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Distributions of dissolved organic and inorganic carbon and radiocarbon in the eastern North Pacific continental margin

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Abstract

Temporal variations in the natural radiocarbon (^{14}C) signatures of dissolved organic and inorganic carbon (DOC and DIC, respectively) in seawater have been studied previously (Druffel, E.R.M., Bauer, J.E., Williams, P.M., Griffin, S., Wolgast, D.M., 1996. Seasonal variability of radiocarbon in particulate organic carbon in the northeast Pacific. *J. Geophys. Res.* 101, 20 543–20 552; Bauer, J.E., Druffel, E.R.M., Williams, P.M., Wolgast, D.M., Griffin, S., 1998. Temporal variability in dissolved organic carbon and radiocarbon in the eastern North Pacific Ocean. *J. Geophys. Res.* 103, 2867–2882) at a long-term time-series station (Sta. M: 32°N, 123W) in the eastern North Pacific located at the eastern edge of the North Pacific abyssal plain. In June 1995 a transect was made from Sta. M inshore to approximately 500 m depth in order to evaluate the distributions of ^{14}C in DOC and DIC from the abyssal plain to the upper continental slope. Concentrations and $\Delta^{14}\text{C}$ values of DOC in mixed layer waters (25 and 85 m) decreased toward the upper slope. In deeper waters, concentrations and $\Delta^{14}\text{C}$ values were in general similar at all three sites. Differences in DOC concentrations and $\Delta^{14}\text{C}$ -DOC between Sta. M and the rise and upper slope sites were explained in part by the mixing of DOC and $\Delta^{14}\text{C}$ along constant density (σ_t) surfaces. However, specific deviations from conservative behavior due to mixing were observed for $\Delta^{14}\text{C}$ -DOC at mesopelagic (~ 700 m) and near-bottom (~ 3600 – 3900 m) depths of the continental rise. Comparable findings are reported for DIC, where σ_t -normalized concentrations and $\Delta^{14}\text{C}$ values in Sta. M, rise and upper slope

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waters were similar, with the exception of slight increases in concentrations and $\Delta^{14}\text{C}$ values in near-bottom waters of the rise. These observations indicate that both DOC and DIC in continental rise and slope surface waters of the eastern North Pacific Ocean margin are comprised of a component of actively upwelled material derived from deeper offshore waters and a component of recent surface-derived material.

Our data suggest that contributions of both ^{14}C -enriched and ^{14}C -depleted DOC and DIC to localized regions of the water column occur in waters of the continental rise and slope. The sources of material having anomalous $\Delta^{14}\text{C}$ values are not certain, but may include organic matter derived from off-shelf and slope transport, particulate organic carbon solubilization, and sediment porewaters. We also examine the relationships between $\Delta^{14}\text{C}$ -DOC and both $\Delta^{14}\text{C}$ -DIC and $\Delta^{14}\text{C}$ of suspended POC measured in a companion study as a means of evaluating similarities or differences in the mechanisms responsible for the distributions of each at all three sites. © 1998 Elsevier Science Ltd. All rights reserved.

1. Introduction

Radiocarbon (^{14}C) natural abundances have been used in studies of marine carbon cycling to determine the sources, residence times, transformations and interactions of both organic and inorganic carbon reservoirs (Williams and Druffel, 1987; Bauer et al., 1992; Druffel et al., 1992). The natural abundance of ^{14}C in dissolved inorganic carbon (DIC, or total CO_2) has been used as an indicator of both the rate of deep-water mass formation and of the circulation times of the deep-water masses between the major ocean basins (Stuiver et al., 1983; Broecker et al., 1996). In both the dissolved and particulate organic carbon pools (DOC and POC, respectively), ^{14}C provides information on the aging of modern, surface-derived organic matter in the oceans as well as on the sources of carbon to the DOC and POC pools in cases where the sources have distinct ^{14}C signatures (Williams et al., 1969; Druffel et al., 1986; Druffel and Williams, 1990; Williams et al., 1992). One outcome of previous work using ^{14}C in DOC has been the identification of an approximate 2000 year greater ^{14}C age of DOC in the deep North Pacific than in the deep North Atlantic (Bauer et al., 1992; Druffel et al., 1992), which may be a result of the conservative transport and aging of refractory DOC during deep-water mass circulation. Another possible explanation for the observed differences in $\Delta^{14}\text{C}$ of DOC in the deep oceans is that the sources and ages of DOC to the deep Atlantic and Pacific may be different.

One region where sources of DOC could have unique ^{14}C signatures is ocean margins, where material derived from terrestrial, riverine, coastal, sedimentary and even petroleum (e.g., from hydrocarbon seeps) sources may be transported to the open ocean (Walsh et al., 1988; Bauer et al., 1990; Moran et al., 1991; Bauer et al., 1992; Guo et al., 1996; Opsahl and Benner, 1997). In ocean margins where terrestrial, riverine, and estuarine inputs are large, seawater DOC has been found to have significantly altered ^{14}C signatures (i.e., both enriched and depleted in ^{14}C) relative to open-ocean DOC (Eadie, 1978; Santschi et al., 1995; Guo et al., 1996). Organic matter in coastal regions also may have ^{13}C signatures that reflect the magnitude of terrestrial and riverine source(s) (Aller et al., 1991), but the greater dynamic range of organic ^{14}C

(ca. -1000 to $+200\%$ vs ~ -30 to -12% for organic ^{13}C , excluding methane) allows it to be used as a more sensitive source indicator (in addition to it being an age indicator) than ^{13}C for studies of marine DOC and POC cycling (Trumbore and Druffel, 1995).

The interactions and exchanges of carbon between the margins (i.e. continental shelf and slope systems) and interiors of the major ocean basins have been studied previously by such programs as the Shelf-Edge Exchange Program (SEEP) (Walsh et al., 1988; Biscaye et al., 1994; Falkowski et al., 1994). In these studies, carbon and elemental pools and fluxes were followed primarily in the forms of sinking and/or suspended particles, both as living biomass and non-living particulate organic matter (POM). However, these and other early studies failed to examine the role of the much larger pool of dissolved organic matter (DOM) (Hedges, 1992) as a component of total carbon fluxes. The potential importance of ocean margins in exporting organic matter to adjacent abyssal sediments has been deduced from sediment trap studies (Walsh, 1989; Biscaye et al., 1994; Falkowski et al., 1994), sediment oxygen and carbon budget balances (Jahnke et al., 1990; Reimers et al., 1992; Anderson et al., 1994; Smith et al., 1994; Sayles et al., 1994) and from both open ocean (Smith and MacKenzie, 1987) and coastal ocean (Smith and Hollibaugh, 1993) water-column production-respiration budgets. However, estimates of water-column transport of DOC, using gradients measured both across ocean margins and between margins and the open ocean, are comparatively few (Bauer and Druffel, 1998, in press).

Many of the earlier studies of carbon fluxes in ocean margins also were conducted in relatively shallow and geographically expansive regions (e.g. the Mid-Atlantic Bight, [cf. Walsh et al. (1988) and Biscaye et al. (1994)] and southern California continental borderland [cf. Landry et al. (1992) and Venkatesan and Kaplan (1992)] where inputs from riverine, terrestrial and urban sources are concentrated. In this study we measured DOC and DIC concentrations and $\Delta^{14}\text{C}$ signatures in both pools in the water column at three sites extending from a well-studied abyssal time-series station (Sta. M; Smith et al. (1994)) to the upper continental slope off the central California coast, a narrow leading-edge margin with minimal freshwater, and presumably terrestrial, inputs. Previous work at Sta. M has helped to establish the degree of temporal variability in DOC, DIC and suspended POC concentrations and $\Delta^{14}\text{C}$ signatures during both normal upwelling and El Niño/Southern Oscillation periods (Druffel et al., 1996; Bauer et al., 1998; Masiello et al., 1998). Our goal in the present study was to establish whether concentrations and $\Delta^{14}\text{C}$ signatures of DOC and DIC in continental rise and slope waters are different from or similar to those at abyssal Sta. M, and whether these differences, similarities and gradients indicate exchanges of carbon across this margin.

2. Materials and methods

2.1. Study sites

A transect consisting of 3 stations extending from abyssal Sta. M to the upper continental slope was conducted in June 1995 (Pulse-26 cruise; Fig. 1). Sta. M has been

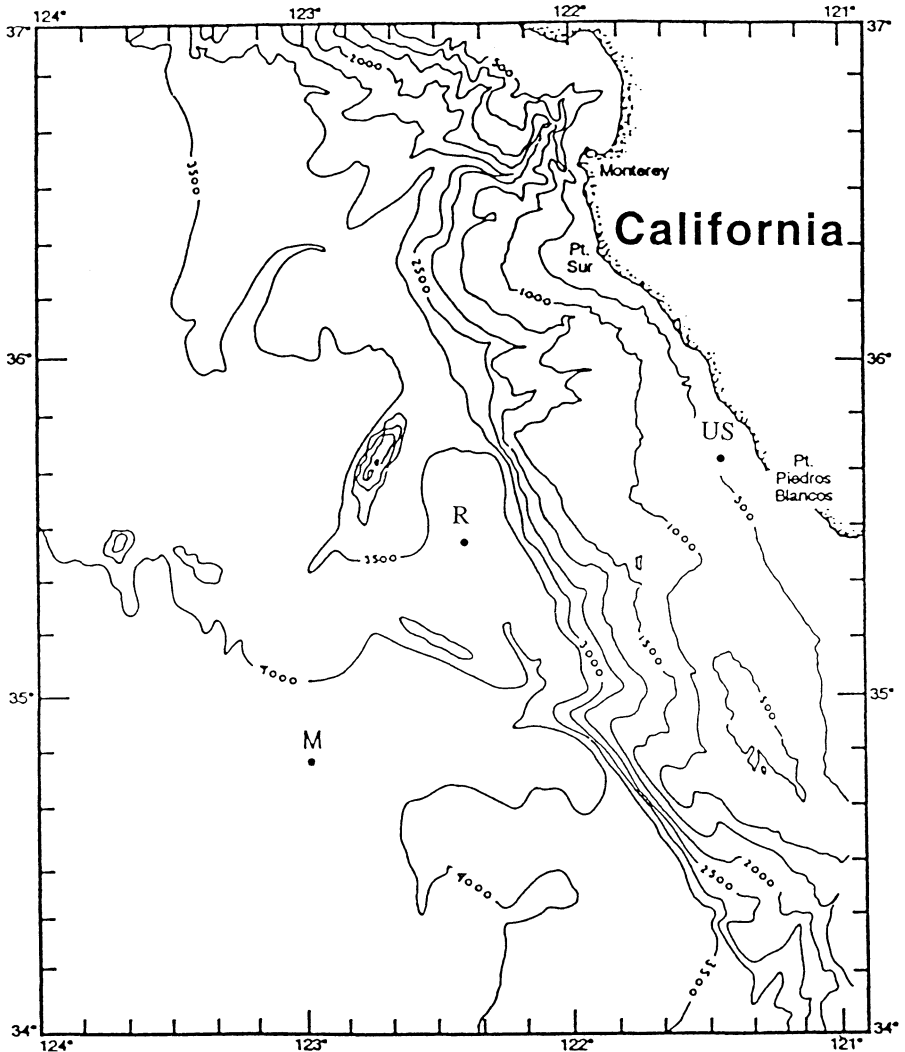


Fig. 1. Locations of Sta. M (M), Rise site (R) and Upper Slope site (US) occupied during June 1995 transect cruise (Pulse-26).

occupied every 3–4 months since 1987 by Smith et al. (1992, 1994) to measure deep ocean POC flux, benthic respiration and other parameters related to food sources and sinks in the benthic boundary layer. It has been occupied 3–4 times per year since 1991 by our group (Druffel et al., 1996; Cherrier et al., 1996; Bauer et al., 1998) for studies of deep ocean DOC and POC cycling. Station M is located at 34°50'N 123°00'W at the base of the Monterey Deep Sea Fan, approximately 220 km west of Point Conception, California. Average water depth is 4100 m. The other two sites

occupied, the Rise site (35°25'N, 122°24'W) and the Upper Slope site (35°40'N, 121°27'W) (Fig. 1), were located in approximately 3800 and 500 m water depth, respectively. The Rise site is the same station occupied by Jahnke et al. (1990) and Reimers et al. (1992), who estimated that a large lateral component of organic carbon was supplied to the sediments there. The distributions and carbon isotopic characterization of suspended POC and sediment organic carbon at each of these sites is described by Druffel et al. (1998).

The stations on this transect are in the region influenced by the south-flowing California Current (Knauss, 1978). Intense wind-driven upwelling occurs in late spring and throughout the summer along this part of the California coast, and upwelling-derived production is carried offshore in the form of discrete plumes or jets of chlorophyll (Smith et al., 1988; Michaelson et al., 1988). Independent long-term studies in this region (Hayward et al., 1994; Lynn et al., 1995; CALCOFI, 1997) indicate that primary production during summer 1995 in this region ranged from approximately 700–1900 mg C m⁻² d⁻¹ near station M and the Rise and Upper Slope stations. This material drives a seasonally varying vertical flux to the deep ocean at Sta. M, with the primary peaks occurring in late spring to mid-summer and secondary peaks in late fall (Smith et al., 1994). The deep (50–600 m above bottom) sinking POC flux to Sta. M in June 1995 ranged from approximately 5–25 mg C m⁻² d⁻¹ (Baldwin et al., 1998).

2.2. Sampling methods

All sampling preparations and sample processing procedures were conducted using organic- and isotopic-contamination-free protocols (Druffel et al., 1992). The depths from which samples were collected at each of the three stations are shown in Tables 1 and 2. Seawater samples were collected on a standard 1/4" o.d. hydrowire using 12- or 30-l Go-Flo bottles that were cleaned with methanol, 10% HCl and double-distilled water, and air-dried in the laboratory prior to use. Go-flos were allowed to "soak" and rinse at the depth of sample collection for approximately 30 min prior to tripping. Bottles were kept closed and covered in the ship's lab at all times in order to reduce contamination from atmospheric and other sources.

Seawater for both total DOC concentrations and $\Delta^{14}\text{C}$ measurements was gravity filtered directly from the sampling bottles through a pre-combusted (525°C) 147 mm diameter Gelman type A/E (1- μm nominal pore-size) glass fiber filter. Samples were collected in pre-combusted 1-l amber glass bottles with Teflon-lined caps for $\Delta^{14}\text{C}$ -DOC measurements. All DOC samples were stored at -20°C in the dark until analysis. In addition to DOC, samples were collected at the same times and from the same Go-Flo bottles for dissolved inorganic carbon (DIC) concentrations and $\Delta^{14}\text{C}$ signatures, details of which are provided in Masiello et al. (1998). Briefly, filtered half-liter samples were dispensed into pre-cleaned glass bottles, poisoned with 100 μl saturated HgCl_2 , sealed with greased (Apiezon N grease) ground-glass stoppers and stored at room temperature until analysis. Other samples collected included those for oxygen, salinity total CO_2 , and alkalinity measurements.

Table 1

Concentrations and isotopic natural abundances of DOC collected during Pulse-26 cruise to Sta. M, the continental rise, and upper slope in June 1995. Also included is standard hydrographic data. The reported error in $\Delta^{14}\text{C}$ values of DOC is for AMS counting statistics which was typically greater than replicate or blank-corrected errors for individual analyses

Depth (m)	Temp. (°C)	Salinity	Oxygen (ml/l)	LJA #	CAMS #	DOC (μM)	$\Delta^{14}\text{C}$ ‰	±	$\delta^{13}\text{C}$ ‰
<i>Sta. M</i>									
25	13.9	32.814	6.16	322	28143	72	− 275	5	− 21.9
85	13.6	33.098	5.70	323	28147	63	− 320	4	− 22.3
256	7.489	33.992	2.64	314	28171	46	− 396	4	− 22.2
475	5.953	34.161	0.70	317	28168	42	− 446	4	− 21.3
722	4.941	34.326	0.13	318	28278	38	− 478	4	− 21.4
1282	3.258	34.497	0.77	315	28170	38	− 468	6	− 21.0
1600	—	34.556	0.90	320	28169	40	− 556	3	− 21.8
2515	1.771	34.652	3.30	312	28201	37	− 568	3	− 21.5
2515	—	—	—	316	28175	36	− 552	3	− 21.8
3466	1.520	34.677	3.12	301	28199	37	− 565	3	− 22.4
4097	1.498	34.690	3.01	324	28192	37	− 549	4	—
<i>Continental Rise site</i>									
22	12.896	33.187	6.46	306	28137	65	− 287	5	—
189	8.077	33.992	2.70	311	28165	46	− 399	4	− 22.4
681	4.759	34.336	0.30	303	28172	41	− 491	3	− 21.5
1628	2.632	34.571	1.26	300	28198	35	− 543	3	− 22.1
2568	1.766	34.657	2.68	319	28176	37	− 568	3	− 21.1
3152	1.627	34.663	3.01	304	28174	39	− 545	3	− 22.1
3601	1.597	34.666	3.11	299	28197	37	− 540	3	—
3884	1.568	34.686	3.34	302	28200	37	− 536	3	− 21.8
<i>Upper Continental Slope site</i>									
25	10.5	33.442	4.93	308	28134	56	− 324	4	− 21.8
100	10.2	33.824	3.14	310	28154	50	− 372	4	− 21.7
259	7.468	34.126	1.62	309	28167	46	− 422	4	− 21.9
419	6.036	34.188	0.73	307	28166	43	− 444	4	− 21.4
483	5.819	34.213	0.70	305	28158	45	− 466	3	− 21.5

2.3. Analytical methods

Concentrations of DOC were measured by high energy ultraviolet (UV) irradiation of seawater samples. The UV determinations of DOC concentration were conducted as a routine part of the measurement of ^{14}C natural abundance of DOC. The DOC was oxidized by a modification of the method described in Williams and Druffel (1987) and Druffel et al. (1992). Seawater samples (650 ml) were transferred to an optically clear quartz vessel, acidified to pH 2–2.5 with 85% H_3PO_4 , and sparged free of inorganic carbon for 45 min with ultra-high purity nitrogen or oxygen. The quartz vessel was designed to interface directly to a vacuum extraction line. Following sparging, the seawater sample was irradiated for 120 min at the focal point of a

Table 2

Concentrations and isotopic natural abundances of DIC collected during Pulse-26 cruise to Sta. M, the continental rise and upper slope in June 1995. Also included is total alkalinity data. The reported error in $\Delta^{14}\text{C}$ values of DIC is for AMS counting statistics which in all cases was greater than replicate or blank-corrected errors. Analytical errors ($\pm 1\sigma$) were: DIC – $\pm 3 \mu\text{M}$; alkalinity – $\pm 4 \mu\text{M}$; $\delta^{13}\text{C}$ – $\pm 0.1\%$

Depth (m)	UCI#	CAMS#	DIC (μM)	$\Delta^{14}\text{C}$ (‰)	\pm	$\delta^{13}\text{C}$ (‰)	Alkalinity ($\mu\text{mol kg}^{-1}$)
<i>Sta. M</i>							
25	1315	24817	2026	68	7	1.6	2206
85	1318	24820	2067	68	5	1.3	2222
256	1500	32430	2245	– 7	7	– 0.2	2274
256	1500	32431	2245	– 10	6	– 0.1	2274
475	1610	32437	2334	– 116	5	– 0.4	2311
722	1498	32429	2386	– 169	6	– 0.6	2348
1282	1614	32441	2416	– 223	5	– 0.6	2395
1600	1615	32442	2414	– 234	4	– 0.3	2415
2515	1620	32447	2418	– 247	4	– 0.3	2439
3466	1504	32435	2398	– 250	5	– 0.1	2423
4097	1625	32452	2394	– 236	4	0.0	2439
<i>Continental Rise site</i>							
22	1501	32432	2076	45	6	1.2	2231
189	1623	32450	2243	– 5	6	– 0.2	2271
681	1621	32448	2383	– 166	5	– 0	2348
1628	1609	32436	2422	– 238	4	– 0.32	2413
2568	1622	32449	2417	– 263	4	– 0.3	2440
3152	1616	32443	2399	– 234	4	0.1	2438
3601	1624	32451	2408	– 242	5	– 0.2	2433
3722	1502	32433	2402	– 232	4	0.0	2442
3884	1612	32439	2407	– 230	4	0.1	2441
<i>Upper Continental Slope site</i>							
25	1617,19	32444	2143	44	7	0.2	2236
25	1619	32446	2143	44	6	0.4	2236
100	1613	32440	2215	12	5	0.1	2252
259	1618	32445	2291	– 52	5	– 0.3	2290
419	1611	32438	2345	– 124	5	– 0.4	2314
483	1503	32434	2353	– 133	5	– 0.5	2325

2400-W, medium pressure mercury arc UV lamp equipped with a reflector assembly (Canrad-Hanovia Co., Newark, NJ). Following irradiation, the CO_2 evolved from the oxidation of DOC was purged from the quartz vessel using ultra-high purity nitrogen, transferred to the vacuum extraction line through a KIO_3^- trap to remove Cl_2 gas, cryogenically purified and quantified using an absolute pressure gauge (MKS Corp.). The CO_2 sample was then split, with $\sim 90\%$ of the volume used for subsequent $\Delta^{14}\text{C}$ analysis and $\sim 10\%$ for $\delta^{13}\text{C}$ analysis, and flame-sealed into 6 mm Pyrex tubes.

Samples for $\Delta^{14}\text{C}$ -DIC analyses were processed according to the methods of McNichol et al. (1994) and Masiello et al. (1998). Briefly, a 250-ml aliquot of the

poisoned, preserved sample was acidified with concentrated phosphoric acid, and the evolved CO₂ gas was purified on a vacuum extraction line prior to sealing into Pyrex tubes for subsequent analyses of both ¹⁴C and ¹³C. Blank CO₂ from sample processing was found to contribute no more than 0.6% of the total sample CO₂, and Δ¹⁴C blank corrections were less than ± 2‰ (Masiello et al., 1998).

The CO₂ samples for Δ¹⁴C measurements of both DOC and DIC were converted to graphite targets in an atmosphere of H₂ over Co catalyst (Vogel et al., 1987; 1989) and analyzed at the Center for Accelerator Mass Spectrometry at Lawrence Livermore National Laboratory. Typical Δ¹⁴C analytical measurement errors (± 1σ) for sample sizes in the range analyzed (~ 250–600 μg C) were ± 3–6‰. All reported Δ¹⁴C values were corrected for fractionation using the δ¹³C values of the samples (Stuiver and Polach, 1977), which were measured with either a VG Micromass 602E or a Finnegan Delta S isotope ratio mass spectrometer having analytical precisions of better than ± 0.1‰. Irradiations of replicate seawater samples collected from the same cruise and depth were within approximately ± 1 μMC (i.e., ± 1σ) of each other with respect to DOC concentrations calculated from CO₂ yields. The Δ¹⁴C values of these replicate samples had a precision within the Δ¹⁴C measurement error of 3–6‰. The total blank for the UV oxidation procedure (including sample handling and sparging) was determined by reprocessing and re-irradiating both seawater and double-distilled water samples that had been previously processed and the DOC removed by UV oxidation. The total processing blank was found to be < 10 μg C (less than 2–4% of the sample size).

3. Results

3.1. DOC concentrations and Δ¹⁴C signatures

Data on DOC and DIC concentrations and isotopic (¹⁴C and ¹³C) natural abundances, and hydrographic data from Sta. M and the Rise and Upper slope stations, are presented in Tables 1 and 2. Concentrations of DOC at the three sites were similar (Fig. 2A) and ranged from 37 μM in Sta. M and Rise deep waters to 72 μM in Sta. M surface waters (Table 1). In deeper waters of the Rise, DOC showed greater variability with depth in DOC concentrations than at Sta. M. In shallower mixed-layer waters, concentrations were greatest at Sta. M followed by the Rise and Upper Slope sites (Fig. 2B). At 25 m depth, the range in DOC was approximately 18 μM across the three sites. Concentrations at Sta. M and the Upper Slope site converged to similar by ~ 250 m; at depths greater than 250 m, DOC concentrations were similar at Sta. M and the Rise site. At the greatest depth sampled at the Upper Slope site (483 m or ~ 15–20 m above the seafloor), DOC concentrations increased slightly (Fig. 2B), possibly due to near-bottom influences.

The Δ¹⁴C-DOC profiles at Sta. M (Fig. 3A) were similar to those observed previously (Bauer et al., 1998). Δ¹⁴C-DOC values across all three sites ranged from

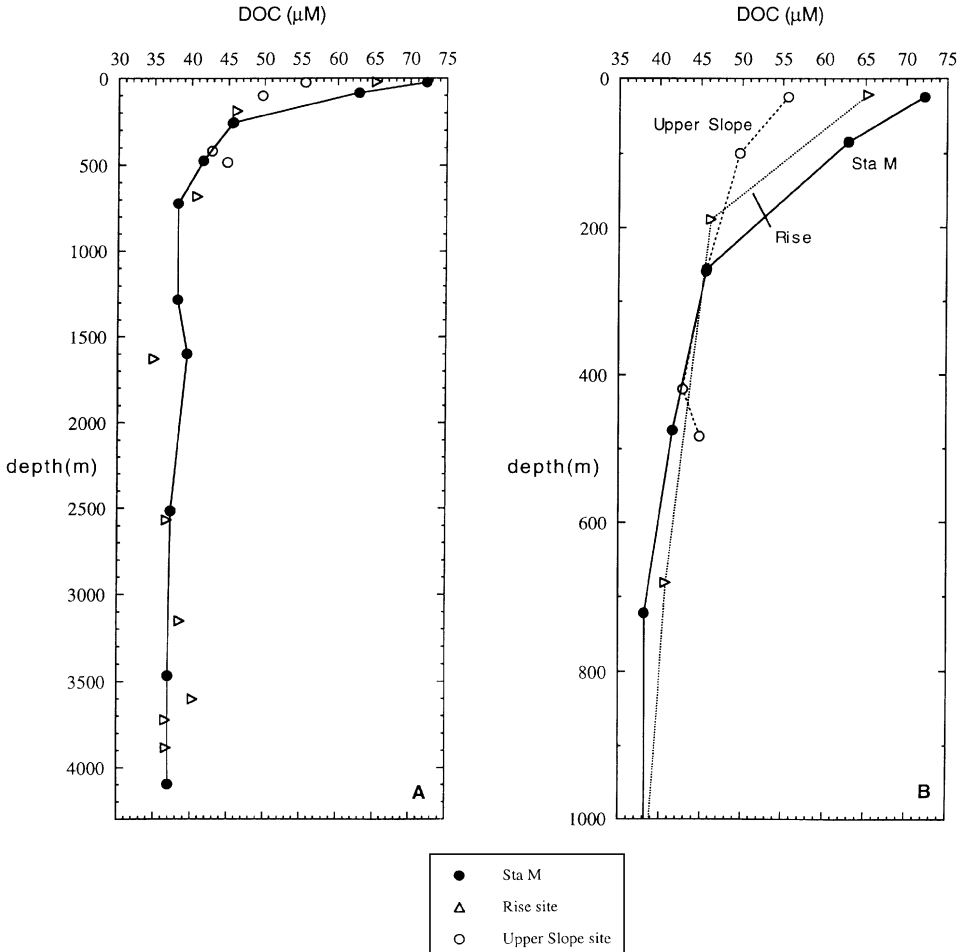


Fig. 2. Concentrations of DOC as measured by high-energy ultra-violet oxidation of 650-ml samples and manometric quantification of CO₂ on a vacuum extraction line for (A) the entire water column and (B) the upper 1000 m of each of the three study sites. Symbols used in all figures are: filled circles – Sta. M; open triangles – Rise site; open circles – Upper Slope site. Typical analytical error for these measurements was ± 1 µM C.

a low of - 576‰ in deep (3600 m) waters to a high of - 275‰ in shallow (25 m) waters (Table 1). Similar to DOC concentrations, Δ¹⁴C-DOC appeared to be more variable throughout the water column at the Rise site than at Sta. M. In waters shallower than ~ 1000 m depth (Fig. 3B), DOC was found to have a small (20–40‰) but significant depletion in Δ¹⁴C at the Rise and Upper Slope sites relative to Sta. M. At 25 m depth, Δ¹⁴C-DOC was significantly greater at Sta. M (- 275‰) and the Rise site (- 287‰) than at the Upper Slope site (- 324‰; Fig. 3B).

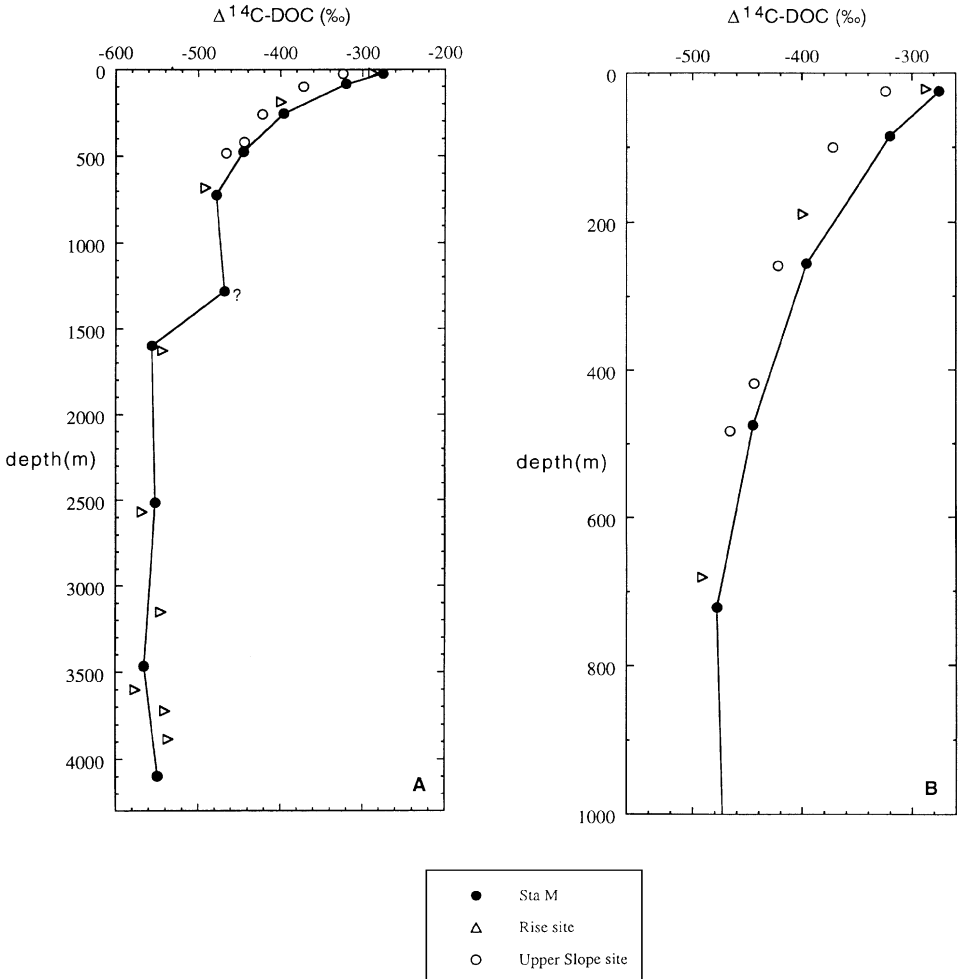


Fig. 3. $\Delta^{14}\text{C}$ values of DOC extracted by high-energy ultra-violet oxidation of 650-ml samples for (A) the entire water column and (B) the upper 1000 m of each of the three study sites.

3.2. DIC concentrations and $\Delta^{14}\text{C}$ signatures

Concentrations of DIC increased with increasing depth and also increased in surface and mesopelagic (to ~ 500 m) waters from Sta. M to the Upper Slope site (Figs. 4A and B). In deep waters at Sta. M and the Rise site, DIC concentrations (Fig. 4A) and $\Delta^{14}\text{C}$ values (Fig. 5A) were similar. However, three of the four samples closest to the seafloor at the Rise site showed small but significantly elevated DIC and $\Delta^{14}\text{C}$ -DIC values compared with the two deepest Sta. M samples (Figs. 4A and 5A, respectively). In shallow waters, the highest DIC values were observed in waters of the

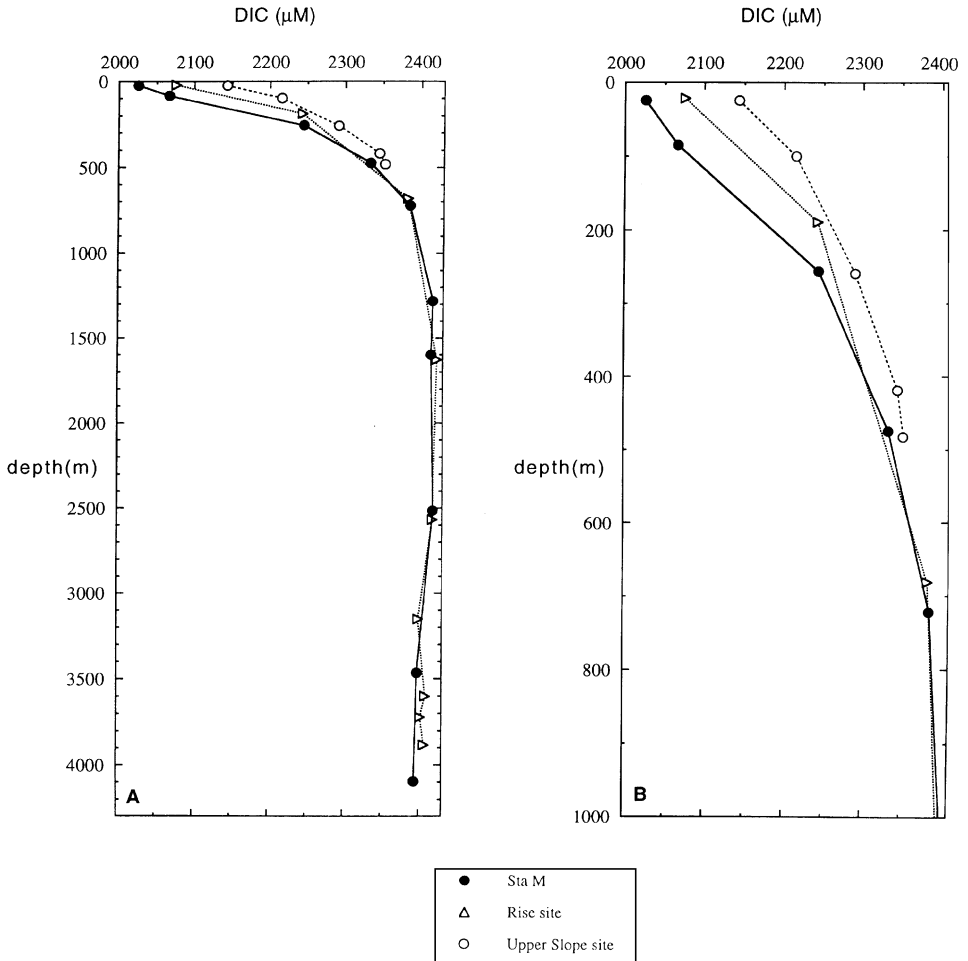


Fig. 4. Concentrations of DIC as measured by acid-sparging 250-ml samples and manometric quantification of CO₂ on a vacuum extraction line over (A) the entire water column and (B) the upper 1000 m of the three study sites. Typical analytical error for these measurements was ± 3 μM C.

Upper Slope and the lowest values were found at Sta. M (Fig. 4B). Thus, DIC appeared to have an inverse relationship to DOC in the upper 200 m of the three sites. The Δ¹⁴C-DIC values were concomitantly highest in Sta. M surface waters and lower and similar in Rise and Upper Slope waters (Fig. 5A and B).

3.3. δ¹³C signatures of DOC and DIC

With the exception of the deepest sample at Sta. M, δ¹³C values of DOC (Table 1; Fig. 6A) ranged between - 22.4‰ and - 21.1‰ across all three sites and there were

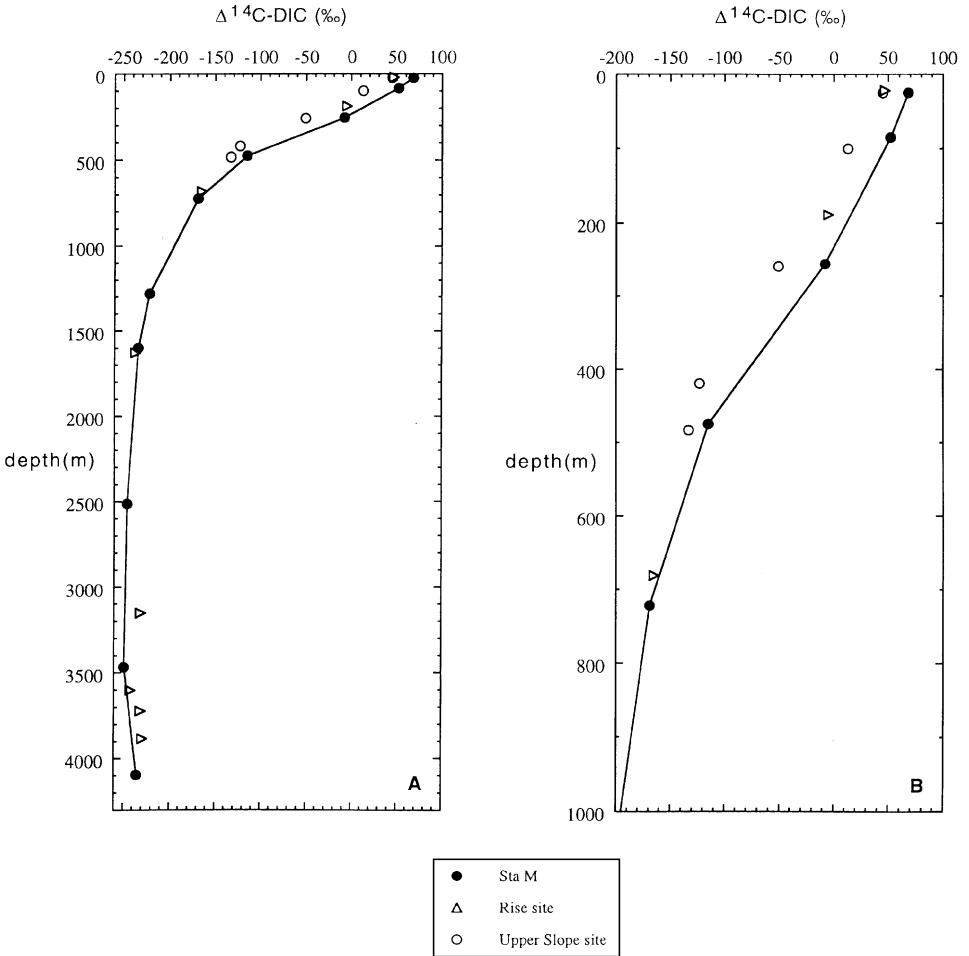


Fig. 5. $\Delta^{14}\text{C}$ values of DIC for (A) the entire water column and (B) the upper 1000 m of the three study sites.

no major apparent differences over most of the water column. At 4097 m depth (~ 2 m above bottom) at Sta. M, the $\delta^{13}\text{C}$ of DOC was significantly elevated (-19.9‰) compared to all other samples (Fig. 6A), suggesting that near-bottom processes may be important here. However, this anomalous $\delta^{13}\text{C}$ value was not concomitantly reflected in significantly different DOC concentrations (Fig. 2A) or $\Delta^{14}\text{C}$ values (Fig. 3A) at this depth at Sta. M. The range of $\delta^{13}\text{C}$ values observed for DOC is virtually identical to that of suspended POC (range in sub-mixed-layer waters: -21.8‰ to -19.7‰); surface waters (25–85 m) at Sta. M were significantly lower ($\delta^{13}\text{C} = \sim -25\text{‰}$) in $\delta^{13}\text{C}$ -POC due to fractionation by phytoplankton (see Table 1 and Fig. 2c in Druffel et al., 1998). It is also interesting to note that in shallow surface waters of all three sites where $\delta^{13}\text{C}$ -POC values are highly depleted (-22.9‰

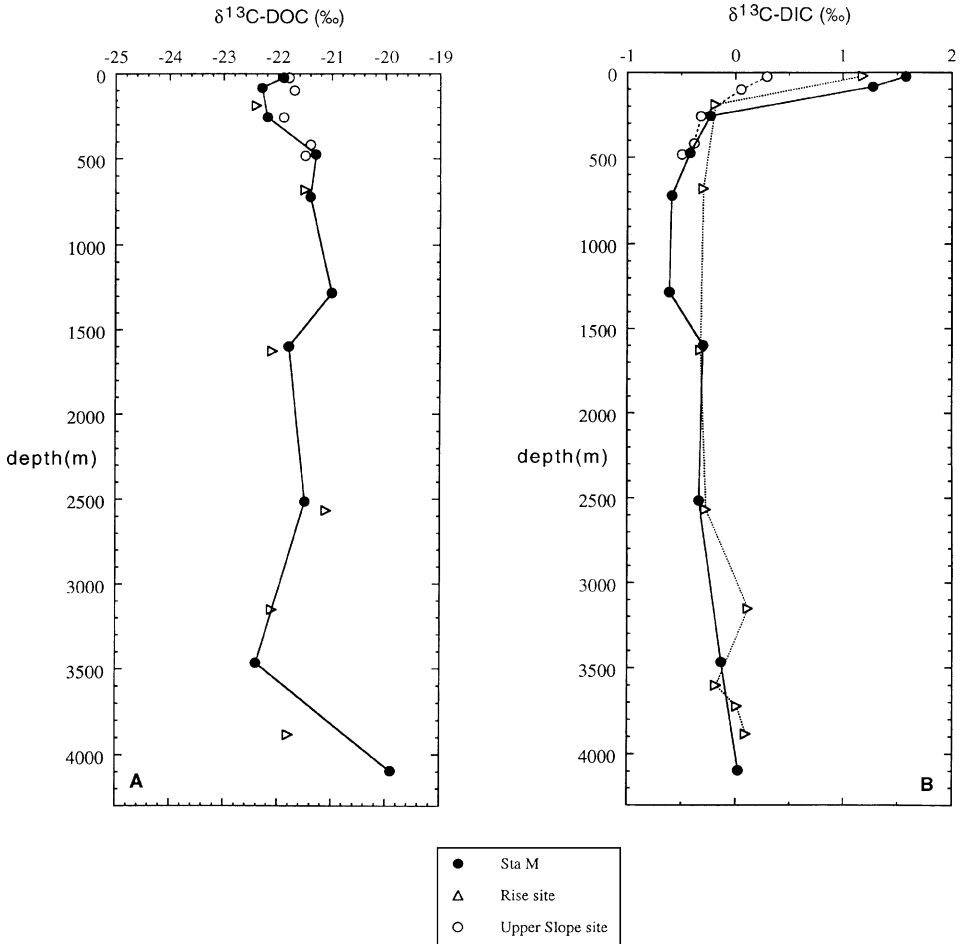


Fig. 6. $\delta^{13}\text{C}$ values of (a) DOC for the entire water column of each of the three study sites, and (b) DIC for the entire water column of each of the three study sites.

to -24.7‰ ; Druffel et al., 1998), there is a small corresponding depletion in $\delta^{13}\text{C}$ of DOC (Fig. 4A). Assuming that the $\delta^{13}\text{C}$ of DOC should resemble that of the POC from which it is derived, this suggests that recently produced DOC (i.e. isotopically light) comprises a relatively small fraction of the total DOC found there, at least for the times at which these samples were collected.

The $\delta^{13}\text{C}$ values for DIC also showed the effects of both upwelling and different rates of primary production at the three sites (Table 2; Fig. 6B). The most enriched $\delta^{13}\text{C}$ values were found in Sta. M surface waters, where inputs of isotopically light deep water were less than at the Rise and Upper Slope sites, and where rates of primary production, which fractionates against ^{13}C , are generally higher

(CALCOFI, 1995). A minimum in $\delta^{13}\text{C}$ was noted at Sta. M between ~ 500 and 1600 m depth (Fig. 6B) in the vicinity of the oxygen minimum (~ 700 m, Table 1; also, Bauer et al., 1998). A similar minimum was not noted for the Rise or Upper Slope sites.

4. Discussion

Ocean margins are regions where the rates of primary production, organic matter sources and fluxes may exceed significantly those of the open ocean (Smith and MacKenzie, 1987; Wollast, 1991; Smith and Hollibaugh, 1993). The factors contributing to these differences include proximity of a given ocean margin to terrestrial and riverine inputs, water column depth and the contribution of sediment particles and solutes to the water column, and seasonal effects of coastal upwelling. In the eastern North Pacific ocean margin, deep ($> \sim 1000$ m) concentrations and radiocarbon signatures of both DOC and DIC exhibited only minor deviations between a site located at the continental rise and that of Sta. M, on the eastern edge of the abyssal plain. However, in shallower ($< \sim 1000$ m) rise waters, DOC concentrations and $\Delta^{14}\text{C}$ -DOC signatures were lower than in Sta. M waters while higher DIC concentrations were associated with lower $\Delta^{14}\text{C}$ -DIC values. A similar but more pronounced pattern also was observed in DOC and DIC at all depths in Upper Slope station waters compared to Sta. M. Previous studies using ^{14}C in ocean margins have been limited primarily to DIC where it has been used to estimate the balance between atmospheric influx of contemporary CO_2 and upwelled, ^{14}C -depleted DIC from deeper waters (Robinson, 1981; Tanaka et al., 1990; Kashgarian and Tanaka, 1991).

Studies that include ^{14}C measurements of DOC or colloidal organic carbon in ocean margins are few (Williams et al., 1992; Guo et al., 1996; Bauer et al., 1998; Bauer and Druffel, in press). Williams et al. (1992) measured $\Delta^{14}\text{C}$ -DOC values in Santa Monica Basin and concluded that they closely resembled ^{14}C -depleted DOC found in the central North Pacific Ocean, suggesting that sources of DOC to Santa Monica Basin from the surrounding California continental borderland were relatively small. In the southern Mid-Atlantic Bight, Guo et al. (1996) observed ^{14}C -depleted ($\Delta^{14}\text{C}$ as low as -709‰) high-molecular weight (> 10 kD) organic matter in near-bottom waters from the shelf and slope. Bauer and Druffel (in press) observed that both high-molecular weight (> 1 kD) and bulk DOC were significantly depleted in ^{14}C in continental slope waters, but significantly enriched in ^{14}C in continental shelf waters. These findings reflect inputs of both old and young forms of DOC (relative to the central North Atlantic) to waters of this trailing-edge margin where freshwater and terrestrial inputs can be substantial (Guo et al., 1996).

Station M has been studied previously with respect to the temporal (seasonal and interannual) variability in DOC, DIC and POC abundances and ^{14}C signatures (Bauer et al., 1998; Druffel et al., 1996; Masiello et al., 1998). Over a 2-year period (1991–1993), changes in the concentrations and $\Delta^{14}\text{C}$ values of DOC could not be differentiated exclusively on the basis of seasonal variations in POC flux rates to the deep ocean (Bauer et al., 1998); however, significant long-term (i.e. months to years)

changes in both concentrations and $\Delta^{14}\text{C}$ values were observed. These changes were especially evident at mesopelagic (ca. 450 and 700 m) depths, close to the oxygen minimum. Positive relationships were found between DOC $\Delta^{14}\text{C}$ values and concentrations at both mixed layer and mesopelagic depths at Sta. M, primarily reflecting diminishing vertical inputs of “recent” DOC through the main thermocline. At abyssal depths ($\geq \sim 1600$ m), however, this relationship was reversed, with $\Delta^{14}\text{C}$ being negatively correlated with DOC concentration and suggesting that there is a source of ^{14}C -depleted DOC to deeper waters, perhaps arising in ocean margin (i.e. rise and slope) regions (Bauer et al., 1998). In addition, previous studies of benthic oxygen utilization at the Rise site (Jahnke et al., 1990; Reimers et al., 1992) have invoked lateral transport of organic carbon from the continental slope to explain the high rates of oxygen utilization observed there.

Significantly elevated concentrations of DOC also were observed at Sta. M when compared to a previously occupied site in the north central Pacific (NCP) in all regions of the water column except mesopelagic depths, where concentrations were lower (Bauer et al., in press). The corresponding $\Delta^{14}\text{C}$ values of DOC at Sta. M were always lower than in the NCP. We have speculated (Druffel et al., 1996; Bauer et al., 1998) that dissimilarities in the size and $\Delta^{14}\text{C}$ signature of the DOC pools at seasonally productive Sta M and the oligotrophic NCP result from differences in DOC sources and sinks between the two regions, as well as from the magnitude of interaction between DOC and POC at these sites. We also have hypothesized that there exists in ocean margins a ^{14}C -depleted source of DOC that may contribute to the observed old ^{14}C ages of DOC in the central gyres (Williams and Druffel, 1987; Bauer et al., 1992; Druffel et al., 1992; Bauer and Druffel, in press). While ocean margins can be net sources of organic carbon and have ^{14}C ages that differ from the open ocean, our transect from the eastern edge of the North Pacific abyssal plain to the upper continental slope does not indicate strong gradients in DOC and DIC in deep waters of the three study sites. This is in agreement with studies of suspended POC at these three sites (Druffel et al., 1998), where resuspension and lateral transport of sediment-derived POC was found likely to be an important factor at the Upper Slope site and in deeper waters of both the Rise site and Sta. M. The gradients in DOC and POC concentrations and $\Delta^{14}\text{C}$ signatures between Sta. M and the NCP (Bauer and Druffel, in press) and the concomitant absence of large gradients between Sta. M and the Upper Slope (this study) suggest that the entire eastern North Pacific margin (rather than specific zones within it) acts as a source region of ^{14}C -depleted organic matter.

Radiocarbon signatures of DIC profiles at Sta. M also have been shown to exhibit seasonal and longer variability (Masiello et al., 1998). The magnitude of variability in $\Delta^{14}\text{C}$ -DIC was found to be greatest (up to 53‰) in surface (25 and 85 m) and upper mesopelagic (~ 450 m) waters, followed by abyssal waters (32 and 23‰ at 3500 and 4050 m, respectively); the lowest variability in ^{14}C -DIC (12‰) was at two intermediate depths (1600 and 2500 m) at Sta. M. Masiello et al. (1998) identified that physical forcing factors are responsible for the high surface and upper mesopelagic variability, and also may account for the relatively high abyssal variability observed at Sta. M. Druffel et al. (1996) and Bauer et al. (1998) were unable to

completely identify all sources of variability in $\Delta^{14}\text{C}$ -POC and $\Delta^{14}\text{C}$ -DOC profiles in deep waters at Sta. M, in spite of the normally strong seasonal signal imposed by productivity and sinking POC fluxes in this region (Hayward et al., 1994; Smith et al., 1994).

Shallower waters (to ~ 250 m for DOC and 500 m for DIC) of the Rise and Upper Slope sites showed distinct and consistent differences in concentrations and $\Delta^{14}\text{C}$ signatures of DOC and DIC from Sta. M (Figs. 2B, 3B, 4B and 5B). Strong upwelling occurs along this part of the U.S. west coast during this time of year (summer) and has previously been observed to result in a depletion in $\Delta^{14}\text{C}$ of DIC (Robinson, 1981). During active upwelling, an upward displacement of isopycnal surfaces occurs closer to the coast, and can potentially result in solutes (e.g. DOC and DIC) from subsurface depths (100–300 m) being mixed with those from surface waters. When DOC concentrations and $\Delta^{14}\text{C}$ signatures from each of our study sites are plotted as a function of seawater density (as σ_t) (Fig. 7A and B, respectively), we find that much of the observed overall difference in the shallow waters is explained by the along-isopycnal mixing of subsurface waters which shoal nearer to the coast (i.e. Upper Slope site). Other regions where upwelling of low-DOC deep waters have been observed include the equatorial Pacific (Peltzer and Hayward, 1996) and high-latitude Southern Ocean waters (Hansell and Waterhouse, 1996; Bauer et al., unpublished data). Carlson et al. (1994) and Ducklow et al. (1995) observed mixed-layer DOC concentrations to decrease dramatically as a result of mixing with low-DOC waters from below the thermocline following the seasonal breakdown of the thermocline in the Sargasso Sea. The level of variability in DOC concentration and $\Delta^{14}\text{C}$ in the eastern North Pacific appears to be at least as great, if not greater, in mesopelagic and deep waters ($\sigma_t \sim 26.5$ – 27.8) between the three sites as in surface waters ($\sigma_t \sim 24.6$ – 26.5) when normalized to seawater density. This suggests that while mixing processes govern most of the variability in surface waters, other processes must play a role in some of the variability observed in deeper waters.

To a first approximation, DOC in the open ocean can be viewed as being comprised of an old, refractory component whose concentration and ^{14}C content are represented by the deep ($> \sim 1000$ m) DOC pool, and a younger component that is absent in deep water but present to varying extents in the upper ocean (Druffel et al. 1992; Carlson et al., 1994; Cherrier et al., 1996). The old, refractory component has been shown to be likely present throughout the water column as a result of the long-lived nature of this fraction (4000–6000 yr) relative to ocean mixing times (~ 1500 yr; Stuiver et al. (1983)). If we assume that surface (22–25 m) waters at the Rise site and Sta. M contain DOC derived from admixtures of deep (using average DOC concentrations and $\Delta^{14}\text{C}$ values from ≥ 1600 m; Table 1) water, we may calculate the $\Delta^{14}\text{C}$ value of the “added” surface-derived component for both sites using the mass balance techniques of Druffel et al. (1992). The general equation used for these calculations is:

$$[\text{DOC}]_{\text{surface}} * \Delta^{14}\text{C}\text{-DOC}_{\text{surface}} = [\text{DOC}]_{\text{deep}} * \Delta^{14}\text{C}\text{-DOC}_{\text{deep}} \\ + [\text{DOC}]_{\text{added}} * \Delta^{14}\text{C}\text{-DOC}_{\text{added}}$$

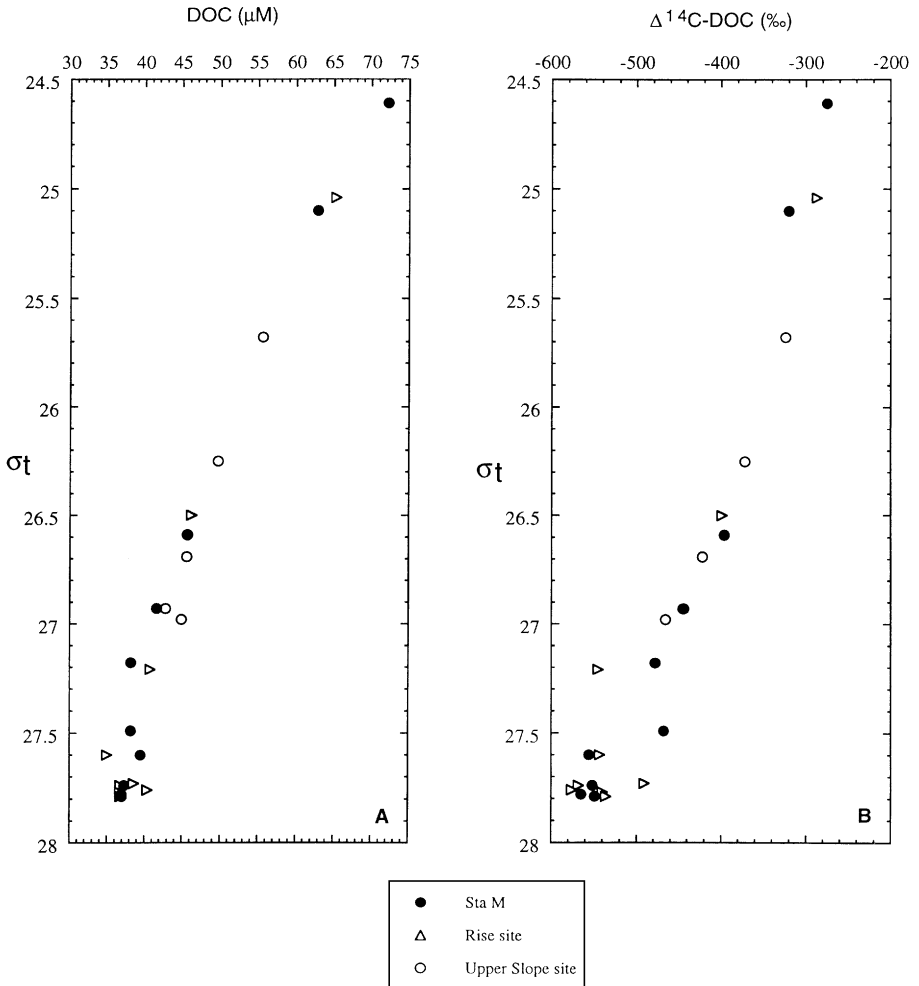


Fig. 7. DOC concentrations and $\Delta^{14}\text{C-DOC}$ values at the three study sites plotted as a function of temperature-corrected density (σ_t). (A) Concentrations for the full range of σ_t over all depths measured; (B) $\Delta^{14}\text{C-DOC}$ values for the full range of σ_t over all depths measured.

where $[\text{DOC}]_{\text{added}}$ is the difference between surface and deep $[\text{DOC}]$, and $\Delta^{14}\text{C-DOC}_{\text{added}}$ is unknown. The values of DOC concentrations and $\Delta^{14}\text{C}$ used are shown in Table 3. Solving for $\Delta^{14}\text{C-DOC}_{\text{added}}$ we obtain $\Delta^{14}\text{C}$ values of the added surface derived component of +36‰ and +61‰ for Sta. M and the Rise site, respectively. Both of these values are close to the $\Delta^{14}\text{C-DIC}$ values observed in surface waters of the Rise site ($\Delta^{14}\text{C-DIC} = +47‰$) and Sta. M ($\Delta^{14}\text{C-DIC} = +68‰$) (Table 2). Thus, surface seawater DOC at both the Rise site and at Sta. M appears to consist of an admixture of ^{14}C -depleted DOC from deep waters (apparently evenly distributed

Table 3

Average values of DOC concentration (in μM) and $\Delta^{14}\text{C}$ -DOC (in ‰) used in mass balance calculations for estimating ^{14}C of “added” surface derived components from all three sites in this study. See text for details of calculations

	Sta. M	Rise	Upper slope
$[\text{DOC}]_{\text{surface}}$	72	65	56
$\Delta^{14}\text{C}\text{-DOC}_{\text{surface}}$	– 275	– 287	– 324
$[\text{DOC}]_{\text{deep}}$	38	38	38
$\Delta^{14}\text{C}\text{-DOC}_{\text{deep}}$	– 556	– 542	– 542
$[\text{DOC}]_{\text{added}}$	34	27	18

both above and below the thermocline) and ^{14}C -enriched DOC from recent surface ocean production.

We can repeat this calculation for Upper Slope surface waters by assuming that they contain DOC derived both from deep ($> \sim 1600$ m) waters similar to those at the Rise site (mean $[\text{DOC}] = 38 \mu\text{M}$ and mean $\Delta^{14}\text{C} = -542\text{‰}$; Table 1), and from some other added component (Table 3). Using the above equation, we calculate by mass balance the $\Delta^{14}\text{C}$ value of the added surface-derived component ($56 \mu\text{M} - 38 \mu\text{M} = 18 \mu\text{M}$) to be $+118\text{‰}$, which is higher than that of present-day DIC ($+45\text{‰}$) at this site (Table 2), but may represent a seasonal average that could be higher than our observed value. The elevated $\Delta^{14}\text{C}$ value of the added component in Upper Slope surface waters also may represent an input of ^{14}C -enriched DOC to these waters. Still, the similarity of the $\delta^{13}\text{C}$ -DOC values in Upper Slope surface waters does not suggest a strong terrestrial source (Table 1).

Concentrations and $\Delta^{14}\text{C}$ signatures of DIC plotted as a function of σ_t instead of depth (Fig. 8A–C and Fig. 9A and B, respectively) indicate that much of the offset in $\Delta^{14}\text{C}$ observed when plotted vs. depth (especially for surface waters where differences of 60–150 μM exist; see Fig. 4B) can be resolved by differences in isopycnal depths between the three sites. However, a residual remains in the offset in DIC (5–15 μM ; see Fig. 8B) such that all Upper Slope and shallow Rise concentrations exceed Sta. M concentrations. This suggests a source of excess DIC nearer to the coast in this region. One possible explanation for this excess DIC is the respiration of organic matter deposited from the water column to slope sediments. Higher rates of respiration occur in slope and rise sediments compared to Sta. M (Jahnke et al., 1990; Reimers et al., 1992), and excess DIC diffuses to the overlying water column. Mixing (presumably dominated by lateral eddy diffusion) of the excess component with seawater leads to a dilution of the sedimentary DIC, but the source is still strong enough to lead to the slight but significant elevated concentrations observed here. Offsets in $\Delta^{14}\text{C}$ -DIC as a function of σ_t between the three sites were not significant except for three of the four deepest Rise samples, which were greater than the average profile using the deepest two Sta. M samples (Fig. 9A and B). The elevated DIC concentrations in Rise and Upper Slope waters, and elevated $\Delta^{14}\text{C}$ values for deep Rise waters, suggest that the source of this excess DIC is respiration of ^{14}C -enriched organic matter in sediments

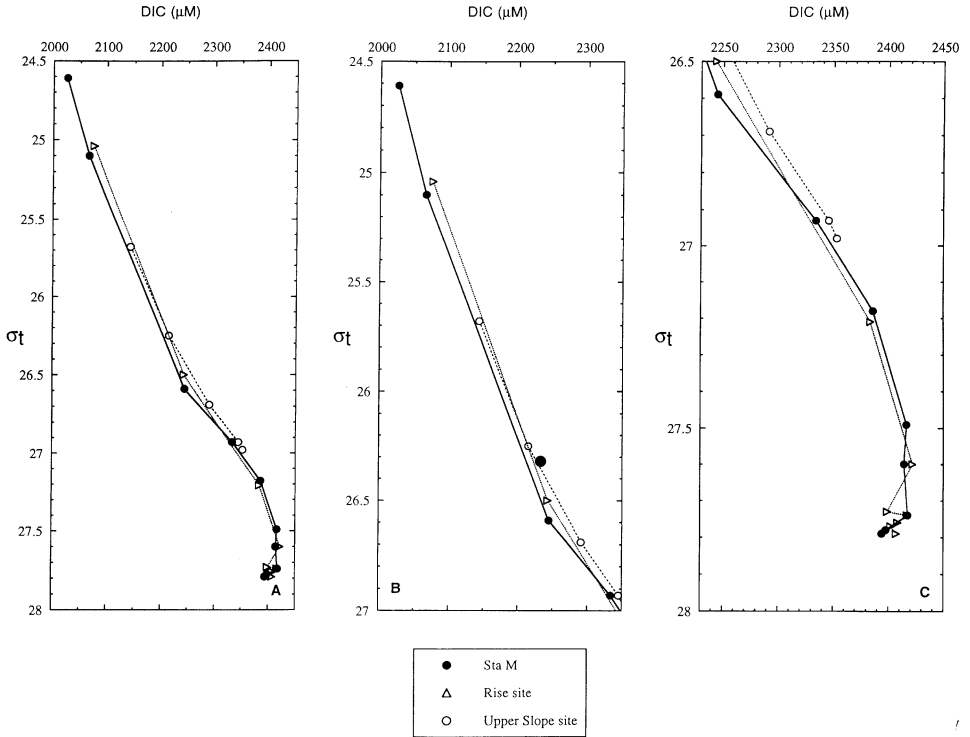


Fig. 8. DIC concentrations at the three study sites plotted as a function of temperature-corrected density (σ_t). (A) Concentrations for the full range of σ_t over all depths measured; (B) Concentrations from the shallowest depth sampled (~ 25 m) to the approximate bottom of the mixed layer (~ 200 – 150 m); (C) Concentrations from the approximate depth of the bottom of the mixed-layer (~ 200 – 250 m) and deeper.

that subsequently diffuses to the water column (Jahnke et al., 1990; Reimers et al., 1992; Bauer et al., 1995).

In general, $\Delta^{14}\text{C}$ -DIC values in the upper and lower mixed-layer (to $\sigma_t < \sim 26.5$; Fig. 9A) at each site differ by considerably less (range: 16‰ at Sta. M to 52‰ at Rise site; Fig. 9A and Table 2) than the corresponding differences in mixed-layer $\Delta^{14}\text{C}$ -DOC (range: 45‰ at Sta. M to 112‰ at Rise site; Fig. 7A and Table 1). The composite relationship between $\Delta^{14}\text{C}$ -DOC and $\Delta^{14}\text{C}$ -DIC for mixed-layer samples at all three sites is shown in Fig. 10A which indicates that the rate of change in $\Delta^{14}\text{C}$ -DOC through the mixed-layer is approximately 1.7 times that of $\Delta^{14}\text{C}$ -DIC. When the entire $\Delta^{14}\text{C}$ -DOC and $\Delta^{14}\text{C}$ -DIC data sets are transformed to ^{14}C concentrations in order to normalize for disparities in DOC and DIC pool sizes (Fig. 10B), we find that below the mixed layer there exists a linear relationship between $[\text{DO}^{14}\text{C}]$ and $[\text{DI}^{14}\text{C}]$. This is primarily a reflection of the greater age (lower ^{14}C) of the deep DOC reservoir due to more extensive recirculation and hence aging compared to the

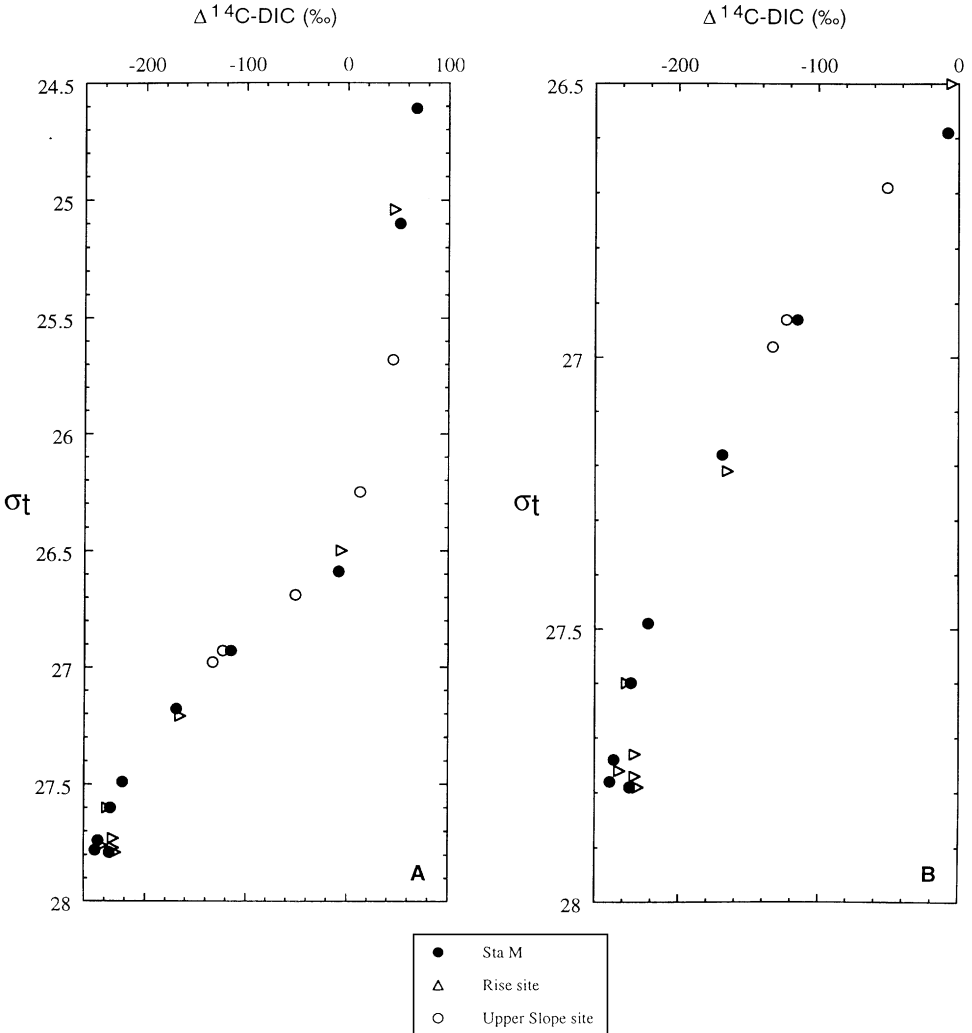


Fig. 9. $\Delta^{14}\text{C-DIC}$ values at the three study sites plotted as a function of temperature-corrected density (σ_t). (A) $\Delta^{14}\text{C-DIC}$ values for the full range of σ_t over all depths measured; (B) $\Delta^{14}\text{C-DIC}$ values from the approximate depth of the bottom of the mixed-layer (~200–250 m) and deeper.

deep DIC reservoir. In the mixed layer, however, while $[\text{DI}^{14}\text{C}]$ is “reset” as a result of atmospheric equilibration, $[\text{DO}^{14}\text{C}]$ shows a broad range of values likely associated with heterotrophic utilization of DOC during recycling of photosynthates.

$\Delta^{14}\text{C-DOC}$ values also showed varying relationships with $\Delta^{14}\text{C-POC}$ values (Fig. 10C; also see Druffel et al., 1998). In the surface and mesopelagic water column, $\Delta^{14}\text{C-DOC}$ varied exponentially with $\Delta^{14}\text{C-POC}$, and the slope of the relationship was smallest at mesopelagic depths and greatest in near-surface waters. This suggests

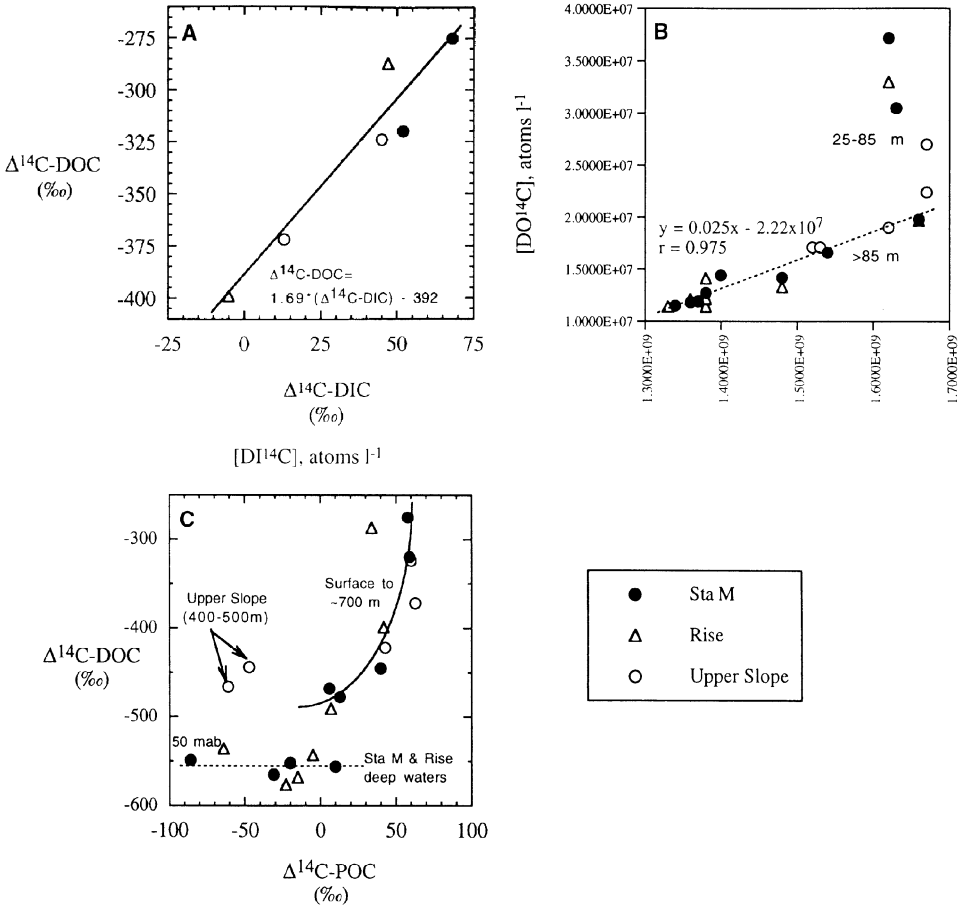


Fig. 10. (A) $\Delta^{14}\text{C-DOC}$ vs $\Delta^{14}\text{C-DIC}$ values for the two shallowest depths sampled at each of the study sites. (B) Abundance of ^{14}C (in atoms of ^{14}C per liter) in DOC vs DIC for all samples measured. (C) $\Delta^{14}\text{C-DOC}$ vs $\Delta^{14}\text{C-POC}$ values for all depths and sites where both DOC and POC were collected.

that there are varying degrees of “coupling” (i.e. either via POC dissolution to DOC or via DOC sorption to POC) between the DOC and POC pools in this region. The selective influence of seafloor processes (e.g. sediment resuspension) on $\Delta^{14}\text{C-POC}$ deeper Upper Slope waters is also apparent (Fig. 10C; Figs. 2A and 3A). The two deepest $\Delta^{14}\text{C-DOC}$ values from the Upper Slope were similar to values at mesopelagic depths of the other two sites, but sediment resuspension of old, reworked POC (Druffel et al., 1998) contributed to the deviation of these samples from either of the other relationships shown.

In deeper samples (i.e. below the main thermocline, $\geq \sim 700$ m) from Sta. M and the Rise site, the relationship between $\Delta^{14}\text{C-DOC}$ and $\Delta^{14}\text{C-POC}$ appeared not to co-vary in the same manner as in upper and mesopelagic regions of the water column

(Fig. 10C). Instead, $\Delta^{14}\text{C}$ -POC showed a larger relative range (i.e. relative to the total range in $\Delta^{14}\text{C}$ -POC values) than $\Delta^{14}\text{C}$ -DOC. In the deepest (~ 50 m above the seafloor) Sta. M and Rise waters, ^{14}C -depleted POC values (Fig. 10C) were also observed in the region of the benthic nepheloid layer (Smith et al., 1994) present at these two sites. In both cases, large deviations in $\Delta^{14}\text{C}$ -POC values were noted without a corresponding change in $\Delta^{14}\text{C}$ -DOC. Thus, $\Delta^{14}\text{C}$ -POC appears to be more sensitive than $\Delta^{14}\text{C}$ -DOC to deep ocean processes such as sediment resuspension and lateral transport, probably due in part to its lower relative concentrations than DOC. In addition, as pointed out by Druffel et al. (1998), POC concentrations and $\Delta^{14}\text{C}$ -POC values do not exhibit the same relationships with σ_t as DOC and $\Delta^{14}\text{C}$ -DOC. This indicates that the mixing processes that predominantly control both DOC (and its $\Delta^{14}\text{C}$) and DIC (and its $\Delta^{14}\text{C}$) in waters extending from Sta. M across the eastern North Pacific continental rise and slope do not control the distributions of suspended POC and its $\Delta^{14}\text{C}$. Processes such as resuspension, lateral transport, etc. may preferentially affect one form of carbon (POC) over another (DOC), not only in parts of the water column associated with these processes, but in a general sense between the entire upper and lower water columns of Sta. M and the continental rise.

5. Conclusions

Measurements of DOC and DIC concentrations and $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ values at three sites extending from abyssal Sta. M (~ 4100 m depth), to the continental rise (~ 3800 m) to the upper continental slope (~ 500 m depth) in summer 1995 revealed variable effects of upwelling on all of these parameters in surface waters. The greatest effects were noted at the Upper Slope site where concentrations and $\Delta^{14}\text{C}$ values of DOC and DIC were significantly lower than the Rise site; values from both the Upper Slope and Rise were in turn lower than values at Sta. M. In deep waters of the Rise and Sta. M, the main differences observed were in the deepest samples where both DIC concentrations and several $\Delta^{14}\text{C}$ -DIC values were slightly elevated at the Rise, indicating that there is a source (most likely from water mass intrusion or sediment respiration) of ^{14}C -enriched DIC to the water column there. Several differences were also observed in the relative distributions of $\Delta^{14}\text{C}$ -DOC and $\Delta^{14}\text{C}$ -POC throughout the water column at all three sites indicating that the mechanisms controlling the ^{14}C contents and inputs of DOC and suspended POC are distinct from one another.

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