Lysocline, Calcium Carbonate Compensation Depth, and Calcareous Sediments in the North Pacific Ocean[1]

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ABSTRACT: An extensive oceanographic investigation has been carried out in the North Pacific Ocean. The purpose of this report is to present the results of two cruises in which we participated and to report additional carbonate data from samples collected for us in the North Pacific. These data are combined with data from the literature to provide an overall picture of the carbonate system in the North Pacific.

The degree of saturation of seawater with respect to calcite and aragonite was calculated from all available data sets. Four selected cross sections, three longitudinal and one latitudinal, and two three-dimensional graphs show that a large volume of the North Pacific is undersaturated with respect to $CaCO_3$. The saturation horizon generally shows a shoaling from west to east and from south to north in the North Pacific Ocean. It was found that the lysocline is at a depth much deeper (about 2500 m deeper) than the saturation horizon of calcite, and several hundred meters shallower than the calcium carbonate compensation depth. Our results appear to support the kinetic point of view on the $CaCO_3$ dissolution mechanisms. Differences in the abundance of the calcareous sediments are explained by differences in the calcium carbonate compensation depth.

THE BEHAVIOR OF THE carbonate system in seawater is one of the most complex topics in oceanography. The system has long interested many oceanographers from various fields because it plays an important role in all three subspheres of the earth (biosphere, lithosphere, and hydrosphere).

An extensive oceanographic investigation has been carried out in the North Pacific Ocean. The purpose of this paper is to present the results of two cruises in which we participated (Chen 1982, Chen et al. 1986) and to report additional carbonate data from samples collected for us in the North Pacific. These data are combined with data from the literature to provide an overall picture of the carbonate system in the North Pacific. Details on data collection are presented elsewhere (Chen et al. 1986) and are not repeated here.

DEGREE OF SATURATION OF $CaCO_3$, LYSOCLINE, AND CALCIUM CARBONATE COMPENSATION DEPTH

It is well known that deep waters of the North Pacific Ocean are the oldest in the world oceans. Hence, the concentration of carbonate ion in the North Pacific Ocean shows the lowest value. Takahashi et al. (1981) found a low mean CO_3^{2-} concentration in the Pacific Ocean and attributed it to the low alkalinity/total CO_2 ratio of this area. The result of low CO_3^{2-} concentration is a lower degree of saturation and shallower calcite and aragonite saturation horizons in the Pacific Ocean than in other oceanic regions.

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The distribution of the degree of saturation of seawater with respect to calcite and aragonite is presented below. The relationships between the saturation state and the calcium carbonate compensation depth and the lysocline are also addressed.

METHOD OF CALCULATING THE DEGREE OF SATURATION

In calculating the degree of saturation of seawater with respect to calcite and aragonite, we need to calculate the carbonate ion concentration from the measured carbonate data. The CO_3^{2-} concentration in seawater at in situ temperature and pressure conditions has been computed from the NOAA Eastern North Pacific CO_2 Dynamics Cruise, 1981 (ENP), and Western North Pacific CO_2 Dynamics Cruise, 1982 (WNP), salinity, temperature, alkalinity, pH, and calcium data. In addition to our two meridional sections along 165° E and 150° W, all GEOSECS (Geochemical Ocean Section Studies, 1973–1974[GS]) and INDOPAC (Scripps Institute of Oceanography, 1976 [IP]) alkalinity and total CO_2 data from the North Pacific Ocean are used to calculate the degree of saturation of seawater. Two cross sections, one longitudinal along roughly 180° W and one latitudinal along roughly 35° N, are selected from GEOSECS stations to demonstrate the distribution of saturation values. The effects of pressure on the dissociation constants for carbonic and boric acids determined by Culberson et al. (1967) and Culberson (1972) are used for the computation. Our expression for the saturation states for calcite and aragonite in seawater is in percent of saturation:

$$\Omega = \frac{ICP}{K'_{sp}} \times 100\%$$

where ICP (ion concentration product) = $[Ca^{2+}] \times [CO_3^{2-}]$ $(M/kg)^2$ and K'_{sp} = apparent solubility product for calcite or aragonite. The apparent solubility products for calcite and aragonite in seawater at 1 atm total pressure determined by Ingle et al. (1973) and by Berner (1976), respectively, are used. In order to obtain the apparent solubility product at in situ conditions, the pressure effects on the solubility products summarized by Culberson (1972) are used.

DEGREE OF SATURATION OF THE SURFACE WATER

Figures 1a and 1b show the correlation of temperature with the degree of saturation of seawater with respect to calcite (Ω_c) and aragonite (Ω_a), respectively, for the surface waters of the North Pacific Ocean. It is evident that all surface waters are supersaturated with $CaCO_3$ (Alekin and Katunin 1973). Linear correlations between Ω_c, Ω_a, and temperature are found in the data sets for both figures. A high degree of saturation is observed in high-temperature Pacific surface seawater (Lyakhin 1968) for calcium carbonate. The degree of saturation is strongly influenced by the titration alkalinity (TA) versus total CO_2 (TCO_2) ratio, which also shows a linear dependence on surface temperature as suggested by Feely et al. (1984).

The isopleths of the surface saturation values are shown in Figures 2a and 2b for calcite and aragonite, respectively. The overall pattern is similar to the temperature distribution, which in turn is influenced by the surface oceanographic circulation and other factors. The Ω_c and Ω_a decrease slightly from west to east. In the western subarctic regions, low Ω_c and Ω_a are also found in the cold Gulf of Alaska. A sharp change occurs from north to south at the region of mixing between the cold Oyashio and the warm Kuroshio around 40° N. A poorly defined area of high Ω_c and Ω_a values occurs near the center of the subtropical gyre west of Hawaii.

Figures 2a and 2b represent mainly summer conditions, and the contours are likely to move southward in winter as surface temperature decreases. For instance, Lyakhin (1970) reported 300–400% saturation of calcite in the Sea of Okhotsk in summer but less than 150% saturation in winter.
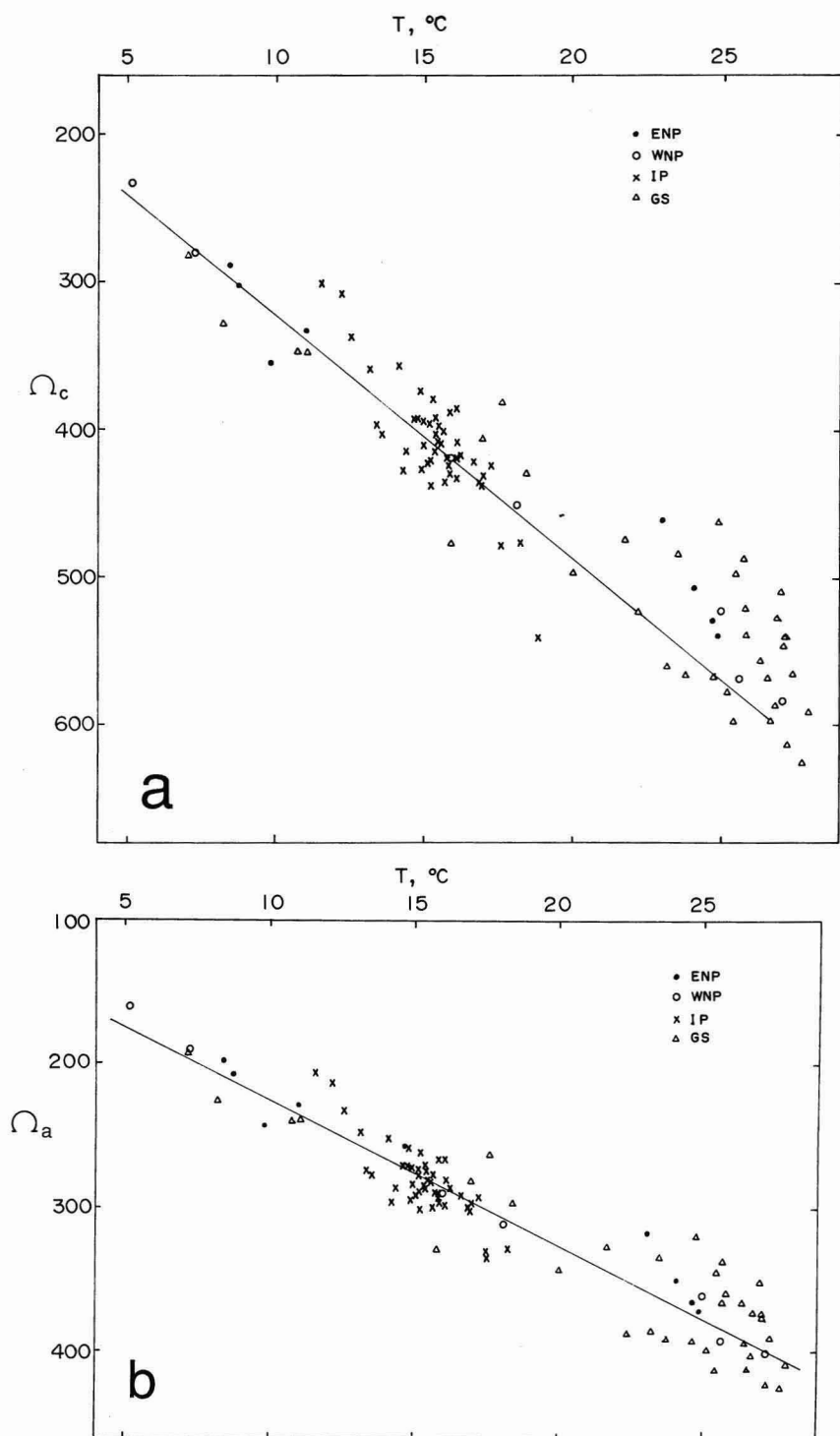
FIGURE 1. Correlation of surface (a) Ω_c and (b) Ω_a with temperature in the North Pacific. The lines are rough fits by eye.
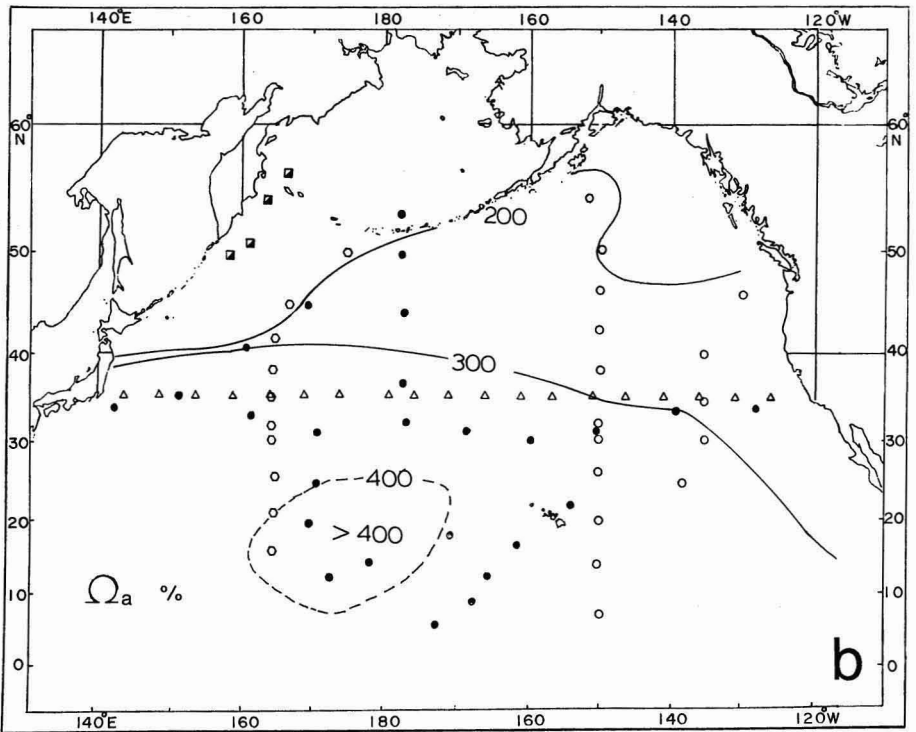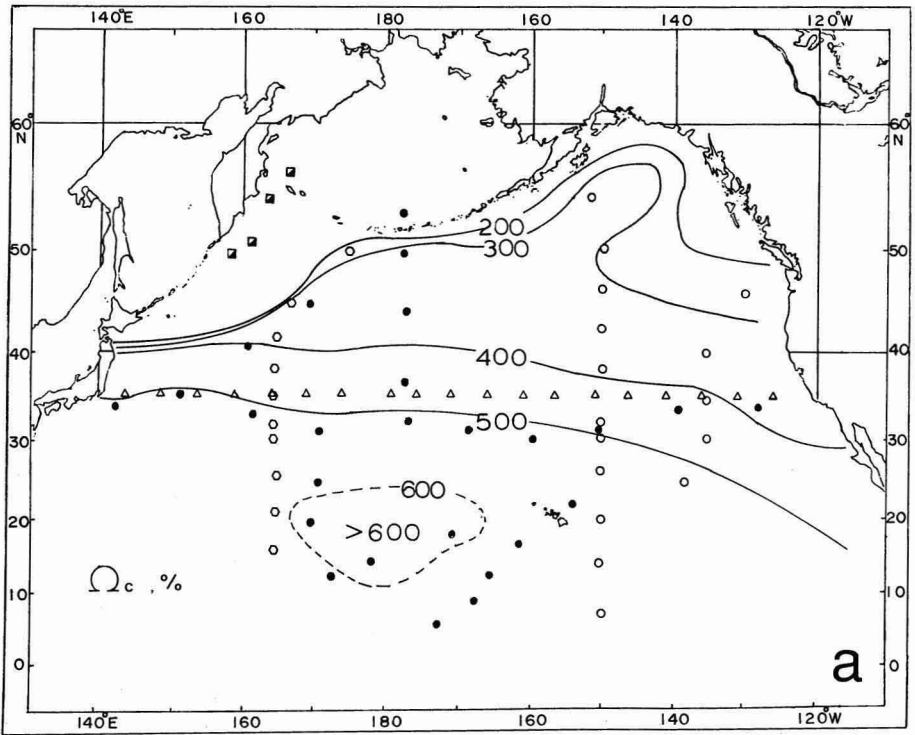
FIGURE 2. Distribution of surface (*a*) Ω_c and (*b*) Ω_a in the North Pacific Ocean. Areas enclosed by dashed circles probably have the highest degree of supersaturation.

VERTICAL DISTRIBUTION OF DEGREE OF SATURATION

Four cross sections of saturation values of calcite and aragonite are plotted from the ENP, WNP, and GEOSECS data.

1. The 35° N Cross Section

The latitudinal cross section along approx. 35° N was selected also by Takahashi (1975), who used TCO_2 data unadjusted for the fossil fuel CO_2 input to calculate saturation values; and only Ω_c was calculated in his paper. The revised carbonate data (Takahashi et al. 1980) are used to recalculate the degree of saturation of both calcite and aragonite. The results are shown in Figures 3a and 3b. A very small variation in the degree of saturation is found in the water column below 1000 m depth. The saturation horizons shoal from west to east because of the general surface circulation pattern in the North Pacific Ocean. The western North Pacific has the deepest saturation horizon at a depth of 1100 m. The intensified western boundary current results in the deepening of the saturation horizon. Characteristic minimum Ω_c and Ω_a layers can be seen across the entire ocean from coast to coast. These minimum layers are relatively narrow and are not shown in Takahashi's (1975) profile. These layers are strongly related to the region of highest partial CO_2 pressure (P_{CO_2}) concentrations in the oxygen minimum layer (Figure 4). The low saturation values found in this layer are caused by oxidation of organic matter, releasing carbon dioxide and reducing the carbonate ion concentration.

2. The 165° E Cross Section

Figure 5a shows the distribution of the saturation values of calcite along 165° E. The saturation horizon deepens from less than 150 m in the north to >3000 m at 25° N. Figure 5b shows the depth of the aragonite saturation horizon at less than 100 m in the north, at 750 m at 30° N, and at 500 m at 15° N. Byrne et al. (1984) studied the dissolution rate of aragonite particulates collected using free-drifting sediment traps during the

Discoverer cruise (Betzer et al. 1984). Their findings revealed that the depth at which a high dissolution rate of aragonite was found is shallower at high latitudes than at midlatitudes. These findings support our saturation profile, which shows a concave structure at mid-latitudes. A similar structure was reported by Betzer et al. (1984).

A sharp change from undersaturation to supersaturation for calcite is found between stations WNP5 and WNP6. The oversaturated water at WNP3 and WNP5 may be the result of the younger age of the seawater in that region. Station WNP6 defined the southern limit of North Pacific deep water that has low oxygen and high nutrients. Such a sharp change of the saturation value from north to south also was observed by Hawley and Pytkowicz (1969) along their 170° W cross section. We do not have sufficiently deep samples at WNP3 and WNP5 to see the distribution of saturation values down to the bottom. A core of low saturation is found as a tongue extending from north to south at a water depth of 700 m.

The saturation horizon also is affected by the surface circulation. A depression of the saturation surface at 30° N is located slightly south of the subtropical convergence zone. The 100% saturation horizon almost reaches a depth of 100 m at 50° N. Again, upwelling must play an important role in governing the depth of this shallow saturation horizon.

Similar results were reported by Feely et al. (1984) for waters above 1000 m.

3. The 180° W Cross Section

Takahashi (1975) selected similar but not identical cross sections. Only the calcite saturation was calculated from the original GEOSECS data set in his paper. As in the 165° E cross section, the contour lines in the surface layer exhibit a concave, downward structure at mid-latitudes (Figures 6a, 6b). The 100% Ω_c contour and 70% Ω_a contour deepen from 300 m at the north to 2200 m at the south. Such a trend may depict the large-scale upwelling phenomenon of the deep water of the northern region. The cores of low saturation values are observed in both Figures
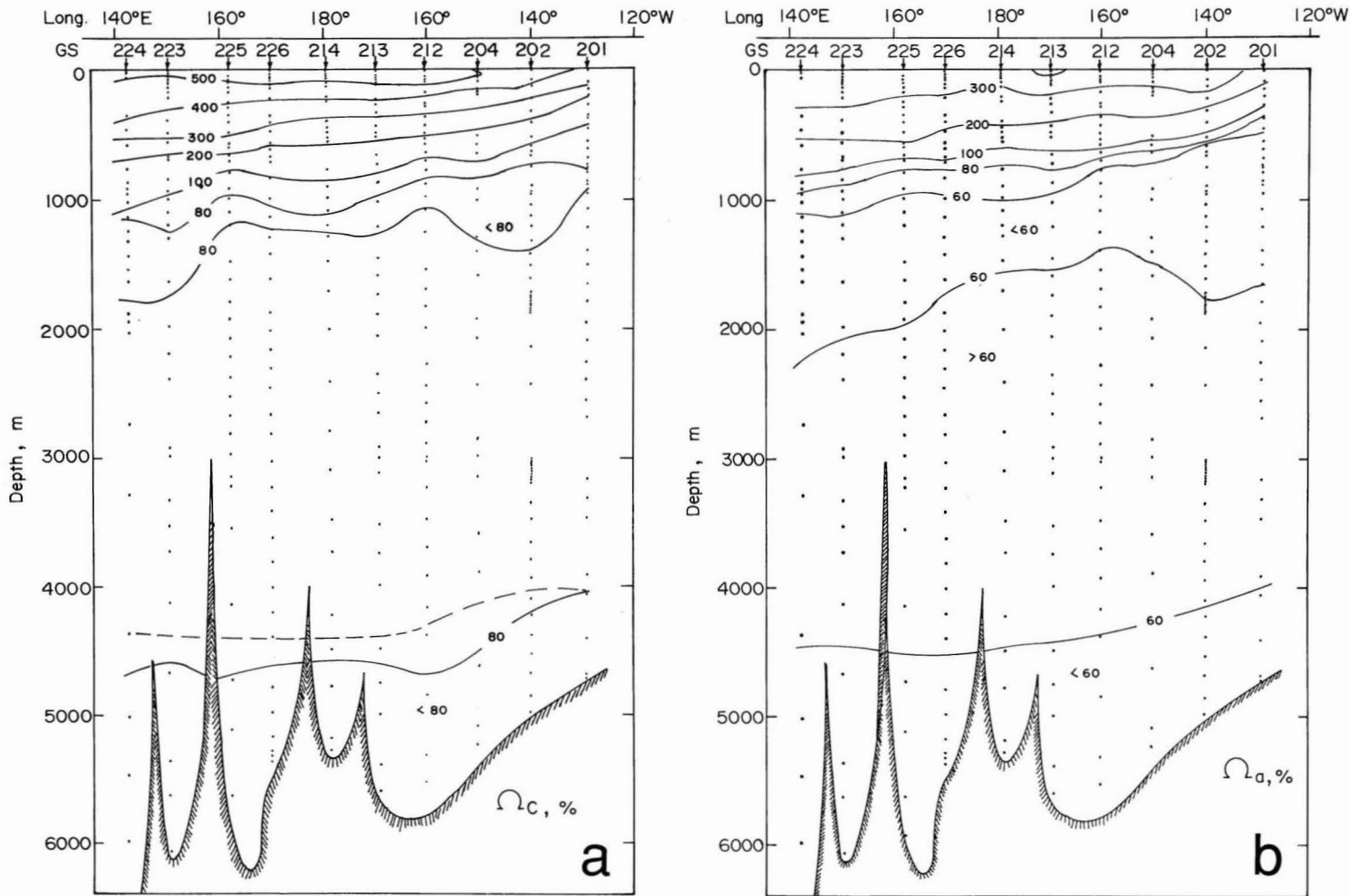
FIGURE 3. Cross section of (a) Ω_c and (b) Ω_a along 35° N. Dashed line represents calcium carbonate compensation depth data (CCD) from Berger et al. (1976).
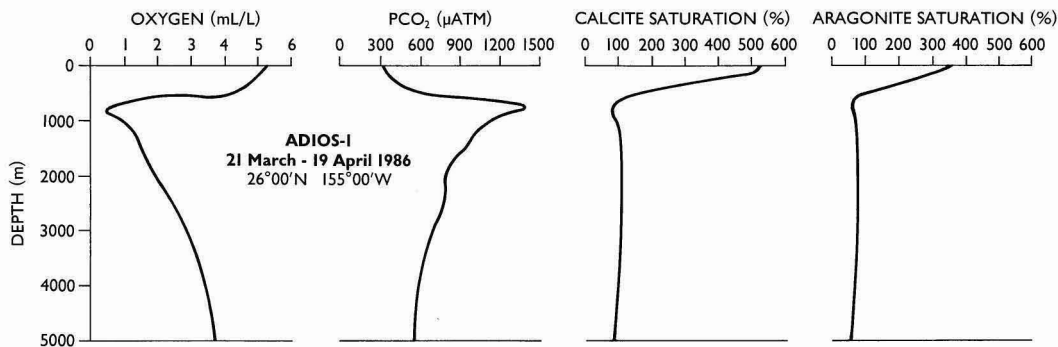
FIGURE 4. Vertical profiles of dissolved oxygen (ml/liter), PCO_2 (μatm), calcite saturation (%), and aragonite saturation (%) at the ADIOS-I (Asian Dust Input to Ocean System-I time series station (26°00′ N, 155°00′ W). The data are a composite of nine vertical profiles taken from 21 March through 19 April 1986. The minima in calcite and aragonite saturation correlate with the maximum in PCO_2.

6a and 6b. Takahashi (1975) did not show this minimum core in his paper.

Similar results for aragonite were reported for waters above 1000 m by Feely et al. (1984).

4. The 150° W Cross Section

This data set was presented by Feely and Chen (1982), who used the estimated calcium concentration to calculate the ionic product for the data above 1500 m. Figures 7a and 7b show our results of recalculated saturation values using measured calcium concentrations for all the samples taken. The distribution pattern is similar to the cross section along 165° E.

Similar results were given by Feely and Chen (1982) for waters above 1.5 km.

5. Subsurface Distribution of the 100% Saturation Horizons for Ω_a and Ω_c

Figures 8a and 8b give three-dimensional representations of the 100% saturation horizons for Ω_c and Ω_a, respectively. The data are based on the NOAA ENP, CNP (Central North Pacific Dynamics Cruise, 1983), and SOCM (Summer Ocean Carbon Monitoring) data sets (Feely, unpublished data), as well as the GEOSECS and INDOPAC data. Interpolation is done by spline fits between data sets. Also given in figure 8b is the depth of the 26.7 σ_t ($\sigma_t = [\rho_t - 1] \times 1000$) surface, which

shows a close correspondence with the 100% saturation horizon. This correspondence indicates that circulation in the upper water column is the major factor controlling the saturation horizons. The figures show that the 100% saturation horizons are shallowest in the cold-water region north of the subarctic front. In the northwest Pacific, the 100% horizon reaches its shallowest depth ($z = 220$ m for calcite and $z = 110$ m for aragonite) at about 50° N, whereas farther to the east the saturation horizons are slightly deeper ($z \approx 290$ m and $z \approx 170$ m, respectively, for calcite and aragonite). The shallow levels of the horizons north of the subarctic front are a result of the general cyclonic circulation within the subarctic gyre, where intensive upwelling produces a doming of the saturation isopleths. In the western Pacific, the deepest 100% saturation horizons are found in the region south of the subtropical front at about 33° N. Intensified vertical mixing induced by the Kuroshio extension results in the deepening of the saturation horizons in this region. As indicated by the oxygen distributions of Reid (1965) and the tritium data of Ostlund et al. (1979), mixing and lateral transport of the North Pacific intermediate water induces greater vertical exchange with upper water than the subarctic waters further to the north.

Another important factor affecting the depth of the saturation horizons is the buildup of TCO_2 relative to TA underneath the high
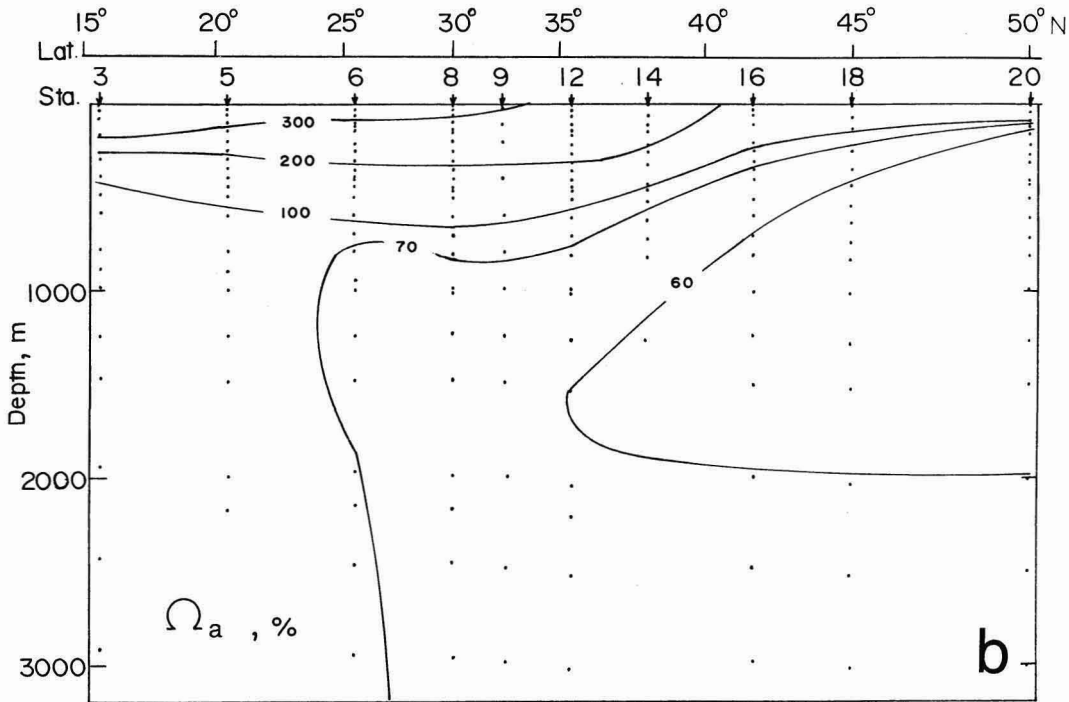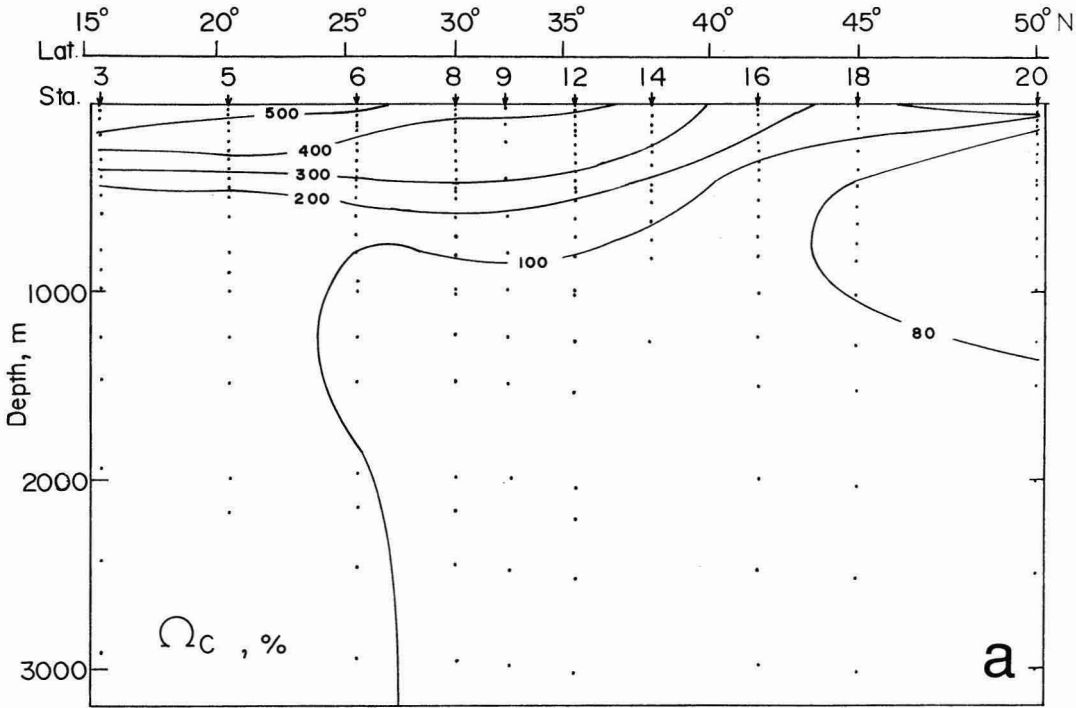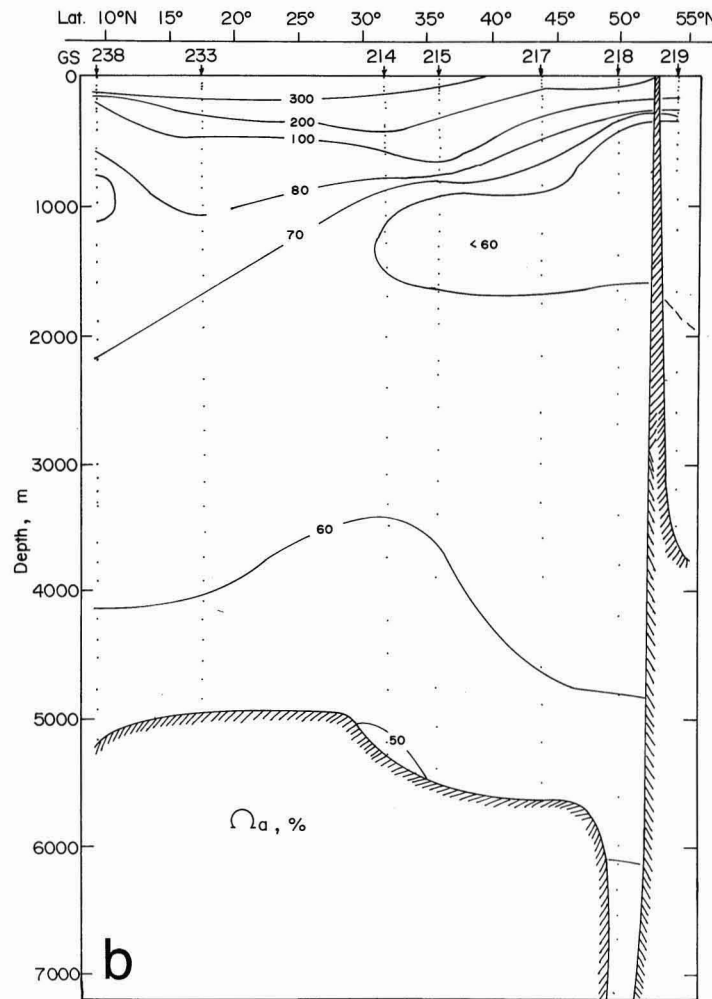
FIGURE 5. Cross section of (a) Ω_c and (b) Ω_a along 165° E.
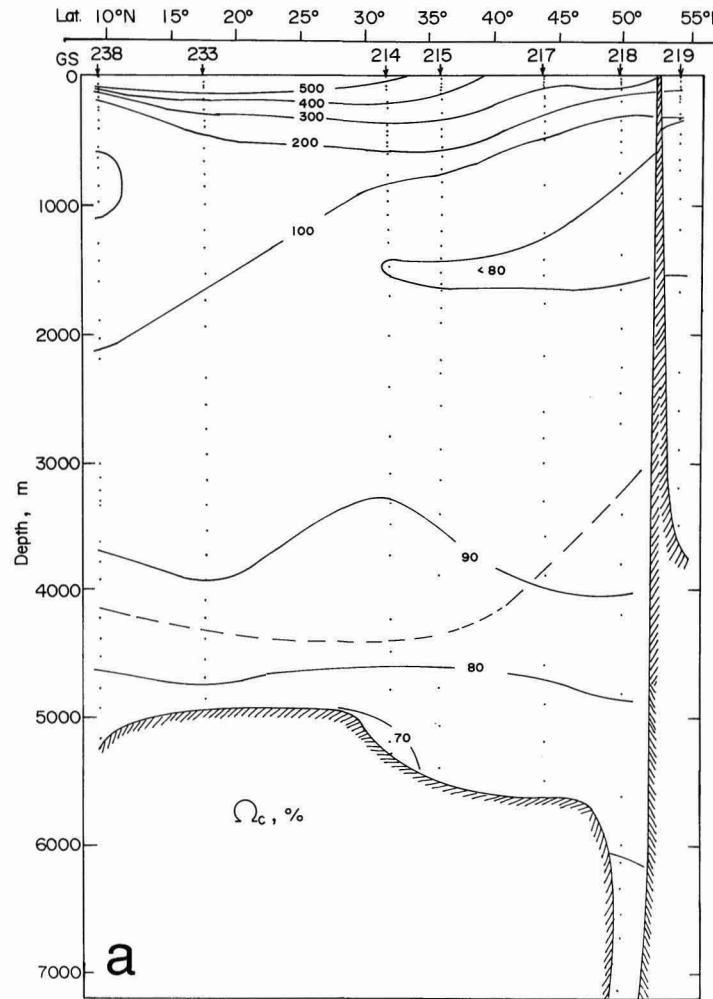
FIGURE 6. Cross section of (a) Ω_c and (b) Ω_a along 180° W. Dashed line represents calcium carbonate compensation depth data (CCD) from Berger et al. (1976).
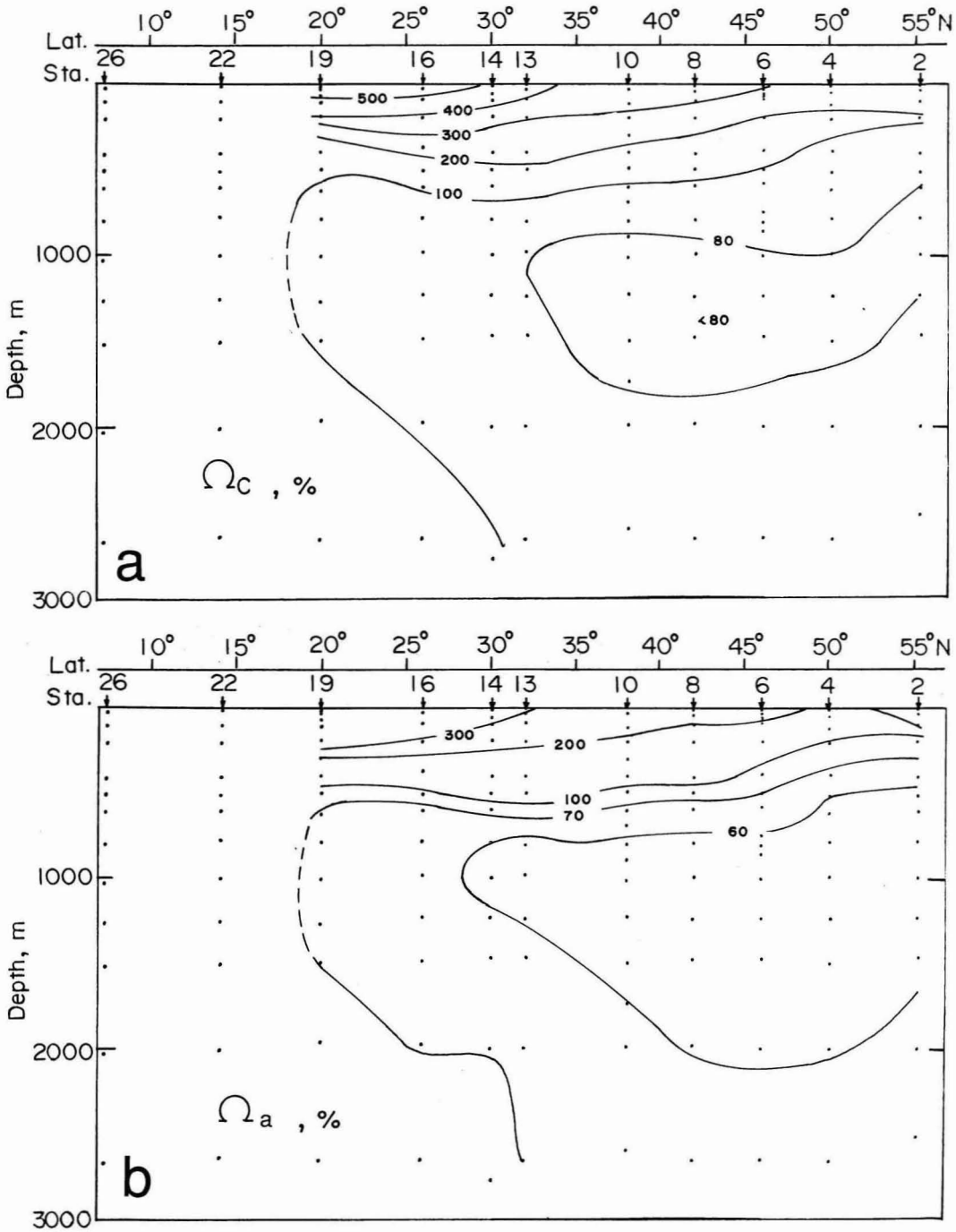
FIGURE 7. Cross section of (a) Ω_c and (b) Ω_a along 150° W.
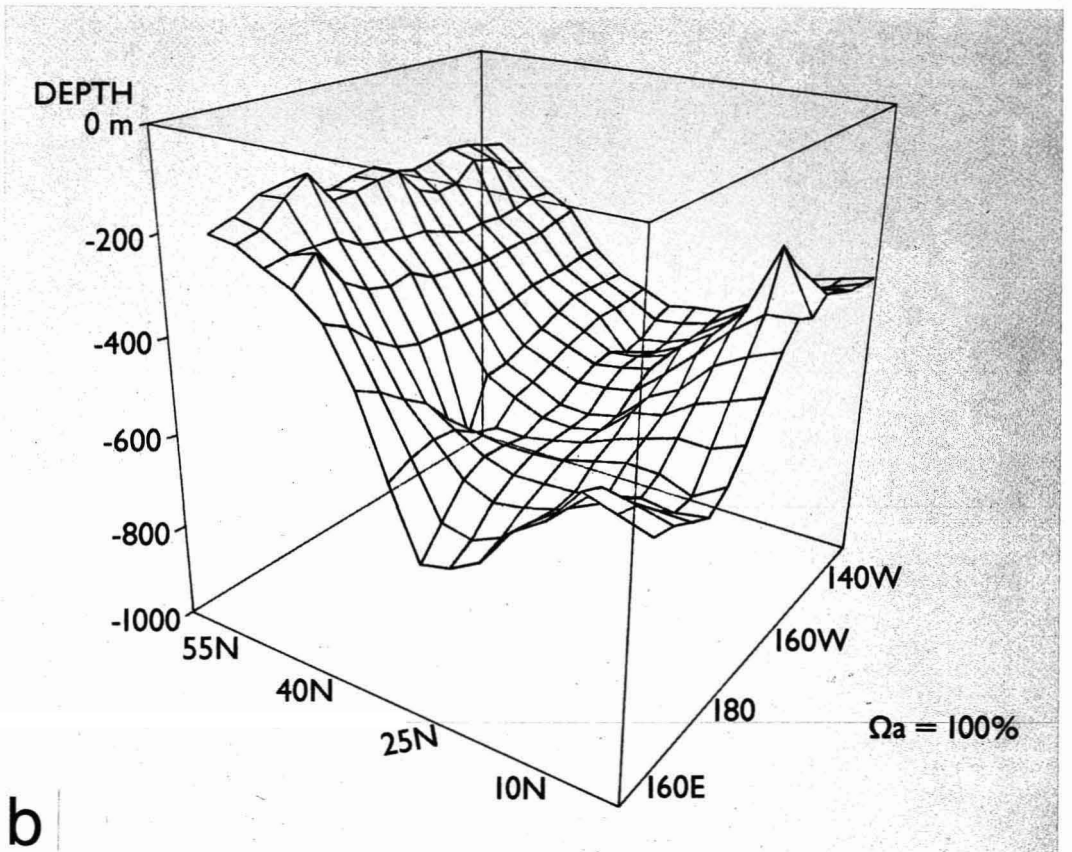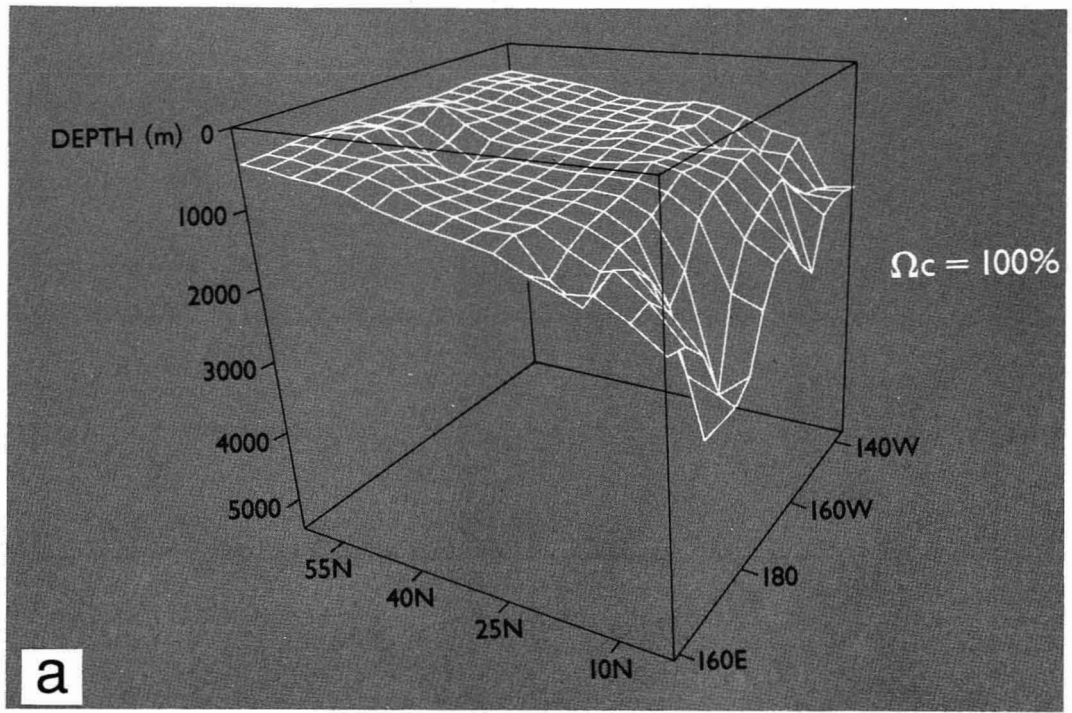
FIGURE 8. Three-dimensional representations of the 100% saturation horizons for (a) Ω_c and (b) Ω_a. For (b), $\sigma_t = 26.7$.

productivity regions, which causes enhanced shoaling of the 100% saturation surfaces. This effect is evidenced by the upward slope of the Ω_c and Ω_a saturation surfaces in the eastern Pacific. The increased acidity resulting from enhanced respiration processes decreases the carbonate ion concentration to the extent that the waters are undersaturated much closer to the surface. This effect is significantly enhanced in highly productive upwelling areas such as those of the equatorial waters of the eastern North Pacific or the nutrient-rich subarctic waters north of the subarctic front.

### THE RELATIONSHIPS AMONG Ω_c, LYSOCLINE, AND CALCIUM CARBONATE COMPENSATION DEPTH

The mechanisms that control the distribution of calcium carbonate-rich sediments are not clear. The relationships among the saturation horizon, lysocline, and calcium carbonate compensation depth (CCD) have been investigated by many workers (Broecker and Takahashi 1978, Edmond and Gieskes 1970, Heath and Culberson 1970, Li et al. 1969, Lisitzin 1972, Pytkowicz 1970, Takahashi 1975). In order to examine these three properties, we need to know the distribution of the saturation horizon, lysocline, and CCD. Peterson (1966) and Berger (1967) carried out in situ experiments of the dissolution of calcite in the central North Pacific Ocean (18°49′ N, 168°31′ W). They found that the rate of dissolution of calcite increases sharply at a depth of about 3700 m. This depth is called the lysocline. Unfortunately, this kind of experiment is scarce. An indirect approach to studying the lysocline was carried out in laboratory experiments by Morse and Berner (1972) and Berner and Morse (1974) by examining the parameter ΔpH, the difference between the pH for the calcite–seawater equilibrium and the seawater pH. Takahashi (1975) suggested a ΔpH value of 0.08 rather than the 0.15 proposed by Morse and Berner (1972), as an indicator of a sharp increase in the dissolution rate of calcium carbonate. According to Berner and Wilde (1972), a ΔpH value of 0.08

is equal to the saturation value of 91% Ω_c. This value is adopted in this report as a qualitative reference for the lysocline depth. The term "qualitative" is used carefully because of the uncertain depth at which $\Omega_c = 91\%$ falls. A large amount of water in the North Pacific has a calcite saturation value of 90–100%. The scattering of data also contributes too much noise for the $\Omega_c = 91\%$ depth to be defined precisely.

In Figures 3a and 6a the CCD according to Berger et al. (1976) is shown. As can be seen, the CCD usually falls at a depth above the 80% Ω_c contour in the mid-latitude region. Figure 6a shows a shoaling of the CCD north of 40° N to a depth where the Ω_c value is larger than 90%. If the 90% Ω_c contour were defined as the lysocline in the North Pacific, then the CCD generally falls at a depth deeper than the lysocline, and the lysocline generally falls at a depth deeper than the saturation horizon of calcite.

Station GS233 is selected for the purpose of discussion of the relationships among Ω_c, the lysocline, and the CCD. This station is selected because it is located near the station at which Peterson's (1966) in situ experiment was carried out. The vertical distributions of carbonate ion concentration and normalized alkalinity at GS233 from the surface to the sea bottom are shown in Figure 9. Even taking into consideration the maximum uncertainty in the determination of the saturation horizon (Plath et al. 1980, Pytkowicz 1983), the saturation horizon at 750 m is still much shallower than the lysocline and CCD depths. The depth of the lysocline ($\sim$3500 m) estimated from ΔpH is consistent with that of Peterson (1966). The CCD at about 4400 m (Berger et al. 1976) is also shown in the figure. The differences in the depths of the three parameters—saturation horizon, lysocline, and CCD—cannot be explained except by kinetics.

The carbonate ion concentration increases slightly below the saturation horizon, but this increase cannot be attributed to the dissolution of $CaCO_3$ alone. Physical conditions and pH values due to the speciation of carbonic acid also affect the concentration of CO_3^{2-} ion. Nevertheless, the general increasing trend of CO_3^{2-} concentration and the decreasing
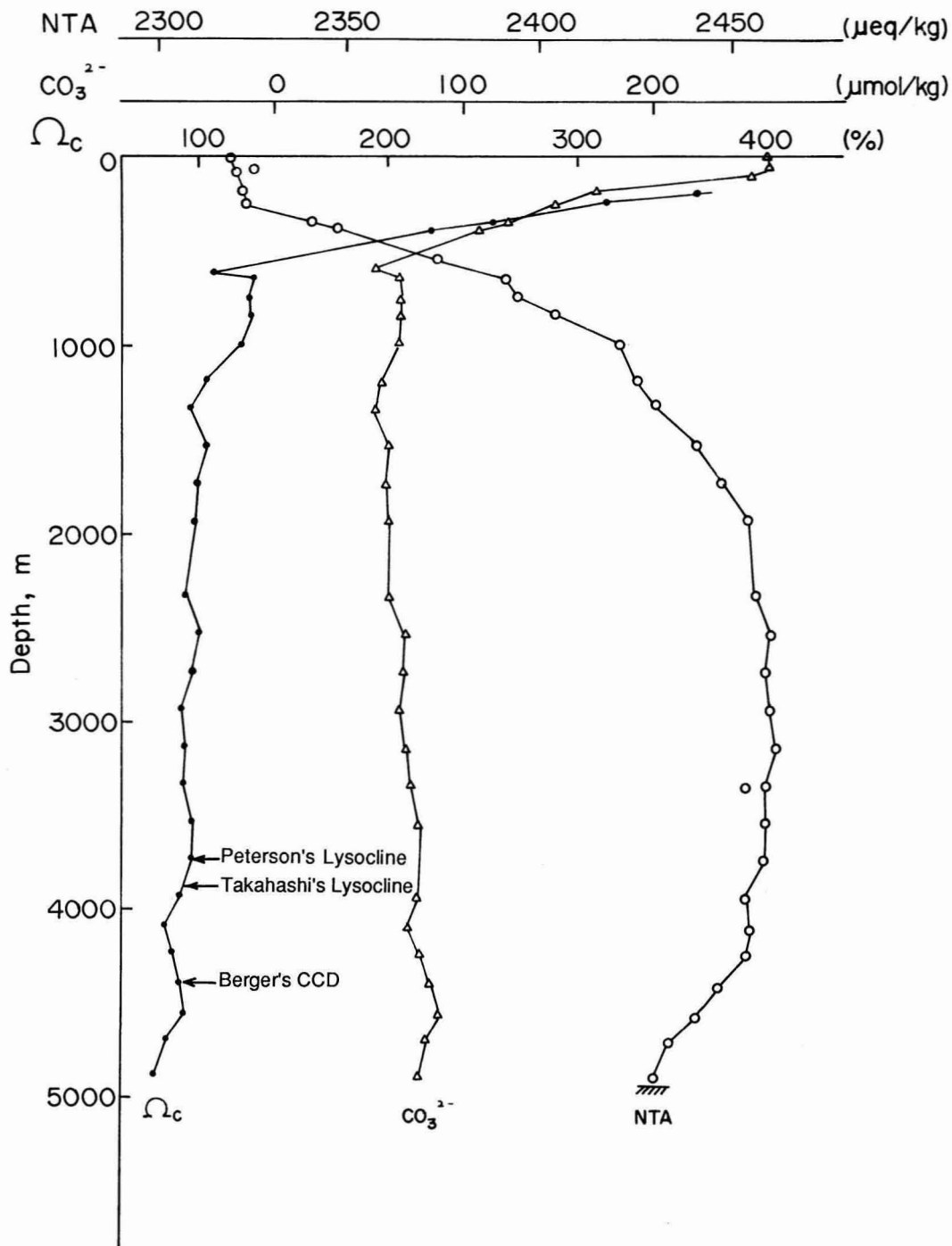
FIGURE 9. Vertical distributions of Ω_c, CO_3^{2-}, and NTA at GS233.

trend of saturation value below the saturation horizon still indicate the importance of the solubility product in the saturation value for deep waters.

Normalized titration alkalinity (NTA = TA × 35/salinity) is independent of temperature, salinity, and pressure effects if measurements are taken per weight unit. Hence, NTA distribution may serve as a better indicator of $CaCO_3$ dissolution than CO_3^{2-} ion concentration. The significance of a maximum NTA layer is noted here. The maximum NTA layer generally lies at a depth between 3000 and 3500 m in the North Pacific and is always shallower than both the lysocline and the CCD. This situation was not expected because the deep lysocline and CCD should imply more $CaCO_3$ dissolution below these depths, and an increasing trend should be found. An explanation for this phenomenon is that the rate of dissolution of $CaCO_3$ is not fast enough to overcome the flushing of the bottom water in that region. The location of GS233 is one of the main passages of the Pacific bottom water, with a relatively high

current speed of about 10 cm/sec originating from the southern ocean (Edmond et al. 1971). The rate of delivery of low-alkalinity bottom water is higher than the rate of in situ production from dissolution of $CaCO_3$.

DISTRIBUTION OF CARBONATE IN SURFACE SEDIMENTS OF THE PACIFIC OCEAN

Berger et al. (1976) provided a detailed map of calcium carbonate percentages in Pacific surface sediments. Their results for the North Pacific are reproduced in Figure 10. In general, carbonate-rich sediments are scarce north of 15° N, presumably due to a predominance of sea floor at or below the calcite and aragonite compensation depths (Figure 3–8). The calcium carbonate compensation depth deepens south of 25° N (Figures 5–8). The deepening corresponds to a narrow zone of transition between low and high surface carbonate content between 15° and 10° N. The calcium carbonate percentages reach more than 80% in
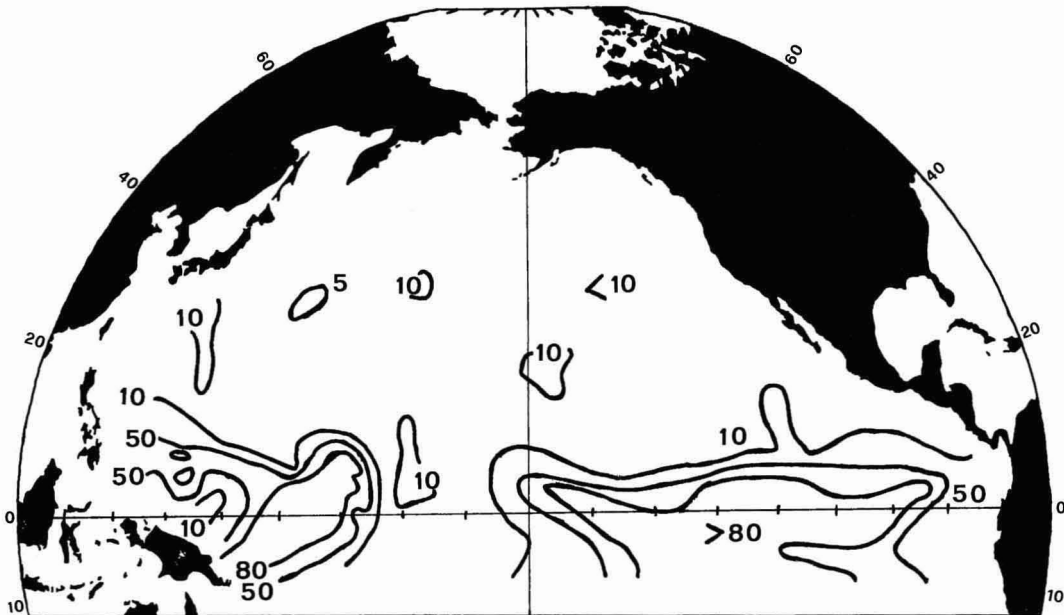

FIGURE 10. A contour map of calcium carbonate percentages in the North Pacific sediments (from Berger et al. 1976).

the equatorial region, reflecting the deepening of the calcium carbonate compensation depth.

CONCLUSION

The degree of saturation with respect to calcite and aragonite was calculated from all available data sets. Four selected cross sections, three longitudinal and one latitudinal, and two three-dimensional graphs show that a large volume of the North Pacific is undersaturated with respect to $CaCO_3$. The saturation horizon generally shoals from west to east and from south to north in the North Pacific Ocean. It was found that the lysocline falls at a depth much deeper (about 2500 m deeper) than the saturation horizon of calcite and several hundred meters shallower than the calcium carbonate compensation depth. Our results appear to support the kinetic point of view on the $CaCO_3$ dissolution mechanisms. The calcium carbonate compensation depth deepens south of 25° N. A narrow zone of transition between low and high surface carbonate content exists between 15° and 10° N. The calcium carbonate percentages reach more than 80% in the equatorial region, reflecting the deepening of the calcium carbonate compensation depth.

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