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Distribution and variations of oceanic carbon dioxide in the western North Pacific, eastern Indian, and Southern Ocean south of Australia

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ABSTRACT

The partial pressure of carbon dioxide (pCO_2) in surface seawater was measured to clarify its time and space variation in the western North Pacific, eastern Indian and Southern Ocean south of Australia during the Southern Cross Cruise in 1968/69, the BIOMASS Cruise in 1983/84 and the Ryofu-maru Cruises in 1984 and 1987. The pCO_2 in surface seawater was high in high latitudes, coastal and equatorial regions and low in the subtropics. The spatial variation of pCO_2 clearly reflects the general circulation of seawater, i.e., vertical mixing of seawater plays an important rôle in determining the pCO_2 in surface seawater.

Two kinds of periodicities during the annual cycle, which are caused by the variations in seawater temperature, vertical mixing and biological activities, were found off Japan's coast. The pCO_2 in surface seawater obtained in the boreal winters of 1969 and 1984 suggests an increase due to the enhancement of CO_2 flux from air to sea. The apparent increase was 20.0 ± 10.6 ppm in the western North Pacific, and 24.4 ± 8.3 ppm in the Southern Ocean, south of Australia.

1. Introduction

Carbon dioxide is a trace gas in the air which is increasing at a rate of 1.5 ppm yr^{-1} (Gammon et al., 1985). This increase appears to be one of the major factors used to determine the Earth's climate. In order to understand the present increase and predict increase in the future, it is necessary to clear up the changing carbon cycle among global carbon reservoirs; atmosphere, ocean and biosphere.

The exchange of CO_2 between the air and ocean is one of the important processes to be investigated in detail. Air/sea interaction clearly has an influence on the rate of increase of atmospheric CO_2. The El Niño event, which occurs every few years, is considered to be related to the variations of the increase rate of atmospheric CO_2 (Bacastow, 1976; 1977a,b; Komhyr et al., 1985). But how El Niño events influence the atmospheric CO_2 increasing rate is a matter of controversy (Machta et al., 1977; Newell and Weare, 1977; Newell et al., 1978; Bacastow et al., 1980).

During the 1982/83 El Niño event, the distribution of pCO_2 in the surface seawater was greatly altered in the western North Pacific; pCO_2 in the area from 7°N to 1°S along 137°E (Equatorial Counter Current and South Equatorial Current) increased with the surface seawater temperature (SST) decrease, whereas from 7°N to 17°N along 137°E (North Equatorial Current) it decreased with SST (Inoue et al., 1987). On the other hand, Feely et al. (1987) have reported that, during 1982/83, El Niño event pCO_2 in the central equatorial Pacific decreased to the levels almost equivalent to those of the atmospheric CO_2 due to the cessation of upwelling. These results suggest the necessity for depicting the large-scale distribution of pCO_2 in the surface seawater.

Generally speaking, surface seawater is

supersaturated with respect to the air CO_2 in the equatorial region, and undersaturated in the subtropics, apart from upwelling areas. However, little is known of the pCO_2 in the surface seawater in high latitudes and coastal seas. The measurements of pCO_2 in both areas are interesting, because the former is the area where intermediate and deep waters are ventilated directly to the atmosphere, and the latter has been related to the CO_2 flux measured by the eddy correlation method (Smith and Jones, 1985).

The time variation of pCO_2 in surface seawater are also required for the quantitative understanding of the CO_2 exchange between air and sea. Short-term and long-term variations due to changes in temperature, vertical mixing, biological activity, CO_2 invasion etc., should also be characterized. Seasonal variations in pCO_2 have been reported only in a limited number of areas. The annual cycle of pCO_2 in the area off Japan, high in summer and low in winter (Inoue et al., 1987), has a much larger amplitude than that of the air (Tanaka et al., 1987).

Secular variation in the pCO_2 in surface seawater is considered to be direct evidence for fossil fuel CO_2 absorption by the ocean. There are only a few reports concerning the secular trend of pCO_2 in the surface seawater (Roos and Gravenhorst, 1984, Takahashi et al., 1983). There seems to be an ambiguity concerning its magnitude. Difficulties in characterizing the short-term variations and different CO_2 measurement methods employed lead to a rather scattered estimation.

Time and spatial variations in the pCO_2 in the surface seawater are now needed to determine the CO_2 exchange between air and sea more quantitatively. This paper reports the distribution of surface seawater pCO_2 in the western North Pacific, eastern Indian and Southern Ocean south of Australia, and compares it with data obtained previously.

2. Observation

The pCO_2 in surface seawater and air was measured on board the R.V. *Hakuho-maru* belonging to the Ocean Research Institute, University of Tokyo, and on board the R.V. *Ryofu-maru* belonging to the Japan Meteorological Agency. The ship tracks of R.V. *Ryofu-maru* and R.V. *Hakuho-maru* are shown in Fig. 1. The Southern Cross Cruise in 1968/69 consisted of 5 legs. Here, we report the results of pCO_2 measurements during leg 3, from Wellington to Sydney and leg 4, from Sydney to Tokyo. The BIOMASS Cruise in 1983/84 was divided into 5 legs. Leg 1 was from Tokyo to Sydney, leg 2 from Sydney to Hobart, leg 3 from Hobart to Fremantle, leg 4 from Fremantle to Cebu, leg 5 from Cebu to Tokyo. The pCO_2 in the surface seawater of the western North Pacific was measured in the winter season from 1981 to 1987 on board the R.V. *Ryofu-maru*. The R.V. *Ryofu-maru* travelled from Tokyo to Naha via 1°S, 137°E, then back to Tokyo. Seasonal variations of pCO_2 in surface seawater studied in the same area (32°N–29°N, 134°E–138°E) are reported by Inoue et al. (1987).

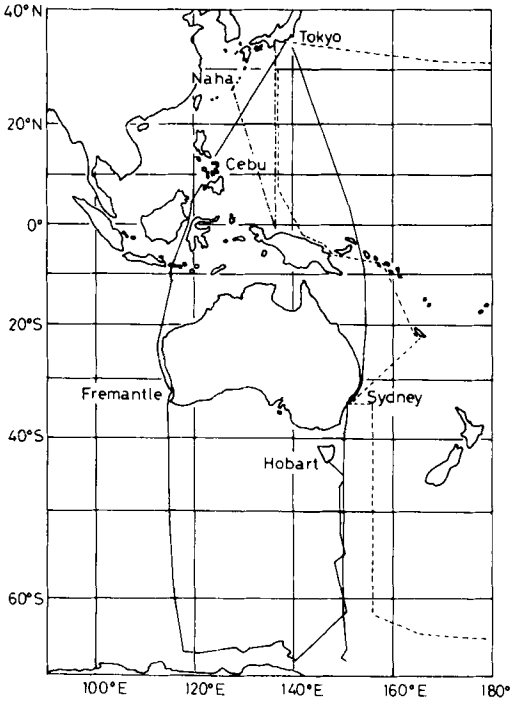

Fig. 1. The solid line shows the BIOMASS cruise of the R.V. *Hakuho-maru* (Tokyo, 24 Nov., 1983–Tokyo, 24 Feb., 1984), the dashed line the Southern Cross cruise of the R.V. *Hakuho-maru* (Tokyo, 14 Nov., 1968–Tokyo, 3 March, 1969), and the dashdotted line the RY8401 cruise of the R.V. *Ryofu-maru* (Tokyo, 20 Jan., 1984–Naha, 8 Feb., 1984).

2.1. Method

The measurement of the pCO_2 in surface seawater was carried out along with that of the air on board the ship as described earlier (Miyake et al., 1974; Inoue and Sugimura, 1986; Inoue et al., 1987). Here, we describe the experimental procedure employed at present, which was modified slightly in January 1987.

The data logger and acquisition system (7V-14 Nihon Denki Sanei, Japan and HP 85B Hewlett Packard USA) were introduced to collect information from the non-dispersive infra-red gas analyzer (NDIR Model 864 or 865, Beckman USA) and thermoelectric thermometer (Pt 100 ohm). The primary record was obtained digitally on a floppy disk, with stripcharts serving as a back-up.

2.2. Measurement of pCO_2 in surface seawater

Seawater was pumped up continuously from the bottom of the ship (about 5 m below the surface) and introduced into an equilibrator. The seawater temperature at the water inlet was recorded on a recorder chart, and the seawater temperature at the CO_2 equilibrator was recorded digitally on the floppy disk.

24 min were allowed in each hour for the observation of 4 standard gases, 18 min for measuring the pCO_2 in the air, 17 min for measuring the pCO_2 in surface seawater, and 1 min for data file creation. To obtain the measurements of CO_2 in the standard gases and background air, each gas was introduced into the NDIR's sampling cell for 4.5 min, and after that, gas flow was stopped. Signal integration was commenced 1 min after a 30-s equilibration time. During the time of CO_2 measurement in the background air, air for measuring pCO_2 in the surface seawater came into contact with the seawater in advance. After the measurement of CO_2 in the background air, air for measuring the pCO_2 in surface seawater was circulated for 15.5 min in a closed circuit consisting of NDIR, a dessicant system, and the CO_2 equilibrator. Signal integration was commenced 1 min after the equilibration time of 30 sec. All the mesurements were carried out at atmospheric pressure. The instrument was designed to keep a constant value of output voltage for several min, even in an environment of high CO_2 concentration. Linear interpolation was used to evaluate any drift in the analyzer response between successive calibrations. The pCO_2 value in the air and seawater was then calculated according to the quadratic least squares.

Correction was made using eq. (1), due to a temperature rise in the inner piping (Gordon and Jones, 1973):

$$\frac{\delta pCO_2}{\delta T} = 4.4 \times 10^{-2}(pCO_2) - 4.6 \times 10^{-6}(pCO_2)^2, \quad (1)$$

where T is the water temperature.

3. Estimation of the pCO_2 in surface seawater during the Southern Cross Cruise

Taking into account the ship track of the Southern Cross Cruise in 1968/69, we estimated the pCO_2 in surface seawater in the region from 44°S to 62°S along 155°E and 9°N to 24°N along 138°E. The observation data of the Southern Cross Cruise was given as the difference in pCO_2 between the seawater and the atmosphere. The CO_2-in-N_2 calibration gases were used during the Southern Cross Cruise. Therefore, carrier-gas effect on an analyser (Beckman 315A, USA) was determined by measuring CO_2-in-air gases of known concentration with CO_2-in-N_2 gases. Because we did not have CO_2-in-N_2 gases prepared by the gravimetric method, we have adopted the value determined by the Takachiho Kagaku Co Ltd., who use CO_2-in-N_2 gases prepared by the gravimetric method. The quadratic least squares, when fitted to the data, yield the following:

$$pCO_2(air) = -18.54 + 1.140pCO_2(N_2) - 2.198 \times 10^{-4}(pCO_2(N_2))^2, \quad (2)$$

where pCO_2(air) is the CO_2 concentration determined by the CO_2-in-air gases, and $pCO_2(N_2)$ is the apparent CO_2-in-air concentration determined by the CO_2-in-N_2 gases. In the first place, the pCO_2(air) in the atmosphere was estimated in order to obtain the pCO_2(air) in surface seawater. The meridional distribution of the pCO_2 expressed as the difference from the South Pole concentration was assumed to be equal to that of Bolin and Keeling (1963), and obtained monthly by data plots for the period from January 1969 to March 1969. We

added this value to the concentration of the South Pole determined by CO_2-in-air gases (Bacastow and Keeling, 1981). Thus, pCO_2(air) in the atmosphere during the Southern Cross Cruise could be estimated by the interpolation of these data.

From eq.(2), the $pCO_2(N_2)$ in the atmosphere during the Southern Cross Cruise was calculated. By adding the $pCO_2(N_2)$ in the atmosphere to the observed difference between seawater and atmosphere, the $pCO_2(N_2)$ in surface seawater could be obtained. The pCO_2(air) in surface seawater was calculated from eq.(2) and corrected for the temperature effect on pCO_2 by eq.(1). The temperature increase in the inner piping was assumed to be the latest average value of Hakuho-maru, 0.3°C.

4. Results and discussion

4.1. Distribution of pCO_2 in surface seawater

It is quite useful to measure the pCO_2 in surface seawater, because by this means, the estimation of the CO_2 exchange between air and seawater is made possible. The CO_2 exchanges between the atmosphere and the ocean are taken as proportional to the difference in pCO_2 (Deacon 1977; Smethie et al., 1985; Andrie et al., 1986). Time and spatial variations in the air pCO_2 were reported in detail (Komhyr et al., 1985), while those of pCO_2 in seawater still lack sufficient data to calculate the CO_2 exchanges between the atmosphere and the ocean. Figs. 2, 3 show the meridional distribution of the pCO_2 in surface seawater observed during the BIOMASS Cruise (Fig. 1). Both figures clearly represent high pCO_2 values in high latitudes, coastal and tropical regions, and low in the subtropics. A peak of pCO_2 in the Coral Sea (10°S 155°E) was caused by a red-tide outbreak of *Tricho desmium*. Carbon dioxide derived from the oxidation of organic material accumulated in the surface weawater, and resulted in high pCO_2.

Three major ocean fronts were observed south of 40°S (Nakai et al., 1986). The subtropical convergence, which is marked as a southern limit of subtropical surface water, was located at 47°S 150°E. Along 115°E, the location of the subtropical convergence could not be defined clearly, but might be at 43.5°S (Nakai et al., 1986). The subantarctic front, characterized by a steep horizontal temperature gradient in the layer between the depth of 200 m and 1000 m, was found at 49°S 150°E and 47.5°S 115°E. The polar front, characterized by the steep temperature gradient of the surface seawater and the northern limit of subsurface temperature minimum (about 2°C), was found at 56.5°S 150°E and 55°S 115°E. According to our observation, the pCO_2 became higher than that of the air to the south of the subantarctic front. Direct ventilation of the intermediate and deep sea to the air has been considered to play an important rôle for the ocean uptake of excess CO_2. Seawater south of the subantarctic front is shown here to release CO_2 into the air during early summer. Because seasonal variation in high latitudes have not been studied yet, whether or not the sea acts as a source throughout the year remains undetermined.

Mean values of the pCO_2 in each zone are listed in Table 1 along with SST and the concentration of nutrients. We conclude that vertical mixing of seawater plays an important rôle in determining the pCO_2 in the surface seawater, and overcomes the temperature effect on pCO_2 due to horizontal mixing.

South of the subtropical convergence, there was only a small difference between 150°E and 115°E, while pCO_2 in the surface seawater from 43.5°S to 40°S which is probably north of the subtropical convergence along 115°E, was about 40 ppm higher than that of 150°E. High pCO_2 with relatively low SST in the north of 43.5°S was probably caused by the northward flow of surface seawater (i.e., the West Australian current).

The pCO_2 off the western coast of Australia was clearly high in comparison with that off the eastern coast. One may expect the effect of coastal upwelling to occur at the eastern boundaries. However, upwelling along the western coast of Australia was not clearly ascertainable (Deacon, 1963). The pCO_2 in surface seawater increased with the temperature rise and an apparent temperature dependence of the pCO_2 was calculated to be $2\%°C^{-1}$. Warming of surface seawater along the Australian continent could explain the increase of pCO_2 if coastal upwelling is suppressed. The relationship between water temperature and pCO_2 is different from eq.(1), which was probably due to CO_2 consumption by biological activity.

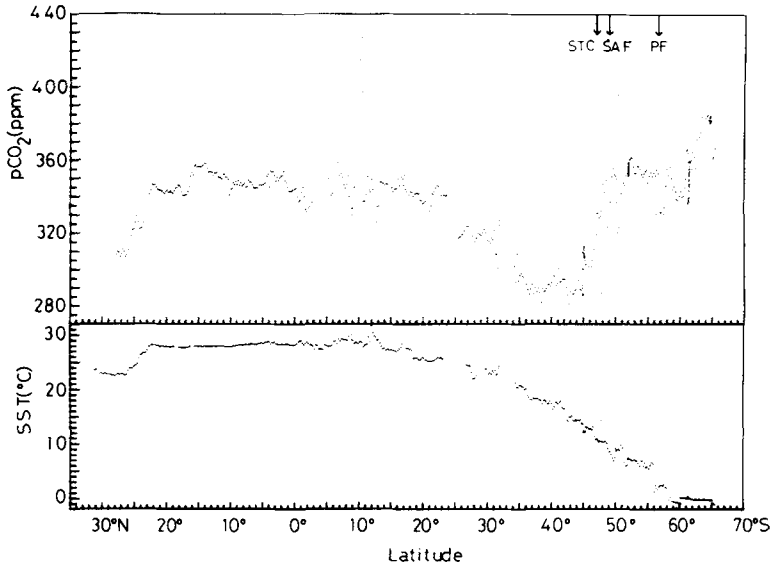

Fig. 2. Meridional distribution of the pCO_2 in the surface sea water during legs 1, 2, and 3 (along 150°E) of the BIOMASS Cruise. STC stands for subtropical convergence, SAF for subantarctic front and PF for polar front.

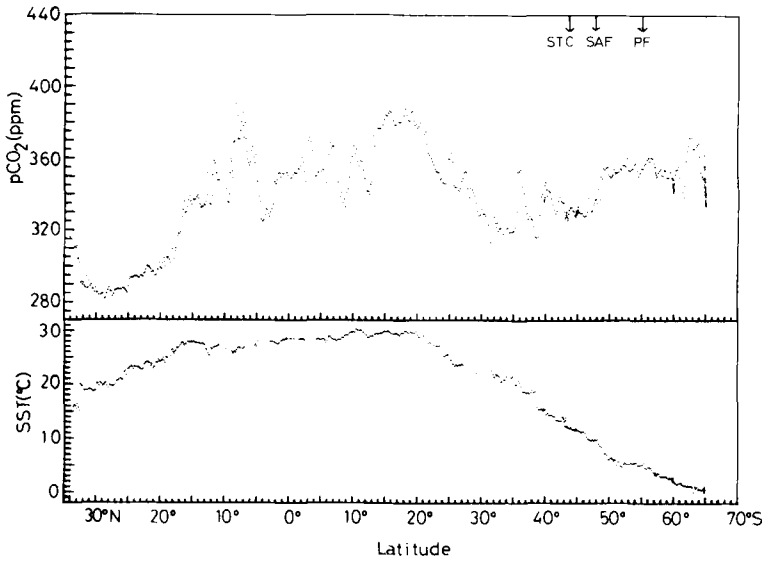

Fig. 3. Meridional distribution of the pCO_2 in the surface seawater during legs 3 (along 115°E), 4, and 5 of the BIOMASS Cruise. STC stands for subtropical convergence, SAF for subantarctic front and PF for polar front.

The pCO_2 in the surface seawater varied from 328.5 ppm to 414.0 ppm in the Java Sea, Celebes Sea and Sulu Sea. Large variations in pCO_2 may be a characteristic of coastal sea, because we also found a large variability of pCO_2 in the coastal area off Japan.

4.2. Time variations in pCO_2 in surface seawater

Continuous measurements of atmospheric CO_2 were begun in 1958 (Pales and Keeling, 1965), and now precise and continuous measurements are conducted all over the world. Meanwhile,

Table 1. *Mean values of the pCO_2 in surface seawater; the area is divided based on the oceanographic fronts; STC means the subtropical convergence, SAF the subantarctic front and PF the polar front*

Date	Latitude	Longitude	pCO_2 (ppm)	SST (°C)	n	NO_3^- (μmol dm^{-3})*
12–13 Dec., 1983	40°S–44°S	150°E	291.4 ± 4.5	15.9 ± 1.4	25	—
13–15 Dec., 1983	44°S–47°S (STC)	150°E	301.4 ± 12.4	13.0 ± 0.8	39	1.9
15 Dec., 1983**	47°S–49°S (SAF)	150°E	332.6 ± 8.7	10.6 ± 1.1	13	—
15–17 Dec., 1983**	49°S–56.5°S (PF)	150°E	350.0 ± 9.0	6.9 ± 1.4	57	17.1
17–23 Dec., 1983**	56.5°S–65°S	150°E	356.0 ± 16.5	0.0 ± 0.7	87	24.2, 30.7
23–27 Dec., 1983**	56.5°S (PF)–65°S	150°E	355.8 ± 13.2	0.5 ± 0.8	136	24.4
27–30 Dec., 1983**	49°S (SAF)–56.5°S	150°E	348.5 ± 7.2	6.6 ± 1.9	77	22.2, 22.7
30–31 Dec., 1983**	47°S (STC)–49°S	150°E	328.6 ± 4.3	10.1 ± 0.3	9	—
31 Dec., 1983–1 Jan., 1984	44°S–47°S	150°E	305.9 ± 12.5	12.7 ± 1.2	36	7.8
8–9 Jan., 1984	44°S–47°S (STC)	150°E	310.3 ± 8.4	13.2 ± 1.3	21	—
9 Jan., 1984**	47°S–49°S (SAF)	150°E	334.0 ± 8.3	9.9 ± 0.5	11	—
9–11 Jan., 1984**	49°S–56.5°S (PF)	150°E	342.9 ± 10.4	7.7 ± 1.8	54	—
11–13 Jan., 1984**	56.5°S–65°S	150°E	353.7 ± 7.4	1.3 ± 1.0	49	—
13–18 Jan., 1984	63°S–65°S	115°E–150°E	352.1 ± 14.2	0.0 ± 0.6	79	28.8
18–23 Jan., 1984	55°S (PF)–65°S	115°E	348.9 ± 9.2	1.9 ± 1.3	106	25.1, 25.7, 26.5
23–25 Jan., 1984	47.5°S (SAF)–55°S	115°E	350.5 ± 6.4	6.5 ± 1.6	37	22.6
25–27 Jan., 1984	43.5°S (STC)–47.5°S	115°E	329.9 ± 2.5	11.4 ± 0.6	55	9.5
27–28 Jan., 1984	40°S–43.5°S	115°E	334.7 ± 5.6	14.0 ± 1.1	31	4.2, 4.4

Data is represented along with 1σ, and n means the number of measurements.
* Concentration at the depth of 0m observed at the hydrocast station.
** Inoue and Sugimura (1986).

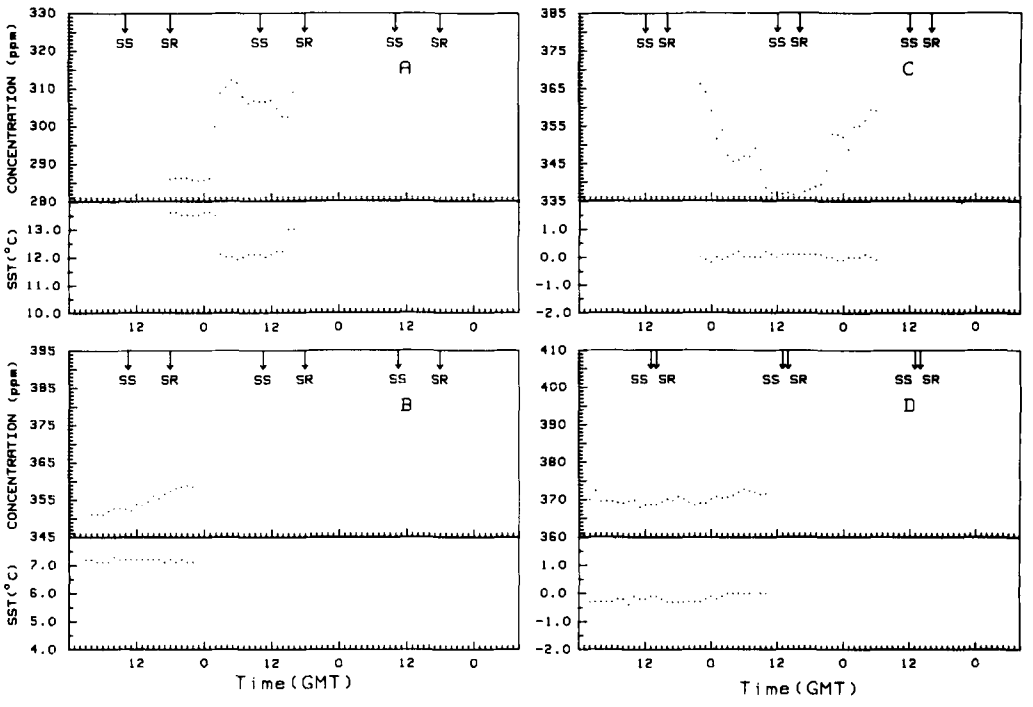

Fig. 4.

time variations in the pCO_2 of surface ocean waters have not yet been characterized. Short-term and long-term variations must be studied, in order to estimate the future increase of atmosphere CO_2 precisely.

4.3. Short-term variation of the pCO_2 in surface seawater

The diurnal variation of pCO_2 in surface seawater was studied at the main hydrocast stations during the BIOMASS Cruise (Figs. 4 and 5). Regular periodicities related to the tide and solar radiation were not observed clearly. Large variations, ranging from 285 ppm to 312 ppm, occurred with the SST change at 45°S 150°E. High pCO_2 was accompanied by relatively low SST. At the other stations, variations in pCO_2 did not correlate well with the SST change. Large fluctuations of pCO_2 appeared in the Antarctic zone, which could be connected with the upwelling and biological activity at the Antarctic divergence.

We have reported that the pCO_2 is high in summer and low in winter off the Japan coast (Inoue et al., 1987). We now report more data in the same region (Fig. 6, Table 2). The long-term shift was removed by assuming a linear time trend, which is estimated to be 1.4 ppm yr^{-1}, as described in the section on secular variation. There are at least 2 kinds of regular periodicities.

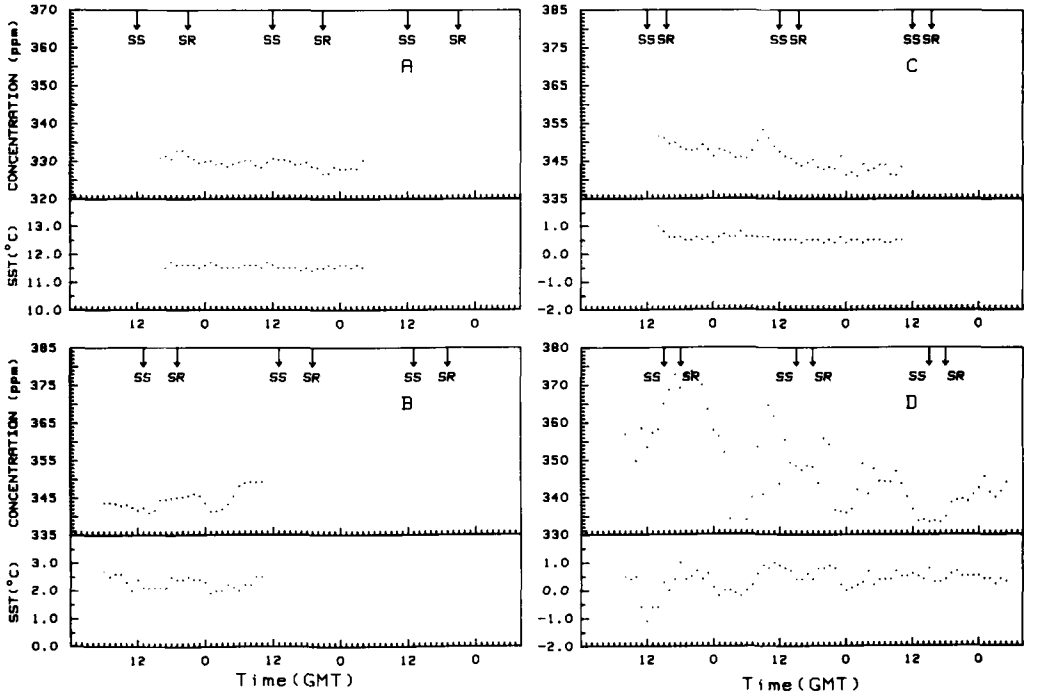

Fig. 5. Diurnal variation of the pCO_2 in surface seawater along 115°E and 150°E during the BIOMASS Cruise. SR stands for sunrise and SS for sunset.
A: 44.5°S 115°E. From 25 Jan., 1984 to 27 Jan., 1984.
B: 60°S 115°E. From 21 Jan., 1984 to 22 Jan., 1984.
C: 61.5°S 150°E. From 24 Dec., 1983 to 26 Dec., 1983.
D: 65°S 115°E. From 17 Jan., 1984 to 20 Jan., 1984.

Fig. 4. Diurnal variation of the pCO_2 in surface seawater along 150°E during the BIOMASS Cruise. SR stands for sunrise and SS for sunset.
A: 45°S 150°E. From 13 Dec., 1983 to 14 Dec., 1983.
B: 52°S 150°E. On 16 Dec., 1983.
C: 61.5°S 150°E. From 18 Dec., 1983 to 20 Dec., 1983.
D: 65°S 150°E. From 22 Dec., 1983 to 23 Dec., 1983.

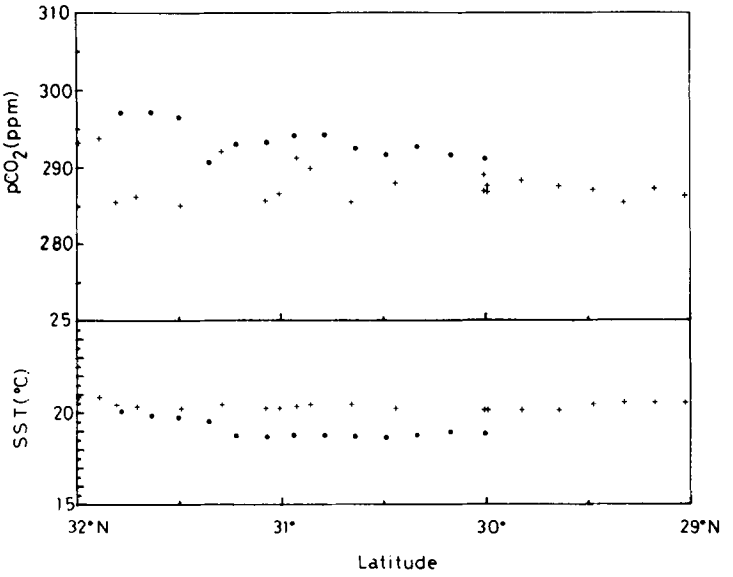

Fig. 6. Seasonal variation of the pCO_2 in surface seawater off Japan's coast (32°N–29°N, 134°E–138°E). ●: 14 Jan., 1987, +: 1 March, 1987.

Table 2. *Seasonal variation of the pCO_2 in surface seawater (29°N–32°N, 134°E–138°E)*

pCO_2 (ppm)	No	pCO_2^{TR} (ppm)	SST (°C)	Date
287.8 ± 2.5	22	283.5	20.3 ± 0.2	19 Jan. 1985*
285.7 ± 1.9	23	284.2	19.9 ± 0.3	16 Jan. 1987
292.2 ± 2.2	14	292.0	18.9 ± 0.3	22 Feb. 1984*
293.6 ± 2.2	13	289.2	19.1 ± 0.5	1 March, 1987
332.0 ± 1.2	9	329.9	24.5 ± 0.8	29 June, 1985*

pCO_2^{TR} means the long-term removed value. We assume the linear time trend and set $t = 0$ for the beginning of 1984.

* Inoue et al., (1987).

One of them shows the increase of the pCO_2 with SST decrease (Fig. 6). The pCO_2 in March was clearly high, irrespective of the low SST, in comparison with that of January. According to the oceanographic data in this area (JMA, 1983, 1984, 1985, 1986), the concentration of NO_3^- has increased 0.47 ± 0.31 μmol dm^{-3} ($n = 4$) for the period from the end of January to the end of February. This means that enhancement of the vertical mixing in winter overcomes the decrease of pCO_2 due to temperature lowering and carbon uptake by the biological activity. In the middle of July, the concentration of NO_3^- in the surface seawater is about 0.01 μmol dm^{-3}, which decreased by two orders of magnitude in comparison with that in February. The difference of the temperature dependence during the annual cycle from eq. (1) is caused by the variations in the vertical mixing and biological activity. Two kinds of regular periodicities clearly exist, though their amplitudes and phases are not yet determined. The seasonal variation mentioned above is different from that in the tropical region of the North Pacific (15°N–20°N) reported by Weiss et al. (1982). The apparent temperature dependence is calculated to be 3.1% °C^{-1}. The amplitude and phase of the pCO_2 variations in surface seawaters hitherto reported (Takahashi et al., 1983, Takahashi, 1987) vary remarkably from place to place.

4.4. Secular variation

It is very important to investigate the long-term variation of the pCO_2 in surface seawater in order to predict the future increase of the atmospheric CO_2. Following the increase of atmospheric CO_2, the net flux from air to sea is expected to increase. However, there are only few data on the ocean's response, based on observation.

One of the authors (YS), has measured the difference in pCO_2 between the atmosphere and the seawater during the Southern Cross cruise in 1968 and 1969 (Fig. 1). The ship track was fairly close to the BIOMASS Cruise in the southern hemisphere, and very close to the *Ryofu-maru* cruise in the northern hemisphere. The method adopted in 1968/69 was basically the same as the present one. The pCO_2 in the surface seawater during the Southern Cross cruise was calculated as described earlier.

An El Niño event occurred in 1969, and affected the increasing rate of atmospheric CO_2 (Bacastow et al., 1980). The variation of pCO_2 in the equatorial Pacific during the El Niño Event has been considered to perturb the rate of CO_2 increase in the air (Bacastow et al., 1980 and Feely et al., 1987). Therefore, in order to elucidate the long-term variation based on data of the Southern Cross Cruise, it is necessary to consider the changes of pCO_2 in surface water in low latitudes.

We have examined the distribution of pCO_2 in the surface water of the western North Pacific in winter seasons since 1981 (Fushimi 1987, Inoue et al., 1987). There have been two El Niño Events since 1981. During the major El Niño Event in 1982/83, which started in June, the pCO_2 in surface seawater was greatly disturbed, as was SST. The pCO_2 increased with SST decrease from 1°S to 7°N along 137°E (equatorial counter current and south equatorial current) and decreased with SST from 7°N to 17°N along 137°E (north equatorial current).

In January 1987, which was the period of the beginning of 1986/87 El Niño Event, anomalies of pCO_2 and SST were relatively small, especially in the region of the north equatorial current. From these results, the anomaly of pCO_2 in January and February 1969 (the beginning of the 1969 El Niño Event) was judged to be small in the area reported here.

In the southern hemisphere, the pCO_2 value observed in 1983/84 was much higher than that of the Southern Cross cruise (Fig. 7), although the

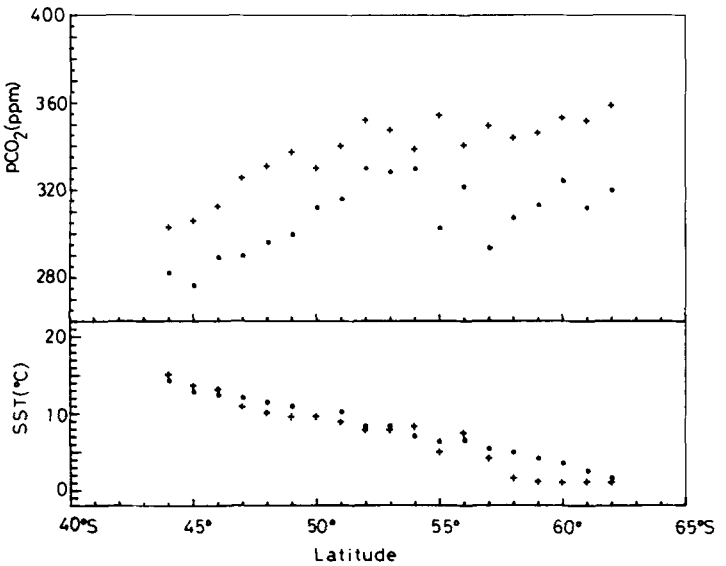

Fig. 7. Secular trend of the pCO_2 in the surface seawater of the Southern Ocean, south of Australia. + : BIOMASS cruise, ● : Southern Cross cruise.

Table 3. *Mean values of the pCO_2 in the surface seawater of the Southern Ocean observed in 1969 and 1984*

	25–29 Jan., 1969			8–13 Jan., 1984		
Latitude (S)	pCO_2 (ppm)*	SST (°C)*	No.	pCO_2 (ppm)*	SST (°C)*	No.
44	282.5 ± 5.8	14.5 ± 0.3	9	302.6 ± 3.0	15.0 ± 0.3	3
45	276.6 ± 11.5	13.0 ± 0.5	5	305.6 ± 6.5	13.6 ± 0.9	8
46	289.1 ± 5.5	12.7 ± 0.3	5	312.2 ± 4.7	13.1 ± 0.3	6
47	290.3 ± 5.1	12.3 ± 0.3	6	325.3 ± 4.0	10.8 ± 0.7	7
48	296.3 ± 5.7	11.6 ± 0.1	7	330.6 ± 0.8	10.0 ± 0.1	5
49	299.9 ± 2.4	11.1 ± 0.1	6	337.0 ± 11.7	9.5 ± 0.5	8
50	312.0 ± 3.8		7	329.7 ± 4.8	9.6 ± 0.1	6
51	316.1 ± 2.6	10.4 ± 0.1	8	339.8 ± 5.7	8.9 ± 0.6	6
52	330.1 ± 4.6	8.1 ± 0.6	5	351.9 ± 5.0	7.8 ± 0.2	6
53	328.6 ± 2.6	8.2 ± 0.1	7	347.2 ± 6.2	7.8 ± 0.2	5
54	330.2 ± 4.4	7.3 ± 0.5	8	338.3 ± 1.7	8.3 ± 0.1	5
55	302.5 ± 3.6	6.5 ± 0.3	5	354.2 ± 7.6	4.9 ± 1.7	11
56	322.0 ± 14.0	6.9 ± 0.4	5	340.2 ± 4.5	7.4 ± 0.3	10
57	294.0 ± 9.0	5.5 ± 0.3	7	349.3 ± 1.3	4.1 ± 2.3	3
58	307.6 ± 11.8	5.0 ± 0.4	5	343.5 ± 2.2	1.5 ± 0.5	4
59	313.0 ± 11.9	4.2 ± 0.3	4	345.8 ± 4.2	1.1 ± 0.1	5
60	324.8 ± 4.7	3.6 ± 0.2	8	353.0 ± 5.4	1.0 ± 0.1	9
61	312.0 ± 10.6	2.8 ± 0.2	5	351.7 ± 2.9	1.1 ± 0.1	7
62	320.1 ± 10.0	1.4 ± 0.4	3	358.6 ± 6.4	1.0 ± 0.2	17

* Data are presented along with 1σ.

short-term variation and longitudinal difference must be considered in order to ascertain the long-term trend.

Hydrographic data from 44°S to 55°S indicate no remarkable difference of seawater structure according to longitude (between 155°E and 150°E). And mentioned above, pCO_2 was nearly equal between 150°E and 115°E in the south of the subtropical convergence. Therefore, it is possible to compare the data between 150°E and 155°E without taking into account the longitudinal variation (Table 3). South of 55°S, SST decreased gradually along 155°E, but steeply along 150°E. The pCO_2 from 55°S to 44°S was observed during the periods from January 25 to January 31 in 1969 and from January 8 to January 13 in 1984. As reported previously, the pCO_2 along 150°E was found to be almost constant for about one month, starting from the middle of December (Table 1). Therefore, the contribution of the short-term variation and longitudinal variation to the observed difference in pCO_2 between 1969 and 1984 is expected to be negligible. During the perod from 1969 to 1984, the increase of the pCO_2 in the surface seawater ranged from 8.1 ppm to 37.1 ppm, with a mean value of 24.4 ppm (Table 3).

In the northern hemisphere, we have compared the data of the Southern Cross Cruise (138°E) with that obtained along 137°E in 1984. We have been conducting pCO_2 observations regularly since 1981 (Fushimi 1987, Inoue et al., 1987). The pCO_2 in 1969 and 1984 are presented along with SST in Fig. 8. The pCO_2 in surface seawater in 1984 is clearly higher than that achieved in 1969. The difference in the pCO_2 from 9°N to 24°N lies between 2.5 ppm and 37.3 ppm, and the mean value is 20.0 ppm. Observation of the Southern Cross cruise was conducted in late February, and that of the *Ryofu-maru* in late January. As mentioned above, little is known about the short-term variation. Weiss et al. (1982) have reported the seasonal variation in the tropical region of the North Pacific (15°N–20°N). As described above, the apparent temperature dependence is $3.1\%°C^{-1}$. Therefore, only the data between 15°N and 20°N are corrected by the assumption of temperature dependence of

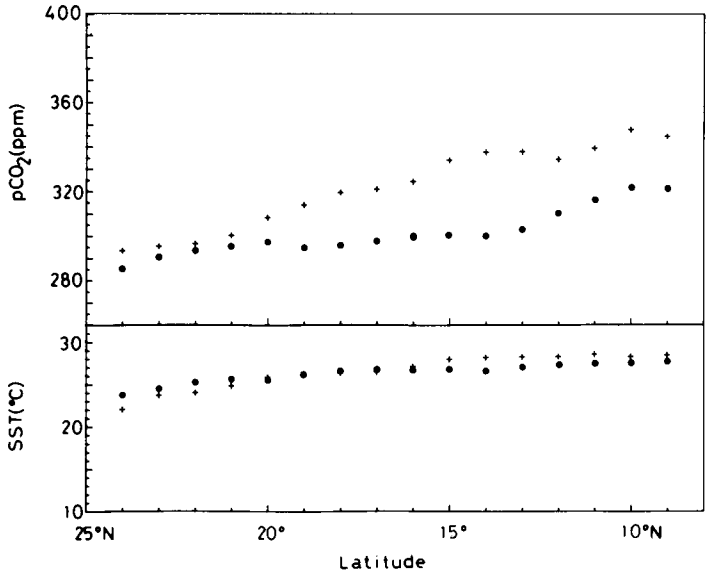

Fig. 8. Secular trend of the pCO_2 in the surface seawater of the western North Pacific. +: RY8401 cruise, ●: Southern Cross cruise.

Table 4. *Mean values of the pCO_2 in surface seawater of the western North Pacific observed in 1969 and 1984; the data of pCO_2 from 15° N to 20° N was corrected (pCO_2^c) by assuming the temperature dependence of 3.1% °C^{-1}*

	24–27 Feb., 1969				24–28 Jan., 1984		
Latitude (N)	pCO_2 (ppm)*	SST (°C)*	No.	pCO_2^c (ppm)	pCO_2 (ppm)*	SST (°C)*	No.
9	321.6 ± 1.3	27.6 ± 0.1	4		344.5 ± 1.5	28.4 ± 0.1	3
10	321.6 ± 1.3	27.6 ± 0.1	3		347.6 ± 2.6	28.2 ± 0.2	9
11	316.1 ± 1.9	27.5 ± 0.1	5		339.1 ± 2.1	28.5 ± 0.1	7
12	310.3 ± 1.0	27.3 ± 0.1	5		334.2 ± 1.1	28.2 ± 0.1	6
13	303.1 ± 1.8	27.1 ± 0.1	4		337.7 ± 4.2	28.2 ± 0.1	6
14	300.1 ± 1.0	26.6 ± 0.1	5		337.4 ± 2.5	28.1 ± 0.2	7
15	300.9 ± 0.1	26.7 ± 0.2	4	312.1	333.9 ± 2.3	27.9 ± 0.2	8
16	300.2 ± 1.2	26.8 ± 0.1	5	303.0	324.3 ± 3.7	27.1 ± 0.3	6
17	298.0 ± 1.0	26.7 ± 0.1	4	295.2	320.9 ± 1.2	26.4 ± 0.1	6
18	295.9 ± 0.7	26.5 ± 0.1	4	294.1	319.3 ± 3.1	26.3 ± 0.1	6
19	295.2 ± 0.9	26.2 ± 0.1	4	295.2	313.8 ± 1.5	26.2 ± 0.1	6
20	297.6 ± 0.9	25.6 ± 0.2	4	299.4	308.2 ± 2.8	25.8 ± 0.2	9
21	295.5 ± 1.5	25.7 ± 0.1	5		300.1 ± 4.5	24.8 ± 0.6	7
22	293.8 ± 0.4	25.2 ± 0.2	4		296.3 ± 1.2	23.9 ± 0.2	6
23	290.5 ± 1.7	24.5 ± 0.4	5		295.1 ± 3.8	23.6 ± 0.9	6
24	285.3 ± 0.4	23.8 ± 0.3	4		293.2 ± 4.6	22.0 ± 0.5	7

* Data are represented along with 1σ.

3.1%°C^{-1} (Table 4). Correction of the pCO_2 based on the temperature dependence of 3.1%°C^{-1} may be a tentative one, because amplitude and phase vary largely from place to place.

Whether the sea acts as a source or not, the pCO_2 in the surface seawater clearly increased. The linear trend approximation gives 1.4 ppm yr^{-1} to 1.6 ppm yr^{-1}, which is nearly equal to the atmospheric CO_2 increase (Gammon et al.,

1985). The increase of pCO_2 ranged from 20.3 ppm to 24.2 ppm during the period from 1969 to 1984. The increase of the pCO_2 in the surface seawater is caused by the transfer of fossil fuel CO_2 to the oceans. From the increase of the pCO_2 in the surface seawater, the amount of CO_2 accumulated in the mixed layer can be roughly estimated. The buffer factor is reported to be 14 in the cold region and 9 in the warm region (Siegenthaler, 1983) and the amount of inorganic carbon in the mixed layer 600 GT-C in the warm region and 300 Gt-C in the cold region (Bolin et al., 1979). The amount of CO_2 accumulated in the mixed layer per year is estimated to be less than 0.5 GT-C. This is the minimum of the amount of CO_2 invasion, because the amount of CO_2 transferred into the deep sea and the response of marine biology are not included. The long-term and short-term variation of the pCO_2 should be studied further in detail in order to understand the air/sea interaction more quantitatively.

5. Summary

Observations of pCO_2 in the surface seawater of the western North Pacific, eastern Indian, and Southern Ocean south of Australia were made during cruises of the Southern Cross (1968/69), the BIOMASS (1983/84), and the *Ryofu-maru* (1984, 1987). The pCO_2 in surface seawater was higher than that of the air in high latitudes, coastal and tropical regions, and lower in the subtropics. Vertical mixing of seawater always plays an important rôle in determining the pCO_2 in the surface.

Diurnal variation was not found clearly, but there exist at least 2 kinds of regular periodicities during the annual cycle off Japan's coast. Based on the data obtained in the boreal winter in 1969 and 1984, clear-cut evidence of an incremental increase of the pCO_2 in surface seawater was indicated. The apparent increase was 20.0 ± 10.6 ppm in the western North Pacific and 24.4 ± 8.3 ppm in the Southern Ocean south of Australia. The linear approximation of the long-term variation gives about 1.5 ppm yr^{-1} as the rate of increase of the pCO_2 in surface seawater, which is nearly equal to that of the atmospheric CO_2. In order to estimate the future secular trend of pCO_2 in surface seawaters more quantitatively, short-term variations should be studied further.

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