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NOTE

Depth distribution of $\delta^{13}C$ of dissolved ΣCO_2 in seawater off eastern Taiwan: effects of the Kuroshio current and its associated upwelling phenomenon

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Abstract—This paper presents depth variations of dissolved O_2, ΣCO_2, alkalinity, and $\delta^{13}C$ of ΣCO_2 in seawater off eastern Taiwan. O_2 decreases from an averaged surface value of approximately 4.6 ml l^{-1} to a minimum of about 2.0 ml l^{-1} at a depth of 1000 m, and then increases gradually to about 3.5 ml l^{-1} near the seafloor. ΣCO_2 concentrations increase from typical values of 1900–2050 μmol kg^{-1} at the surface to 2100–2300 μmol kg^{-1} at depths of more than 1000 m, corresponding to an increase in alkalinity from about 2250 μeq kg^{-1} at the surface to 2450 μeq kg^{-1} in the bottom water. $\delta^{13}C$ of ΣCO_2 at the surface averages about +0.25% and becomes lighter with increasing depth to approximately −1.0%. The observed depth distributions of O_2, ΣCO_2, alkalinity and $\delta^{13}C$ of ΣCO_2 are thus similar to those reported in the open ocean and can be attributed to O_2 utilization and CO_2 production during organic decomposition. On a close examination, however, rates of O_2 decrease and ΣCO_2 and alkalinity increases are less profound than those observed in the open ocean, particularly in the Pacific. Furthermore, values of $\delta^{13}C$ measured are invariably lighter than those reported in the Pacific. The discrepancy is attributed largely to the presence of the Kuroshio current and its associated upwelling phenomenon in the continental margin off eastern Taiwan. Copyright © 1996 Elsevier Science Ltd.

1. INTRODUCTION

In spite of its non-conservative behavior and dynamic nature, $\delta^{13}C$ of dissolved ΣCO_2 in seawater has been used in combination with other parameters as an additional tracer to study the fate and pathway of carbon in the oceans. For instance, Deuser and Hunter (1969) found that $\delta^{13}C$ minimum was correlated to dissolved O_2 minimum at all depths below the top 200 m of the water column in the Atlantic, and the phenomenon can be attributed to the oxidation of organic matter that is depleted in ^{13}C. They further postulated that the observed isotopic shift in the dissolved inorganic carbon could be used to assess the contribution of organic carbon that was oxidized. Craig (1970) showed that

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the $\delta^{13}C$ of dissolved inorganic carbon in the South Pacific decreased from +2.2‰ at the surface to +0.5‰ in the bottom water, and there was a $\delta^{13}C$ minimum corresponding to the ΣCO_2 maximum and the dissolved O_2 minimum at a depth of approximately 2500 m. He then estimated that the increase in deep-water ΣCO_2 from particulate flux would be derived about 30% from planktonic carbonate shells and 70% from organic matter. Soon after these findings, similar depth profiles of $\delta^{13}C$ and estimates of carbonate vs organic contributions have been reported in both the Atlantic and the Pacific (Kroopnick *et al.*, 1970, 1972; Kroopnick, 1974a,b, 1980, 1985).

However, these studies had been mainly conducted in the open oceans, with scant attention to the marginal sea thus far. This paper presents systematic measurements of $\delta^{13}C$ in the water column off eastern Taiwan, a unique marginal sea area characteristic of the existence of the Kuroshio current and its associated upwelling phenomenon.

2. EXPERIMENTAL METHODS AND PROCEDURES

A total of 19 hydrographic stations off eastern Taiwan was investigated during cruises in April 1989 (stations R1–R7 for O_2) and September 1990 (stations 1–12 for ΣCO_2, alkalinity and $\delta^{13}C$), respectively (Fig. 1). All water samples were collected at various depths with 2.5-l Niskin bottles using a rosette sampler aboard the RV *Ocean Research I*. Except for O_2 measurements, samples used for alkalinity (500 ml), ΣCO_2 (500 ml) and carbon isotope analyses (300 ml) were added with 1 ml of $HgCl_2$ immediately after sampling and stored at 5°C in darkness to prevent biological production of CO_2 (Sackett and Moore, 1966; Kroopnick *et al.*, 1972).

Measurement of dissolved O_2 concentration was performed aboard the ship using the Winkler titration method with minor modifications (Carpenter, 1965). Upon return to the laboratory, concentrations of ΣCO_2 and alkalinity were determined by the conventional titration techniques (Bradshaw *et al.*, 1981). Precision estimated from replicates was ±0.1 ml l^{-1}, ±5 μmol kg^{-1} and ±3 μeq kg^{-1} for O_2, ΣCO_2 and alkalinity, respectively. In-situ measurements of chlorophyll *a* fluorescence in seawater were performed with a fluometer (Sea Tech Inc.) mounted on CTD and Rosette assemblies (Gong *et al.*, 1993). The fluorescence output from the fluometer had been calibrated with the known chlorophyll *a* concentrations and a proportional factor of 5 was applied to the fluometer for the seawater concentration range of chlorophyll *a* in the region ($<10 \mu g\, l^{-1}$; Chen, 1992). The averaged standard error of measurements was better than ±0.1 $\mu g\, l^{-1}$.

Procedures of extracting ΣCO_2 from seawater for isotopic measurement consist of acidifying the seawater, then collecting and purifying the evolved CO_2 from samples under vacuum. Approximately 3 ml of 100% H_3PO_4 was introduced into the side arm of a custom-made reaction vessel, which could be closed with a high vacuum valve. Next, approximately 60 ml of water sample was transferred into the vessel without protection from contact with air. The procedure took only a few seconds and had been shown previously by Presley and Claypool (1971) to result in an insignificant atmospheric contamination. The vessel was then assembled on the vacuum line to remove the remaining air from the spare space of the vessel.

Because of the difficulty of freezing the larger-size sample vessels in the vacuum line and the frequent crack of glass vessels during the thawing of water after being deep frozen, the removal of air from the reaction vessel in this study was performed without freezing the water samples. There was an attempt to carefully evacuate the air out from the head space,
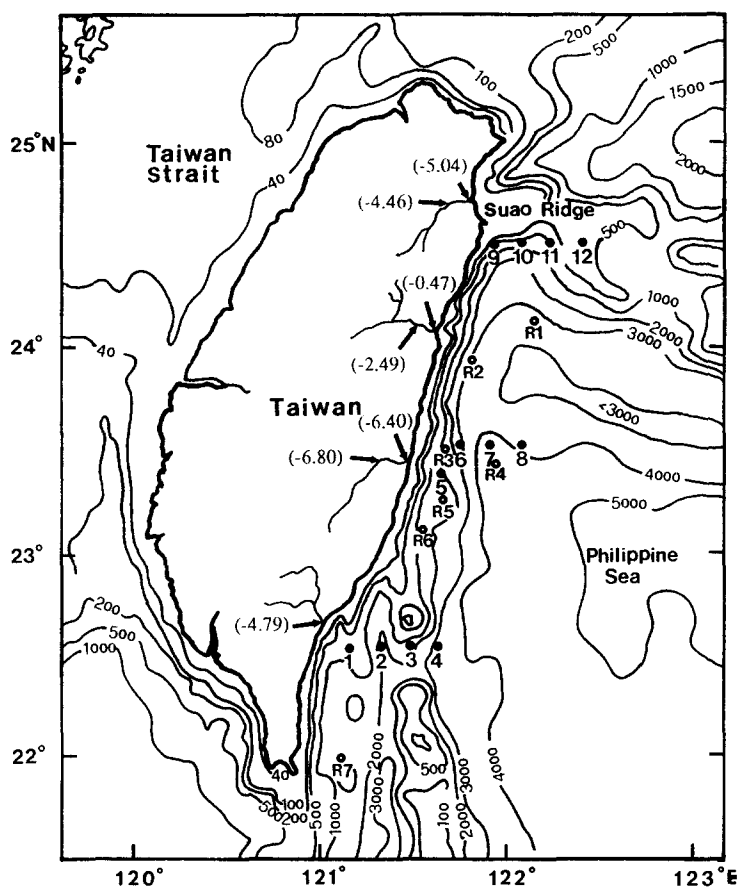
Fig. 1. Bathymetric map showing the sampling locations (stations R1–R6 for dissolved O_2 measurement only; stations 1–12 for ΣCO_2, alkalinity and $\delta^{13}C$ analyses). Data of additional $\delta^{13}C$ measurements for river water samples are shown in parenthesis.

but a vigorous bubbling was often observed and thus made the direct pumping method unsatisfactory. Consequently, the removal of air was furnished by opening repeatedly (usually three times) the vessel to a large space on the line until an incipient degassing is barely observed. After the air was expelled, the vessel was closed and removed from the line. The vessel was tilted to allow the reaction of the water with the acid and placed in a water bath at 50°C with agitation for 20 min. The vessel was then re-assembled on the vacuum line. The CO_2 gas was collected with a liquid nitrogen trap after complete removal of the water vapor by a slurry of a mixture of dry ice and alcohol. In this study, the extraction of CO_2 from water samples usually gave a yield of better than 92%, with the exception of a few samples. The percentage of yield was shown by Sackett and Moore (1966) not to affect the $\delta^{13}C$ value from a given water sample.

Isotopic analysis was performed with a VG Sierra 903 mass spectrometer. Results of isotopic measurement were expressed with the conventional δ notation and reported as per mil (‰) difference relative to the PDB standard (Craig, 1957). A total of more than

150 seawater samples including replicates was measured. Replicates of full procedural analysis showed the precision was variable but generally better than ±0.15‰.

As mentioned previously, the procedure for removal of the air from the vessels might result therefore in a slight loss of dissolved CO_2 from water samples and yields an enrichment of $\delta^{13}C$ in the remaining water due to the known isotopic fractionation of carbon between CO_2 and HCO_3^- in the water (e.g. $\alpha = 1.0096$ at 10°C, Deuser and Degens, 1967; Friedman and O'Neil, 1977). The following two experiments had been carried out to evaluate such a potential problem on the $\delta^{13}C$ measurements, and necessary corrections were made accordingly. In the first experiment, the CO_2 that was outgassed from the water sample was collected, and it was added to that was liberated from the seawater sample after reaction with acid, and then analyzed the combined CO_2 for $\delta^{13}C$. The second experiment first involved the evacuating of another home-made vessel that had filled with acid and had a rubber septum through which the water samples can be injected into the vessel using a syringe. These two methods ensured a complete recovery of all CO_2 in water. Results from these two experiments showed that the $\delta^{13}C$ of CO_2 prepared by the procedure used in this study consistently exhibited an enrichment of ~0.6‰. As a consequence, a corresponding correction of 0.6‰ was applied to the $\delta^{13}C$ values analyzed in this study.

3. RESULTS AND DISCUSSION

The continental margin off eastern Taiwan (Fig. 1) is characteristic of an abrupt change in seafloor topography due to the island-arc/continent collision, and in many areas the bottom slope drops abruptly from the coast to more than 3500 m within a distance of only 40–50 km. (Fig. 1; Chai, 1972; Wu, 1978; Tsai, 1986; Sheu and Huang, 1989; Huang *et al.*, 1992). The region is also known for the Kuroshio current to develop and intensify to a strong western Pacific boundary current after leaving its source area east of Luzon island (Nitani, 1972). The presence of the Kuroshio water off eastern Taiwan can be readily seen in hydrographic profiles due to its relatively high salinity and temperature compared with the overlying and underlying water (Liu, 1983; Liu *et al.*, 1988; Wong *et al.*, 1991). The depth interval of the main path of Kuroshio flowing northward along the east coast of Taiwan ranges approximately from 100 m to nearly 500 m, with a maximum velocity of about 3 knots.

The abrupt change in bathymetry also causes the upwelling phenomenon in the continental margin east of Taiwan (Bodvarsson, 1976; Fan, 1980). Persistent upflow of deep water below the main stream of the Kuroshio, however, is found mainly in regions off southeastern and northeastern Taiwan, where the Kuroshio encounters the continental shelf. Patches of cold water from depth in southeast Taiwan were first identified in a satellite Gemini X photograph by Emery and Stevenson (1972), and later were confirmed by shoreward rises in isotherms, isohalines and chemical hydrographic data (Hung, 1975; Bodvarsson, 1976; Liu, 1983; Wong *et al.*, 1991; Liu *et al.*, 1992). Recently, the striking difference in these properties between the Kuroshio and its adjacent waters off northeast Taiwan was used to study the upwelling mechanism and its associated exchange processes in the region off northeast Taiwan (Liu and Pai, 1987; Wong *et al.*, 1991; Liu *et al.*, 1992). The extension and intensity of the upwelling are further shown by these authors to vary both spatially and temporally.

Measurement of dissolved O_2 at seven hydrographic stations (R1–R7; Fig. 1) was used

to demonstrate the general distribution of O_2 in the water column off eastern Taiwan (Fig. 2). As can be seen, O_2 averages about 4.6 ml l^{-1} at the surface, and then decreases progressively to a minimum of approximately 2.0 ml l^{-1} at 1000 m. Below this depth, O_2 increases gradually to about 3.0 ml l^{-1} near the seafloor. The observed O_2 profiles in this study are consistent with previous observations and are mirror images of the nutrient distributions (Liu *et al.*, 1988; Wen *et al.*, 1989; Wong *et al.*, 1991; Gong *et al.*, 1992). Furthermore, they are similar to those observed in the open ocean despite that dissolved O_2 minima in waters off eastern Taiwan are less distinct than those in the central Pacific, as reported by Kroopnick *et al.* (1972) and Kroopnick (1985).

Figure 3a and b shows the vertical distributions of dissolved ΣCO_2 concentrations measured at stations 1–12. ΣCO_2 increases gradually from a consistent surface value of about 1950 μmol l^{-1} to nearly 2300 μmol l^{-1} at a depth of 2000 m with minor variation. Thus, the increase in ΣCO_2 from the surface to 1000 m is accompanied by a decrease in the total dissolved O_2. Below this depth, both dissolved ΣCO_2 and O_2 increase with depth. Ranges of ΣCO_2 variations with depth are comparable to those observed in the north Pacific (cf. Kroopnick, 1985), yet their concentrations are much higher than those reported in the north Atlantic (e.g. 2300 vs 2160 μmol kg^{-1} at a water depth of 2000 m; Kroopnick *et al.*, 1972).

Alkalinity increases rapidly from an averaged surface value of about 2250 μeg kg^{-1} to about 2300 μeq kg^{-1} at 100 m, remains constant between 100 and 500 m, and then increases gradually to approximately 2450 μeq kg^{-1} towards the seafloor (Fig. 3c and d). The depth
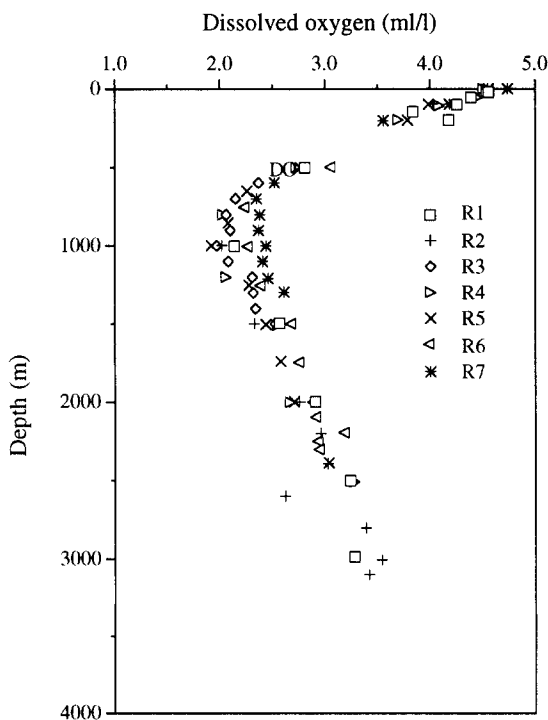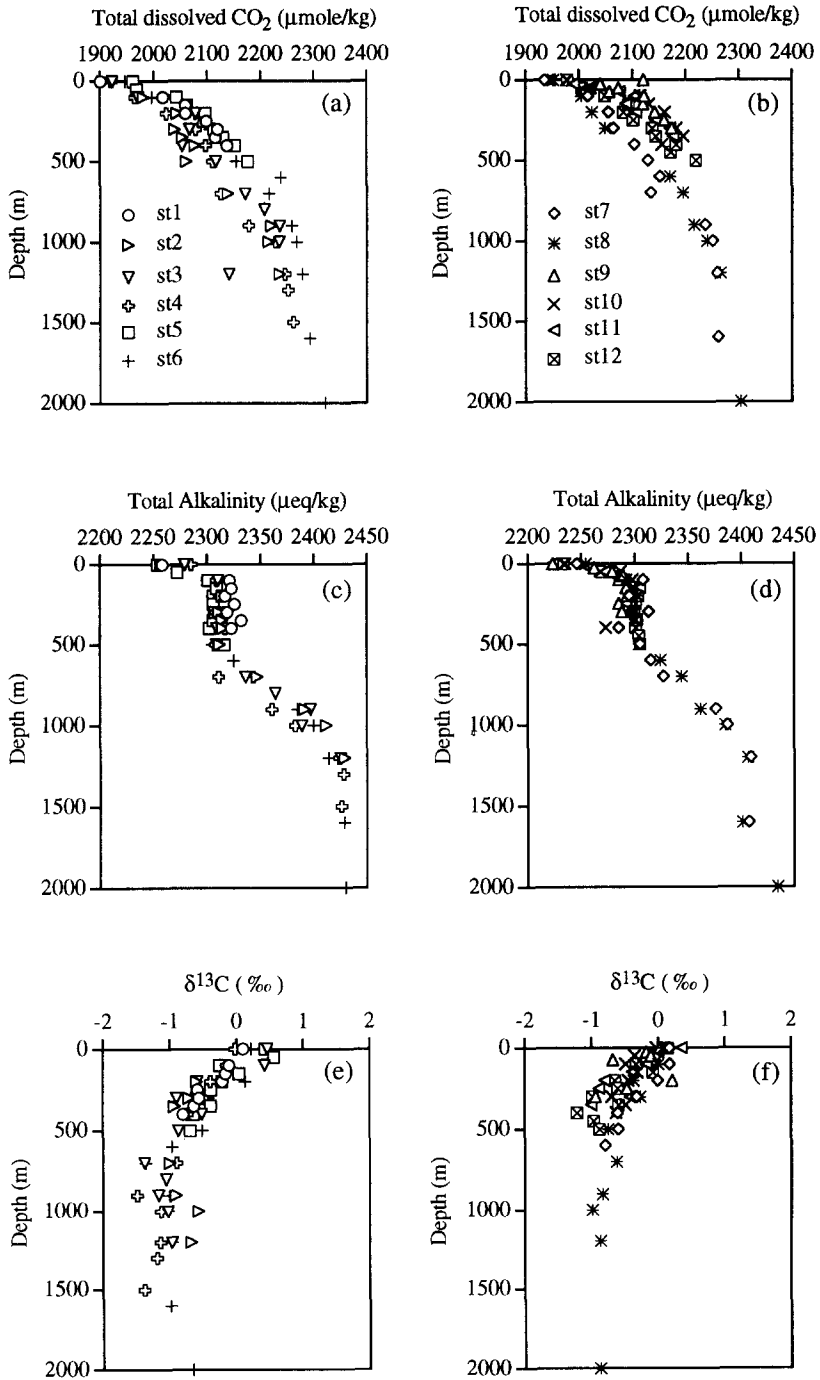

Fig. 2. Depth distributions of dissolved oxygen in seawater off eastern Taiwan. Note that the O_2 minimum locates at the depth of approximately 1000 m.

distribution of constant alkalinity thus appears to coincide with the depth interval of the main path of the Kuroshio. Carbon isotopic composition of ΣCO_2 in the water columns generally decreases from 0 to +0.5‰ at the surface to −1.5 to −1.0‰ near the seafloor with minor variations (Fig. 3e and 3f). The change in $\delta^{13}C$ distribution with increasing depth is similar to that observed in open oceans, yet the gradient is less distinct than that in the Pacific (Kroopnick, 1985). For instance, Kroopnick *et al.* (1970) reported a decrease in $\delta^{13}C$ from 2.2‰ to −5‰ from the surface to 1000 m, while $\delta^{13}C$ in waters off eastern Taiwan varies from 0.5‰ at the surface to approximately −1.0‰ at 1000 m (Fig. 3e and f). Furthermore, values of $\delta^{13}C$ measured in the surface waters off eastern Taiwan are invariably lower than those reported in the north Pacific by a maximal difference of approximately 1‰. The discrepancy can be attributed to the addition of ^{13}C-depleted biogenic CO_2 via upwelling and/or advection of water from depth.

Figure 4a shows a cross-plot of alkalinity vs ΣCO_2 in the waters off eastern Taiwan. As seen, the lack of an increase in alkalinity with increasing ΣCO_2 between 100 and 500 m indicates a production of CO_2 from organic degradation. Organic detritus in the ocean is readily oxidized and decomposed to form CO_2 as it is settling through the water column. This organically derived CO_2 is known to deplete in $\delta^{13}C$ (Williams and Gordon, 1970) and adds to the deep water that then surfaces as upwelling takes place. Figure 4b shows that except for a few data, $\delta^{13}C$ in waters off eastern Taiwan is correlated fairly well to ΣCO_2. Therefore, the correlation suggests that the depth distribution of $\delta^{13}C$ observed in these water columns are the results of the addition of ^{13}C-depleted CO_2 from in-situ decomposition of organic matter.

Nonetheless, factors affecting the observed depletion of $\delta^{13}C$ in the surface waters off eastern Taiwan, other than the addition of the ^{13}C-depleted waters from deep, need further discussion. First, effects of latitudinal change in $\delta^{13}C$ of atmospheric CO_2 that may be the cause of the observed depletion of ~1% in the surface water can be neglected because the present data and those used for comparison are all from stations between 20 and 40°N in the western Pacific and no appreciable change in $\delta^{13}C$ of atmospheric CO_2 is found (Kroopnick, 1985). Furthermore, dissolved CO_2 in river water is known to deplete in ^{13}C relative to seawater and has a range of $\delta^{13}C$ of approximately −3 to −10‰ (Tan, 1989; and references therein). Therefore, the inflow and mixing of this riverine water with offshore seawater would avert a decrease in the $\delta^{13}C$ observed. To elucidate this potential effect, an additional seven river water samples from runoffs in eastern Taiwan were collected and analyzed for their ^{13}C compositions. Results (cf. Fig. 1) show that they all fall in the range of typical riverine water previously established in the literature. Two samples of heavier $\delta^{13}C$ values (i.e. −0.47 and −2.49%) may be attributed to the contribution of solutions from the dissolution of carbonate rocks in the drainage basin (Lee and Teng, 1986). A cross-plot of $\delta^{13}C$ vs salinity in waters between 0 and 100 m (Fig. 4c), however, reveals that they are not significantly correlated. It should be pointed out that using salinity as a conservative tracer to characterize results of mixing of riverine water with seawater in

Fig. 3. Depth distributions of dissolved ΣCO_2 (a and b), total alkalinity (c and d) and $\delta^{13}C$ of ΣCO_2 (e and f) in seawater off eastern Taiwan. Note that the depth interval between 100 and 500 m showing consistent values of total alkalinity coincides with the main path of the Kuroshio in the region off eastern Taiwan.
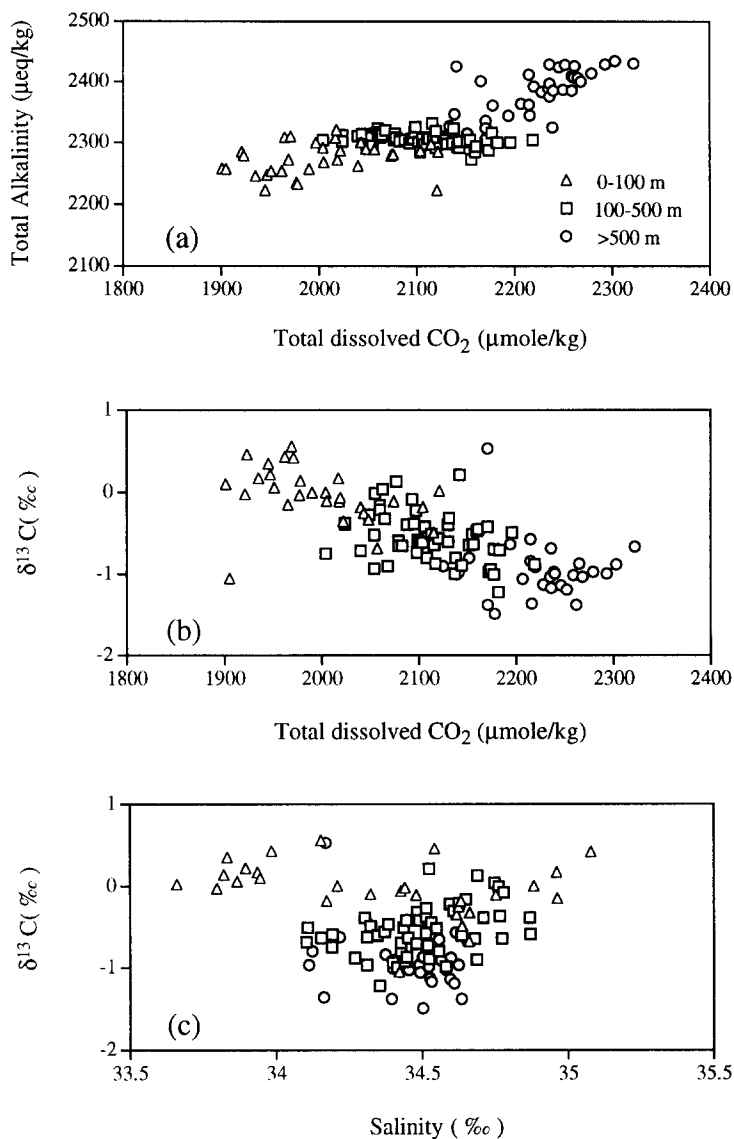
Fig. 4. Plot of total alkalinity (a) and $\delta^{13}C$ of ΣCO_2 (b) vs dissolved ΣCO_2 and $\delta^{13}C$ of ΣCO_2 vs salinity (c) in seawater off eastern Taiwan. Note that waters between 100 and 500 m are characterized by a nearly constant alkalinity with increasing CO_2 concentrations, indicating a biogenic origin of these CO_2.

the region off eastern Taiwan is practically infeasible because the emergence of subsurface high-salinity Kuroshio water from depths during the upwelling would disrupt the salinity gradient that is normally observed in the mixing of river and sea waters alone. In spite of this, a slightly gradual increase in salinity from 33.8 to 34.5 in the surface waters was observed with increasing distance from the coast (stations 1–4), indicating the input of riverine water from Taiwan island. As a consequence, although there is lack of significant
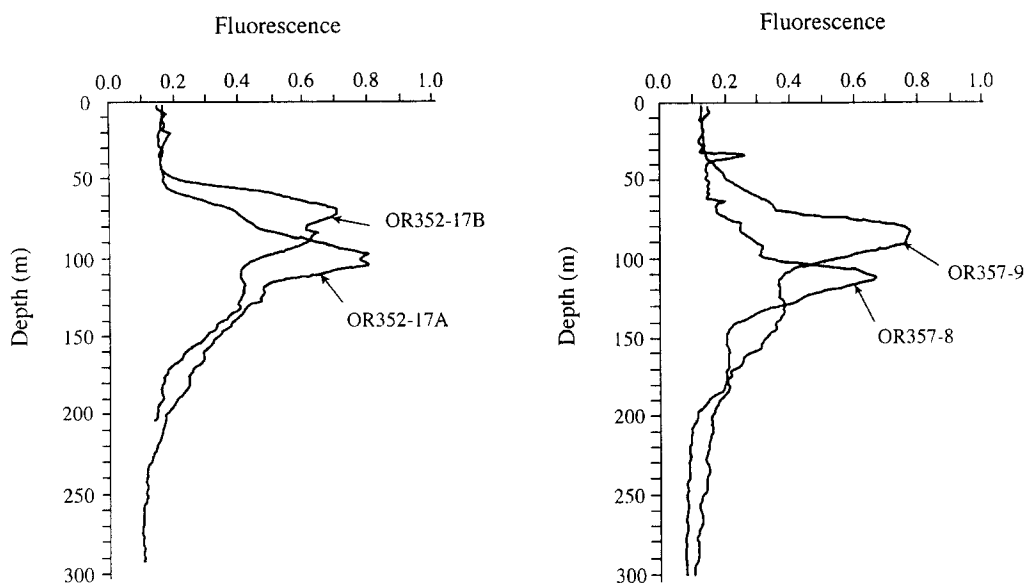
Fluorescence   Fluorescence

Fig. 5. Distributions of fluorescence in water columns off eastern Taiwan (station locations: OR352-17A and -17B, 123°10′E 25°00′N; OR357-8, 122°00′E 21°45′N; OR357-9, 122°30′E 21°45′N).

correlation between the $\delta^{13}C$ and salinity, it is conceivable that the outflow of freshwater from Taiwan island would contribute, to a lesser extent, to the lighter $\delta^{13}C$ observed near the surface.

Moreover, the isotopic compositions of dissolved ΣCO_2 in surface waters in the ocean can be influenced by the biological activity (Kroopnick, 1974a,b). Marine organisms preferentially utilize the light carbon (^{12}C) isotope and leave behind the heavy carbon (^{13}C) in the ambient water. In this way, the $\delta^{13}C$ of dissolved ΣCO_2 in seawater will become heavier. As a consequence, high productive water in the surface ocean generally shows more positive $\delta^{13}C$ values of dissolved ΣCO_2 than those surrounding waters characteristic of a low productivity. As mentioned previously, the surface water (<100 m) overlying the main path of the Kuroshio in the region off eastern Taiwan is known to be deplete in nutrients. As an indirect measure of biological productivity, values of fluorescence were recently shown by Gong et al. (1993) to be linearly correlated to the chlorophyll a in these waters. In the present study, four casts have been carried out of an in-situ fluorescence measurement in the surface 0–300 m waters at three stations (Fig. 5). As shown, the upper 50–100 m water has relatively lower fluorescence values as compared to the underlying Kuroshio water itself. The low fluorescence values measured in this study suggests a low productive surface water in the region off eastern Taiwan and thus may further contribute to the lower $\delta^{13}C$ values measured.

4. CONCLUSIONS

In short, this study presents systematic measurements of $\delta^{13}C$ of total dissolved CO_2 at various depths of seawater off eastern Taiwan, with pertinent data on alkalinity, O_2 and ΣCO_2. Results show that the low $\delta^{13}C$ values at the surface (<100 m) and the persistent

depletion of approximately 1‰ of $\delta^{13}C$ below 100 m can be largely accounted for by the emergence and mixing of the ^{13}C-depleted, subsurface Kuroshio water from depth. Additional factors that may further act as controls on the observed depletion of $\delta^{13}C$ at surface are the inflow of freshwater from Taiwan island and the prevailed low productive surface water overlying the Kuroshio in the continental margin off eastern Taiwan. Although the discussion of these processes is constrained by a lack of quantitative estimates, that such mechanisms may operate is neither surprising nor novel. Kroopnick (1985) reported a similar depletion of approximately 1% in the surface water of GEO-SECS stations 222 and 224, where the Kuroshio current flows through the western Pacific off Japan after leaving the eastern coast off Taiwan. Furthermore, it should be mentioned that, unlike in the deep ocean, where various water masses usually are well-defined and vertical settling of organic remains dominates their transports, the present data are from a very unique hydrographic regime where the upwelling and its associated transport and flux may be constantly operating throughout the water column, and organic matter would have been recycled many times before accumulation in sediments, thus making attempts to quantify the fraction of organic contributions and its subsequent changes in $\delta^{13}C$ from different water sources in this study difficult.

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