

## **Distribution of Dissolved Organic Carbon in the Continental Margin off Northern Taiwan**

JIA-JANG HUNG<sup>1</sup> and PO-LING LIN<sup>1</sup>

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### **ABSTRACT**

Analytical precision of dissolved organic carbon (DOC) in seawater using the high-temperature catalytic method (HTCO) and distributions of DOC in the continental margin off northern Taiwan were investigated. The DOC concentration of Kuroshio water off northeastern Taiwan varied from 122-136  $\mu\text{M C}$  at the surface to 65-74  $\mu\text{M C}$  at the bottom. The concentration decreased sharply downward in the upper 200 m of the water column and then decreased slightly toward the bottom. The pattern showed neither seasonal variation nor a significant difference from that observed in the water off southeastern Taiwan. The concentration of DOC was found to be inversely correlated with apparent oxygen utilization (AOU) in the Kuroshio water. The slope, however, was smaller than that predicted from the Redfield stoichiometry, indicating that only a small fraction (<25%) of AOU was derived from the oxidation of DOC. Distributions of DOC were found to vary, both temporally and spatially in the continental shelf and slope waters.

Although the distributions were relatively patchy in the inner and middle shelves, they were influenced apparently by the upwelled Kuroshio subsurface water in the outer shelf and slope regions. DOC may have been diluted in the intensive upwelling regime but enhanced on the margin of the upwelling center. Terrestrial DOC was largely confined to areas close to the continent. DOC was higher in near bottom waters, indicating that it diffused from sediments and/or was released from resuspended sediments. Thus, DOC varied non-conservatively across the continental margin through various biogeochemical processes. The horizontal gradient of surface DOC decreasing from the southern East China Sea (ECS) to the N. Pacific may reflect a similar trend of surface biological productivity and suggest the potential export of DOC out of the ECS continental margin as well.

(Key words: Dissolved organic carbon, Continental margin, Kuroshio, Northern Taiwan)

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<sup>1</sup> Institute of Marine Geology, National Sun Yat-Sen University, Kaohsiung, Taiwan, R.O.C.

## 1. INTRODUCTION

The study of the distribution of DOC in marginal seas is highly desirable because it may act as an important sink of atmospheric carbon dioxide and play an important role in determining the carbon flux from continental margins to open oceans (Toggweiler, 1989; Sharp, 1991; IGBP, 1992). However, DOC data in continental margins so far have been limited as compared to those from open oceans due to the fact that DOC in marginal seas are highly dynamic or variable. The scarcity is further impeded by the reality of "missing DOC" proposed by Sugimura and Suzuki (1988). Although this significant fraction of new-DOC has been proven to be inconclusive (Hedges *et al.*, 1993; Suzuki, 1993), the issue remains interesting and challenging to marine scientists, and a precise and accurate measurement of DOC needs to be sought. During the Seattle DOC comparison workshop, instrument blanks, especially for the HTCO method, was ascertained to be the source of disagreement in the data reported by different groups of analysts. A standard deviation of >50% from the mean value of 34 analyses on four reference natural water samples was found (Hedges *et al.*, 1993; Sharp *et al.*, 1994). It was generally agreed that a careful assessment of the blank should be made prior to DOC measurements and further interpretations.

Recently, Sharp (1993) initiated an interlaboratory comparison of DOC measurements and provided participants with samples that had been measured by his own group, so that each individual analyzer had an opportunity to see how well the blank water and reference samples were measured. In taking this opportunity, the authors were able to re-evaluate the instrument blank and compare the results with the previous data (Hung and Chang, 1992). Meanwhile, a more rigorous protocol for sample collection and preparation for DOC analyses has been applied throughout this study. The purposes of this study are (i) to improve the precision of DOC analyses, (ii) to document the spatial and temporal distributions of DOC in the KEEP study area, and (iii) to explore the influence of the mixing between the Kuroshio and ECS shelf waters upon the DOC distributions.

## 2. MATERIALS AND METHODS

This study was performed on board the *R/V Ocean Researcher I* during three cruises, OR-314 (April 16-22, 1992), OR-338 (Dec. 3-8, 1992) and OR-365 (Aug. 29-Sept. 1, 1993). The sampling stations of each cruise, depicted in Figure 1, are in the southern ECS continental margin off northern Taiwan in the KEEP study area. The measurement of DOC followed mainly the HTCO (Shimadzu TOC 5000) method (Hung and Chang, 1992) with some modifications on instrument conditions and analytical procedures in order to improve precision. The blank water used to determine the instrument blank was made from the irradiation of Q-H<sub>2</sub>O in quartz tubes with a UV lamp (550 W) for 12 h, in which 0.5% (v/v) of 30% H<sub>2</sub>O<sub>2</sub> was added to enhance carbon oxidation. This blank water (usually containing 4-6  $\mu$ M C) was first preconcentrated via freeze-drying and then determined by sealed combustion. This value was consistent with those reported by Benner and Strom (1993) and Peltzer and Brewer (1993). Before any standard calibration or sample measurement was made, a new Pt/Al<sub>2</sub>O<sub>3</sub> catalyst in the combustion tube was thoroughly conditioned with this blank water until a constant carbon value was detected (Figure 2). On the top of the catalyst, 8 g of muffled (850°C, 2 h) CuO wire was placed to improve the peak shape in order to increase the reproducibility of peak integration (Sharp *et al.*, 1994).



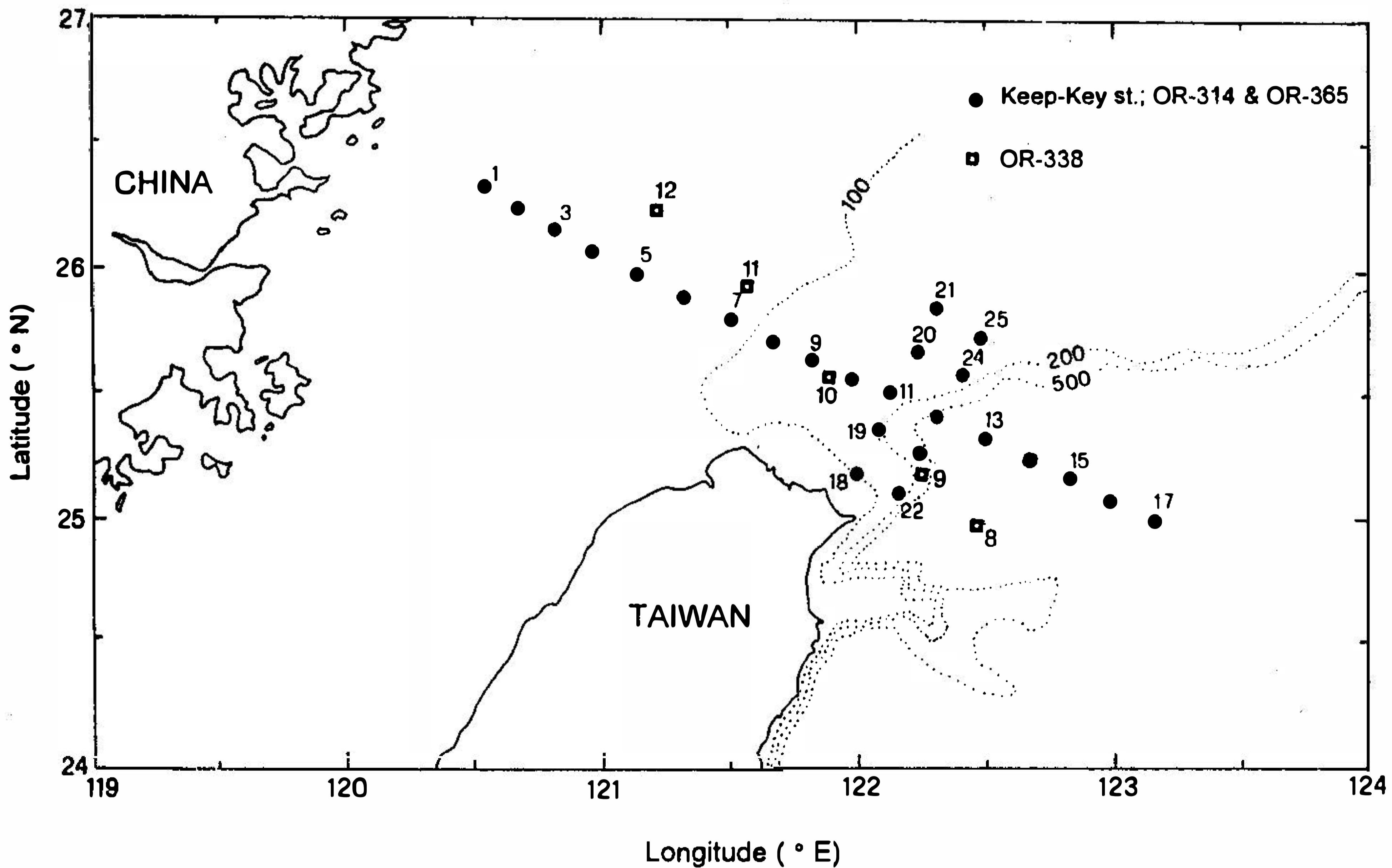


Fig. 1. The study area and sampling locations.

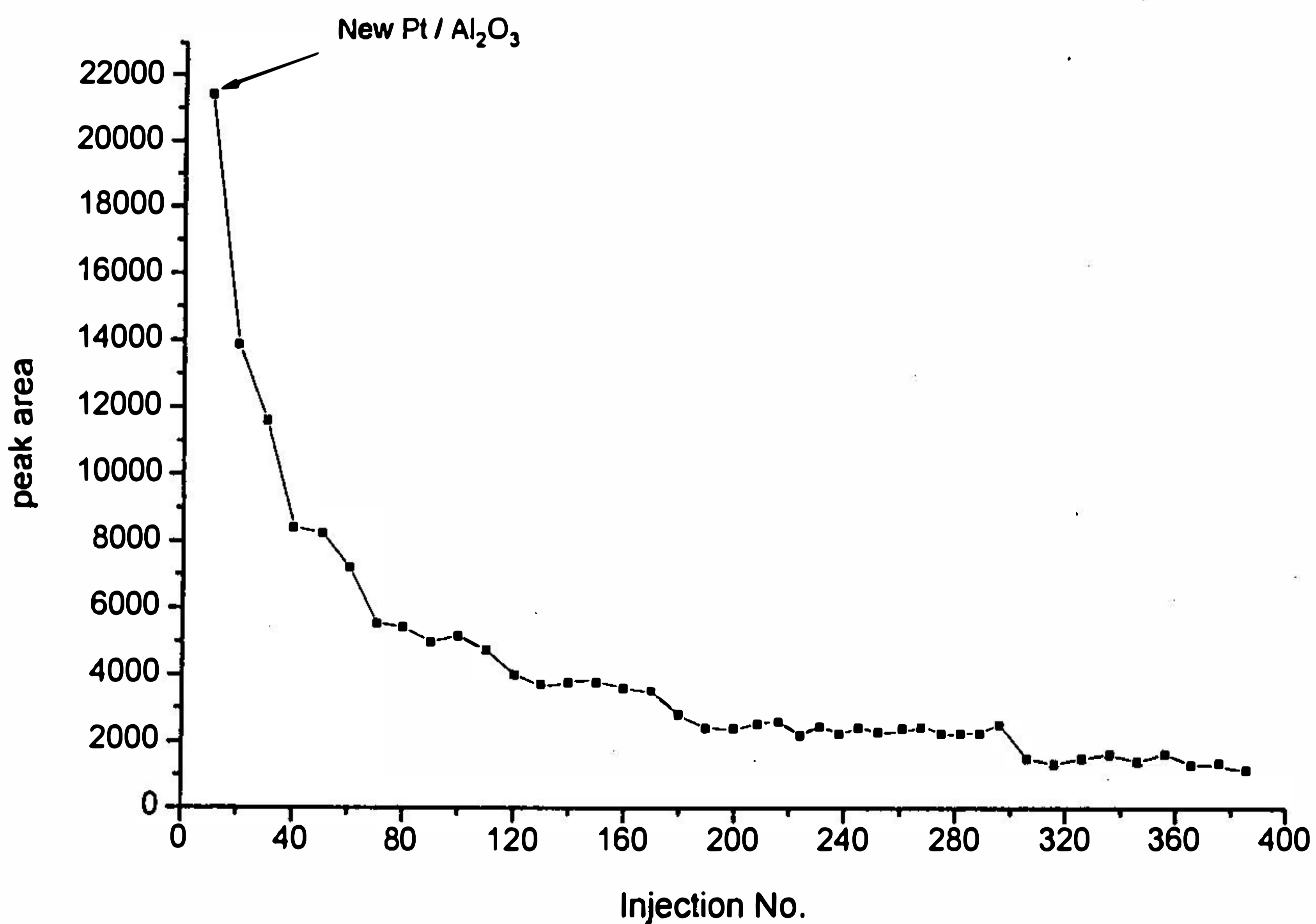


Fig. 2. The change of carbon content (peak area) during repeated injections by combusting a new catalyst (Pt/Al<sub>2</sub>O<sub>3</sub>).

Three reference samples, LCW ("zero" carbon water), POW (deep Pacific Ocean seawater) and SPOW (glucose spiked POW), provided by Jonathan Sharp at the University of Delaware, USA, were measured using the HTCO method with the instrument conditions mentioned above, and the results were reported for an interlaboratory comparison. In this study, all reference samples were analyzed in duplicate. The measurement (counts) of LCW

was used as a blank to correct for the measurements of the POW and SPOW samples. Concentrations of DOC in POW and SPOW thus measured were used to compare with those recommended values.

Seawater samples were collected with Go-flo bottles mounted on a CTD(SeaBird SBE11) /Rosette sampler. Water temperature and salinity were recorded from the CTD unit, and dissolved oxygen data (courtesy of K. K. Liu, National Taiwan Univ.) were adopted from the Chemical Oceanography Data Bank of the *R/V Ocean Researcher I* Regional Instrument Center, National Science Council. AOU was calculated from the difference between the oxygen solubility and the observed dissolved oxygen concentration. Chlorophyll *a* data in the OR-365 cruise were calculated from the fluorescence recorded with a Sea Tech fluorometer (Gong *et al.*, 1993). For the DOC analyses, one liter of each sample was drained from the Go-flo into a Pyrex glass bottle and then immediately filtered aboard through the pre-combusted (450°C, 4 h) 25-mm glass-fiber filter (Whatman GF/F), which was mounted on an ultra-cleaned, all-Pyrex-glass vacuum filtration unit. The filtration was performed using an oilless vacuum pump, and the ambient air was filtered (Gelman, 5 $\mu$ m) to remove aerosols. After the first 800 ml of filtrate was discarded, a subsequent 100 ml of filtrate was transferred to the Pyrex glass bottle which was acid-cleaned and pre-rinsed with filtered seawater. The filtered seawater was immediately acidified with 0.4 ml of subboiled HCl and then stored at 4°C. Upon return to the laboratory, the DOC in seawater was analyzed with 20 ml of acidified seawater being purged in a Pyrex glass test tube with ultrapure air for 8 min to remove dissolved CO<sub>2</sub>. The 100  $\mu$ l of de-carbonated seawater was injected into the oxidation column by means of an automatic syringe. Each sample was run at least through 5 injections, and abnormal signals were deleted to ensure that the standard deviation of repeated injections was less than 2%. The concentration of DOC in seawater was calculated according to the slope of the calibration curve, which was derived from running 1-4 ppm of potassium hydrogen phthalate in UV-irradiated natural seawater. The instrument blank was applied to correct the DOC concentrations in samples throughout the study.

### 3. RESULTS AND DISCUSSION

#### 3.1. Intercalibration

Concentrations of DOC in the interlaboratory comparison samples (POW and SPOW) were calculated according to the following equation:

$$\mu\text{M C DOC} = (\text{sample peak area} - \text{average LCW peak area})/\text{calibration slope} \quad (1)$$

The average LCW peak area is the measured signal of LCW water from the well-conditioned instrument. It actually contains the instrument blank and a small amount of DOC (3-5  $\mu$ M C) present in the LCW. The calibration slope is the least-square fitted linearity between peak areas and the standards which were prepared by dissolving potassium hydrogen phthalate (Merck) in natural seawater. This slope, however, was nearly identical to those found in the standards containing 3.5% NaCl and Q-H<sub>2</sub>O (Figure 3). DOC of the POW and SPOW samples measured by this study, therefore, was consistent with those reference data and is shown in Table 1. The signal of LCW is equivalent to 14.9  $\mu$ M C. The instrument blank is approximately 10  $\mu$ M C after the signal of LCW was corrected for residue DOC (3-5  $\mu$ M C)



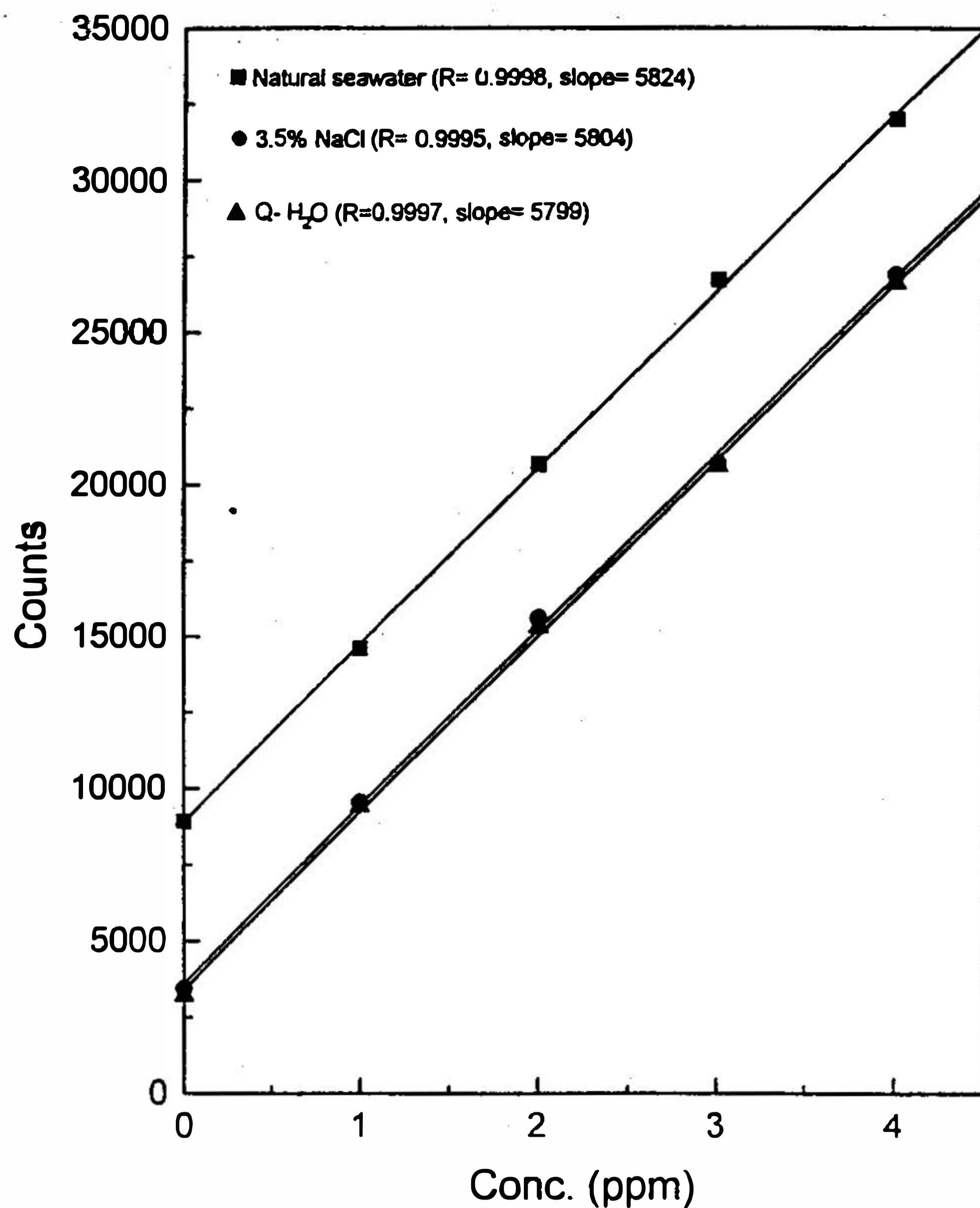


Fig. 3. The standard calibration of DOC in different metrics.

Table 1. Results of DOC measurements for intercomparison samples.

Sample	Recommended value $\mu\text{M C}$ (N = 5)*	Measured concentration $\mu\text{M C}$ (N = 2)*
LCW	3 - 5	$14.9 \pm 0.4^{\#}$
POW	$35.5 \pm 0.7$	$35.7 \pm 0.3$
SPOW	$85.3 \pm 0.4$	$81.7 \pm 1.4$

\*N: sample number

# Concentration derived from LCW (3-5  $\mu\text{M C}$ ) and instrument blank; instrument blank is approximately equal to 10  $\mu\text{M C}$ .

in the LCW. This instrument blank was lower than that (14.8  $\mu\text{M C}$ ) found previously (Hung and Chang, 1992). The relative deviations of the results in this study to the references were 0.6% and 4.2%, respectively, for the POW and SPOW samples. In spite of the possibility for a partial decomposition of spiked glucose (SPOW) during sample delivery to the laboratory at ambient temperature, the agreement between these determinations and reference data is excellent for the POW and very well for the SPOW. The error of DOC measurement in natural seawater, therefore, is estimated to be 4.2%.

Since the instrument blank may change over time, it was first checked with UV-irradiated Q-H<sub>2</sub>O ( $4 \pm 2 \mu\text{M C}$ ) prior to the routine DOC determination. The DOC concentrations in the samples were calculated according to the following equation:

$$\mu\text{M C DOC} = (\text{seawater peak area} - \text{blank H}_2\text{O peak area})/(\text{calibration slope}) + 4 \mu\text{M C} \quad (2)$$

As shown in the equation, the precise determination of the instrument blank is therefore necessary to obtain a reliable DOC measurement in seawater. For instance, the DOC analysis with a scrutinized blank correction (Sharp *et al.*, 1994) was found to be more accurate than that conducted without a rigorous blank consideration (Hedges *et al.*, 1993).

### 3.2. DOC in Kuroshio Water

The concentration of DOC in the Kuroshio water off northeastern Taiwan (Keep-Key sts. 16 & 17) during Cruise OR-314 (04/16-22/1992) and Cruise OR-365 (08/29-09/01/1993) varied from 122-136  $\mu\text{M C}$  in surface to 65-74  $\mu\text{M C}$  in bottom water (Figure 4-A). The concentration of DOC decreased sharply with depth in the upper 200m of the water column and then remained nearly constant toward the bottom. This distribution pattern and concentration range were similar to those observed in the Kuroshio water off southeastern Taiwan during Cruise OR-255 (09/1990), in which even samples had been frozen for almost two years before analyses (Figure 4-B). However, the DOC concentrations were lower than those observed in the Equatorial Pacific (145  $\mu\text{M C}$ -115  $\mu\text{M C}$ ; Martin and Fitzerwater, 1992) and the N. Pacific (150-105  $\mu\text{M C}$ ; Tanoue, 1992), but higher than those reported in the N. Pacific by Benner *et al.* (1992) (41-82  $\mu\text{M C}$ ) and Ogawa and Ogura (1992) (85  $\mu\text{M C}$ ). Although concentrations of DOC determined with the HTCO method were different in various regimes of the N. Pacific, concentrations of DOC in the Kuroshio water measured in this study fell well within the range reported in literature. The surface DOC maximum in the Kuroshio water was most likely due to the increasing activity of phytoplankton rather than the entrainment of DOC of terrestrial origin. The downward decrease in DOC in the water column, however, was mainly caused by microbial decomposition. Such a conclusion is supported by the supersaturation of oxygen (data not shown here) in the euphotic layer, and by the fact that DOC is largely recycled in the upper 200 m of water column which appears to be freshly produced by marine organisms since terrestrial DOC is known to be bio-resistant (Sharp, 1991). The distributions shown in Figures 4-A&B may further suggest that DOC in the Kuroshio water does not vary significantly in time and space as the Kuroshio passes along the eastern coast off Taiwan. DOC is correlated inversely with AOU in stations 16 & 17 ( $R=0.7286$ ,  $p<0.01$ ) of the OR-314 cruise. The DOC-AOU slope was calculated to be -0.1333 after the pairs of oxygen supersaturation in the euphotic layer were rejected (Figure 5). The slope was much smaller (in absolute value) than -0.768 predicted from the Redfield stoichiometry (106 mole DOC/138 mole DO). A similar result (DOC/AOU  $\ll$  -0.768) was also found by Kepkay and Wells (1992) and Martin and Fitzerwater (1992) although different slopes were reported. The phenomenon would, therefore, indicate that only a small fraction (<25%) of dissolved oxygen in the Kuroshio middle and deep waters was consumed by the oxidation



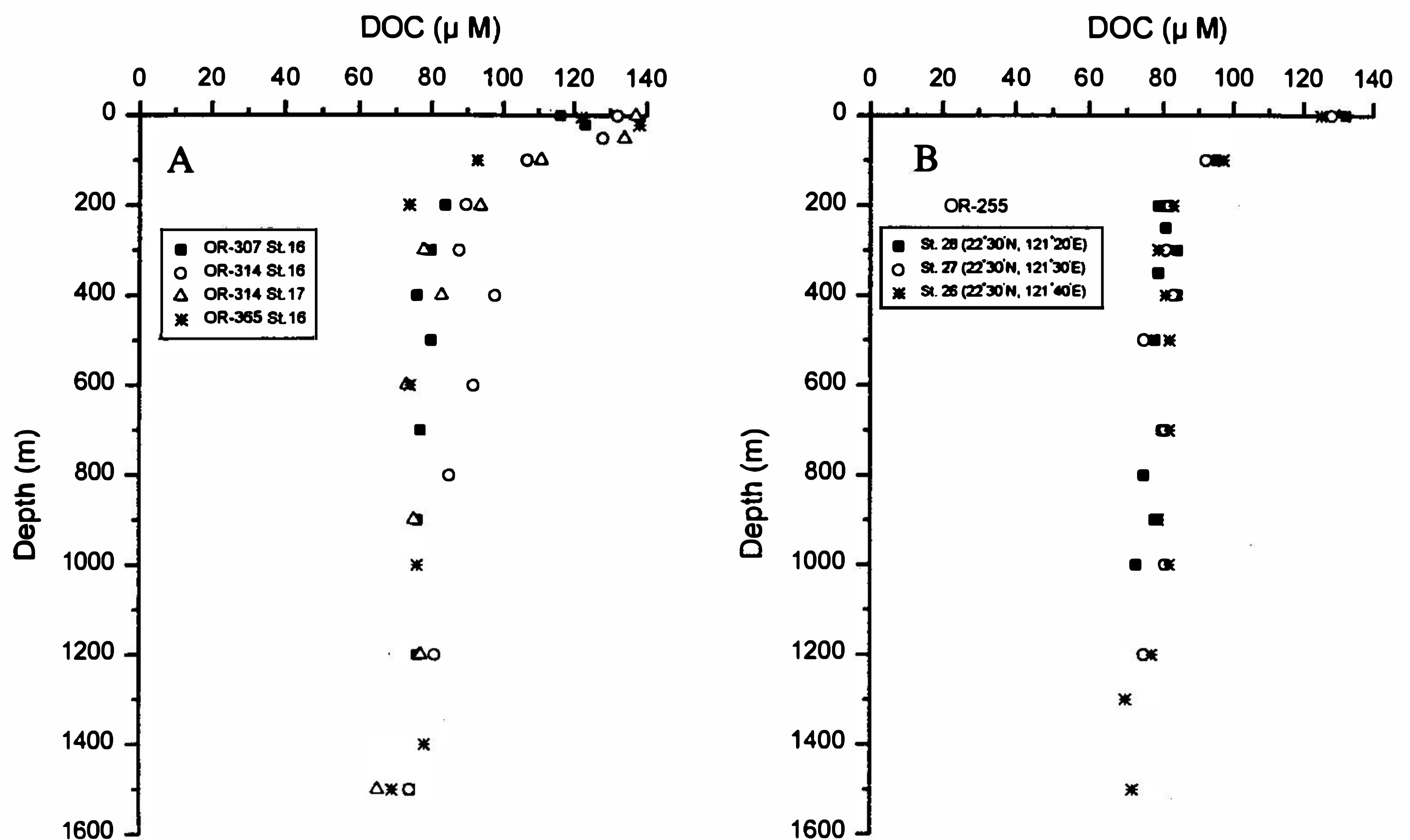


Fig. 4. (A) The distribution of DOC with depths in the Kuroshio water off north-eastern Taiwan. (B) The distribution of DOC in the Kuroshio water off southeastern Taiwan.

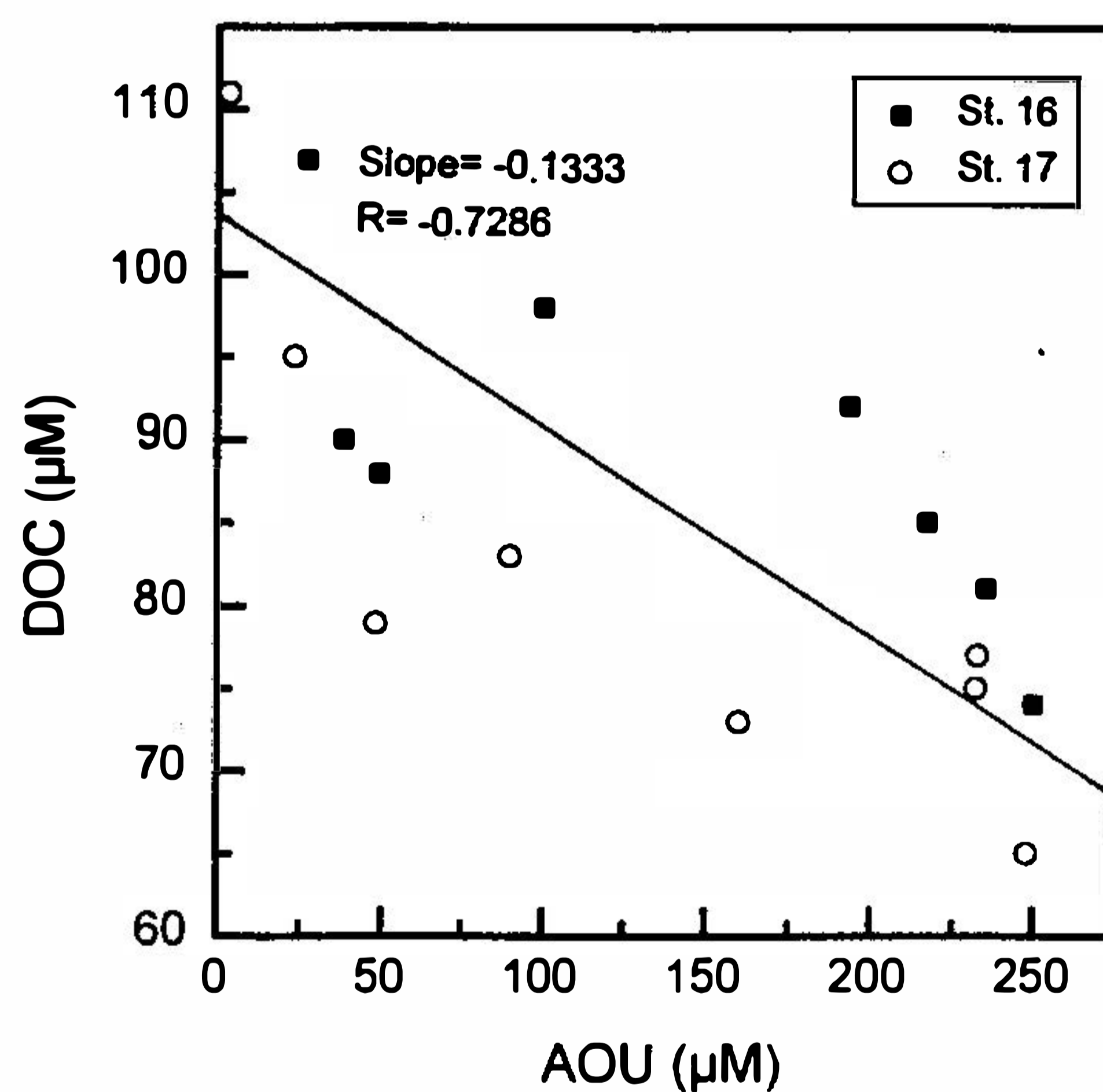


Fig. 5. The correlation between AOU and DOC in the Kuroshio water.

of the downward transport of labile DOC. In other words, more than 75% of AOU arises from the oxidation of sinking and/or laterally transported particulate organic carbon (POC).

### 3.3. Spatial and Temporal Distributions of DOC in Shelf and Slope Waters

Distributions of DOC in the southern ECS off northern Taiwan vary remarkably in time and space as seen from the results (Figures 6A-C, 8 & 9) of the OR-314 (spring), OR-338 (winter) and OR-365 (summer-autumn) cruises. The wide range of DOC concentration was

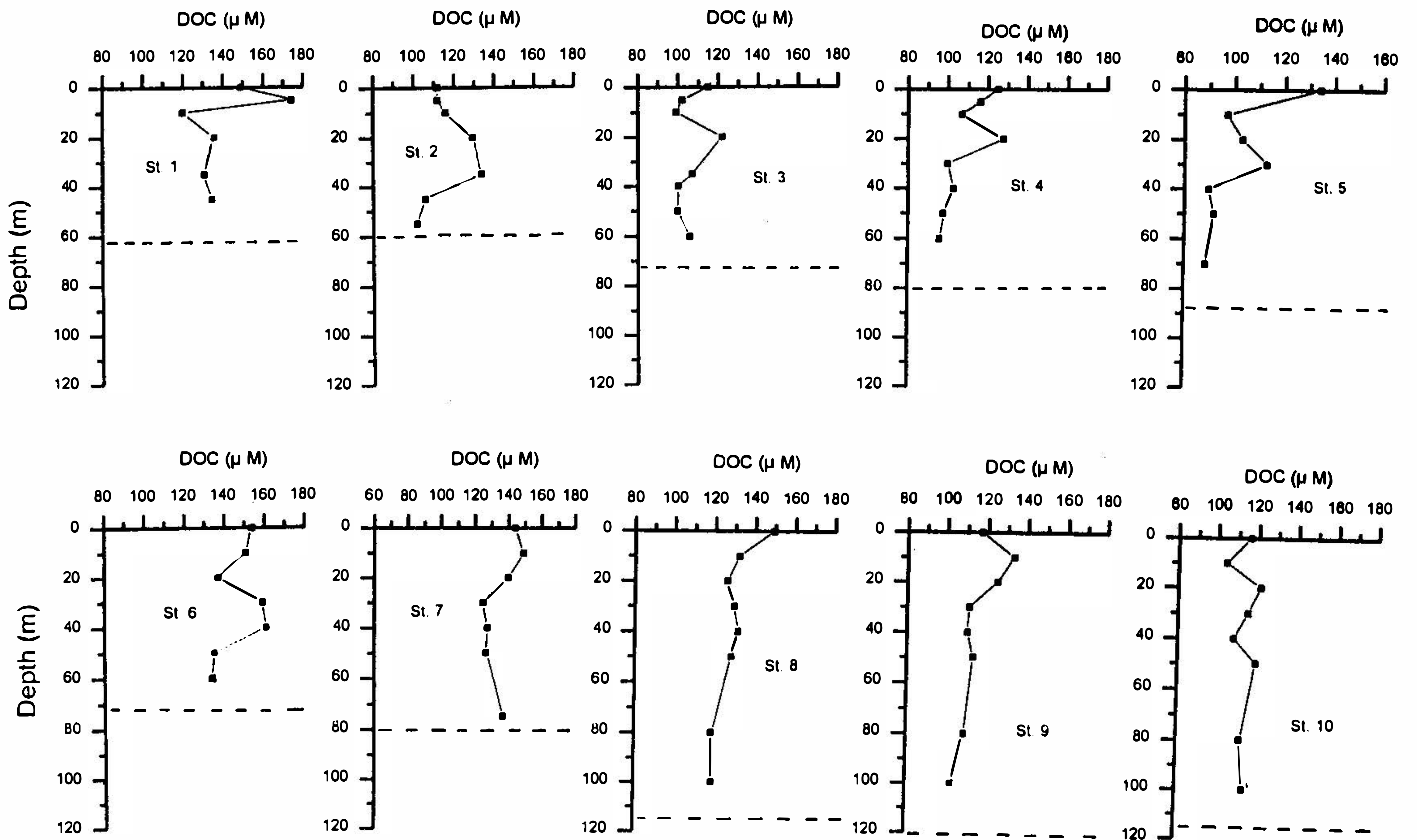


Fig. 6-A. The distribution of DOC in stations 1-10 along the main transect of the Keep-Key (the OR-314 cruise). Dash lines are bottom depths.

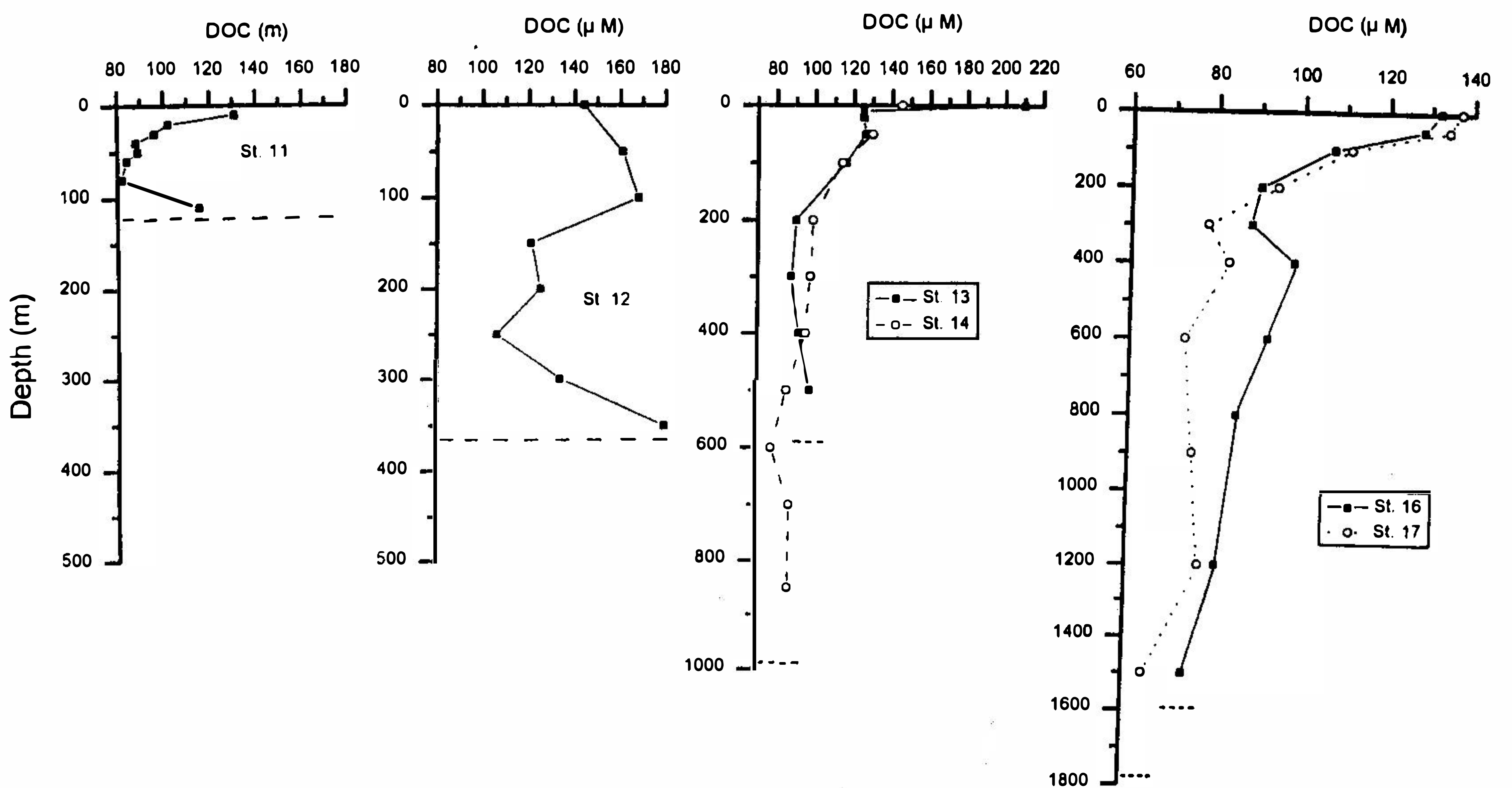


Fig. 6-B. The distribution of DOC in stations 11-17 along the main transect of the Keep-Key (the OR-314 cruise). Dash lines are bottom depths.



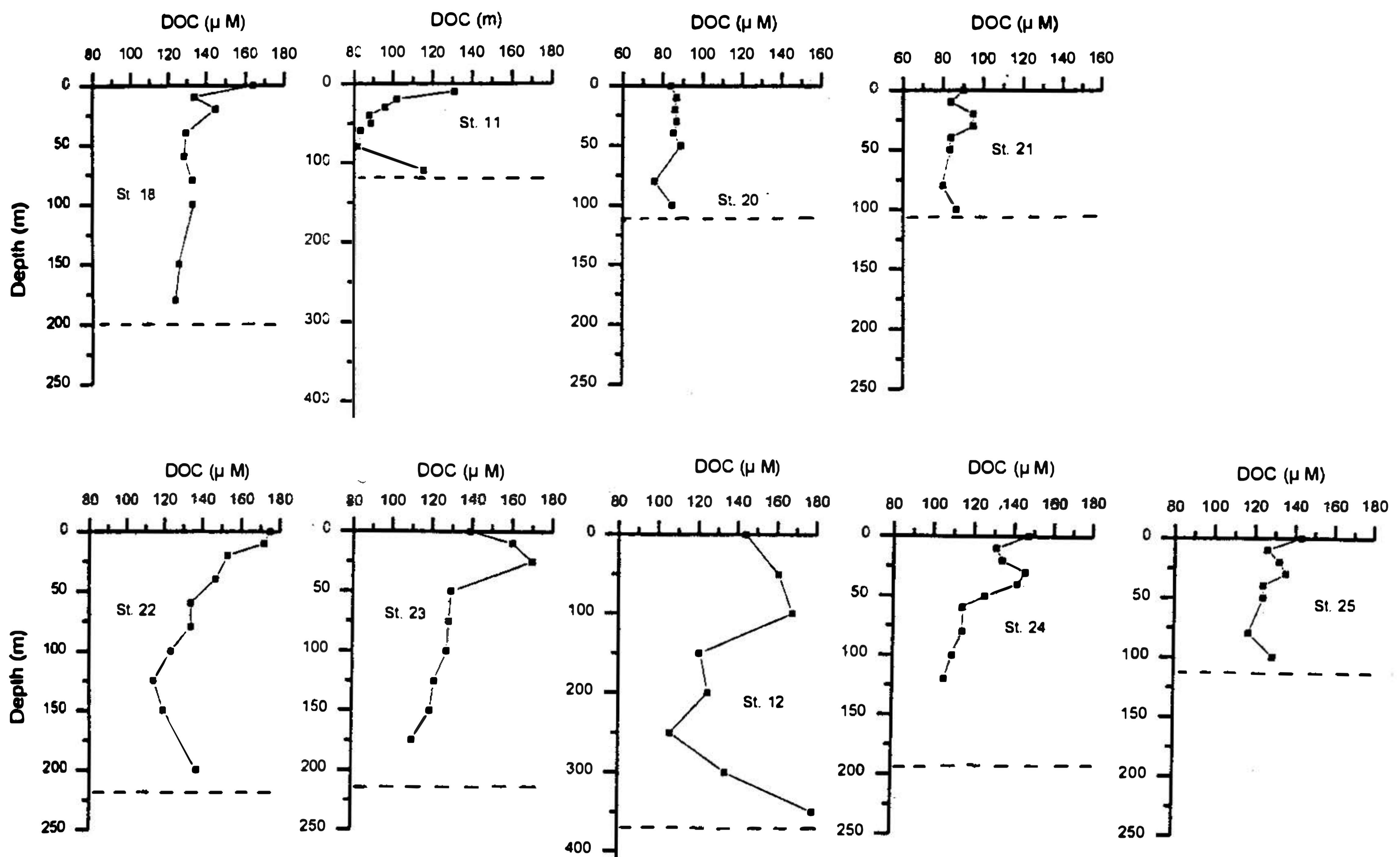


Fig. 6-C. The distribution of DOC in stations 18-25 along two short transects of the Keep-Key (the OR-314 cruise). Dash lines are bottom depths.

apparently associated with the dynamic variation of water mass in the area. The exchange phenomena between the ECS-shelf and Kuroshio waters as well as the characteristics of water masses in this area have been described elsewhere (Chern *et al.*, 1990; Hsueh *et al.*, 1992, 1993; Liu *et al.*, 1992a, b; Chuang *et al.*, 1993). In the OR-314 cruise, DOC was relatively high (120-175  $\mu\text{M C}$ ) in station 1 (Figure 6-A) which is close to mainland China. The water mass at station 1 shows relatively low temperature ( $< 18^\circ\text{C}$ ) and salinity ( $< 33$  psu) reflecting the characteristics of the Zhejian coastal water which is greatly influenced by the Changjiang runoff (Beardsley *et al.*, 1985; Gong *et al.*, 1994). The contribution of terrestrial DOC in station 1 thus must be greater than it is in other stations. Concentrations of DOC, however, were lower at stations 2, 3, 4 and 5 than at station 1 and other seaward stations, which may have resulted from the decrease in the terrestrial influence and relatively low activity of phytoplankton during the night (0320-0817 AM). A slight increase in DOC was found at stations 6-8 and 12-14 (Figure 6-B) which were located symmetrically on the flank of the upwelling center (sts. 9-11). A general consensus is that phytoplankton may have had a greater opportunity to grow on the edge of the upwelling water than at the center during the intensive upwelling. The enriched nutrients brought by the upwelling water may have further retarded the exudation of DOC by phytoplankton. More importantly, the DOC in the upwelling center may have been diluted by the DOC-depleted water that upwelled from the Kuroshio subsurface.

The distributions of DOC in two short transects normal to Taiwan Island showed a similar trend (Figure 6-C). The concentration was greatest at stations near the island and then decreased gradually with increasing distance from the coast. This may indicate that Taiwan Island can be an important source of terrestrial DOC. The DOC measured in the transect of

stations 22-25 has had slightly greater concentrations than that of stations 18-21, implying the possible edge-effect of the Kuroshio upwelling. Apparently, the spatial variation of DOC in the region was primarily governed by biological activities and upwelling, except that DOC at stations near mainland China and Taiwan Island were obviously influenced by terrestrial inputs.

In general, the maximal DOC was found at depths between 0 and 50 m. This is thought to be related to phytoplankton activity because the chlorophyll *a* maximum was also found at subsurface depths in the KEEP area (Chen, 1992). Although neither phytoplankton nor chlorophyll *a* distribution was obtained in Cruise OR-314, a statistically significant correlation was found between DOC and chlorophyll *a* ( $R=0.5371$ ,  $p<0.01$ ) in the shelf and upper layer of the slope in the OR-365 cruise (Figure 7). The greater content of chlorophyll *a* indicates more abundant phytoplankton which would have released a greater amount of DOC into the water. A high concentration of DOC was also found for samples near the bottom. The excess DOC may have diffused from the sediment and/or have been derived from resuspended sediment because a nepheloid layer was persistently observed in the Keep-Key shelf area (J. J. Hung, unpublished results).

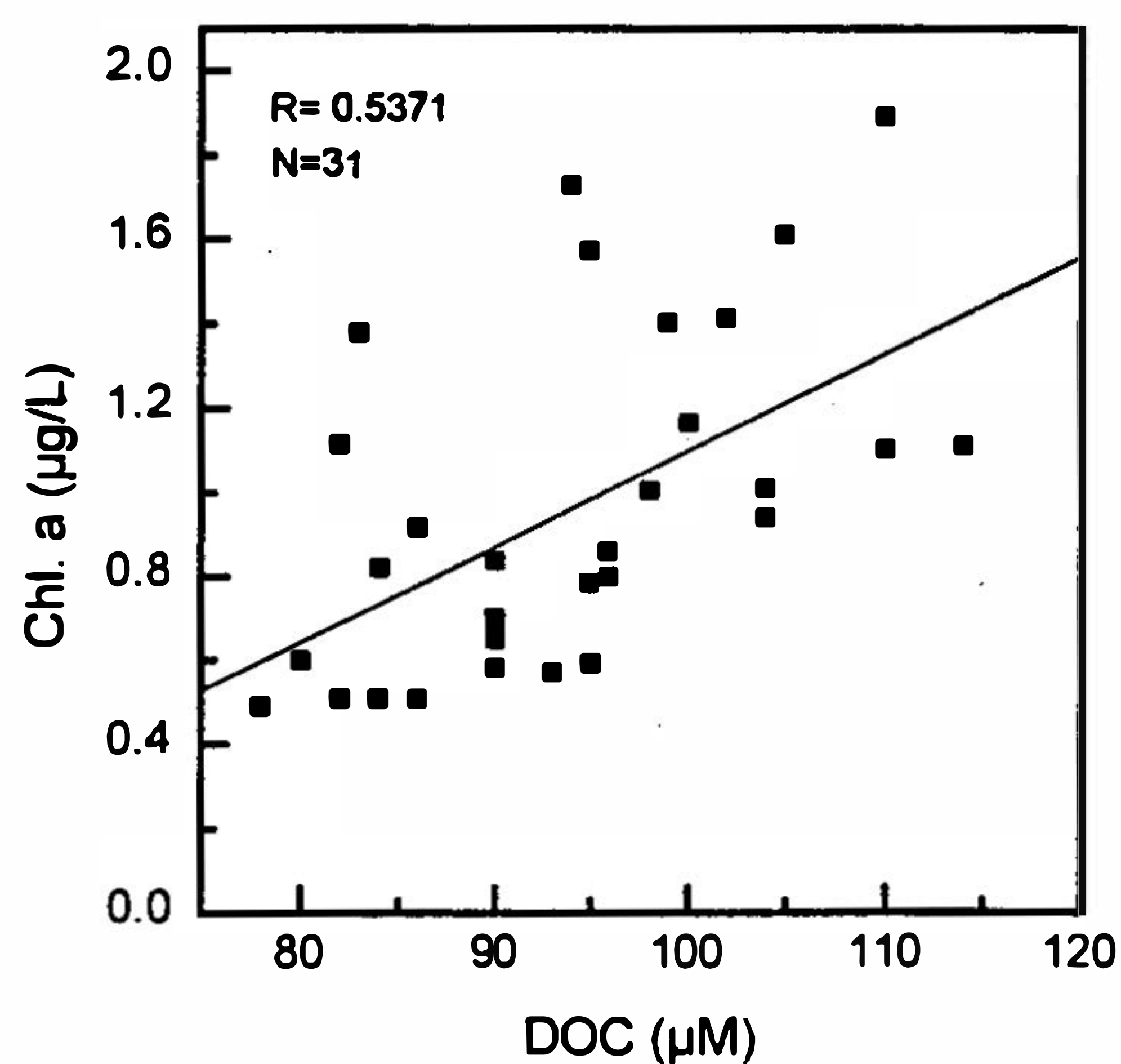


Fig. 7. The plot of DOC vs. chlorophyll *a* for shelf samples of the OR-365 cruise.

When the distribution of DOC in the spring season (the OR-314 cruise) is compared with that in the winter (the OR-338 cruise) and summer-autumn (the OR-365 cruise) seasons, the DOC in both winter (Figure 8) and summer-autumn (Figure 9) seasons was apparently lower than that in the spring season. Meanwhile, the fact is that a relatively high concentration of DOC appeared at station 1 during the OR-314 cruise but was not found during the OR-365 cruise when the influence of terrestrial input may have been minimal as revealed by salinity (34.219-34.325 psu). Since the upwelling in the KEEP area is a persistent event throughout the whole year (Liu *et al.*, 1992b), and the upwelling center was generally found around the shelf break during all the seasons of this study as judged from the distribution of temperature and nutrients (not shown here), the distributions of DOC in both winter and summer-autumn are also strongly influenced by the upwelling, and DOC is likely diluted by the upwelling water.



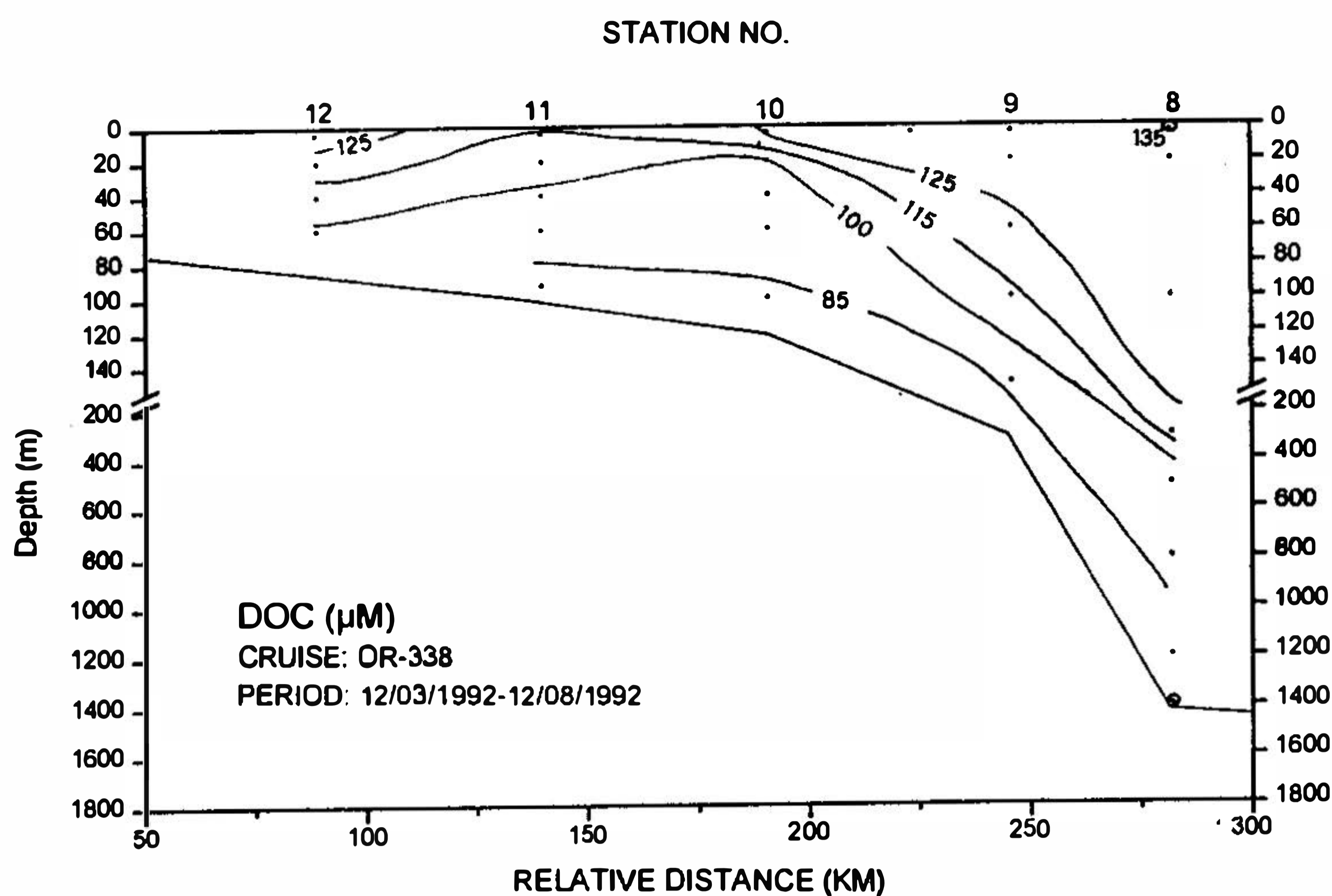


Fig. 8. The contours of DOC along the transect of the OR-338 cruise.

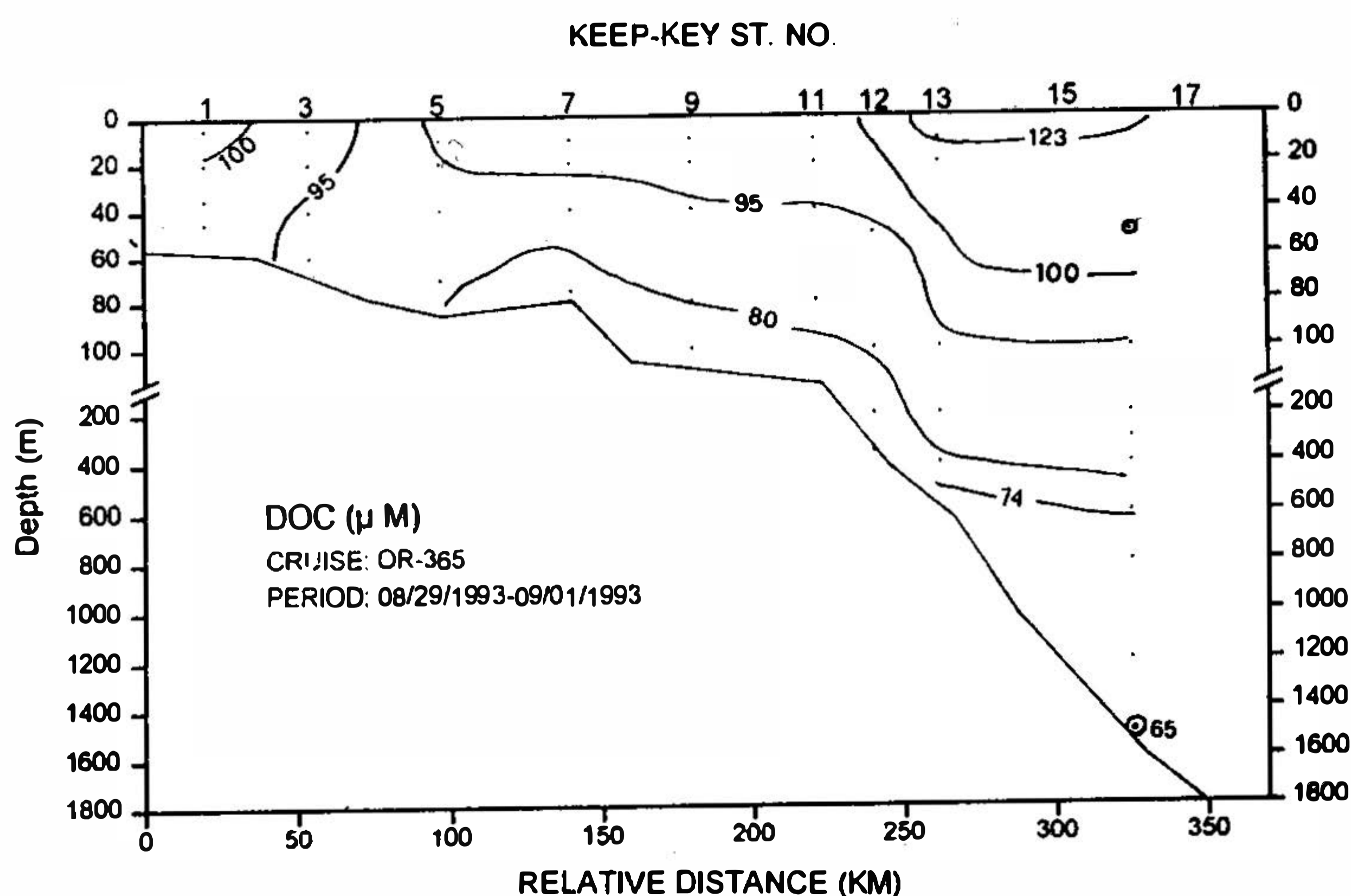


Fig. 9. The contours of DOC along the main transect of the Keep-Key during the OR-365 cruise.

Biogeochemical processes, rather than the simple physical mixing, in regulating DOC distributions in the KEEP area can be illustrated by the DOC-salinity relationship in Figure 10. Although previous studies have shown that DOC is most likely conservative in estuaries (Mantoura and Woodward, 1992; Hung and Chen, 1992) and coastal oceans (Yan *et al.*, 1991; Guo *et al.*, 1994), DOC distributions in this area are apparently non-conservative as seen from two mixing end-members: the DOC in coastal water (CW) at station 1 (average:  $149 \mu\text{M C}$ ) and that in the Kuroshio subsurface water (KSBW, ca.  $80 \mu\text{M C}$ ). A significant addition was observed especially in the surface of the shelf break zone which is strongly influenced by upwelling. This is also true even though the mixing was due to intrusion from the Kuroshio surface water (KSW) containing  $118\text{-}136 \mu\text{M DOC}$ . The DOC which behaved conservatively must have been recycled over time longer or at least equivalent to the residue

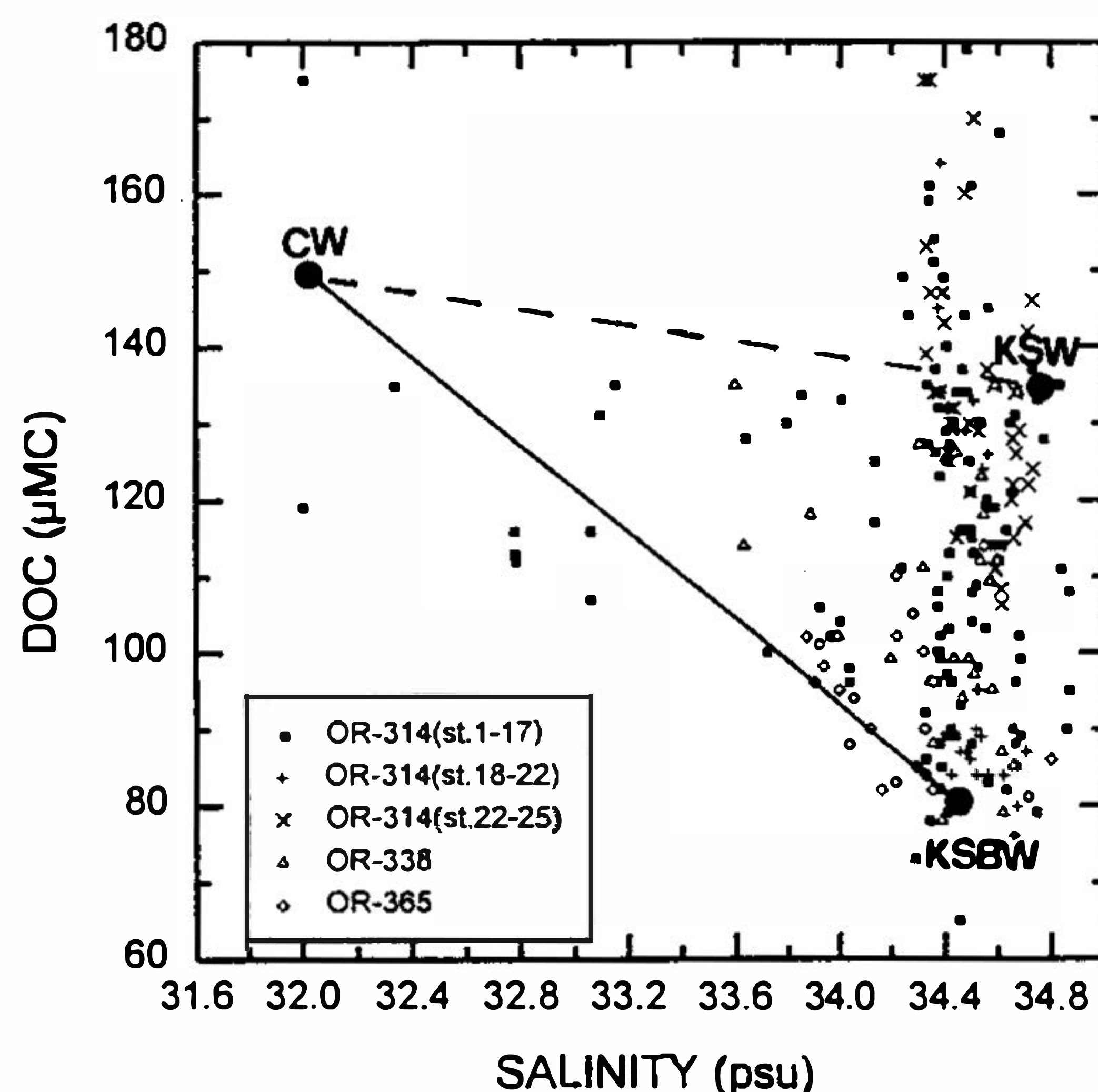


Fig. 10. The DOC-salinity plot from April 1992 to Aug.-Sept. 1993. Solid circles indicate the mean concentrations of possible mixing end-members, CW (coastal water, st.1), KSW (Kuroshio surface water) and KSBW (Kuroshio subsurface water). Solid and dash lines are conservative mixing relationships between CW and KSBW and between CW and KSW, respectively.

time of most estuarine and certain coastal waters. In contrast, in the KEEP area, DOC may have been recycled in situ and its residue time was shorter than that of water mass. The significantly recycled DOC in the surface water of the KEEP area may also indicate that DOC is relatively labile and largely biological origin. This also can be inferred from the patchy distributions of surface DOC which are likely related to biological activity. In effect, the contribution of terrestrial DOC is significant only in areas near lands as judged from the DOC distributions (Figures 6A-C). If the surface concentration of DOC in the N. Pacific open ocean is estimated to be 72-85  $\mu\text{M C}$  (Benner *et al.*, 1992; Ogawa and Ogura, 1992; Sharp, 1993), the horizontal gradient of DOC decreasing from the southern ECS continental margin to the N. Pacific open ocean may imply a similar trend for the surface biological productivity, and also suggest the potential export of DOC out of marginal ECS to the N. Pacific Ocean. Nevertheless, the contribution from terrestrial sources may not be significant. Additionally, DOC diffused from bottom sediments and/or derived from sediment resuspension can also be important in regulating DOC distributions in the shelf bottom water.

#### 4. SUMMARY

- (1) The catalysts and instrument blank of a high-temperature catalytic oxidation system should be fully conditioned and carefully examined, respectively, before a standard calibration and sample determination are performed to precisely measure DOC in natural seawater, cautions should be taken to prevent contamination during the transferring, filtering and storage of the samples.
- (2) The distribution of DOC in the Kuroshio water was relatively constant, both spatially and temporally, ranging from 122-136  $\mu\text{M C}$  in surface to 65-74  $\mu\text{M C}$  in deep water. This distribution is mainly controlled by biologically-mediated processes.



- (3) The distribution of DOC in the shelf and slope water of the Keep-Key area was variable in time and space. DOC can be largely derived from biological activity and bottom sediments and, to a less extent, affected by terrestrial inputs. Biogeochemical processes other than physical mixing were responsible for the distribution of DOC in the continental shelf and slope of the KEEP area.

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