

Carbon Dioxide in Surface Ocean Waters¹

3. Measurements on Lusiad Expedition 1962–1963

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Direct measurements of carbon dioxide in equatorial surface waters during Lusiad Expedition, 1962–1963, indicate that high concentration relative to atmospheric CO₂ is associated with eastward-flowing equatorial undercurrents in the Indian and Atlantic oceans. In the tropical Indian Ocean the measurements are sufficient to construct the areal distribution during two seasons. A broad band of high CO₂ occurs during the northeast monsoon but not during the southwest monsoon. An undercurrent is observed only during the former period. Adjacent to the equatorial zone the CO₂ concentration is generally lower than equilibrium with the atmosphere in all oceans. In the South Indian Ocean the concentration rises sharply southward on approaching the northern limit of the antarctic circumpolar current. In the western North Pacific Ocean the concentration attains very low values in the east-flowing extension of the Kuroshio Current. In general, low concentration in surface waters exists when rapid cooling occurs and when withdrawal of CO₂ by marine organisms exceeds replenishment by physical transport; high concentration exists when rapid warming occurs and when upwelling or vertical mixing supplies more CO₂ than can be withdrawn by plants or can escape to the atmosphere.

INTRODUCTION

At present, about half of the carbon dioxide released by the burning of coal and petroleum is accumulating in the atmosphere [*Pales and Keeling*, 1965]. The remainder is being absorbed by the oceans and possibly to some extent by land plants [*Hutchinson*, 1954, p. 389]. If chemical equilibrium were achieved between atmospheric CO₂, inorganic carbon dissolved in ocean water, and carbonate sediments on the ocean bottom, the oceans could absorb most of the CO₂ from combustion [*Bolin and Eriksson*, 1959, p. 133]. Under present conditions, however, the rate of injection of CO₂ into the air by combustion is too rapid for the atmosphere and the oceans to approach equilibrium. We are therefore obliged to document the transient response of the atmosphere and oceans to the burning of fossil fuels, if we are to explain the present rise in atmospheric CO₂ and make a reliable prediction of future trends.

A major step in documenting this response is to determine the present distribution of CO₂ gas in surface ocean water. Direct measurements on oceanic expeditions since 1957 [*Leahy*,

1961; *Takahashi*, 1961; *Hood et al.*, 1963; *Ibert*, 1963; *Keeling et al.*, 1965] indicate that the concentration of CO₂ in surface water of the Atlantic and Pacific oceans varies in a complicated way with geographic location. Although these published data are too few to reveal time variations, we expect the concentration to vary with season as well. Clearly we need to improve our understanding of contemporary conditions. To this end we now report a survey of the North Pacific, the Indian, and the Atlantic oceans that extends considerably the areal coverage of oceanic CO₂ and for the tropical Indian Ocean furnishes data to assess seasonal variability.

EXPERIMENTAL PROCEDURE

Measurements of CO₂ were made on the research vessel *Argo* during Lusiad Expedition, the second of three expeditions made by the Scripps Institution of Oceanography between 1961 and 1965 as part of the International Indian Ocean Expedition. Equipment and methods closely duplicated those of Downwind and Monsoon expeditions, as described by *Keeling et al.* [1965]. The output from a nondispersive infrared gas analyzer was continuously moni-

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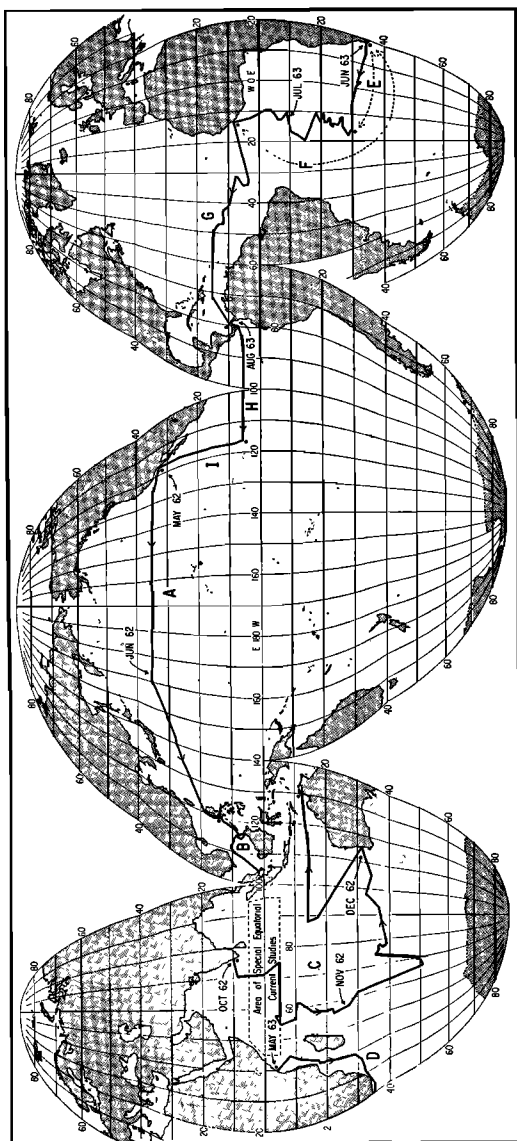


Fig. 1. Ship track for Lusiad Expedition.

tored on a stripchart recorder, calibrated in parts per million of dry air by volume (ppm). The average amplitude of the instrument noise was 1 ppm.

Atmospheric CO_2 was measured directly by drawing in air at mast height (10 to 20 meters above sea level) through intake cups protected against ocean spray. Water vapor was removed before the air was analyzed by passing the gas stream through a cold trap maintained below -40°C by a refrigerator. The CO_2 in surface

ocean water was determined by recording the concentration of CO_2 in a closed volume of air that was circulated continuously so as to approach equilibrium with a constantly renewed supply of water obtained from an intake in the ship's hold 2 to 3 meters below the surface. An improved version of the equilibrator used on Monsoon Expedition was employed.

The concentration of CO_2 in the air and ocean water, as reported below, is expressed as a volume fraction (mixing ratio) in ppm. This quantity is the partial pressure of CO_2 divided by the total pressure exerted in the atmosphere or ocean water by the gases that pass through the infrared analyzer. Methods for computing the partial pressure for the air and water are described by Keeling *et al.* [1965, pp. 6088-6091].

DESCRIPTION OF CONTINUOUS DATA

A total of 14,231 atmospheric air and 12,040 ocean surface water measurements, each representing a 5- to 20-minute average of the original continuous record, have been processed. Where the ship traversed a predominately east-west course, the data were averaged over longitudinal intervals, otherwise they were averaged over latitudinal intervals. For the South China Sea and equatorial crossings in the Indian and Atlantic oceans, 1° intervals were used. Elsewhere $2\frac{1}{2}^\circ$ intervals were employed. These averages are assembled in tables similar to Tables 1 and 2 of Keeling *et al.* [1965] and are contained in a report by Keeling and Waterman [1967].² This report also presents all relevant calculations and supporting data for the Lusiad Expedition and for two earlier expeditions reported by Keeling *et al.* [1965].

The ship's track is shown in Figure 1 divided into nine sections, A through I. The complicated track patterns in the equatorial Indian Ocean are shown in Figures 2 and 3. Observations of the concentration of CO_2 and of sur-

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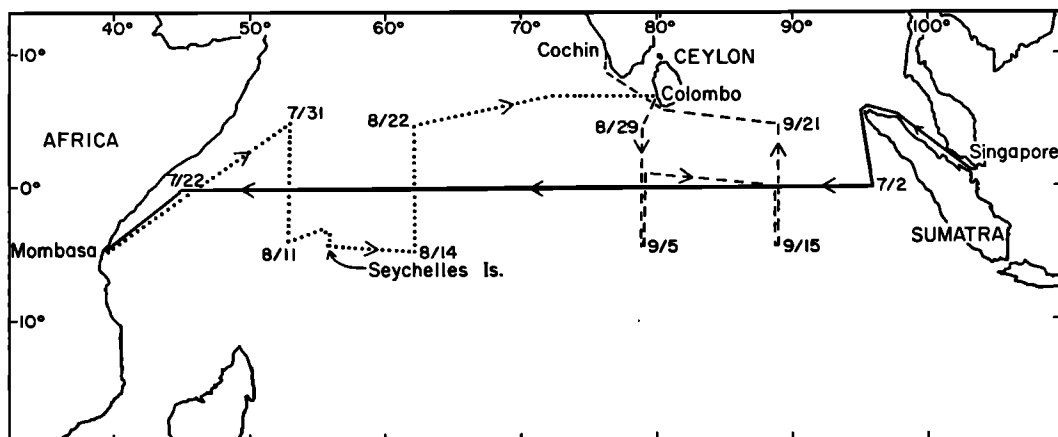


Fig. 2. Ship track for first equatorial current study, June 26 to September 24, 1962.

face water temperature are plotted versus latitude or longitude in Figures 4 to 16.

Section A, across the North Pacific Ocean from San Diego to Manila (Figure 4), is both the northernmost and the longest east-west profile so far obtained. The atmospheric CO_2 concentration appears, as it does in all sections, as an almost regular horizontal line; the CO_2 concentration in the water varies almost symmetrically about a sharp minimum at 175°E longitude.

Section B (Figure 5) occurs in relatively shallow water of the South China Sea from Manila to Singapore. The CO_2 concentration in the water relative to equilibrium with the atmosphere is high throughout the section.

In the first of two detailed studies in the equatorial Indian Ocean, during the southwest

monsoon, the CO_2 concentration in the water was everywhere above equilibrium. Along the equator (Figure 6) maxima were observed near 70°E over the mid-Indian and Carlsberg ridges and near 50°E over the western slope of the Somali basin. North-south cross sections show a trend toward lower values southward (Figure 7).

Section C reveals that this latter trend is a major feature of the South Indian Ocean (Figure 8). Strikingly low values were observed over a broad zone from 25° to 45°S . South of 45° the CO_2 concentration in the water rises abruptly. On the run northward from Kerguelen Island (49°S) the mean values do not depart markedly from the values obtained on the run southward, nor is the retracing as close as was observed at the same latitudes in the

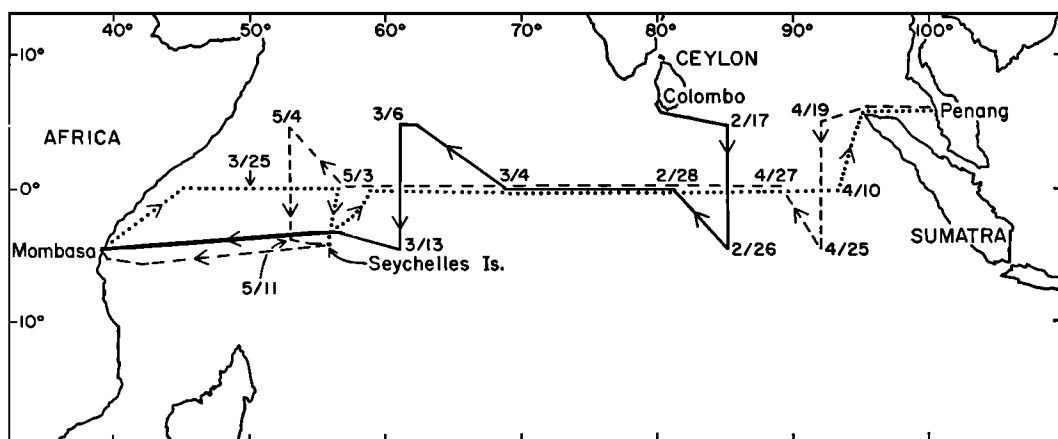


Fig. 3. Ship track for second equatorial current study, February 16 to May 15, 1963.

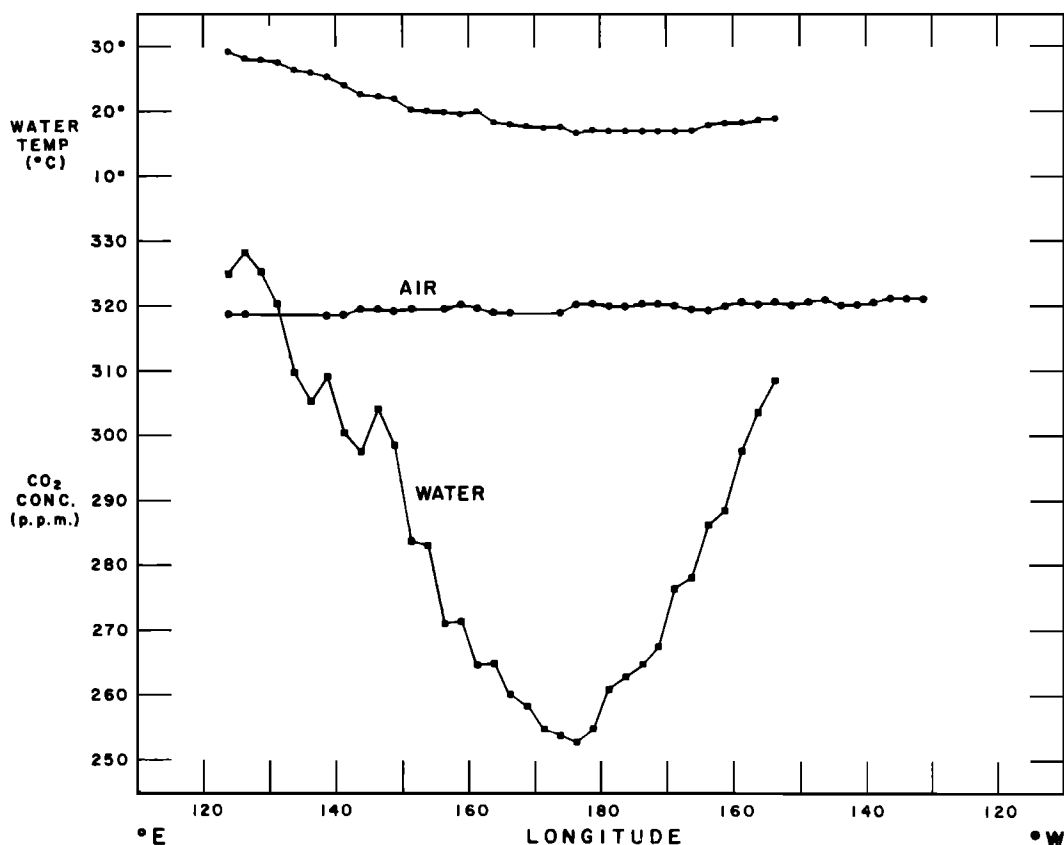


Fig. 4. CO_2 concentration and water temperature near the ocean surface as a function of longitude, section A, North Pacific Ocean, May 18 to June 9, 1962.

Pacific Ocean during Downwind Expedition [Keeling *et al.*, 1965].

In the second equatorial study, which included the season of the northeast monsoon, four cross-sectional traverses and three runs along the equator were made (Figure 3). North-south cross sections show a variety of features individually (Figure 10) but, viewed together, show a steady trend toward higher CO_2 concentration westward. This trend shows even more clearly in three traverses along the equator (Figure 9).

In the final section in the Indian Ocean from Mombasa to Cape Town, section D, the CO_2 concentration in the water declined steadily southward. The trend is strikingly similar to that observed east of Malagasy Republic (Madagascar) seven months earlier (Figure 8).

In the Atlantic Ocean (sections E, F, and G; Figures 12 to 14) the CO_2 concentration in

the water shows a narrow band of high values between 6°S and 1°N . The concentration is

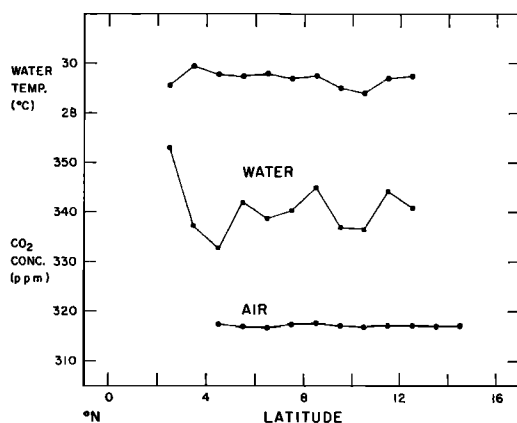


Fig. 5. CO_2 concentration and water temperature near the ocean surface as a function of latitude, section B, South China Sea, June 14-24, 1962.

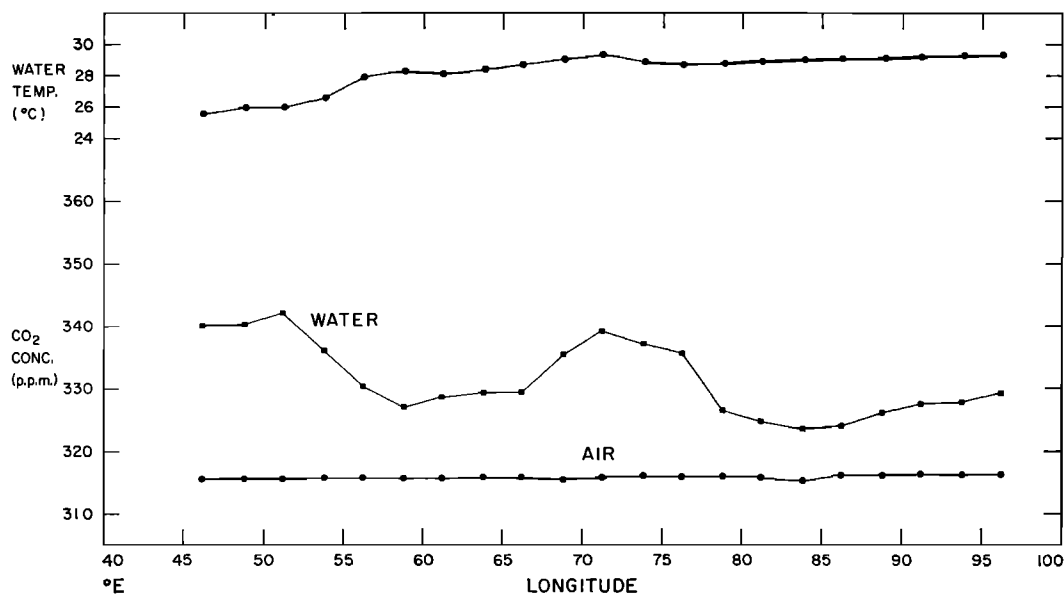


Fig. 6. CO_2 concentration and water temperature near the ocean surface as a function of longitude along the equator, Indian Ocean, July 2-27, 1962.

close to atmospheric equilibrium in most other regions traversed, except south of 25°S , where it diminishes rapidly southward, and in the Caribbean Sea, where a sharp peak occurs over the Beata ridge (75°W).

On the final leg of the expedition, in the Pacific Ocean from Panama to San Diego (sections H and I, Figures 15 and 16), the CO_2

concentration in the water rises from east to west near 5°N with values near atmospheric equilibrium for more than half of section H (Figure 15). It declines sharply from 5° to 10°N , then increases approaching the continental shelf of North America (Figure 16). The distribution closely approximates the distribution obtained in the East Pacific in October

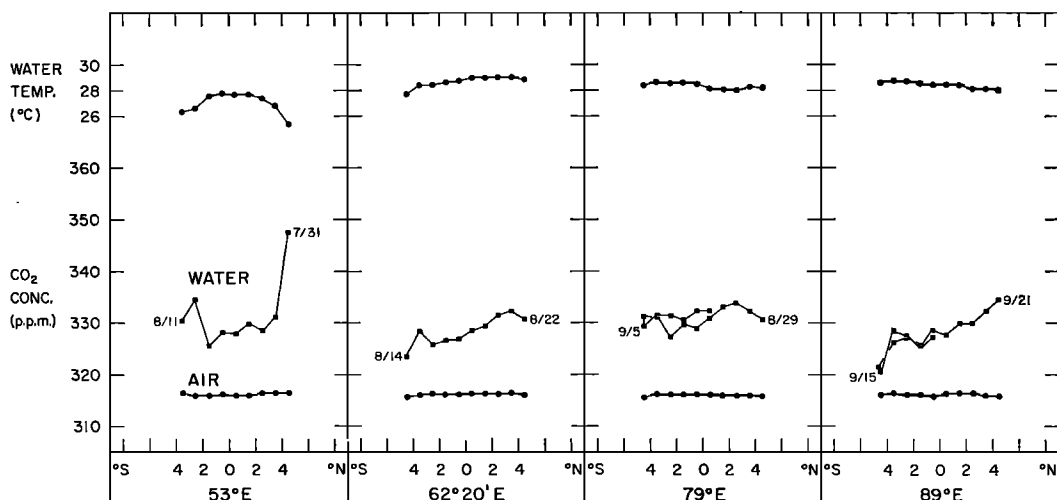


Fig. 7. CO_2 concentration and water temperature near the ocean surface as a function of latitude along four equatorial cross sections shown in Figure 2, Indian Ocean, July to September 1962. The date (month/day) is shown for the beginning and end of each run.

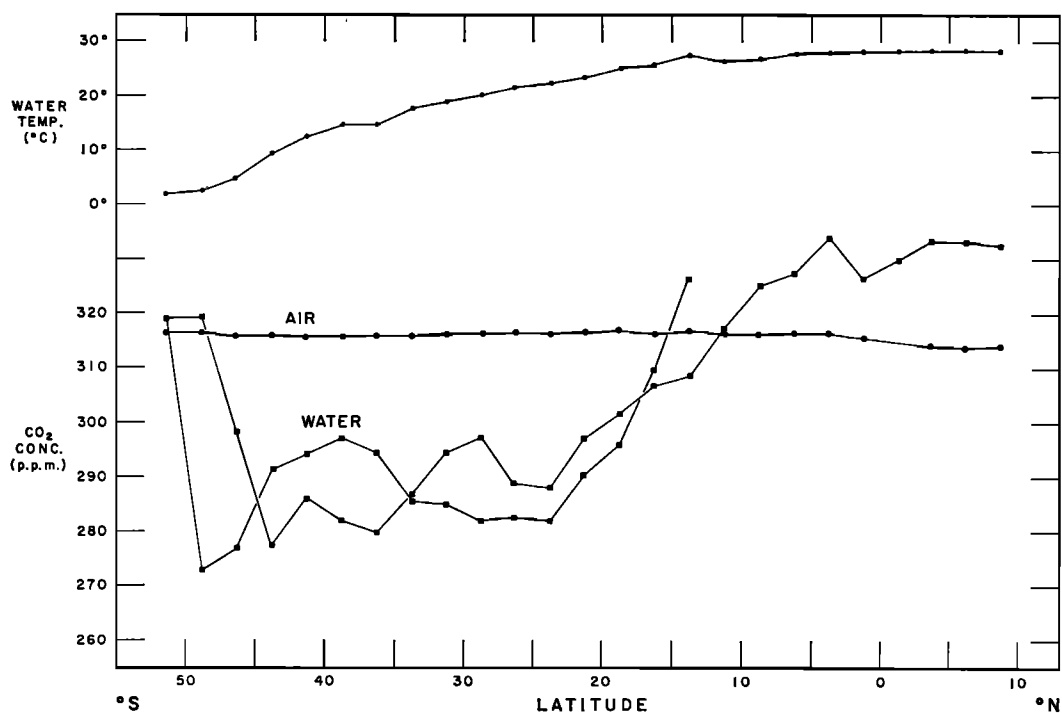


Fig. 8. CO₂ concentration and water temperature near the ocean surface as a function of latitude, section C, Indian Ocean, October 5 to December 21, 1962.

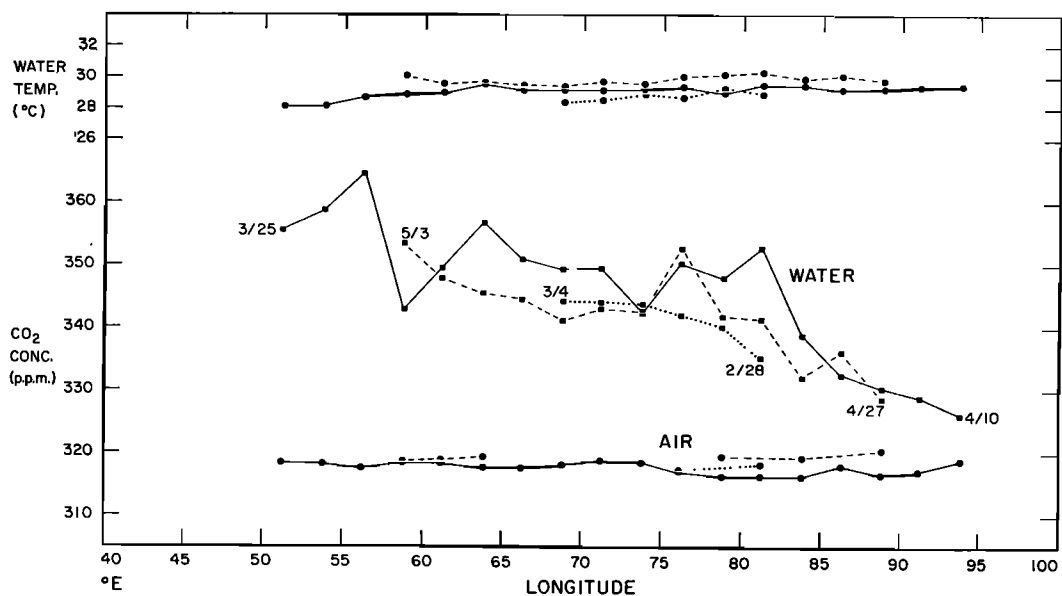


Fig. 9. CO₂ concentration and water temperature near the ocean surface as a function of longitude along the equator, Indian Ocean, February to May 1963. The date (month/day) is shown for the beginning and end of each of three runs.

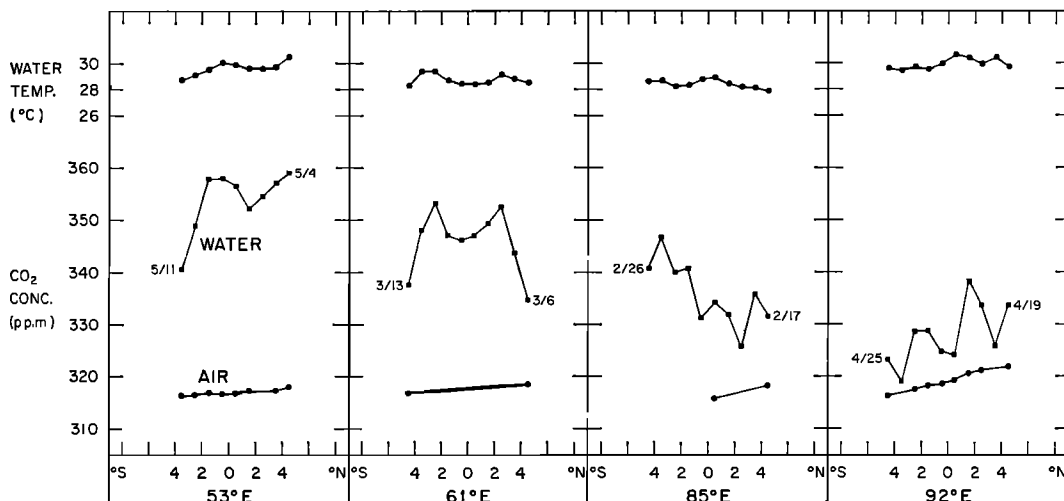


Fig. 10. CO_2 concentration and water temperature near the ocean surface as a function of latitude along four equatorial cross sections shown in Figure 3, Indian Ocean, February to May 1963. The date (month/day) is shown for the beginning and end of each run.

1957 on Downwind Expedition [Keeling *et al.*, 1965, p. 6091].

DISCUSSION

The data of Lusiad Expedition are very extensive, but the area investigated was also very extensive. We cannot accurately determine the surface distribution of oceanic CO_2 , except in a few limited regions where relationships are clearly revealed. More often we must guess whether the ship's track tended to run parallel

to the gradient (perhaps true for sections A, C, D, and F; Figures 4, 8, 11, and 13) or parallel to lines of equal concentration (likely for sections E, G, and the western half of section H; Figures 12, 14 and 15). In most waters visited the CO_2 concentration was measured for the first time, and the data must be assessed solely on their own merit. In this article we selected for discussion a few well-defined features to illustrate the typical character of the distribution so far observed. We shall give major attention to the Indian Ocean, where the coverage is most detailed.

Across the North Pacific Ocean (section A,

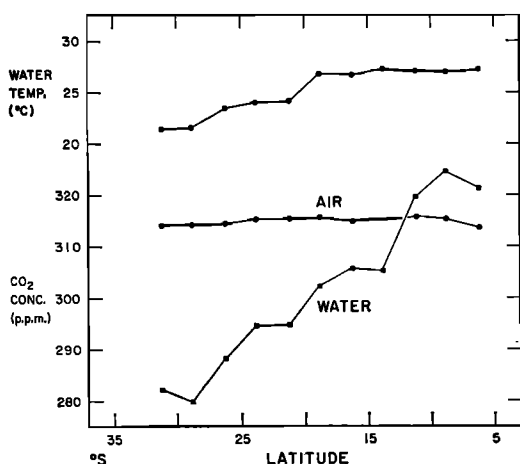


Fig. 11. CO_2 concentration and water temperature near the ocean surface as a function of latitude, section D, Mozambique Channel, May 18-26, 1963.

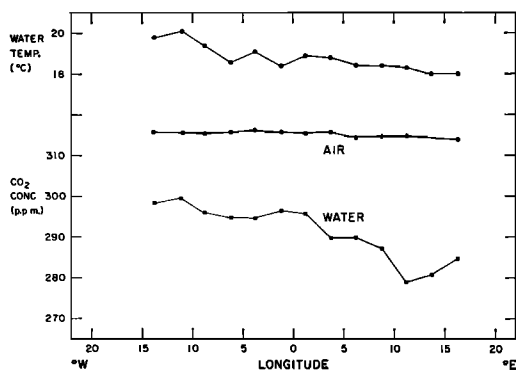


Fig. 12. CO_2 concentration and water temperature near the ocean surface as a function of longitude, section E, Cape Town, South Africa, to 15°W, June 4-14, 1963.

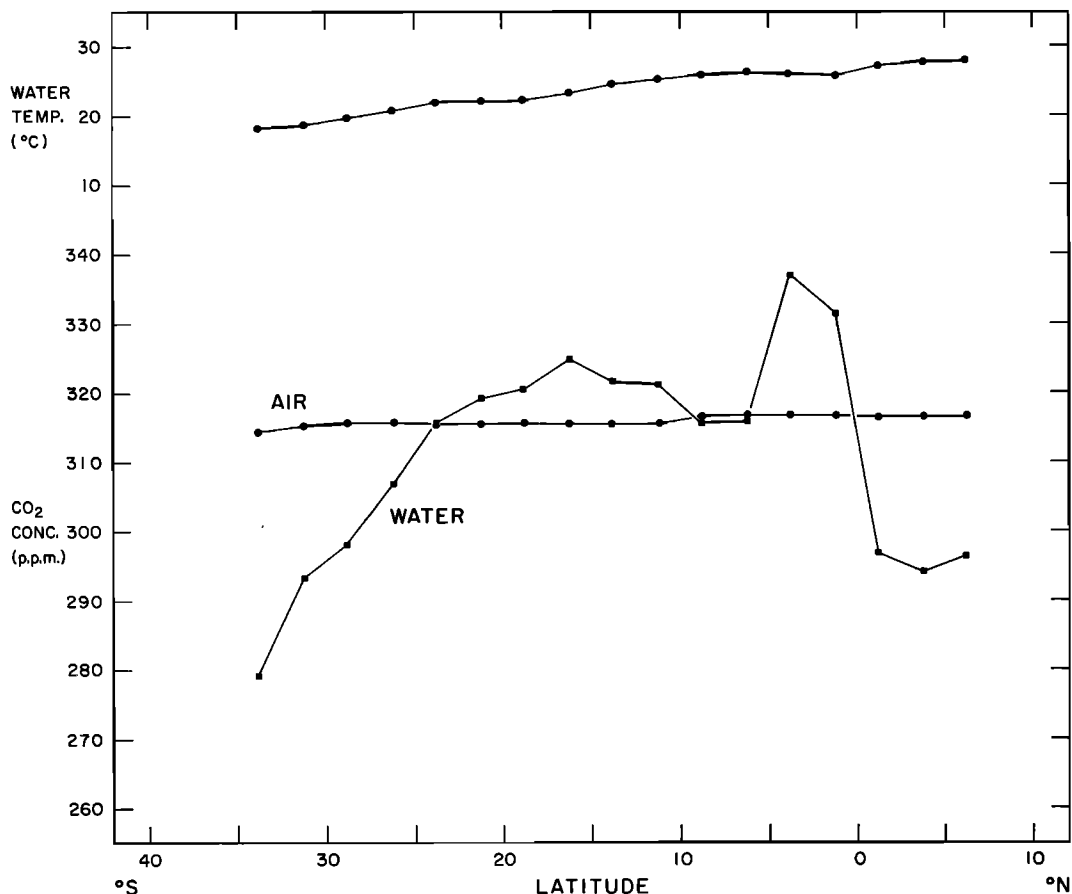


Fig. 13. CO₂ concentration and water temperature near the ocean surface as a function of latitude, section F, Cape Town, South Africa, to Freetown, Sierra Leone, June 4 to July 8, 1963.

Figure 4) the oceanic CO₂ data reveal a single sink for atmospheric CO₂ 10,000 km wide. The center shows the lowest concentration so far observed anywhere in the Pacific Ocean. We attribute this sink principally to the cooling of surface water carried northward and eastward by the Kuroshio Current and its extension [Sverdrup *et al.*, 1942, p. 721]. The Kuroshio transports warm surface water, with an initial temperature of about 28°C, from the tropical west Pacific to the vicinity of Japan. A major branch extends eastward into the open ocean as a relatively strong current along 34°–36°N as far as 175°E [Defant, 1961, p. 570]. Farther east the rate of flow diminishes and water derived from the Kuroshio gradually mixes with subarctic water. While flowing

northward and eastward the water cools 10° to 15°C in about 150 days.

For all of Lusiad section A west of the observed CO₂ minimum at 35°N, 175°E the CO₂ concentration follows closely a prediction based on cooling. From west to east the temperature decreased by 12°C; the CO₂ concentration, by 70 ppm. The CO₂ decrease predicted by cooling is 90 ppm [Harvey, 1955; Tables 25, 26]. (We assume a chlorinity of 19‰ and an alkalinity of 2.32 millequivalents.)

East of 175°E, where Kuroshio water gradually loses its identity, the CO₂ concentration increased rapidly toward equilibrium with the atmosphere while the temperature remained close to the observed minimum.

Absorption of CO₂ from the atmosphere

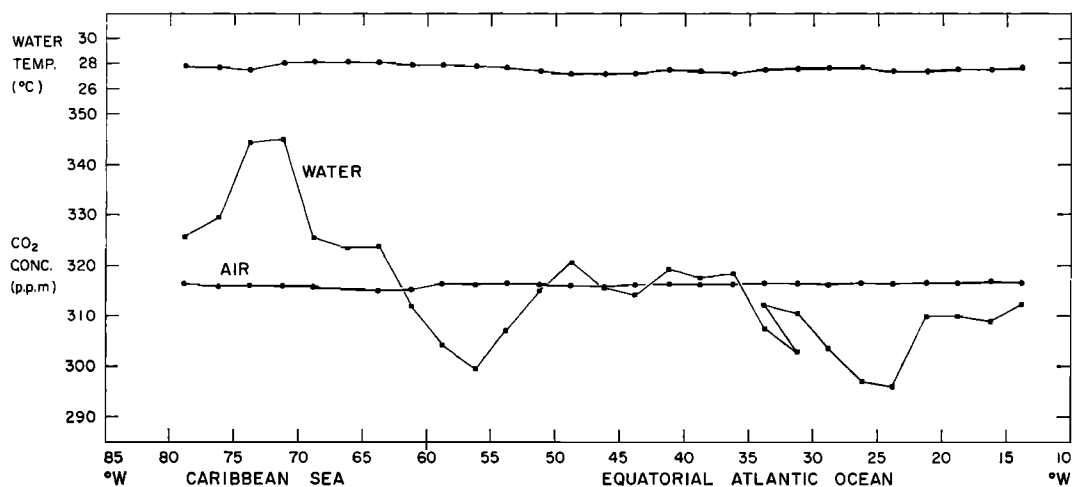


Fig. 14. CO₂ concentration and water temperature near the ocean surface as a function of longitude, section G, Freetown, Sierra Leone, to Panama Canal, July 11-30, 1963.

accounts satisfactorily for a smaller decrease in CO₂ concentration at 175°E than is predicted by cooling. The depth of the mixed layer in the Kuroshio Current is about 50 meters [Sverdrup *et al.*, 1942, pp. 131-132]. If we assume that the CO₂ from the atmosphere penetrated to this depth, the amount of absorption deduced from the observed change in temperature and CO₂ concentration, using the tables of Harvey [1955], is 0.12 mM. If we accept as the absorption rate 7 moles cm⁻² atm⁻¹ yr⁻¹ in accordance with an atmospheric residence time of radiocarbon of five years [Broecker, 1963; Skirrow, 1965, p. 316], we obtain a value of

$\frac{1}{2}$ year for the time required for the water to cool from 29°C off the coast of Formosa to 17° near the temperature minimum at 175°E.

Although the distribution of CO₂ west of 175°E may be due to physical processes alone, as the above calculation suggests, the calculated amount of absorption is uncertain to at least a factor 2. We cannot rule out the possibility that the CO₂ concentration is influenced by the presence of relatively high concentrations of plankton, as mapped by Reid [1962, pp. 300-

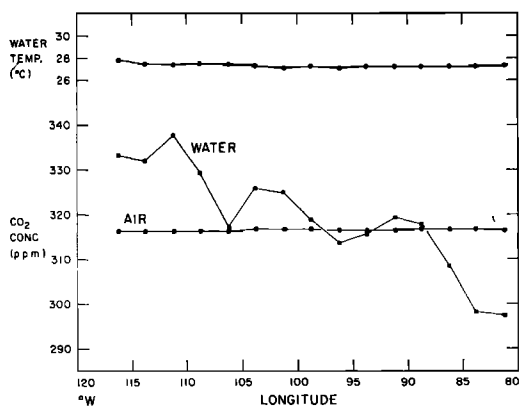


Fig. 15. CO₂ concentration and water temperature near the ocean surface as a function of longitude, section H, Panama Canal to 117°W, August 2-10, 1963.

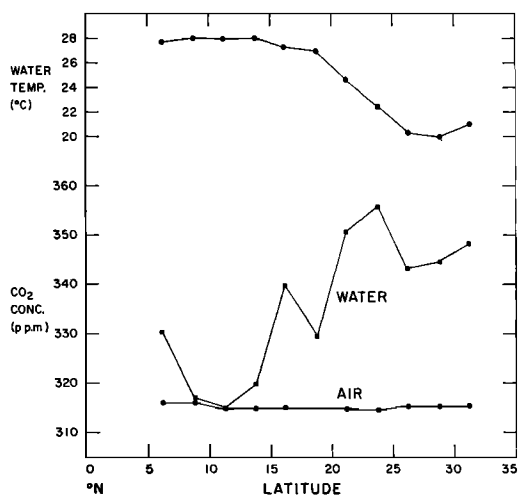


Fig. 16. CO₂ concentration and water temperature near the ocean surface as a function of latitude, section I, 5°N to San Diego, August 10-15, 1963.

301]. Furthermore, the observations were made in May, when the northern oceans often show intense bursts of photosynthetic activity, and the observed position and strength of the CO₂ sink may have been partly determined by time variations in productivity.

Most of the Indian Ocean measurements south of 5° were made during a single season (November and December 1962; see Figure 8), but one track six months later furnished data (Figure 11) that accord so well with the earlier results that we are led to propose that the CO₂ in surface waters of the subtropical South Indian Ocean is without strong seasonal or east-west variation. We thus infer a broad zone of low concentration in temperate and subtropical latitudes, which gives place gradually to high concentrations near the equator and terminates abruptly southward close to the northern limit of the antarctic circumpolar current [Wyrski, 1960, p. 159]. Rising values on approaching the Antarctic Current were also observed southeast of New Zealand in March 1961 [Keeling *et al.*, 1965] and are probably characteristic of the antarctic polar front.

In the equatorial Indian Ocean continuous CO₂ measurements were obtained during two 3-month studies of the equatorial current system [Knauss and Taft, 1964; Taft and Knauss, 1967]. The data are sufficient to sketch the distribution of CO₂ in surface water from June to September 1962, during the southwest monsoon, and from February to May 1963, toward the end of the northeast monsoon, and at the onset of the southwest monsoon. The average distributions for the two periods (Figures 17 and 18) indicate that major changes in CO₂ occur as the wind regime and oceanic circulation change with the seasons. During the southwest monsoon the concentration almost everywhere diminished southward from the northern limits of measurement; during the northeast monsoon a band of high CO₂ extended over two-thirds of the equatorial zone.

Direct velocity measurements, made at the time of the CO₂ measurements, indicate that a steady east-flowing equatorial undercurrent, about one-third as strong as the Pacific undercurrent [Knauss, 1966], was present at depths between 50 and 200 meters across most of the Indian Ocean during the northeast monsoon but was either absent or weak and variable at

other times. North-south cross sections prepared by Taft and Knauss [1967] indicate that the northeast monsoon was accompanied by a spreading of the isopleths for temperature, oxygen, and inorganic phosphate in the thermocline near the equator. During the southwest monsoon, when the undercurrent was absent or weak and variable, no equatorial spreading of the isopleths was observed. Iso-pleth spreading near the equator occurs at all seasons in the Pacific Ocean, a circumstance that Knauss [1963] attributes to upwelling or vertical mixing associated with the undercurrent.

In the eastern tropical Pacific Ocean a band of high CO₂ concentration is found in the surface water near the region of the undercurrent. Keeling *et al.* [1965] attributed this band to the upward transport of CO₂ from CO₂-rich subthermocline waters, the transport at least in part owing to the same mechanism that causes the isopleths of oxygen and temperature to spread. In the equatorial Atlantic (see below) the same features occur and the same mechanism evidently operates. In the Indian Ocean vertical CO₂ transport appears to be synchronized with an intermittent undercurrent. A band of high CO₂ in surface equatorial waters is thus strong evidence for the presence of an undercurrent and vice versa.

That the CO₂ in surface waters of the Indian Ocean adjusts rapidly to changes in vertical transport is indicated by the sharp decrease in CO₂ from spring to summer. The exchange of CO₂ across the ocean atmosphere interface is too slow [Skirrow, 1965, p. 315] to account fully for this rapid adjustment, and it may be due partly, if not principally, to a northerly flow of surface waters driven by the southwest monsoon that displaces the band of high-CO₂ water with surface water of lower concentration lying to the south. It may also be due partly to photosynthetic activity that reduces the CO₂ after the vertical transport ceases.

A clearly defined region of high CO₂ concentration was observed in the east equatorial Atlantic in July 1963 (Figure 9). North of 2°S it is associated with low surface water temperature, an association that also occurs north of the equator in the eastern equatorial Pacific Ocean [Keeling *et al.*, 1965] and suggests upwelling. Vertical cross sections of temperature, salinity, and oxygen prepared from data ob-

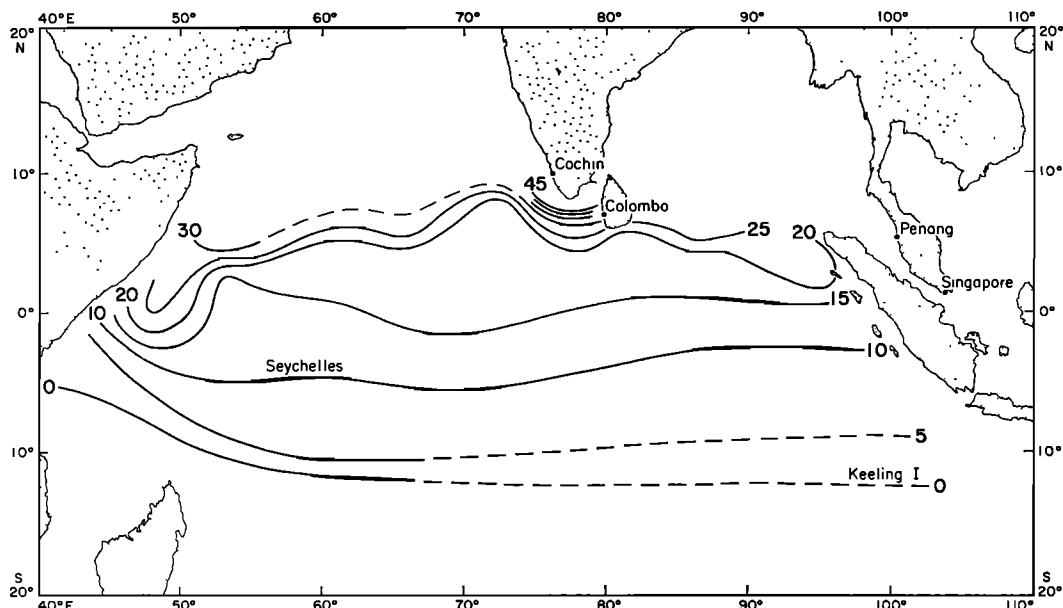


Fig. 17.

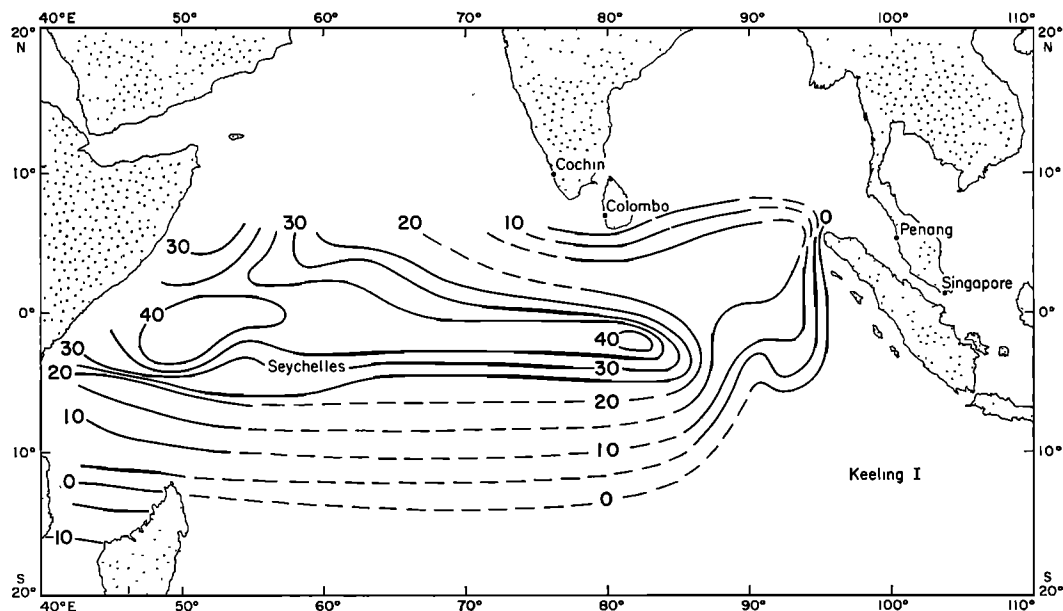


Fig. 18.

Figures 17 and 18 show the areal distribution of carbon dioxide in surface water of the equatorial Indian Ocean during two seasons, as indicated by measurements from June 28 to September 24, 1962 (Figure 17), and February 16 to May 15, 1963 (Figure 18). Concentrations are expressed as departures in ppm from 315 ppm. The latter value is approximately the concentration of atmospheric CO_2 as indicated by atmospheric measurements made at the time of the water measurements. Solid lines are drawn where the data control is good; dashed lines, where it is poor.

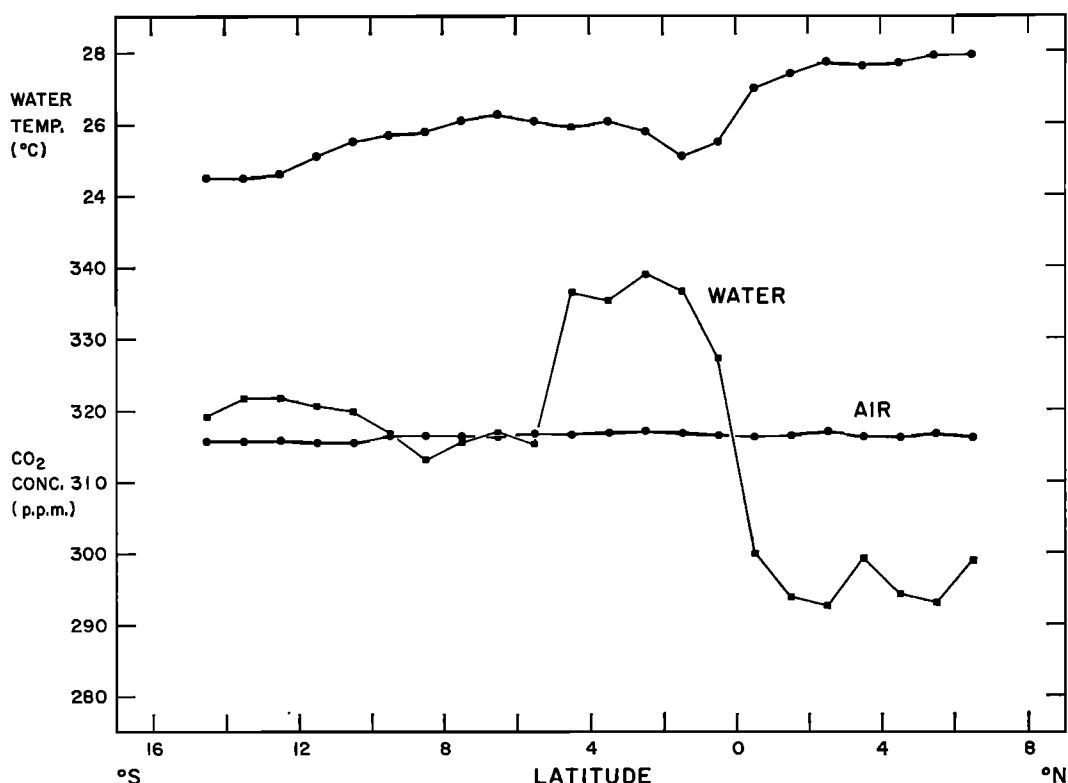


Fig. 19. The concentration of CO_2 near the ocean surface in the atmosphere and in ocean water and surface ocean water temperature as a function of latitude in the eastern Atlantic Ocean, June 28 to July 7, 1963.

tained at the time of the CO_2 measurements [Reid, 1964] show features characteristic of the Atlantic equatorial undercurrent as described by various investigators [Metcalf *et al.*, 1962; Newman and Williams, 1965; Rinkel *et al.*, 1966].

It is perhaps not too early to hypothesize a general mechanism to explain the distribution of CO_2 in surface water, even though we cannot judge it adequately without more knowledge of water motion, chemical concentrations, and biological activity of ocean water. Plants withdraw CO_2 from surface water during photosynthesis. Decaying organisms sink below the thermocline and release CO_2 to the subsurface water. Surface waters are thus persistently depleted of the CO_2 , whereas the deep waters are enriched. Where upwelling or vertical mixing promotes upward transport of CO_2 from the deep water, we encounter surface concentrations higher than equilibrium. Conversely,

where physical transport by the water is slight, we may expect CO_2 concentrations in the surface water to be less than equilibrium with the atmosphere. North-south surface currents which promote warming or cooling of surface water modify the distribution, because the CO_2 concentration in ocean water depends on temperature and because temperature equilibration with the atmosphere proceeds more rapidly than CO_2 equilibration. The data from Lusiad Expedition are to a high degree consistent with this proposed mechanism, which is discussed in greater detail in the article that follows [Keeling, 1968].

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