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CHEN-TUNG A. CHEN

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CHEN-TUNG A. CHEN

Institute of Marine Geology National Sun-Yat Sen University Taiwan, Kaoshiung, Republic of China

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ABSTRACT

Concurrent CTD, salinity, O₂, nutrients, pH, calcium, total alkalinity, total CO₂ and pCO₂ were obtained in the Weddell Sea in the late austral winter of 1981 as part of the Weddell Polynya Expendition aboard the Soviet icebreaker SOMOV. These data represent the initial concentrations of chemicals in the most important source region of world ocean bottom waters at the time they were formed. These winter Weddell Sea data were analyzed along with summer data reported in the literature, and the results indicate that: large variations in chemical concentrations occur across the Polar Front; pCO₂ in the Weddell Sea surface water is only slightly lower than the atmospheric value; mixing dominates the distribution of chemical properties; pH, O₂, total CO₂ and pCO₂ are useful in identifying the sources of waters in the Weddell Sea, whereas calcium and alkalinity are not; deep waters are not homogeneous but show discontinuities; the upwelled Weddell Sea Deep Water makes up approximately 1/3 of the surface layer.

Keywords? Weddell Sea, salinity, temperature, oxygen, nitrate, pH, total alkalinity, total CO₂, PCO₂, calcium, Polynya, mixing, C-13, silicate, phosphate

I. Introduction

The three major oceans, the Atlantic, Pacific and Indian, are linked by the Southern Ocean, where free interocean circulation tends to equalize the physical and chemical characteristics of the major oceans. In essence, deep waters from the three oceans flow to the Southern Ocean and mix there. The resultant relatively homogeneous water becomes the major source of the Antarctic Bottom Water (AABW), which spreads back out into the deep world oceans.

The Weddell Sea is considered the major source of the AABW; we must know the chemical constituents of the Weddell Sea water, therefore, in order to understand their biogeochemical cycles from the global point of view. Unfortunately, few extensive geochemical sampling programs have been made in the Weddell Sea (Weiss et al., 1979). As a result, interpreting variations of the geochemical properties of many elements in the deep oceans is difficult, because we do not know the characteristic properties of the water near its origin. Furthermore, the scant geochemical data in the Weddell Sea were all collected in summer, and whether the summer data are representative of the mainly winter-formed deep waters is uncertain (Chen and Pytkowicz, 1979; Chen, 1982a; 1988; Chen et al., 1990).

This work was undertaken as part of the Weddell Polynya Expedition (WEPOLEX) in the late austral winter of 1981. The pH and total alkalinity (TA) of seawater samples were measured aboard ship and seawater samples were collected for shore-based measurements of calcium. The pH and TA data were used to calculate total CO₂ (TCO₂) and CO₂ partial pressure (pCO₂). These data are here combined with CTD, salinity, O₂, nutrients, and C-13 data reported elsewhere (Huber et al., 1983; Kroopnick, 1985) to describe water mass characteristics in the Weddell Sea and nearby regions:

II. Outline of the WEPOLEX Expedition

Concurrent pH, calcium and TA samples were collected on the research vessel, *Mikhail Somov*, of the Arctic-Antarctic Research Institute (AARI) of Leningrad, USSR. The expedition started in Montevideo, Uruguay, on 9 October 1981, reaching the ice edge near 5°E at 56°30'S on 20 October. The southernmost point was reached at 62°20'S on 3 November. The ship left the ice edge at 57°30'S near the Greenwich Meridian on 14 November and returned to Montevideo on 24 November. The station locations are shown on Fig. 1. Details of the expedition and vertical station locations are given in Chen (1984).

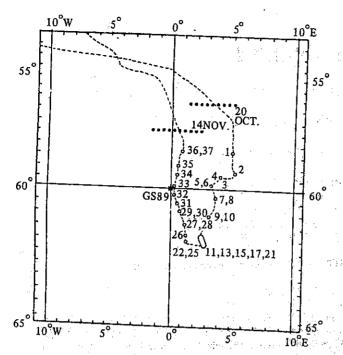


Fig. 1. Location of the WEPOLEX vertical stations. Dotted lines at 20 Oct. and 14 Nov. locate the ice edge.

III. Experimental Technique

Underway surface samples were collected with a bucket. The temperature was immediately measured to ± 0.01°C with a mercury thermometer. Salinity was measured with a Guildline Salinometer, usually within 24 hours. Vertical samples were collected with a twelve-bottle 1.7-liter Rosette sampler accompanying the CTD.

The pH samples were all determined at 25 ± 0.02°C with a combination electrode within 30 minutes. NBS 4.004 (0.05 molal KHC₈-H₄O₄) and 7.415 (0.008695 molal KH₂PO₄ and 0.03043 molal Na₂HPO₄) buffers were used to calibrate the electrode. In addition, a NBS 6.863 (0.025 molal each of KH₂PO₄ and Na₂HPO₄) buffer plus three buffers prepared by V. Fedorov of AARI, 4.01 (0.05 M KHC₈H₄O₄), 6.86 (0.025 M each of Na₂HPO₄ and KH₂PO₄) and 9.18 (0.01 M Na₂B₄O₇ H₂O), were measured and the results agree well with the prepared values (Table 1). The reproducibility of the pH measurements is better than ± 0.003 units for replicate samples. The electrode drift was determined approximately every two weeks. The largest drift was found to be 0.001 unit/day, and correction was made to the measured values.

Alkalinity was determined at $25 \pm 0.02^{\circ}$ C with a Radiometer TTT61 Digital Titrator with a reproducibility of better than $\pm 4 \mu$ eq/kg for replicate samples. Some samples were also measured using the method of Culberson et al., (1970) with similar precision. No syste-

Table 1. Standardization of the pH Electrode and the Intercalibration of the NBS and Soviet Buffers

| Buffers | Measured pH | Difference |
|------------------------------|----------------|---|
| 4.004 (NBS) | 4:004 | (standard) -0.012 0.000 0.001 (standard) 0.007 |
| 4.01 (Soviet) | 3,998 | |
| 6.86 (Soviet) | 6.860 | |
| 6.863 (NBS) | 6.864 7.415 | |
| 7.415 (NBS) 9.18 (Soviet) | | |
| | 9.187 | |

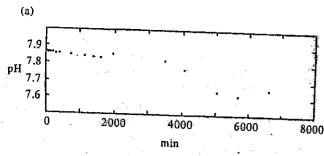
matic difference was found between these two sets of data. All samples were stored in amber plastic bottles, and the alkalinity measurements were accomplished within 12 hours after samples were aboard.

Stored calcium samples were determined using the method of Tsunogai et al. (1968) and 01son and Chen (1981). The precision of the measurements is 5μ mol/kg but the overall precision of the Ca data is $\pm 15 \mu$ mol/kg, because some samples were frozen and irreversibly altered (Chen, 1984).

IV. pH, Titration Alkalinity, Total ${\rm CO_2}$, Nutrients and ${\rm CO_2}$ Partial Pressure in the Surface Water

The values of pH (at 25°C) in surface waters obtained between 12 October and 19 October seemed to decrease as temperature decreased. As the ship moved into the pack ice, the pH values decreased sharply by roughly 0.03 units (Poisson and Chen, 1987). No diurnal pH variability was observed: the pH values of several samples collected at a small polynya (2.0°E, 60.0°S) on 27 October remained essentially constant for over thirty hours (Fig. 2). The pH value for a stored sample (stored in amber plastic bottles at room temperature) also varied little for four hours (Fig. 2), indicating low bacterial activity. The biological activity for waters below the pack ice was also found to be low (Marra and Boardman, 1984).

The surface water pH values in the ice field remained essentially constant: a total of 82 surface measurements within the pack ice yielded an average pH of 7.851 ($1\sigma = 0.009$) at an average temperature of -1.82°C ($1\sigma = 0.091$ °C). The surface pH values, however, increased sharply again by approximately 0.03 units on 14 November when the ship left the pack ice. In the open ocean the pH values also increased steadily with temperature (Poisson and Chen, 1987). The November pH values are indistinguishable from the October values south of the Polar Front, possibly because the primary



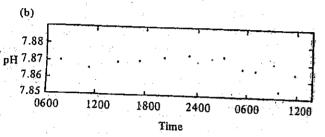


Fig. 2. (a) Variation of pH with time for a stored surface sample; (b) pH values for samples collected from the same site but at different hours of the day.

productivity observed in the eastern Weddell Sea is too low (Balech et al., 1968; Marra et al., 1982) to cause a significant change in pH in the fast moving and vertically well-mixed water. The pH values increase at a faster rate with increasing temperature in November north of the Polar Front (2°C). Perhaps the primary productivity in late October and early November was high enough, especially on and near the Argentine shelf, to decrease the total CO₂ concentration of the water, resulting in higher pH values in the vertically stratified water (Chen, 1984).

The normalized surface TA (NTA = TA x 35.0/

salinity) values seemed to increase steadily with decreasing temperature (Chen and Millero, 1979; Chen and Pytkowicz, 1979), although there seemed to be a smaller change south of the Polar Front. No significant change was detected in and out of the ice field, or between October and November (Poisson and Chen, 1987).

This author calculated the TCO₂ and pCO₂ values using the measured pH and TA values. The contributions of species such as phosphate (PO₄) and silicate (SiO₂) to the alkalinity correction were accounted for. The measured phosphate and silicate values were used when available (Huber et al., 1983); otherwise, these concentrations were calculated according to the following equations based mainly on the GEOSECS and the Islas Orcadas (Huber et al., 1981) data in the South Atlantic (Chen, 1984; Figs. 3-4).

 $NPO_4 = PO_4 \times 35.0/salinity$

= 1.7
$$\mu$$
 mol/kg $T < 4^{\circ}$ C
= 2.2 - 0.13 T/μ mol/kg 4° C $\leq T \leq 16^{\circ}$ C
= 0.1 μ mol/kg $T > 16^{\circ}$ C

 $NSiO_2 = SiO_2 \times 35.0/salinity$

= 60
$$\mu$$
 mol/kg $T < 2^{\circ}$ C = 99.4 - 19.7 $T \mu$ mol/kg 2° C $\leq T \leq 5^{\circ}$ C = 1 μ mol/kg $T > 5^{\circ}$ C

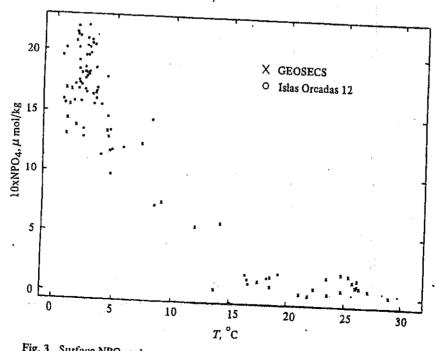


Fig. 3. Surface NPO₄ values vs. seawater temperature in the South Atlantic Ocean.

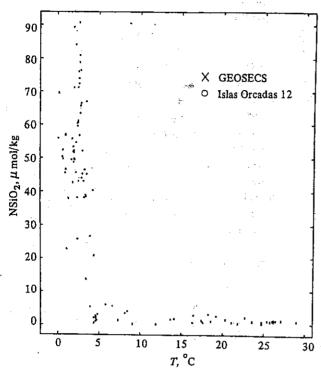


Fig. 4. Surface NSiO₂ values vs. seawater temperature in the South Atlantic Ocean.

The normalized TCO_2 values (NTCO₂ = TCO_2 x 35.0/salinity) increase almost linearly with decreasing temperature (Poisson and Chen, 1987), but at a faster

rate in November, possibly as a result of primary production in late October and early November. The general circulation around Antarctica is such that the southwards flowing deep waters upwell near the continent, bringing with them high concentrations of alkalinity, total CO2 and nutrients. A portion of the Antarctic surface water then spreads northward away from Antarctica. In the mean time, the surface water being warmed loses alkalinity, total CO2 and nutrients as a result of biological productivity. Higher temperatures may also enhance the formation of CaCO₃, either biologically or inorganically, further reducing TA and TCO2. The breaks between 2°C and 4°C in the nutrients/temperature correlations correspond roughly to the region of the Polar Front. Why pH, NTA, NTCO2 and nutrients should correlate with temperature, however, is not clear. The normalized GEOSECS nitrate values correlate with temperature as follows (Fig. 5):

 $NNO_3 = NO_3 \times 35.0/salinity$

= 23
$$\mu$$
 mol/kg
= 30.7 - 1.92 $T \mu$ mol/kg
= 0 μ mol/kg
 $T < 4^{\circ}$ C
 4° C $\leq T \leq 16^{\circ}$ C
 $T > 16^{\circ}$ C

The calculated pCO₂ in open ocean surface waters ranged from 256 to 365 μ atm. The October values seem

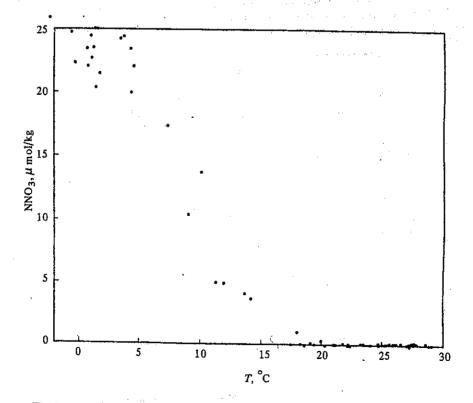


Fig. 5. Surface NNO3 values vs. seawater temperature in the South Atlantic Ocean.

to fall within 326 \pm 7 μ atm north of the Polar Front. The values are distinguishably lower (311 \pm 7 μ atm) near the Polar Front but again higher (319 \pm 8 μ atm) south of the front. The November values seem to decrease steadily with increasing temperature (from 320 μ atm near ice edge) at the rate of 1.6% per °C. The values under the ice ranged from 332 to 365 μ atm, with an average of 354 \pm 9 (10) μ atm (Chen, 1984, 1988). Theoretically, pCO₂ should increase with increasing temperature (Copin-Montegut, 1988). Our data indicate biological consumption.

Takahashi and Chipman (1982) also measured pCO2 during the WEPOLEX expedition. They reported pCO_2 between 307 and 340 μ atm in the open ocean surface waters. The surface values under ice ranged between 313 and 340 μ atm with an average of 325 \pm 7 μ atm. The CO₂ partial pressure in the atmosphere was $341.3 \pm 1.4 (1\sigma) \mu$ atm on the average, slightly greater than the seawater values in the open ocean but similar to values in the ice field. The open water values obtained during the WEPOLEX expedition in the austral winter of 1981 are generally similar to those of previous expeditions, such as the GEOSECS (Geochemical Ocean Section Studies) Atlantic Expedition in January, 1973, the GEOSECS Pacific Expedition in February - March, 1974, and the GEOSECS Indian Ocean Expedition in February, 1978. These observations indicate that the surface waters of the Antarctic Ocean are not a strong sink for atmospheric CO2, in spite of their low temperatures, and are only slightly undersaturated during the late austral winter and most likely throughout the year. Waters beneath ice can not exchange freely with the atmosphere.

V. pH in the Subsurface Waters

The vertical pH distributions (Fig. 6) generally show a pronounced minimum near the broad maximum temperature (T_{max}) and salinity layer (S_{max}) (Fig. 7). The S_{max} layer is usually only 150 m below the T_{max} . A large portion of the T_{max} and S_{max} water at a typical station such as Stn 34 comes from modified Circumpolar Deep Water (CDW). By the time the CDW signal is incorporated into the Weddell Gyre from the Circumpolar Ocean and becomes Weddell Deep Water (WDW), it is characterized by high Apparent Oxygen Utilization (AOU) but low pH values. In deeper layers, the waters are modified by the Antarctic Bottom Water (AABW), and temperature, salinity and AOU decrease with depth, whereas pH values increase monotonically (Chen, 1982c; Gordon et al., 1984, Chen, 1984).

In a few cases, such as at Stn 36/37, the core of the warm water layer (250-1000 m) is warmer and saltier than that observed at Stn 34 and represents fresh CDW input from just north of the Weddell Gyre. The AOU

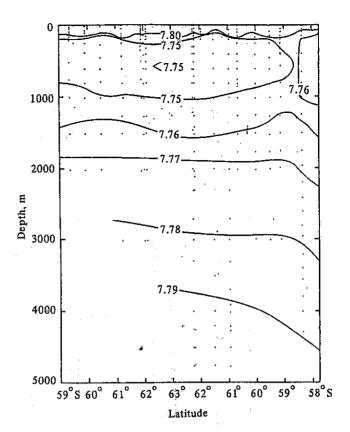


Fig. 6. Cross-section of the WEPOLEX pH data (at 25°C).

values, whose calculation was based on the oxygen solubility given in Chen (1981), are lower, and a broad maximum in pH is found below the near-surface minimum (Fig. 7). This situation suggests that not as much decomposition of organic material has occurred in the circumpolar S_{max} water at Stn 36/37 than has occurred within the Weddell Gyre, represented by Stn 34. A second pH minimum is found at a much greater depth (1200 m) at Stn 36/37, and pH again increases monotonically toward the bottom.

A composite θ/pH plot for all WEPOLEX station samples is shown in Fig. 8. Waters within the Weddell Gyre all fall on approximately the same linear θ /pH correlation line, and the ones above this line all have large fresh input from CDW. The linearity of the θ/pH relation, a mirror image of what was found for θ /AOU correlation, suggests little in situ organic carbon production or decomposition (Poisson and Chen, 1987; Chen, 1988). There is a change in slope at about 0.1° C and σ_4 of 46.06, similar to what was observed on the θ /oxygen plot (Gordon et al., 1984). A σ_4 of 46.06 may roughly correspond with a stability maximum reported by Reid et al. (1977) and Reid (1989) and is close to the upper boundary for AABW as defined by Schlemmer (1978). The waters less dense than about 46.06 and 46.07 in σ_4 have entered from the Drake Passage and the waters

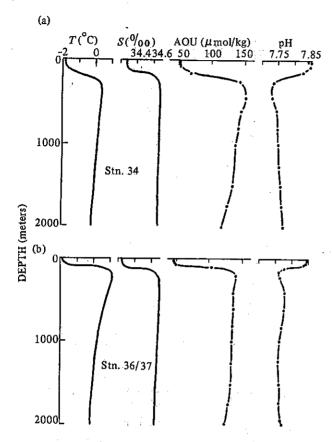


Fig. 7. (a) vertical distributions of temperature, salinity, Apparent Oxygen Utilization (AOU) and pH (25°C) for a typical WEPOLEX station, Stn 34 and (b) a non-typical station, Stn 36/37. Data below 2000 m have been omitted (taken from Chen, 1982b).

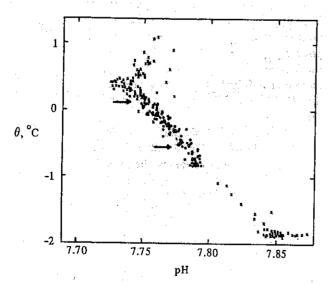


Fig. 8. Composite WEPOLEX θ data plotted vs. pH. Crosses and open circles represent, respectively, samples above and below the S_{max} layer. The arrows indicate slight changes in slope.

denser than 46.06 in σ_4 are from along the shelf of the Weddell Sea (Reid, 1989). Deep water colder than 0.1°C

possesses a slightly steeper slope, suggesting some contribution from deep waters richer in CDW. Another small break seems to be present at approximately -0.6°C and σ_4 of 46.16 with even steeper slope below. Waters denser than a σ_4 of 46.16 call for an even larger input from the CDW and yet start to show an excess CO2 signal (Chen and Rodman, 1990). Gordon and Huber (1984) reported that the warm cells of CDW are eroded quickly in a single winter season. The excess heat brought up by CDW is lost to the atmosphere through the ice, but the excess salt is not necessarily compensated for by increased fresh water introduction. As a result, the pycnocline is destablized the most with a greater than normal injection of warm CDW, hence increasing the chance of deep convection. In the mean time, the water mixes with some surface water and picks up excess CO2. This may explain why the bottom water contains a large portion of warm CDW and some excess CO2.

It has been difficult to identify water masses in the Southern Ocean, because waters are relatively homogeneous, without clearly identifiable structures. The breaks in the θ/pH correlations may be useful in defining water mass boundaries (Chen and Rodman, 1985).

It should be noted that all pH values within the linear range fall within an envelope approximately 0.014 pH units wide (Fig. 8). This spread includes both calibration and sampling errors, so the real station-to-station difference is probably small. In addition, these data clearly demonstrate that, contrary to some reports, pH can be measured rather precisely. Byrne et al. (1988) also demonstrated that pH can be measured accurately.

VI. Titration Alkalinity and Calcium in the Subsurface Waters

Both TA and calcium seem to behave conservatively, as expected (Weiss et al., 1979), because polar marine organisms are mainly siliceous, and little production or dissolution of $CaCO_3$ occurs in the Weddell Sea. The normalized TA (NTA) and calcium (NCa = $Ca \times 35.0$ /salinity) concentrations remain essentially constant (except for a very weak maximum in NTA near 0°C) and show little variation with depth or temperature (Poisson and Chen, 1987). The spread of the composite NTA values (average value 2386 μ eq/kg) is approximately 10 μ eq/kg, which includes sampling and measurement errors. The station-to-station difference, therefore, is probably also small.

Calcium measurements were made for only five stations (9, 27, 28, 36 and 37) with stored samples, some of which were frozen, unexpectedly, during transit. Calcium carbonate precipitates could have formed when the water froze, and some particles might not have redis-

solved when the ice melted. Also, some seawater might have been pushed out of the bottles when ice formed, again changing the overall composition of the sample. The composite NCa results, with an average value of 10240 μ mol/kg, have a large scatter (\pm 15 μ mol/kg vs. the precision with unfrozen samples, \pm 5 μ mol/kg). Consequently, whether our calcium measurements represent real values is questionable. It is obvious, however, that no deep water maximum in NTA and NCa is found in the Weddell Sea, as is commonly found in the low and mid-latitudes (Chen, 1982c; Chen et al., 1982). Because the NTA and NCa values remain constant throughout the water column, they cannot be used as a water tracer in the Weddell Sea (Chen, 1984; Poisson and Chen, 1987).

VII. TCO₂ and pCO₂ in the Subsurface Waters

The TCO_2 of the Wdddell seawater also seems to mix conservatively below the S_{max} layer (Poisson and Chen, 1987; Chen, 1988). The normalized TCO_2 values calculated from pH and TA data for all WEPOLEX stations below the S_{max} layer are plotted vs. θ in Fig. 9. A linear correlation is observed with a standard deviation of 6 μ mol/kg. Since the standard deviation of the least-squares fit is only slightly larger than our analytical precision of \pm 5 μ mol/kg, station-to-station variation is

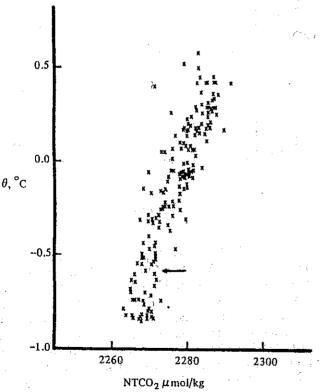


Fig. 9. θ/NTCO₂ correlation for all WEPOLEX smaples below the maximum salinity layer. The arrow indicates a slight change in slope.

minimal. A slight change in slope occurs at -0.6°C.

A linear regression of the normalized total CO2 values of three GEOSECS (January, 1973) stations in the vicinity yields a standard deviation of 15 μ mol/kg. The best fit of the GEOSECS data (Takahashi et al., 1980) yields 2286.0 μ mol/kg at 0.2°C and 16.4 μ mol/kg lower at -0.8°C. The best fit of the WEPOLEX data yields 2281.5 at 0.2°C but only 14.4 μ mol/kg lower at -0.8°C. The difference between these two sets of data is smaller than the uncertainty in the analytical procedures and, therefore, does not necessarily indicate temporal variations occurring between 1973 and 1981. More and more excess CO₂ is expected to penetrate into the AABW before it is found in the warmer water originating largely from the old CDW. It would be interesting to observe in the future whether the NTCO2 value at -0.8°C showed an increase relative to the 0.2°C water.

The θ /calculated pCO₂ correlation (Fig. 10) is a mirror image of the θ /pH correlation. There is a maximum near the S_{max} layer. Below it, the pCO₂ decreases roughly linearly with decreasing temperature, but a steeper slope is observed below -0.6°C. The measured pCO₂ values (Fig. 11; Takahashi, 1982) scatter badly because of operational difficulty during WEPOLEX. Nevertheless, the values agree reasonably with the calculated values.

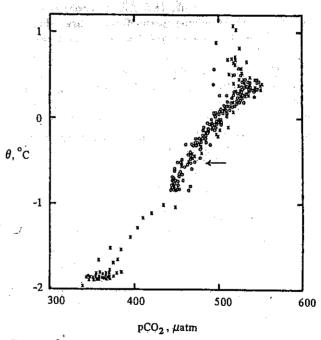


Fig. 10. θ/pCO_2 correlation for all WEPOLEX samples. Crosses and open circles denote, respectively, samples above and below the maximum salinity layer. The arrow indicates a slight change in slope.

VIII. Carbon-13 in the Subsurface Waters

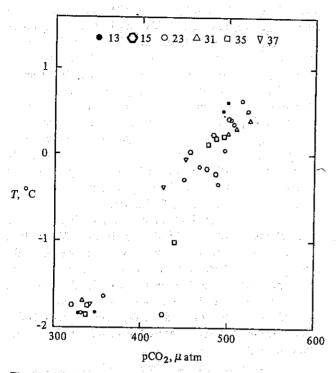


Fig. 11. T/pCO₂ correlations below the maximum salinity layer based on Takahashi's (1982) pCO₂ measurements during WEPOLEX.

The carbon-13 values (Kroopnick, 1985) again indicate conservative mixing in the Weddell Sea. For instance, two linear segments representing data above and below the S_{max} layer, respectively, are observed on the S/δ^{13} C plot (Fig. 12). This plot is similar to the S/NO plot (Broecker, 1974; Fig. 13).

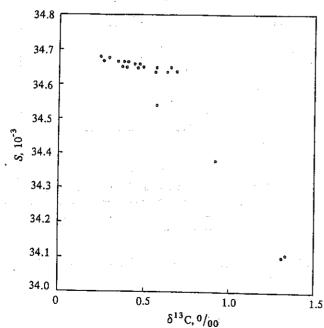


Fig. 12. Salinity vs. δ^{13} C at GEOSECS Stn 89.

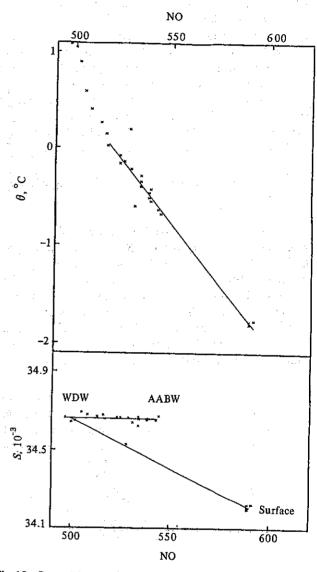


Fig. 13. Potential temperature and salinity vs. NO at WEPOLEX Stn 36/37.

The θ/δ^{13} C correlation is shown in Fig. 14. This plot does not look like the θ/NO plot (Fig. 13) but is a mirror image of the $\theta/NTCO_2$ plot (Poisson and Chen, 1987). The water with the minimum potential temperature is the remnant winter water (Chen, 1988). Assuming that this remnant winter water is a mixture of the surface water left over from summer and the upwelled WDW, this author obtained a surface/WDW mixing ratio of 63/37 based on the δ^{13} C data. This ratio agrees well with the value 68/32 based on nitrate data (Chen, 1984; Poisson and Chen, 1987), 75/25 based on oxygen data (Gordon et al., 1984) and 69/31 based on 3 He data (Schlosser et al., 1987).

XI. Conclusion

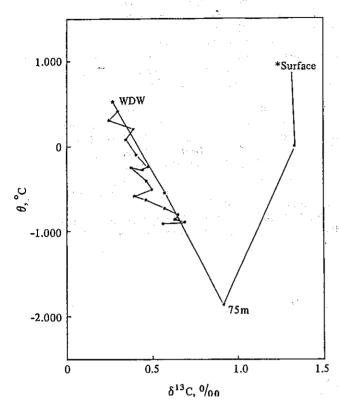


Fig. 14. Potential temperature vs. δ^{13} C at GEOSECS Stn 89.

The values of pH in surface waters of the southern South Atlantic Ocean and the Weddell Sea decrease with decreasing temperature, but NTA, NTCO₂, NNO₃, NPO₄ and NSiO₂ increase with decreasing temperature. The surface pCO₂ values remain constant in October but increase with decreasing temperature in November.

The pH and δ^{13} C correlate linearly with θ for subsurface waters with higher pH and δ^{13} C values at lower temperatures. NTCO₂ and pCO₂ also correlate linearly with θ for subsurface waters but with higher values at higher temperatures. NTA and NCa do not correlate with temperature for subsurface waters. The δ^{13} C data suggest a surface/WDW mixing ratio of 63/37.

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威得海之描述性海洋化學

陳鎭東

國立中山大學海洋地質研究所

摘 要

1981年冬天作者參加了威得海冰中湖 (polynya) 探勘研究,工作人員利用蘇聯破冰船蘇莫號 (Somov) 首度取得了電導、溫度、壓力(CTD)、鹽度、溶氧、營養鹽、pH、鈣、總鹼度、總二氧化碳及二氧化碳分壓之冬季資料。這些數據代表了在全球海洋底層水形成區於形成時之原始濃度。利用所取得冬季資料與文獻中已有之夏季資料相比得知:極區鋒面 (Polar Front)兩側之化學性質差異頗大;威得海表水之二氧化碳分壓僅較大氣中之分壓稍低;水團之混合主導化學性質,如pH,溶氧、總二氧化碳及二氧化碳分壓之分佈,而上述數據有助於研判威得海中水團之來源;鈣及總鹼度資料無法用於分辨水團;深水並非完全混合均勻,而顯現若干不連續面;湧昇至表層之威得海深層水占表層水成份之三分之一