Dissolved inorganic radiocarbon in the North Pacific Ocean and Sargasso Sea

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Abstract

We present radiocarbon measurements of dissolved inorganic carbon (DIC) in depth profiles from reoccupations of our central North Pacific Ocean and Sargasso Sea (SS) sites. From 1985 to 1999, an increase in $\Delta^{14}C$ values of 6–17‰ was measured between 1150 and 2400 m depth in the North Central Pacific. Natural changes in deep circulation are likely responsible for variability of $\Delta^{14}C$ signatures in the deep ocean, though we cannot rule out the presence of bomb ^{14}C at this depth range. Bomb ^{14}C had increased in the deep SS from 1989 to 2000; this is the result of southward transport of North Atlantic Deep Water (NADW) to this site.
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1. Introduction

Oceanic dissolved inorganic carbon (DIC) is the largest pool of exchangeable carbon on Earth (36,000 GtC). It is important to know the timescales of mixing and ventilation of deep ocean waters because most of the excess CO_2 produced by fossil fuel and biomass burning will eventually be stored in the oceans. Early radiocarbon measurements in

seawater revealed several hundred-year transit times of deep water masses in the world's oceans (Bien et al., 1963; Broecker et al., 1960; Stuiver et al., 1983). Global data sets amassed during programs such as the Geochemical Ocean Sections Study (GEOSECS) and the World Ocean Circulation Experiment (WOCE) have been used for quantitative studies of the oceanic carbon cycle (Key, 1996; Stuiver et al., 1983).

Since the production of bomb ^{14}C in the late 1950s and early 1960s, it has been possible to measure short-term exchange of carbon, e.g. the transfer of CO_2 across the air–sea interface. The level of bomb ^{14}C in a given carbon pool is a reflection of the turnover time of the carbon with respect to exchange with the atmosphere. By the 1970s, bomb ^{14}C was detected to depths of several

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hundred meters in the non-polar oceans (Ostlund and Stuiver, 1980; Stuiver and Ostlund, 1980). The deep northern North Atlantic contained bomb ^{14}C throughout the entire water column, introduced during North Atlantic Deep Water (NADW) formation. By 1991, penetration of bomb ^{14}C extended to 1000 m depth in the Pacific (Key, 1997; Stuiver et al., 1996).

The Δ^{14}C data reported here show an increase in values in the deep waters of both the North-central Pacific (NCP) and the Sargasso Sea (SS) (Druffel et al., 1992). Change in circulation is the likely mechanism responsible for the higher Δ^{14}C values in the upper part of the deep NCP. Remineralization of bomb-laden particulate organic matter from the surface is unlikely to have caused a measurable increase in the Δ^{14}C values of DIC in the deep North Pacific or North Atlantic.

2. Methods

Water samples were collected from the NCP site (31°N, 159°W, bottom depth 5820 m) on the Avon cruise from 26 May to 12 June 1999 and from the SS site (31°50′N, 63°30′W, bottom depth at 4500 m) on the SarC cruise from 14 to 29 June 2000. The NCP site is 1000 km north of Hawaii and the SS site is 100 km southeast of Bermuda. Radiocarbon and δ^{13}C measurements of suspended particulate organic carbon collected during these cruises were reported previously (Druffel et al., 2003), and radiocarbon and abundance data of dissolved organic carbon will be reported separately (Bauer et al., in preparation; Loh et al., 2004). The NCP and SS sites were occupied earlier, from 6 June to 3 July 1987 (Eve cruise) and from 29 May to 20 June 1989 (Hydros-6 cruise), respectively, and isotope results were reported separately (Druffel et al., 1992).

Seawater samples were collected in 1999 and 2000 using 12-L or 30-L Go-flo bottles deployed on a hydrowire. Samples for DIC Δ^{14}C and δ^{13}C analyses were filtered through glass fiber filters (1 μm effective pore size) directly into 1-L glass containers and poisoned with a saturated solution of mercuric chloride. Separate samples were filtered and poisoned in the same manner for alkalinity and total CO$_2$ ([DIC]) analyses. For the DIC Δ^{14}C and δ^{13}C analyses, the samples were acidified and sparged of CO$_2$ gas according to published techniques (McNichol et al., 1994). Carbon dioxide was

converted to graphite using hydrogen gas and cobalt metal at 550 °C (Vogel et al., 1987).

The Δ^{14}C measurements from the Avon 1999 cruise were made at the National Ocean Sciences Accelerator Mass Spectrometry Facility (NOSAMS) of the Woods Hole Oceanographic Institution (WHOI), and those from the SarC 2000 cruise were made at the W.M. Keck Carbon Cycle AMS Laboratory at the University of California, Irvine (UCI). The DIC Δ^{14}C values were reported according to standard techniques (Stuiver and Polach, 1977) and have a total uncertainty determined from replicate seawater analyses of ± 3.9‰. The δ^{13}C measurements were performed at NOSAMS with a total uncertainty of ± 0.1‰. Alkalinity and [DIC] measurements were obtained by closed vessel titration of large volume (∼100 ml) samples using an auto-mated titration system (Bradshaw et al., 1981; Brewer et al., 1986) in the laboratory of D. McCorkle (WHOI). Alkalinity and [DIC] measurements were determined using a nonlinear curve fitting approach (DOE, 1994) and standardized using certified refer-ence materials obtained from Andrew Dickson (Scripps Institution of Oceanography). The standard deviation of pairs of replicate analyses of culture water was 4 μeq/kg for alkalinity and 6 μmol/kg for [DIC]. The alkalinity results were consistently high by 25–40 μeq/kg compared with other cruises, which we attribute to the unusually long storage times of the samples prior to analysis (>9 mos); for this reason, the alkalinity results were not reported.

3. Results

The DIC Δ^{14}C values of samples collected from the 1999 NCP site ranged from 95‰ (at 3 m depth) to −243‰ (at 3221 m) (Fig. 1a). Values remained low (average −239.0±2.9‰, $n = 4$) between 2400 and 3600 m, and increased to an average of −215±6‰ ($n = 5$) between 4200 and 5770 m (50 m above bottom). Values of [DIC] were low in the upper 100 m (2050±6 μmol/kg, $n = 4$), reached a max-imum at 1300 m (2382 μmol/kg) and decreased slightly in deep waters (Table 1).

The DIC Δ^{14}C values obtained for samples from the SS site in 2000 (Fig. 2) decreased from 81‰ at 3 m to a low of −72‰ at 1005 m (Antarctic Intermediate Water (AAIW)); values increased to a secondary maximum (−32‰) at 1513 m, and decreased to −84‰ at 50 m above bottom. The [DIC] values were low in the upper 50 m (2099–2107 μmol/kg), highest in AAIW (2203–2221 μmol/kg between 850 and
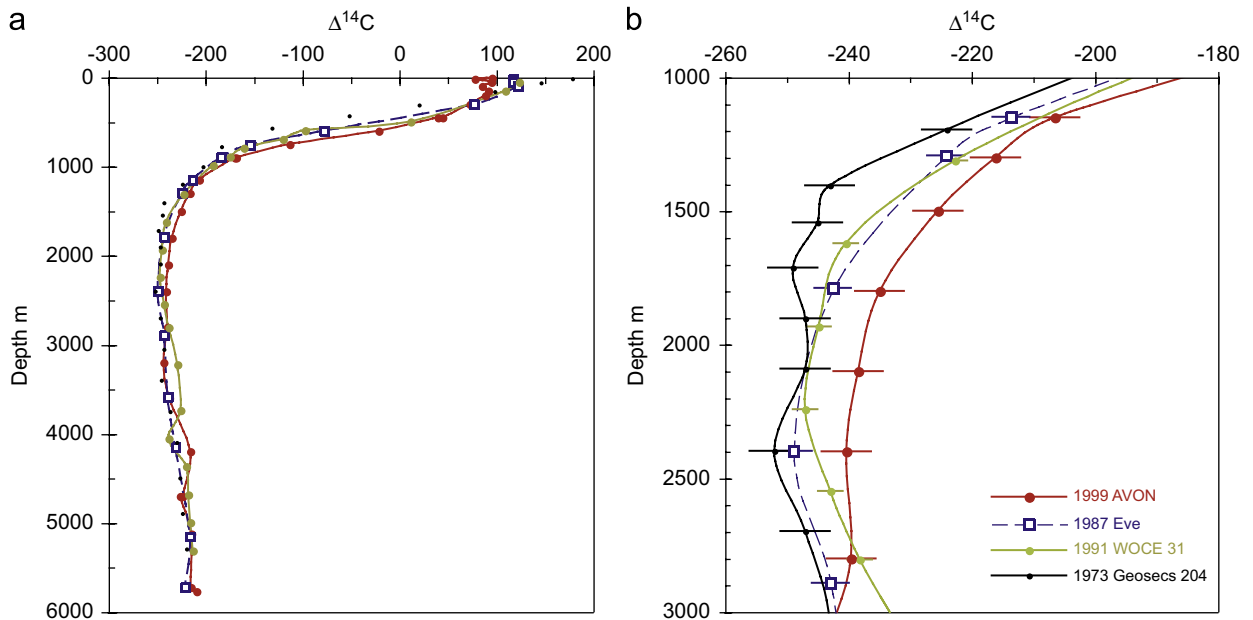
Fig. 1. $\Delta^{14}C$ values (‰) of DIC from the NCP site (31°N, 159°W) during Avon (June 1999) and Eve (June 1987) cruises (a) for 0–6000 m and (b) for 1000–3000 m. Also shown are the $\Delta^{14}C$ profiles obtained previously during GEOSECS (September 1973, Stn 204, 31°N, 150°W) (Ostlund and Stuiver, 1980) and WOCE (March 1991, Stn 31, 30°N, 152°W) (Key, 1996; Stuiver et al., 1996).

1300 m) and slightly lower in NADW (2182–2201 μmol/kg at 1500–3600 m) (Table 2).

The DIC $\delta^{13}C$ values in the NCP were highest in surface waters (1.08‰ at 3 m), decreased to a minimum of −0.84‰ at 1141 m, and increased with depth to 5770 m (0.22‰) (Fig. 3a). At the SS site, the DIC $\delta^{13}C$ values were highest in surface water (1.09‰ at 20 m), decreased to a minimum (−0.21‰) at 1005 m, and increased to 0.7–0.9‰ in deep water (Fig. 3b).

4. Discussion

In the following discussion, we compare the isotopic measurements of samples reported for the recent cruises to the NCP and SS sites with those obtained from earlier cruises.

4.1. North Central Pacific

Surface $\Delta^{14}C$ values in June 1999 were about 40–50‰ lower than those measured in samples from the Eve cruise in June 1987 (Druffel et al., 1992) (Fig. 1a). This reduction reflects the decrease of bomb ^{14}C in DIC in subtropical surface waters since the early 1970s (Druffel, 1987). In contrast, $\Delta^{14}C$ values between 600 and 2400 m depth were sig-

nificantly higher in 1999 than those in 1987 (Fig. 1a, b). The average $\Delta^{14}C$ difference at depths between 600 and 900 m (38 ± 21‰, $n = 3$) was greater than that between 1150 and 2400 m (8 ± 1‰, $n = 4$). Comparison of our DIC $\Delta^{14}C$ data with profiles obtained during GEOSECS in September 1973 (Stn 204, 31°N, 150°W) (Ostlund and Stuiver, 1980) and WOCE in March 1991 (Stn 31, 30°N, 152°W) (Key, 1996; Stuiver et al., 1996) is also plotted with our data in Fig. 1a, b. These sites are located 860 and 680 km east of our NCP site, respectively, and some spatial variability of $\Delta^{14}C$ may be expected. In the upper 100 m, $\Delta^{14}C$ values were highest during 1973 (146–178‰) and lowest during the 1999 cruise (77–95‰). Between 400 and 2400 m, the 1999 values are higher than $\Delta^{14}C$ values from similar depths for any of the previous cruises (Fig. 1a, b). Additionally, the depth of the $\Delta^{14}C$ minimum appears to have deepened with time, from about 1700–2400 m in 1973, to 2200–2400 m in 1987–1991, to 3200 m in 1999 (Fig. 1a, b).

To more clearly illustrate the change of $\Delta^{14}C$ with time, $\Delta^{14}C$ measurements from nine depth ranges (50–2400 m) are plotted versus time of collection (Fig. 4). Data from an earlier cruise (Alcyone October 1985) to the NCP site (Druffel et al., 1989) are also included. At ~50 m depth, $\Delta^{14}C$

Table 1
Radiocarbon, δ^{13}C and DIC concentration measurements for water samples collected from the Avon cruise to the NCP site in June 1999

Avon UCID no.	Event no.	Depth (m)	Therm. pres.	Δ^{14}C (‰)	δ^{13}C (‰)	[DIC] (µmol/kg)
4222	141	3		95.3	1.08	2042
4229	141	20		77.8	0.90	2051
4202	142	50	48	94.9	1.00	2054
4226	142	100	88	85.5	0.84	2055
4213	179	150	158	91.5	0.66	2090
4215	147	200	199	88.5	0.46	2060
4211	179	300	311	72.8	0.44	2101
4201	147	450	457	44.3	0.06	2143
4204	187	450	456	39.9	0.22	2143
4205	179	600		−21.1	−0.09	2219
4216	164	750	762	−112.9	−0.46	2271
4214	146	900		−168.4	−0.65	
4197	187	900	953	−170.0	−0.66	
4203	164	1150	1141	−206.5	−0.84	2374
4212	164	1300		−216.1	−0.54	2382
4193	178	1500	1589	−225.4	−0.51	2345
4207	178	1800	1843	−234.9	−0.35	2377
4206	178	2100	2098	−238.4	−0.28	2362
4225	152	2400	2450	−240.3	−0.35	2371
4219	152	2800	2791	−239.5	−0.18	2384
4218	152	3200	3221	−243.4	−0.16	2342
4217	169	3600	3710	−237.4	−0.12	2347
4192	169	4200	4214	−215.7	0.03	2315
4189	169	4700	5043	−223.7	0.01	2338
4673	169	4700	5043	−226.2	−0.20	
4196	148	5120	5270	−214.6	0.13	2341
4194	148	5720	5906	−214.9	0.08	2345
4190	183	5770		−211.4	−0.06	2346
4191	183	5770		−207.0	0.22	

values decrease from 1973 to 1999, and at 150 m a maximum in 1985 is apparent. At 300 m and deeper, Δ^{14}C values generally increase with time. Least squares fits of the data from 900, 1150, 1800 and 2400 m depth (Fig. 4 inset) reveal increases of 35‰, 17‰, 14‰ and 12‰, respectively, during the 26-year period. These increases in Δ^{14}C are > 3 times the 1-σ uncertainty of our measurements (3.9‰), and thus statistically significant. Could this increase be due to penetration of bomb ^{14}C below the main thermocline?

From data compiled as a part of the Global Ocean Data Analysis Project, Key et al. (2004) showed that bomb radiocarbon had penetrated no deeper than 1000 m in the North Pacific by 1991. They found a weak bomb ^{14}C signal at 1000 m only in the region of intermediate water formation in the far northwest Pacific. The data presented here (Fig. 4) suggest that, by 1999, Δ^{14}C values were higher in the North central Pacific basin, as deep as 2400 m.

Is it possible that the 12–17‰ increase in the deep ocean DIC Δ^{14}C values represents natural variability? The ranges of Δ^{14}C values taken over a 2-year period at 1600 and 2500 m depth at Stn M in the NE Pacific were 16‰ ($n = 7$) and 18‰ ($n = 6$), respectively (Masiello et al., 1998). The ranges of Δ^{14}C values measured in samples from our cruises to the NCP site at 1800 and 2400 m in 1985 and 1987 were 8‰ ($n = 2$) and 4‰ ($n = 4$), respectively. The Stn M site is a more productive, coastal site, and comparison with the midgyre NCP site may not be valid. Roussenov et al. (2004) used an isopycnic circulation model to show that DIC Δ^{14}C values in the deep Pacific are controlled by lateral transport of bottom water from the south and balance between advection–diffusion and decay of ^{14}C in the vertical. They found that a strengthening of diapycnic mixing causes an overall increase in Δ^{14}C values over the North Pacific basin similar to the increase that we see at the NCP site. Thus, it is possible that shifts in deep circulation could cause
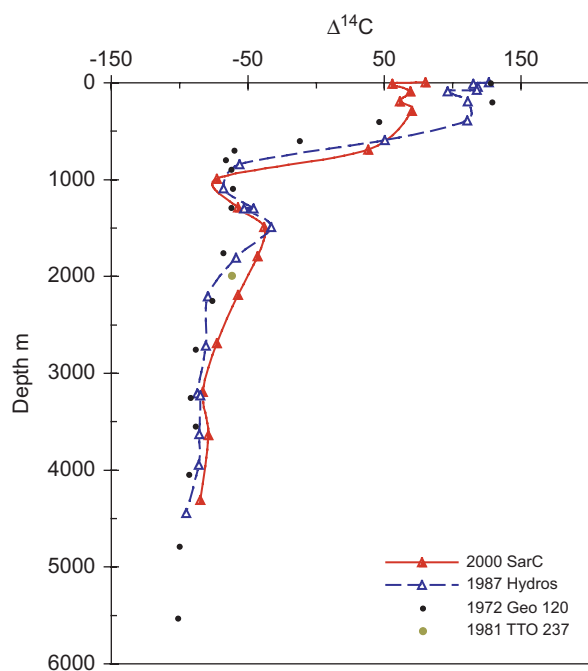
Fig. 2. $\Delta^{14}C$ values of DIC from the SS site (31.5°N, 63.5°W) during the SarC (June 2000) and Hydros (June 1989) cruises. Also shown are the $\Delta^{14}C$ profiles presented previously during GEOSECS (1973, Stn 120, 33°16′N, 56°33′W) (Stuiver and Ostlund, 1980) and TTO in 1981 (Stn 237, 33°8′N, 56°30′W) cruises (Ostlund, 1981).

changes in $\Delta^{14}C$ of the order that we observe at 1150–2400 m depth.

Is it possible that the increase in $\Delta^{14}C$ is due to input of bomb ^{14}C? The two possible sources of bomb ^{14}C would be from remineralization of surface-derived particulate organic carbon (POC) to DIC, and physical mixing of bomb-laden upper waters into the deeper layers of the ocean. First, we calculate the change in deep DIC $\Delta^{14}C$ expected from the input of remineralized bomb-laden POC from surface waters. The amount of DIC in the water column from 1000 to 2000 m with 1 m^2 area is 2.93×10^4 gC/m^2, assuming an average [DIC] of 2370 μmol/kg, and an average density of 1030 kg/m^3. The potential change of $\Delta^{14}C$ by remineralization of POC using the average global flux rate of sinking POC at 1000 m depth of 2.8 gC/m^2 (Martin et al. (1987) for 26 years = 73 gC/m^2), with a $\Delta^{14}C$ of 89‰, is only 0.75‰, which is much smaller than the observed 12–17‰ difference. This calculation assumes only vertical transport of remineralized POC and does not include lateral transport of material. As a note, we could neither use the potential alkalinity method (Rubin and Key, 2002) to estimate the actual bomb ^{14}C concentration (alkalinity measurements were too high), nor could we use Broecker's silica method (Broecker et al.,

Table 2
Radiocarbon, $\delta^{13}C$ and DIC concentration measurements for water samples collected from the SarC cruise to the SS site in June 2000

SarC UCID no.	Event no.	Depth (m)	Therm. pres.	$\Delta^{14}C$ (‰)	$\delta^{13}C$ (‰)	[DIC] (μmol/kg)	Salinity (‰)
	115	3		80.6	1.03	2101	36.552
4882	115	20		56.6	1.09	2099	36.564
	107	50	45			2107	36.669
	107	85				2120	36.644
4874	107	100	98	69.9	0.79	2124	36.613
4888	107	200	205	61.9	0.83	2125	36.646
4886	153	300		71.0	0.86	2126	36.585
	123	400	398			2148	36.477
	123	600	605			2169	36.025
4902	153	700	695	38.8	0.62	2183	35.683
	135	850				2221	35.222
4887	153	1000	1005	−71.9	−0.21	2213	35.085
	135	1100				2208	35.083
4879	135	1300	1301	−56.7	0.68	2203	35.167
4883	135	1500	1513	−43.3	0.74	2196	34.992
4898	212	1500	1492	−31.6	0.77	2182	35.050
4889	143	1800		−42.0	0.84	2194	34.950
4892	181	2200		−56.4	0.91	2193	34.968
4904	143	2700	2736	−75.4	0.93	2193	34.947
4894	212	2700	2725	−69.0	0.90	2196	35.053
4897	181	3200	3245	−82.4	0.81	2183	34.939
4900	151	3650		−78.1	0.86	2201	34.898
4899	151	4320		−84.1	0.80	2212	34.913

a

b
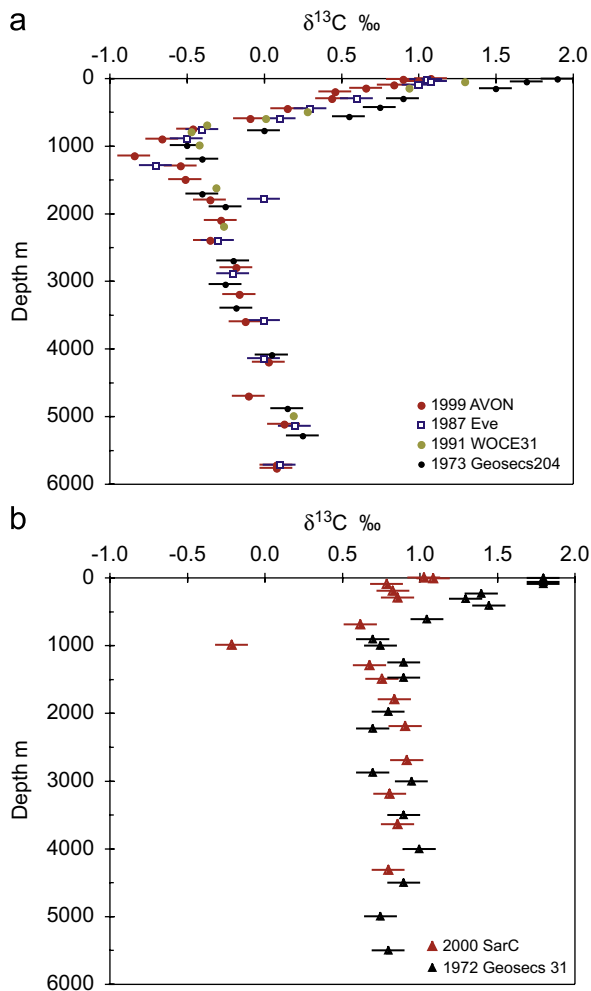

Fig. 3. $\delta^{13}C$ values of DIC from (a) the NCP site and others as per Fig. 1 caption, and (b) the SS site and GEOSECS Stn 31 (27°0′N, 53°32′W).

1995) because silicate was not measured in our water samples. Second, it is difficult to attribute physical mixing of upper waters into the deep waters as a source of bomb ^{14}C without adopting unrealistically high cross isopycnal mixing rates. Thus, we conclude that the increase in the $\Delta^{14}C$ signature between 1150 and 2400 m depth does not reflect bomb ^{14}C addition to the deep Pacific. Horizontal variability in $\Delta^{14}C$ of up to 14‰ is observed below 1000 m at WOCE stations along 152°W line and 134°W line near our NCP station in 1991 (Stuiver et al., 1996; Key et al., 2004). Therefore, we believe that the increase in the $\Delta^{14}C$ values with time represents changes in circulation and mixing in the deep waters, causing shifts in the baseline $\Delta^{14}C$ signature.

Important processes contributing to $\delta^{13}C$ of DIC are (1) mixing, (2) transfer of CO_2 between air and sea, (3) remineralization of organic matter to CO_2, and (4) removal of CO_2 during photosynthesis in the surface water. At the NCP site, DIC $\delta^{13}C$ values in the upper 1100 m were lower in 1999 than during any of the earlier cruises (Fig. 3a). This depletion is evidence of the ^{13}C Suess Effect, which is the input of ^{13}C-depleted CO_2 from fossil fuels and terrestrial biomass to the main thermocline (McNichol and Druffel, 1992; Quay et al., 1992). The $\delta^{13}C$ values in the surface decreased by 0.8‰ from 1973 (Kroopnick, 1985) to 1999 and at 1100 m by about 0.3‰ (Fig. 3a). The decrease of $\delta^{13}C$ in the surface of the North Pacific was about 0.02‰ y^{-1} from 1970 to 1990 (Quay et al., 1992). This is similar to our results (0.8‰/26 yr = 0.03‰ y^{-1}) from 1973 to 1999, and is consistent with an increased rate as anthropogenic CO_2 input increases with time.

4.2. Sargasso Sea

The $\Delta^{14}C$ value of SS surface water sampled in 2000 was about 40‰ lower than that obtained 11 years earlier at this site (Fig. 2). In contrast, at 1800 and 2200 m, $\Delta^{14}C$ values were significantly higher (by 19 ± 4‰) during 2000 than those obtained in 1989, and the same or slightly higher (7 ± 3‰, $n = 4$) below 2200 m depth (Fig. 2).

A comparison of our data in 2000 with a profile obtained during GEOSECS in March 1973 (Stn 120, 33°16′N, 56°33′W) (Stuiver and Ostlund, 1980) and one depth (1994 m) from TTO in October 1981 (Stn 237, 33°8′N, 56°30′W) (Ostlund, 1981) are also shown in Fig. 2 (both located 670 km to the east of our SS site). In the upper 200 m, $\Delta^{14}C$ values were highest during 1973 (128–130‰) and lowest in 2000 (57–81‰). At 1800, 2700 and 4400 m, the $\Delta^{14}C$ values in 2000 were significantly higher than those from similar depths for the previous cruises (Fig. 5). Similar to the NCP, surface $\Delta^{14}C$ values decreased from 1973 to 2000. At 600 m and deeper, $\Delta^{14}C$ values increased with time. Least squares fits of the data from 1800, 2700, 3600 and 4400 m revealed increases of 25‰, 15‰, 9‰ and 16‰, respectively, over this 27-year period. These $\Delta^{14}C$ increases show the progressive penetration of bomb ^{14}C with time into the deep water column of the SS.

The primary source of bomb ^{14}C in the deep SS is from NADW that is produced in the Norwegian and Greenland Seas. Previous studies have shown that tritium and chlorofluorocarbons were
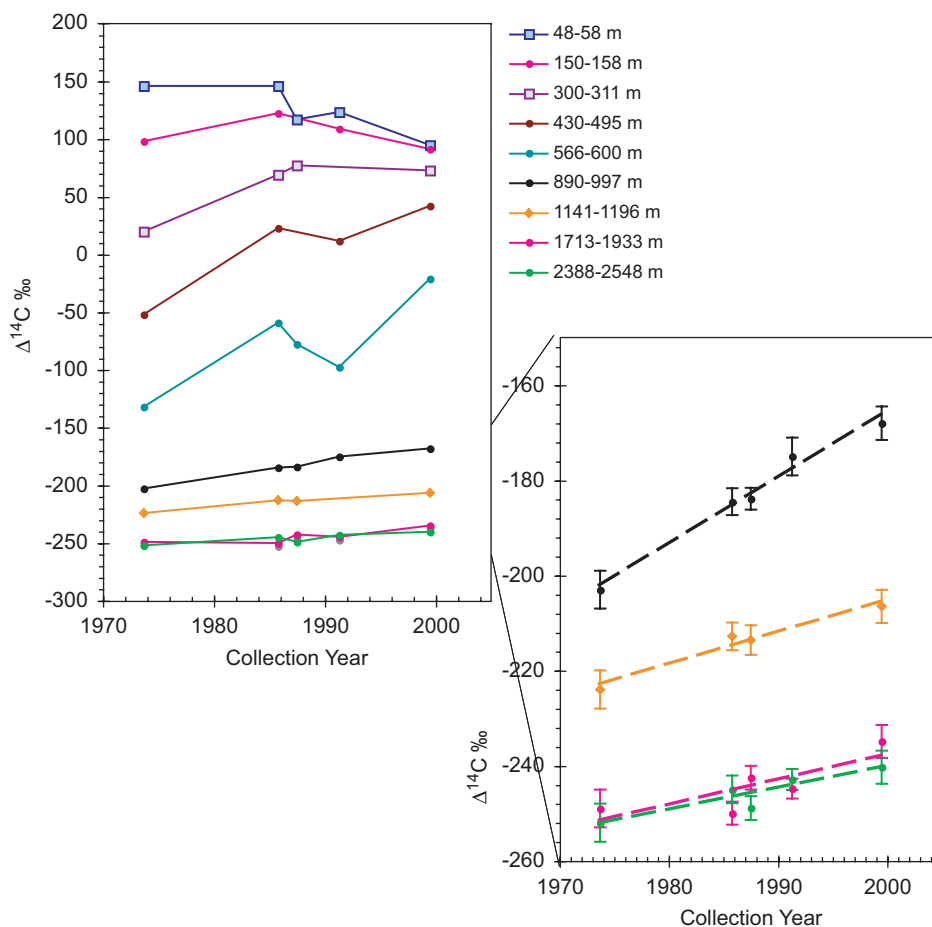
Fig. 4. $\Delta^{14}C$ values of DIC from the NCP (as per Fig. 1 caption with additional data from Alcyone cruise in October 1985) plotted as a function of time of sample collection. Results are binned in depth ranges defined in the legend. Inset shows $\Delta^{14}C$ results for the depth bins at 900, 1150, 1800 and 2400 m with least squares fits (dashed lines) shown. Results from deeper depths show no statistically significant trends over the time series. Linear correlations, r, for these four depth bins are 0.99, 0.98, 0.82 and 0.92, which represent correlations significant to the 99.9‰, 98%, 90% and 95% confidence levels, respectively.

incorporated into NADW and transported south to the subtropics via deep thermohaline circulation (Ostlund and Rooth, 1990; Smethie et al., 1986). As shown above for the NCP site, remineralization of surface-derived POC to DIC is an unlikely source of bomb ^{14}C to the deep SS, unless POC fluxes and dissolution rates are seriously underestimated.

At the SS site, DIC $\delta^{13}C$ values in the upper 1500 m were lower during 2000 than those obtained from Stn 31 (27°0′N 53°32′W) during the 1972 GEOSECS cruise (Kroopnick, 1985) (Fig. 3b) (no $\delta^{13}C$ measurements were available from Stn 120). From 1972 to 2000, the $\delta^{13}C$ values had decreased by about 0.8‰ in the surface and by about 0.5‰ between 200 and 700 m. At 1000 m, the 2000 value appears anomalously low ($-0.2‰$),

though there is no obvious reason to discount this measurement. As in the NCP, the decrease of $\delta^{13}C$ values indicates the presence of ^{13}C-depleted atmospheric CO_2 from anthropogenic sources. Kortzinger et al. (2003) reported that the mean $\delta^{13}C$ decrease in the upper 1000 m of the North Atlantic from 1950 to 1993 was $0.026 \pm 0.002‰\,y^{-1}$. Data from Fig. 3b show a similar decrease from 1972 to 2000 of $0.015\text{–}0.026‰\,y^{-1}$ in the upper 700 m.

5. Conclusion

Bomb ^{14}C has continued to increase in the deep waters of the SS from 1973 to 2000. What is surprising is the increase in $\Delta^{14}C$ to a depth of $\sim$2400 m in the NCP, because anthropogenic tracers
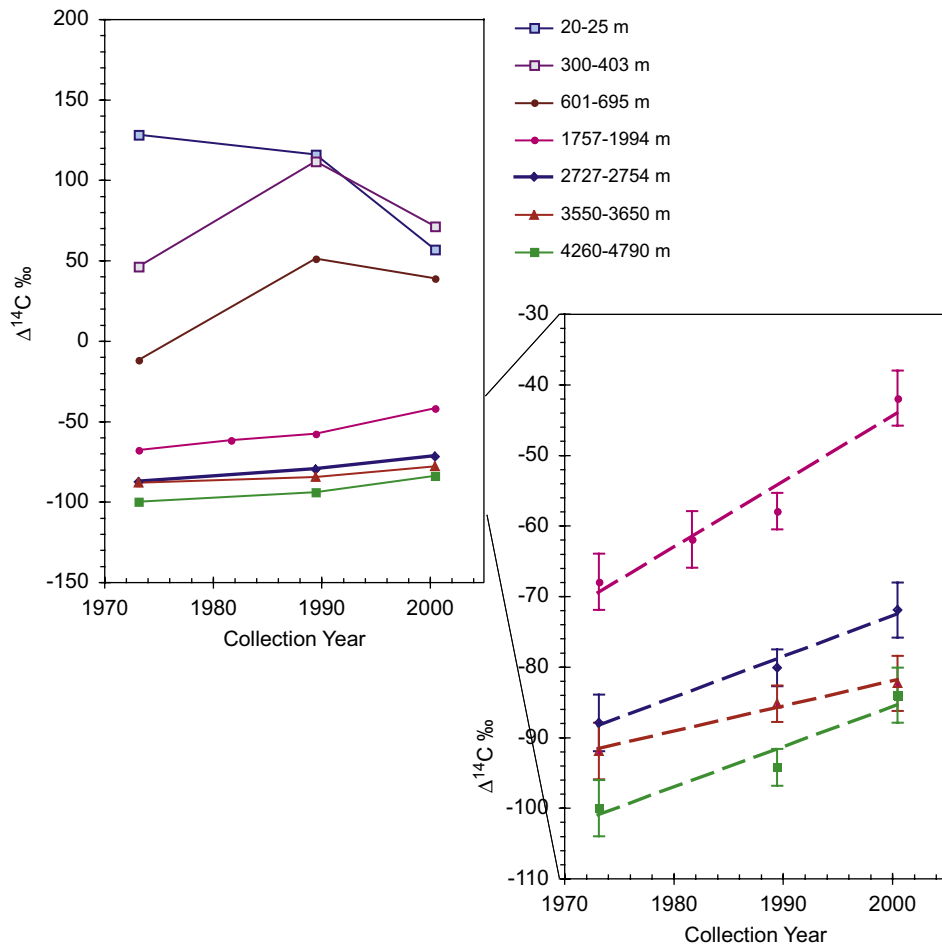
Fig. 5. $\Delta^{14}C$ values of DIC from the SS (as per Fig. 2 caption) plotted as a function of time of sample collection. Results are binned in depth ranges defined in legend. Inset shows $\Delta^{14}C$ results for the depth bins at 1800, 2700, 3600 and 4400 m with least squares fits (dashed lines) shown. Linear correlations, r, for these four depth bins are 0.97, 0.96, 0.99 and 0.96, which represent correlations significant to the 95%, 95%, 99% and 95% confidence levels, respectively.

(e.g., bomb ^{14}C, tritium and CFCs) have been found only to a depth of about 1000 m in the North Pacific by the 1990s (Key et al., 2004). It does not seem feasible that remineralization of POC from the surface could provide a sufficient source of bomb ^{14}C to the deep North Pacific. Instead, we conclude that changes in deep water circulation patterns offer a plausible explanation of the increase in $\Delta^{14}C$ that we observe in the deep waters of the North Pacific Ocean.

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