) in intermediate waters. In deep waters (below 1000 m), the observed ¹⁴C concentrations were similar to GEOSECS values (except for Station 7). While a rapid decrease in ¹⁴C concentrations was observed from the surface (¹⁴C approximately 100‰) down to 1000 m (¹⁴C approximately –200‰), the concentrations below 1000 m were almost constant. The bomb-produced ¹⁴C inventory had increased by more than 20% over the 24-yr period studied.
- The variability of bomb ¹⁴C inventories in the water column over the 24-yr period (ΔI_{bomb}) was estimated at $(32 \pm 5) 10^{12} \text{ atom m}^{-2}$. The amount of bomb ¹⁴C in surface water had decreased to $(9.4 \pm 3.0) 10^{12} \text{ atom m}^{-2}$; conversely, the amount of bomb ¹⁴C in intermediate water had increased to $(41 \pm 4) 10^{12} \text{ atom m}^{-2}$. However, no variation of I_{bomb} was found in deep water over the period. The average annual flux of bomb ¹⁴C has been estimated to be $(1.3 \pm 0.3) 10^{12} \text{ atom m}^{-2} \text{ yr}^{-1}$.
- The results suggest that excess bomb ¹⁴C in surface water of the southwestern Pacific Ocean was transported northward by advection by the Kuroshio Current and Kuroshio Extension and then transported into the intermediate layer by North Pacific Intermediate Water (NPIW), which flows southward from the subarctic area to the tropics.

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