

# Variations and distributions of CO<sub>2</sub> in and over the equatorial Pacific during the period from the 1986/88 El Niño event to the 1988/89 La Niña event\*

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## ABSTRACT

Measurements of pCO<sub>2</sub> in the background air and seawater of the equatorial Pacific were made during the period from January 1987 to February 1989 to study variations in CO<sub>2</sub> flux between the sea and the air. In January and February 1987, which was within the period of 1986/88 ENSO event, the surface water pCO<sub>2</sub> of the central equatorial Pacific decreased due to the reduction of upwelling, but less dramatically than during the 1982/83 ENSO event. In January and February 1989, the pCO<sub>2</sub> of the central equatorial Pacific increased with the SST decrease, due to the enhancement of upwelling during the La Niña event. Changes in surface water pCO<sub>2</sub> along the equator were well approximated by the effect on pCO<sub>2</sub> of the temperature, salinity, biological activity and the CO<sub>2</sub> exchange between the sea and the air. In January and February 1987, the CO<sub>2</sub> flux in the equatorial Pacific was estimated to be 0.4 Gt-C yr<sup>-1</sup>, which was low compared to the non-ENSO periods, but remained positive. In January and February 1989, it increased to the level of 1 Gt-C yr<sup>-1</sup>, mainly due to the increase of surface seawater pCO<sub>2</sub> during the La Niña event. The large interannual variation in CO<sub>2</sub> flux could affect the growth rate of atmospheric CO<sub>2</sub>. For the period from January 31, 1987 to January 31, 1989, the concentration of atmospheric CO<sub>2</sub> has increased considerably (4.8 ppm). The  $\delta^{13}\text{C}$  of atmospheric CO<sub>2</sub> suggests the enhanced releases of CO<sub>2</sub> from the biosphere for the period between 1988 and 1987, and a relatively important role of air/sea CO<sub>2</sub> exchange between 1989 and 1988.

## 1. Introduction

The distribution and the growth rate of atmospheric CO<sub>2</sub> have been found to vary in association with the El Niño/Southern Oscillation (ENSO) phenomena (Bacastow, 1976, 1977; Machta et al., 1977; Bacastow et al., 1980; Keeling et al., 1985; Komhyr et al., 1985; Conway et al., 1988). A maximum peak of atmospheric CO<sub>2</sub> concentration near the equator disappeared during the 1982/83 ENSO event, and then returned to its normal magnitude by 1984 (Conway et al., 1988). During the 1982/83 ENSO period, high

atmospheric CO<sub>2</sub> growth rates in early 1983 followed low growth rate in 1982 (Conway et al., 1988; Tanaka et al., 1987a). The characteristic change in atmospheric CO<sub>2</sub> growth rate during the ENSO events correlates well with the Southern Oscillation Index (SOI). Time lags giving the maximum correlation between the atmospheric CO<sub>2</sub> growth rate and the SOI tend to be shorter near the equator and longer toward the poles (Machta et al., 1977; Conway et al., 1988), which suggests that the fluctuation of the atmospheric CO<sub>2</sub> content originates in the equatorial Pacific region.

It is not yet clear how the global atmospheric CO<sub>2</sub> content is altered by the ENSO events. Variations in CO<sub>2</sub> flux of the equatorial Pacific, which are proportional to the differences in pCO<sub>2</sub> between the sea and the air ( $\Delta\text{pCO}_2$ ) and local wind speed, have been considered to be one of the

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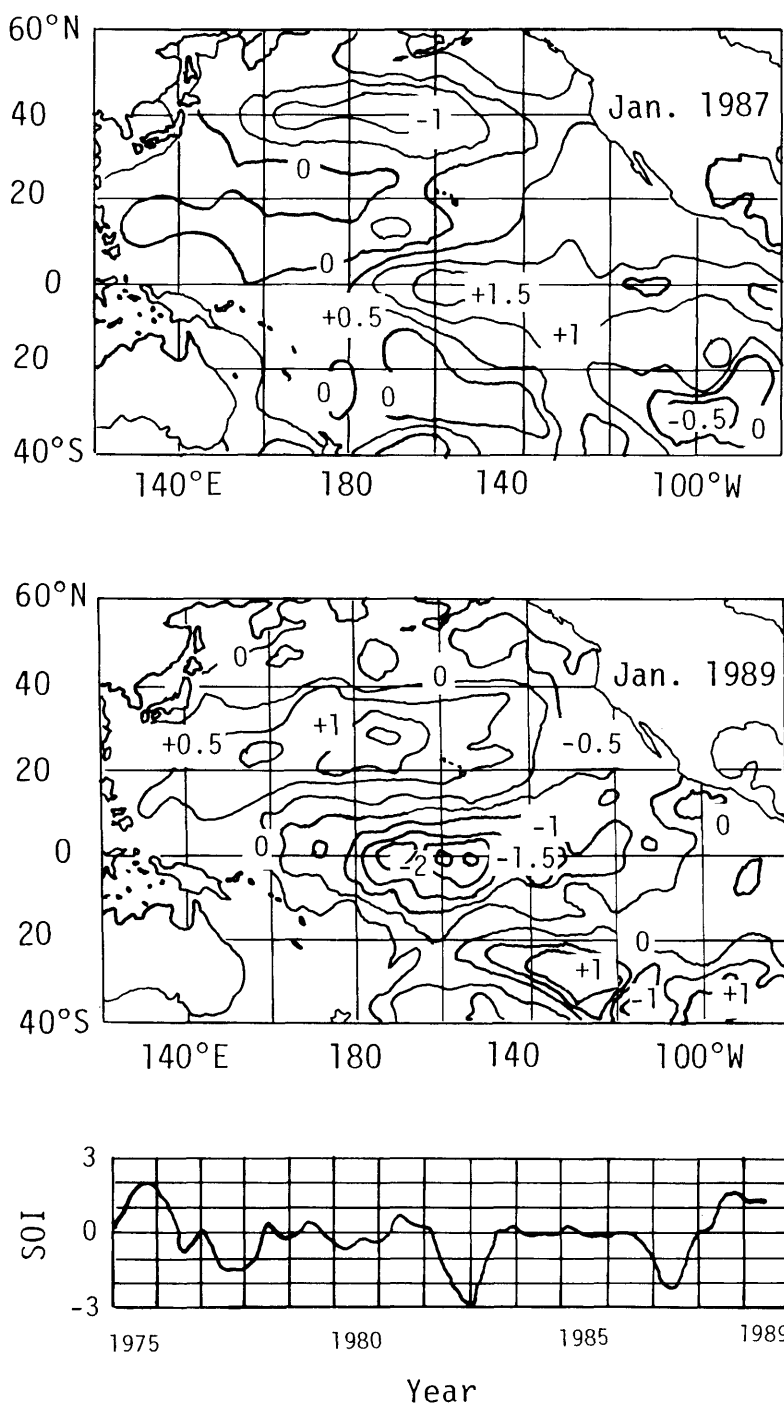


Fig. 1. SST anomalies in January 1987 and in January 1989. A: SST anomalies in January 1987 (MRCS, 1987a). B: SST anomalies in January 1989 (MRCS, 1989). C: SOI (MRCS, 1989).

major causes (Weare et al., 1976; Newell and Weare, 1977; Newell et al., 1978). We found that during the 1982/83 ENSO event (January 1983) the surface water pCO<sub>2</sub> from 7°N to 1°S along 137°E had increased by about 10–30 μatm along with the sea surface temperature (SST) decrease (Fushimi 1987, Inoue et al., 1987), due to the enhancement of vertical mixing in the western equatorial Pacific; however, Feely et al., (1987) have reported that during the 1982/83 ENSO event (April 1983) the surface water pCO<sub>2</sub> near the equator of the central and eastern equatorial Pacific decreased to near equilibrium with the atmosphere due to the cessation of upwelling in this region. Although large scale variations in pCO<sub>2</sub> take place during the ENSO event, the contribution of CO<sub>2</sub> flux from the sea to the air in the equatorial Pacific to the atmospheric CO<sub>2</sub> growth rate remains ambiguous. There are only limited air/sea pCO<sub>2</sub> data for the equatorial Pacific which could be utilized to determine the variability of CO<sub>2</sub> flux from the sea to the air on interannual timescales.

In mid-1986, positive anomalies of SST in the central and eastern equatorial Pacific became larger than +1°C, and extended from the central Pacific to the coast of South America by the end of year (1986/88 ENSO event). Fig. 1 shows the SST anomalies in January 1987 (MRCS, 1987a), when measurements of pCO<sub>2</sub> were conducted in the central and western equatorial Pacific. Following the end of 1986/88 ENSO events in March 1988, the SST in the eastern equatorial Pacific decreased rapidly, negative SST anomalies in the central and eastern equatorial Pacific became greater than –1°C, and the SOI increased to a level almost equal to the level in 1975 (Cold event or La Niña event, Fig. 1). The SST anomalies in January 1989 (Fig. 1) suggests a stronger than normal upwelling in the eastern and central equatorial Pacific between 120°W and 180° (MRCS, 1989a).

During the period from January 1987 to February 1989, shipboard measurements of pCO<sub>2</sub> in the air and surface seawater have been made. During these cruises, we have also collected air samples for post-cruise measurements of the carbon and oxygen isotopic ratio of atmospheric CO<sub>2</sub>. Here we report the pCO<sub>2</sub> distribution between 15°S and 15°N during the period from January 1987 to February 1989, and discuss the CO<sub>2</sub> flux out of equatorial Pacific along with sur-

face water pCO<sub>2</sub> distribution, and the concentration and δ<sup>13</sup>C of atmospheric CO<sub>2</sub>.

## 2. Sampling and analysis methods

### 2.1. Measurements of oceanic and atmospheric CO<sub>2</sub>

We have been measuring CO<sub>2</sub> concentration in and over the western North Pacific every boreal winter since 1981 (Inoue et al., 1987; Inoue and Sugimura, 1988a, b). During the period from January 1987 to February 1989, measurements of pCO<sub>2</sub> in the equatorial Pacific (Fig. 2) were made on board the R.V. Ryofu-maru, R.V. Natsushima, R.V. Kaiyo-maru, R.V. Hakuho-maru, M.V. Yashirogawa-maru and M.V. Wellington-maru along with atmospheric and hydrographic measurements. The data on which this paper is based are available in a summarized form on floppy disks from the authors and will be made available from the Japan Climatic Data Center, Japan Meteorological Agency (JMA) in the future.

The CO<sub>2</sub> concentration was determined in a manner similar to the methods reported earlier (Inoue et al., 1987). Therefore, a short description is given here. The data logger and acquisition system were introduced to collect information from the non-dispersive infra-red gas analyzer (NDIR analyzer, Model 864 or 865, Beckman) and thermometer (Pt-100 ohm) which was used to measure seawater temperature in an equilibrator. The NDIR analyzer was thermostated and the electric line of NDIR analyzer was divided into two parts; the electric voltage and frequency for units of chopper motor, detector, etc., were stabilized (Takasago 330AF) so as to avoid the effects of other instruments, and electric power for the temperature control units of NDIR analyzer was supplied by the other electric lines. Seawater was pumped up continuously from the bottom of the ship (about 5 m below the surface) and introduced into the equilibrator. The seawater temperature at the water inlet was recorded on a recorder chart, and at the equilibrator was recorded digitally on a floppy disk.

24 min were allowed in each hour for the observation of four standard gases (420, 360, 330, 280 ppm of CO<sub>2</sub> in dry air), 18 minutes for measuring the CO<sub>2</sub> in the background air, 17 minutes for measuring the CO<sub>2</sub> in the air equilibrated

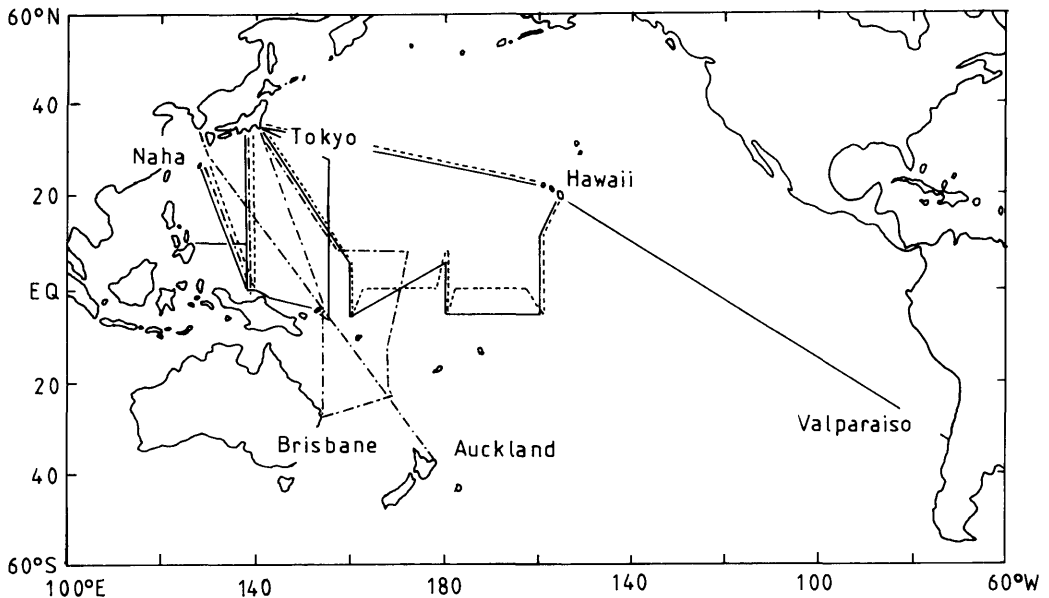


Fig. 2. Cruise track of  $p\text{CO}_2$  measurements during the period from January 1987 to February 1989. Solid line means the cruise conducted in 1987, dash-dotted line in 1988, and dotted line in 1989.

with seawater, and one minute for the data file creation on the floppy disk. Standard gases and background air were introduced into the NDIR analyzer's sample cell for 4.5 minutes at a flow rate of  $0.5 \text{ l min}^{-1}$  and gas flow was stopped. For measuring the oceanic  $\text{CO}_2$  air was circulated for 15.5 minutes in a closed circuit consisting of NDIR analyzer, desiccant system (perma pure dryer and  $\text{Mg}(\text{ClO}_4)_2$ ), and the equilibrator, and then the flow of the air equilibrated with seawater was stopped. Because the pressure of the NDIR analyzer's cell is slightly higher than the atmospheric pressure in the experimental room, it was decreased to the same level in the experimental room, it was decreased to the same level in the experimental room by opening the solenoid valve. The length of tubing ( $> 2.5 \text{ m}$ , o.d.  $0.635 \text{ mm}$ ) is long enough to prevent the ambient air from diffusing back into the NDIR's cell. Signal integration was commenced 1 minutes after the equilibrium time of 30 s. All measurements were carried out at the pressure equal to the atmospheric pressure in the experimental room. In order to evaluate any drifts in the NDIR analyzer, output voltages of 4 standards at the time of sample air  $\text{CO}_2$  measurements were determined by linear inter-

polation between two successive calibrations. The mole fraction in dry air was calculated according to a quadratic least squares fit to the 4 standards. The  $p\text{CO}_2$  was calculated using the water vapor pressure and surface atmospheric pressure observed during the cruise. If the meteorological elements were observed every 3 or 4 h, we obtained the data every hour by cubic spline interpolation.

The effect of the temperature rise in the tubing on the  $p\text{CO}_2$  in surface seawater was corrected using the equation reported by Gordon and Jones (1973).

$$\frac{d(p\text{CO}_2)}{dT} = 4.4 \times 10^{-2}(p\text{CO}_2) - 4.6 \times 10^{-6}(p\text{CO}_2)^2. \quad (1)$$

The working standard gases were calibrated before and after the cruise against the primary standard gases ( $\text{CO}_2$  in air) prepared in the same way as reported by Tanaka et al., (1987a; gravimetric method, Nippon Sanso, Japan). A duplicate analysis of the same air revealed that the standard deviation of the analytical results is less than 0.2 ppm for Model 864 and 0.05 ppm for

Model 865 in the laboratory on land and a little larger (0.1 ppm–0.5 ppm) during the shipboard analysis. It depends mostly on the weather conditions and vibrations due to the ship engine. Considering the temperature rise of seawater in the tubing and pressure broadening effect due to the supersaturation of oxygen which sometimes occurs, the precision of oceanic CO<sub>2</sub> measurements was estimated to be better than 2.0 ppm. But, the precision of CO<sub>2</sub> concentration in the central equatorial Pacific during January and February 1989 was somewhat worse than 2.0 ppm, because the concentration of CO<sub>2</sub> in the air equilibrated with seawater was higher than the highest concentration of standard gas. During the other cruises, almost all the seawater pCO<sub>2</sub> values were lower than that of the highest standard gas.

We were using the primary standard gases prepared by the gravimetric method by Takachiho Kagaku (Japan) before April 1987. The concentration reported here is derived from the standard gases of Nippon Sanso. The relationship between Nippon Sanso scale and Takachiho scale is as follows:

$$x\text{CO}_2^{\text{NS}} = 0.247 + 1.028(x\text{CO}_2^{\text{TK}}) - 7.650 \times 10^{-5}(x\text{CO}_2^{\text{TK}})^2 \quad (2)$$

where  $x\text{CO}_2^{\text{NS}}$  is the CO<sub>2</sub> mole fraction in dry air based on the Nippon Sanso standards, and  $x\text{CO}_2^{\text{TK}}$  based on the Takachiho Kagaku standards.

We will report the difference in CO<sub>2</sub> concentration between Nippon Sanso and the 1985 WMO calibration scale in the future.

Air samples were collected in 5 l glass flasks (Pyrex glass, 130 mm dia) with teflon O-ring stopcocks at both ends. The CO<sub>2</sub> concentration in the air sample was determined by NDIR analyzer (Beckman Model 865) and the CO<sub>2</sub> in the sample air was then extracted to measure  $\delta^{13}\text{C}$  at the Geochemical Laboratory of Meteorological Research Institute in Tsukuba, Japan (Inoue et al., 1987). Results of carbon isotopic ratio are expressed as the per-mil deviation from the PDB standard. NBS20, NBS19, NBS18, NBS17 and NBS16 supplied by the National Bureau of Standards (USA) are in use along with our laboratory standards. For comparison with  $\delta^{13}\text{C}$  data by other laboratories, we quote the  $\delta^{13}\text{C}$  of various standards determined by NBS20

( $-1.06\text{‰}$ ): NBS16:  $-41.69\text{‰}$ ; NBS17:  $-4.47\text{‰}$ ; NBS18:  $-5.02\text{‰}$ ; NBS19:  $1.94\text{‰}$ . To detect the  $\delta^{13}\text{C}$  long-term trend of atmospheric CO<sub>2</sub>, care must be taken to evaluate the long-term and short-term variation (Mook et al., 1983) in mass spectrometer and isotopic standards. Using isotopic standards, day-to-day and long-term trend were estimated.

Corrections due to N<sub>2</sub>O, which interferes with the isotope measurements (Mook and van der Hoek, 1983) have not been applied in this report. On our mass spectrometer, the beam intensity of the mass 44 ion of N<sub>2</sub>O<sup>+</sup> is 71 % compared to that of CO<sub>2</sub><sup>+</sup> at an equal inlet pressure.

## 2.2. Data selection of atmospheric CO<sub>2</sub> concentration

There are two main factors which affect the output voltage of the NDIR analyzer other than natural variability; anthropogenic source (stack gases from the ship itself), and instability of the NDIR analyzer due to rough weather. Obvious contaminated data, as reported by the analogue recorder charts, were excluded from the edited data set. If the difference in CO<sub>2</sub> concentration between two successive measurements ( $\Delta\text{time} = 1\text{ h}$ ) was larger than 1 ppm, both values were flagged, as being either contaminated or not representative of background air. The average value at each degree of latitude was calculated, and data falling outside the one standard deviation are rejected. The present data includes the contribution of regional sources and sinks to the CO<sub>2</sub> concentration, because the time span of CO<sub>2</sub> measurement at each latitude interval was usually in the range from several to several tens of hours.

## 2.3. Data selection of oceanic carbon dioxide

Data selection of oceanic CO<sub>2</sub> must be carried out with care, because the pCO<sub>2</sub> in surface seawater exhibits a great variability. First, the analogue recorder charts were inspected for instrumental problems and events of laboratory air contamination, as done for editing the atmospheric CO<sub>2</sub> record. Next, we checked the temperature relationship between the seawater inlet of the ship and the equilibrator. The temperature rise was usually less than 1.5°C for the observations reported here. If the sea water warming in the tubing changes by more than 0.5°C between two successive measurements,

both measurements were flagged as suspect and unrepresentative of oceanic  $\text{CO}_2$  measurement. The average value of  $\text{pCO}_2$  in surface seawater at each degree of latitude was calculated using the remaining data and the data falling outside of two standard deviations were excluded.

#### 2.4. Data selection of carbon isotopic ratio of atmospheric $\text{CO}_2$

During the atmospheric sampling, the  $\text{CO}_2$  concentration was being continuously measured on board the ship. We have compared the  $\text{CO}_2$  concentration in the sample air to that of the continuous records, and data were excluded if the difference between two measurements was larger than the sum of standard deviation at each latitude and 0.5 ppm which is estimated to be equal to the NDIR instrumental precision under rough weather conditions. The carbon isotopic data which were contaminated by room air during the  $\text{CO}_2$  extraction and sample tube handling have also been excluded.

### 3. Results and discussion

#### 3.1. The $\text{pCO}_2$ distribution of the central and western equatorial Pacific in January and February 1987

In January and February 1987, measurements of  $\text{pCO}_2$  were made in the central and western equatorial Pacific (Figs. 2 and 3), a time when positive SST anomalies appeared in the central and eastern equatorial Pacific. (Fig. 1). The  $\text{pCO}_2$  of surface seawater is lower than the air  $\text{pCO}_2$  in the region between  $10^\circ\text{N}$  and  $15^\circ\text{N}$  and higher in the equatorial region. The distribution of  $\text{pCO}_2$  along  $160^\circ\text{W}$  was markedly different from those of  $137^\circ\text{E}$ ,  $160^\circ\text{E}$  and  $180^\circ$  (Fig. 3).

Along  $160^\circ\text{W}$ , the surface water  $\text{pCO}_2$  was generally above  $380 \mu\text{atm}$  between  $3^\circ\text{N}$  and  $5^\circ\text{S}$ , with a broad maximum near the equator. For latitudinal sections west of  $161^\circ\text{W}$ , the surface water  $\text{pCO}_2$  shows a fairly constant distribution (from  $332 \mu\text{atm}$  to  $360 \mu\text{atm}$ ), without a clear maximum near the equator.

The  $\text{pCO}_2$  along  $160^\circ\text{W}$  was low in comparison with the  $\text{pCO}_2$  during the non-ENSO periods. Feely et al. (1987) reported the higher degree of supersaturation of oceanic  $\text{CO}_2$  in the central

equatorial Pacific in March and April 1984. During the 1982/83 ENSO event, the  $\text{pCO}_2$  between  $7^\circ\text{N}$  and  $1^\circ\text{S}$  along  $137^\circ\text{E}$  has increased as the SST decreased, while it decreased with the SST decrease between  $7^\circ\text{N}$  and  $17^\circ\text{N}$  (Fushimi 1987; Inoue et al., 1987). In January 1987, however, the  $\text{pCO}_2$  along  $137^\circ\text{E}$  showed a pattern similar to those of non-ENSO periods (Inoue and Sugimura, 1988b). During the 1982/83 ENSO event, the surface water  $\text{pCO}_2$  in the central and eastern equatorial Pacific decreased considerably, as did the concentration of nutrients (Feely et al., 1987).

The present results clearly indicate that the surface water  $\text{pCO}_2$  in the central equatorial Pacific decreased during the 1986/88 ENSO event, but less dramatically than during the 1982/83 ENSO event.

#### 3.2. The $\text{pCO}_2$ distribution of the equatorial Pacific during the period from June 1987 to November 1988

During the period from June 1987 to November 1988, five cruises were conducted in the Pacific. Four of these were cruises which crossed the equator in the western Pacific, and one in the eastern equatorial Pacific (Fig. 2).

The oceanic  $\text{CO}_2$  north of  $10^\circ\text{N}$  along  $160^\circ\text{E}$  and  $160^\circ\text{W}$  and north of  $5^\circ\text{N}$  along  $137^\circ\text{E}$  was undersaturated with respect to the air  $\text{CO}_2$  during the boreal winter, but in June and July 1987 the  $\text{pCO}_2$  increased to the level nearly equal to or slightly higher than that of the air  $\text{pCO}_2$  (Fig. 4).

Thermodynamic changes in  $\text{pCO}_2$  with the SST (Eq. (1)) and the salinity (Weiss et al., 1982)

$$\frac{d \ln(\text{pCO}_2)}{d \text{Sa}} = 0.08620 - 1.272 \times 10^{-3}(\text{Sa}) \quad (3)$$

where Sa mean the salinity, are calculated between July 1987 and January 1987, and January 1988 and July 1987, and plotted in Figs. (4) and (5). Between summer and winter changes in nutrient concentration in surface seawater along  $137^\circ\text{E}$  are usually less than  $0.1 \mu\text{mol kg}^{-1}$  for  $\text{NO}_3^-$  (ROO, 1989 and 1990). It is suggestive of a minor role for biological activity in changing surface water  $\text{pCO}_2$ . Amplitudes of the  $\text{pCO}_2$  change between summer and winter are larger than calculated from the SST and salinity change north of  $5^\circ\text{N}$  and nearly equal south of  $5^\circ\text{N}$ . Around  $5^\circ\text{N}$ , there

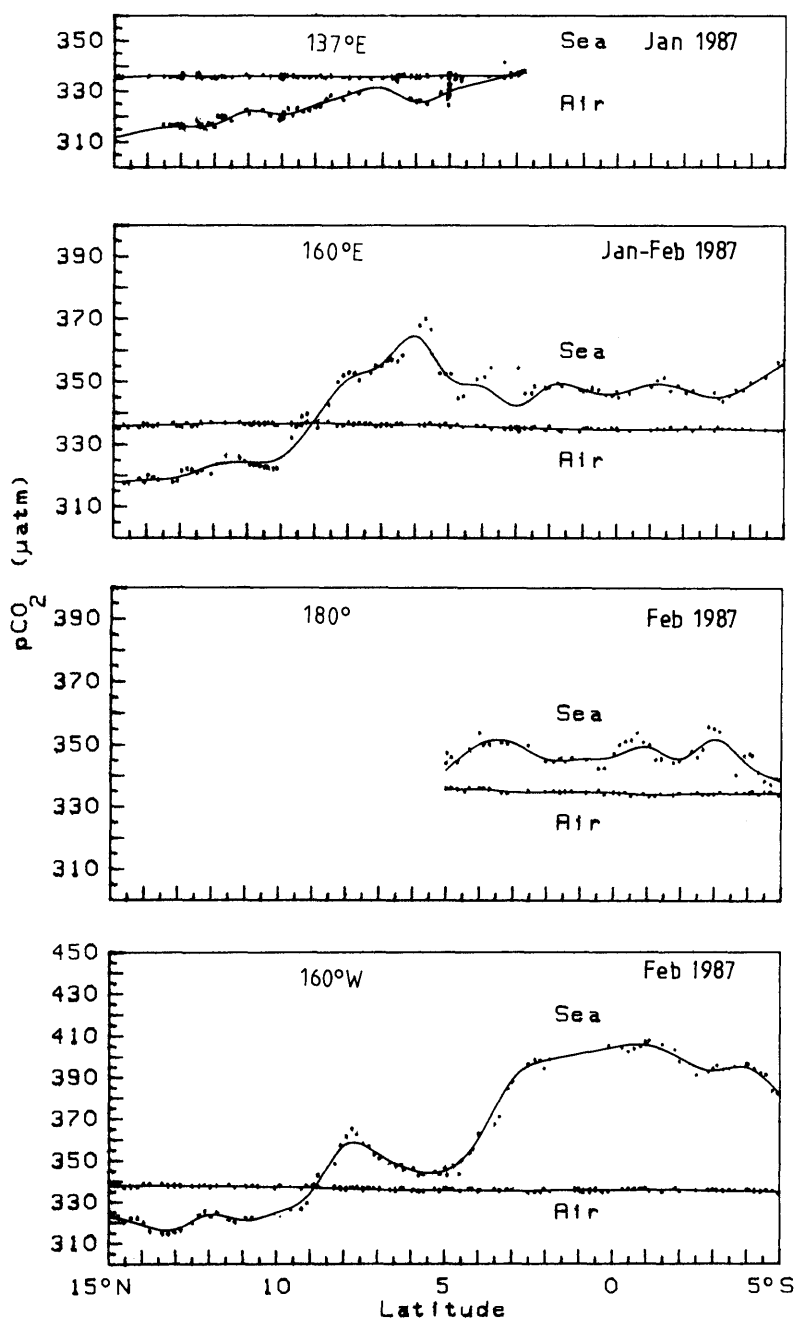


Fig. 3. The pCO<sub>2</sub> distribution along four north-south transects in and over the western and central equatorial Pacific in January and February 1987. The smooth curves are cubic spline fits to the average of each degree of latitude. Cruises were conducted from January 20 (15°N) to January 26 (3°N) along 137°E, from January 29 (15°N) to February 1 (5°S) along 160°E, from February 10 (5°S) to February 12 (5°N) along 180°, and from February 16 (5°S) to February 20 (15°N) along 160°W.

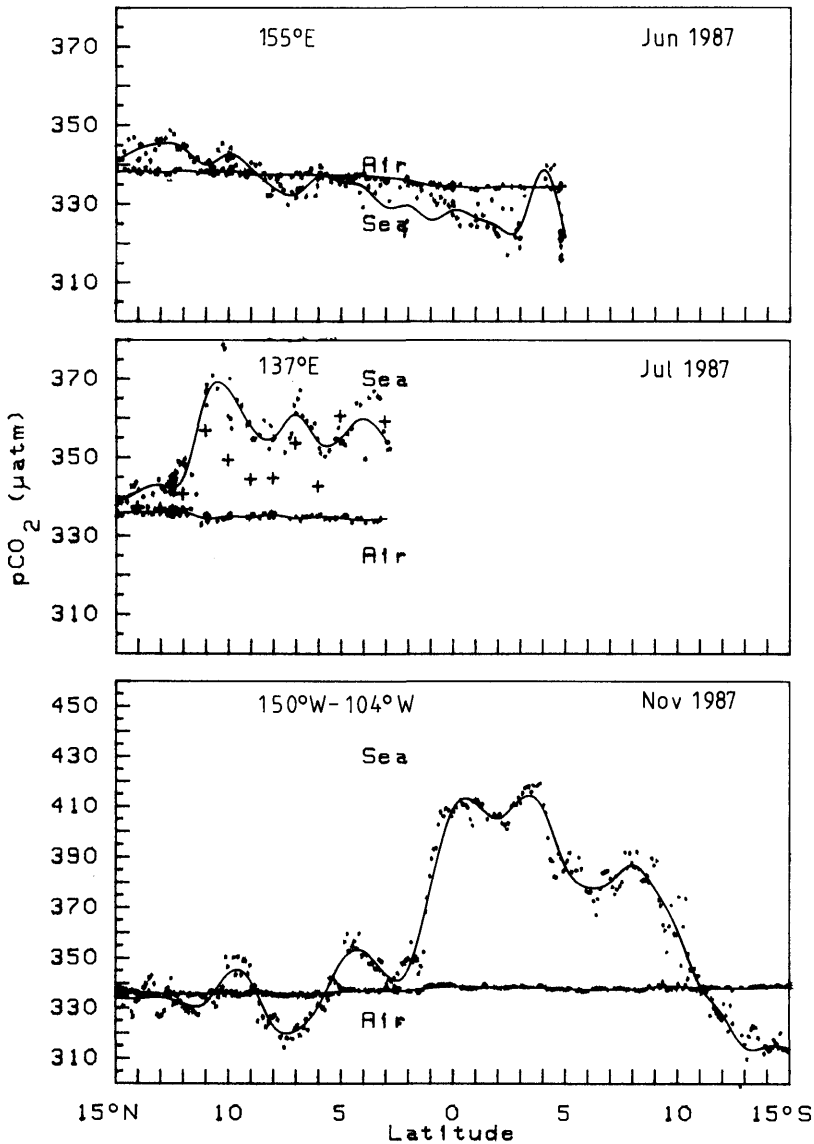


Fig. 4. The  $p\text{CO}_2$  distribution in and over the western and eastern equatorial Pacific in June, July and November 1987. The smooth curves are cubic spline fits to the average of each degree of latitude. Cruises were conducted from June 15 ( $15^\circ\text{N}$ ) to June 20 ( $5^\circ\text{S}$ ) along  $155^\circ\text{E}$ , from July 2 ( $3^\circ\text{N}$ ) to July 19 ( $15^\circ\text{N}$ ) along  $137^\circ\text{E}$ , and from November 16 ( $15^\circ\text{N}$ ,  $150^\circ\text{W}$ ) to November 28 ( $15^\circ\text{S}$ ,  $104^\circ\text{W}$ ). + : the expected  $p\text{CO}_2$  in July 1987 due to changes in SST and salinity from January 1987 (see text).

exists a boundary between the North Equatorial Current and the Equatorial Counter Current (MDJMA, 1988, 1989). This may be caused by the  $\text{CO}_2$  exchange between the sea and the air, but there are still insufficient data to discuss this annual variation in oceanic  $\text{CO}_2$ .

The observed seasonal variation in  $p\text{CO}_2$ , being high in summer and low in winter, displays the same tendency as has been observed in the eastern subtropical gyre of the North and South Pacific (Weiss et al., 1982) and off the coast of Japan (Inoue and Sugimura, 1988a). Takahashi (1987)

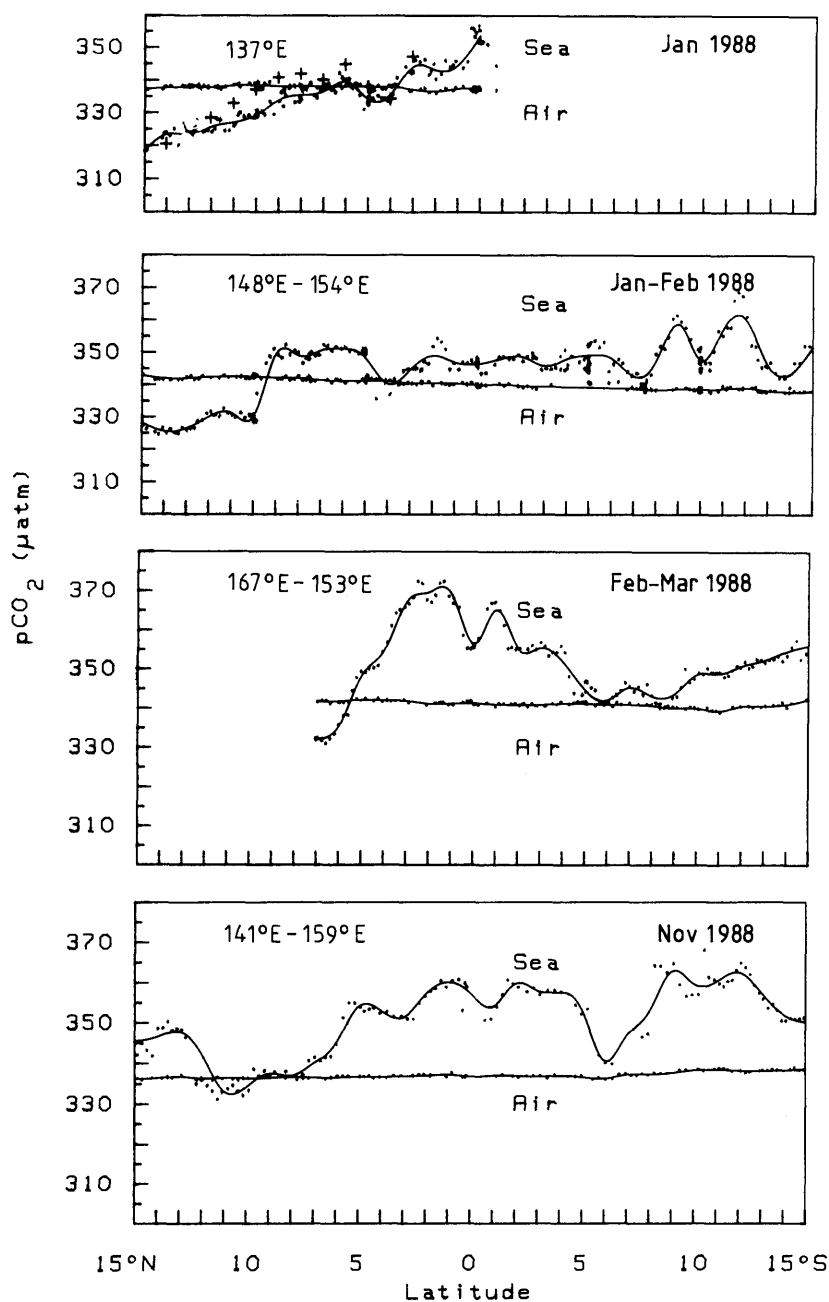


Fig. 5. The pCO<sub>2</sub> distribution along four north-south transects in and over the western and central equatorial Pacific from January to March in 1988, and in November 1988. The smooth curves are cubic spline fits to the average of each degree of latitude. Cruises were conducted from January 23 (15°N) to January 30 (0°) along 137°E, from January 29 (15°N) to February 6 (15°S) along 152°E, from February 27 (15°S, 167°E) to March 18 (15°N, 153°E), and from November 4 (15°N, 141°E) to November 9 (15°S, 159°E). + : the expected pCO<sub>2</sub> in January 1988 due to changes in SST and salinity from July 1987 (see text).

has reported the seasonal variations in  $p\text{CO}_2$  in the northern North Pacific with high  $p\text{CO}_2$  in winter and low  $p\text{CO}_2$  in summer, a pattern opposite to the present results and likely resulting from vertical mixing and biological activity rather than from temperature variations.

During the period from late October to December in 1987, the  $p\text{CO}_2$  was measured from Tokyo to Valparaiso via Hawaii (Fig. 2). From Hawaii to Valparaiso the  $p\text{CO}_2$  in surface seawater was higher than that of the air  $p\text{CO}_2$  between  $5^\circ\text{N}$  and  $11^\circ\text{S}$  (Fig. 4). Near the equator the  $p\text{CO}_2$  in the surface water became maximum ( $74 \mu\text{atm}$ ), but during the non-ENSO events the  $p\text{CO}_2$  was reported to be larger than  $90 \mu\text{atm}$ . (Keeling, 1968, Tans et al., 1990). As described above, during the 1982/83 ENSO event the  $p\text{CO}_2$  in surface seawater of the eastern equatorial Pacific decreased to a level almost in equilibrium with the air. The SST near the equator along  $127^\circ\text{W}$  was  $29.6^\circ\text{C}$  in April 1983 (Feely et al., 1987), which was about  $4^\circ\text{C}$  to  $5^\circ\text{C}$  higher than November 1987. In November 1987, positive SST anomalies of  $+2^\circ$  occurred near  $120^\circ\text{W}$ , and a broad region of positive SST anomalies larger than  $+1^\circ\text{C}$  occurred from  $20^\circ\text{S}$  and  $20^\circ\text{N}$  (MRCS, 1987c).

In November 1987, the  $p\text{CO}_2$  in the eastern equatorial Pacific decreased with the reduction of upwelling, but the surface water supersaturation remained higher than that observed during the 1982/83 ENSO event.

During the period from January to March in 1988, measurements of  $p\text{CO}_2$  of the western North Pacific and western South Pacific were made along three north and south transects (Fig. 5). The oceanic  $p\text{CO}_2$  along  $152^\circ\text{E}$  was undersaturated with respect to the air  $\text{CO}_2$  in the subtropical zone of the Northern Hemisphere, while supersaturated slightly in the subtropical zone of the Southern Hemisphere (Fig. 5), consistent with a seasonal variation driven primarily by heating and cooling. In March 1988, the  $p\text{CO}_2$  measurements were made in the western equatorial Pacific (Fig. 5), which is at the time of the end of 1986/88 ENSO event. From February to March 1988, the positive SST anomalies between  $+0.5^\circ\text{C}$  and  $+1^\circ\text{C}$  disappeared in the western equatorial Pacific (MRCS, 1988a, b). A clear maximum of the  $p\text{CO}_2$  (about  $370 \mu\text{atm}$ ) showed up near the equator coincident with the SST minimum. This means that the recovery of the equatorial supersaturation of sur-

face water  $p\text{CO}_2$  occurred quickly. In November 1988, the surface water  $p\text{CO}_2$  values in the western equatorial Pacific were higher than the air  $p\text{CO}_2$  between  $10^\circ\text{N}$  and  $15^\circ\text{S}$  (Fig. 5). The  $p\text{CO}_2$  had increased by about  $13 \mu\text{atm}$  compared to February 1988.

### 3.3. The $p\text{CO}_2$ distribution of the central and western equatorial Pacific in January and February 1989

In January and February 1989, measurements of  $p\text{CO}_2$  were made in the western and central equatorial Pacific, the same area where we measured the  $p\text{CO}_2$  in January and February 1987 (Fig. 2). In the subtropical area, the surface water  $p\text{CO}_2$  along  $160^\circ\text{W}$  was similar to that in January and February 1987, and along  $160^\circ\text{E}$  somewhat higher. In the eastern and central equatorial Pacific, however, a remarkable change in  $p\text{CO}_2$  took place during the La Niña event (Fig. 6). Along  $180^\circ$  and  $160^\circ\text{E}$ , the  $p\text{CO}_2$  south of  $5^\circ\text{N}$  increased largely along with the SST decrease, as compared to the sections made during the 1986/88 ENSO event. In January and February 1987 the  $p\text{CO}_2$  along  $160^\circ\text{W}$  south of  $3^\circ\text{N}$  was relatively high as mentioned above, and in January and February 1989 it increased further to a level of about  $450 \mu\text{atm}$ . The pattern of  $p\text{CO}_2$  distribution among three north-south transects showed very little longitudinal gradient, unlike January and February 1987.

Along  $137^\circ\text{E}$ , the  $p\text{CO}_2$  did not vary during the La Niña event, even though anomalies of seawater temperature higher than  $7^\circ\text{C}$  was found at a depth of  $150\text{ m}$  at  $6^\circ\text{N}$  following the La Niña event (MDJMA 1989). The latitudinal distribution of  $p\text{CO}_2$  along  $137^\circ\text{E}$  did not change during either the 1986/88 ENSO event or the La Niña event in 1988/89.

In the equatorial Pacific, the  $p\text{CO}_2$  increases with decreasing SST, while in the equatorial Atlantic the  $p\text{CO}_2$  increased with SST during the zonal flow from the east to the west (Andrieu et al., 1986; Smethie et al., 1985). We will discuss the longitudinal distribution of surface water  $p\text{CO}_2$  in the central equatorial Pacific in Section 3.5.

### 3.4. $\text{CO}_2$ flux of the central and western equatorial Pacific in January 1987 and January 1989

The  $\text{CO}_2$  flux ( $F(\text{CO}_2)$ ) of the equatorial Pacific was computed from eq. (4) for two periods,

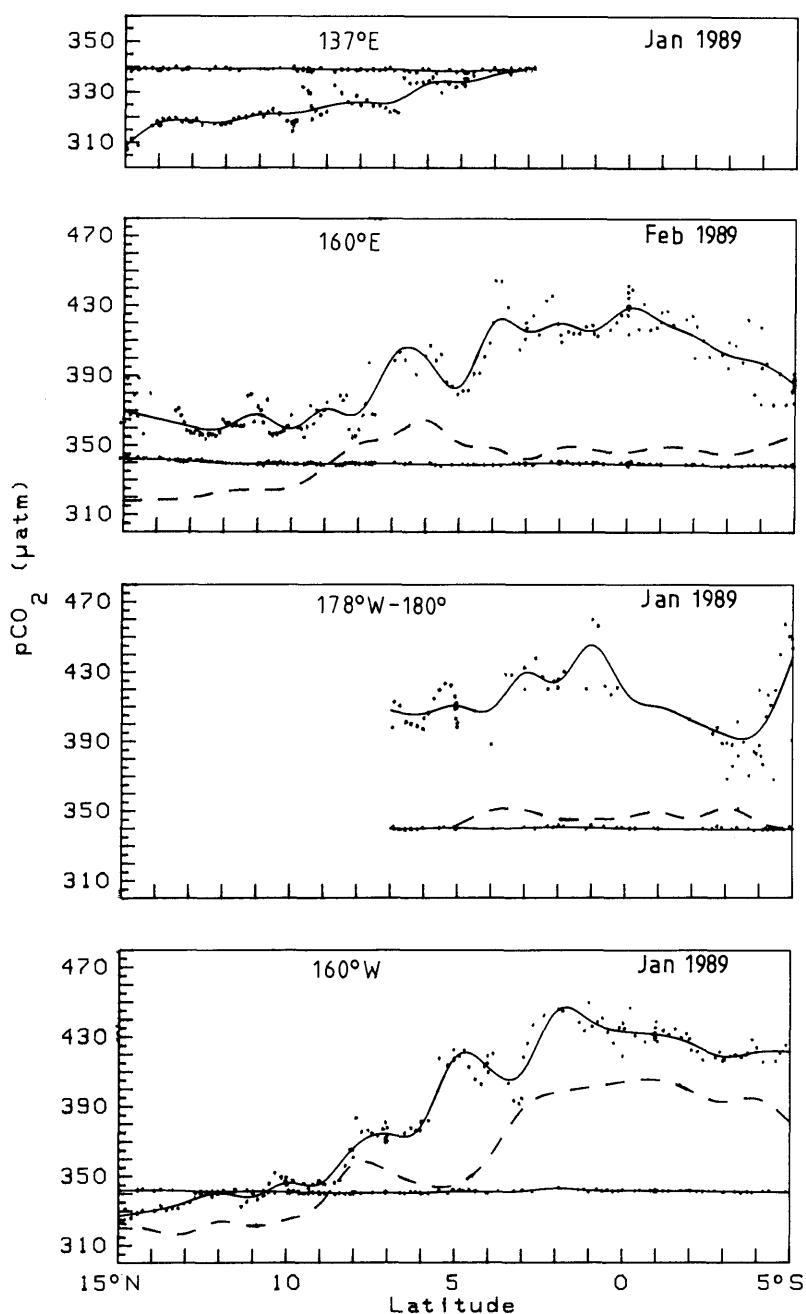


Fig. 6. The pCO<sub>2</sub> distribution along four north-south transects in and over the western and central equatorial Pacific in 1989. The smooth curves are cubic spline fits to the average of each degree of latitude. Solid line represents the data in Jan/Feb 1989, and dashed line in Jan/Feb 1987. Cruises were conducted from January 25 (15°N) to January 30 (3°N) along 137°E, from February 7 (5°S) to February 16 (15°N) along 160°E, from January 25 (5°S) to January 29 (7°N) along 180°, and from January 13 (15°N) to January 16 (5°S) along 160°W.

January and February 1987 and January and February 1989.

$$F(\text{CO}_2) = kS \Delta p\text{CO}_2 = E \Delta p\text{CO}_2 \quad (4)$$

where  $k$  is the gas transfer piston velocity of  $\text{CO}_2$  between the sea and the air,  $S$  is the solubility of  $\text{CO}_2$  in seawater and  $E$  is the gas transfer coefficient. Weiss (1974) reported the empirical formula for the solubility of  $\text{CO}_2$  in seawater. In this paper, the salinity was taken to be  $34\text{‰}$ . The gas transfer coefficient  $E$  (moles of  $\text{CO}_2$  year $^{-1}$  m $^{-2}$   $\mu\text{atm}^{-1}$ ) was suggested by Takahashi et al., (1989).

$$E = 0.016[U - 3], \quad (5)$$

where  $U$  is the wind speed and  $E$  is taken to be zero for  $U \leq 3 \text{ m s}^{-1}$ .

According to Liss and Merlivat (1986) and Etcheto and Merlivat (1988), the gas transfer piston velocity  $k$  is a function of wind speed and Schmidt number.

$$k = 0.17U_{10}(600/\text{Sc})^{2/3} \quad (6a)$$

$$0 < U_{10} \leq 3.6 \text{ m s}^{-1},$$

$$k = [2.85U_{10} - 9.65](600/\text{Sc})^{1/2} \quad (6b)$$

$$3.6 < U_{10} \leq 13 \text{ m s}^{-1},$$

$$k = [5.9U_{10} - 49.3](600/\text{Sc})^{1/2} \quad (6c)$$

$$U_{10} > 13 \text{ m s}^{-1},$$

where  $U_{10}$  is the wind speed at 10-m height and  $\text{Sc}$  the Schmidt number, defined as the ratio between the molecular viscosity of water and the molecular diffusivity of  $\text{CO}_2$  in water (Jahne et al., 1984). Here, in order to compute the  $\text{CO}_2$  flux, we use both eq. (5) and eqs. (6). We divided the equatorial Pacific, the rectangular area between  $5.5^\circ\text{S}$  and  $5.5^\circ\text{N}$ , and  $134.5^\circ\text{E}$  and  $80.5^\circ\text{W}$ , into  $1^\circ$  by  $1^\circ$  boxes. We interpolated and extrapolated the observed  $p\text{CO}_2$  into regions where observation were not made.

As described above (3.1), in January and February 1987 the equatorial  $p\text{CO}_2$  west of  $161^\circ\text{W}$  showed low and constant distribution, without the clear maximum near the equator. But, the surface water  $p\text{CO}_2$  along  $5^\circ\text{S}$  changed steeply between  $160.5^\circ\text{W}$  and  $160^\circ\text{W}$  ( $30 \mu\text{atm}$ , Inoue and Sugimura, 1988b). Therefore, we assume the steep

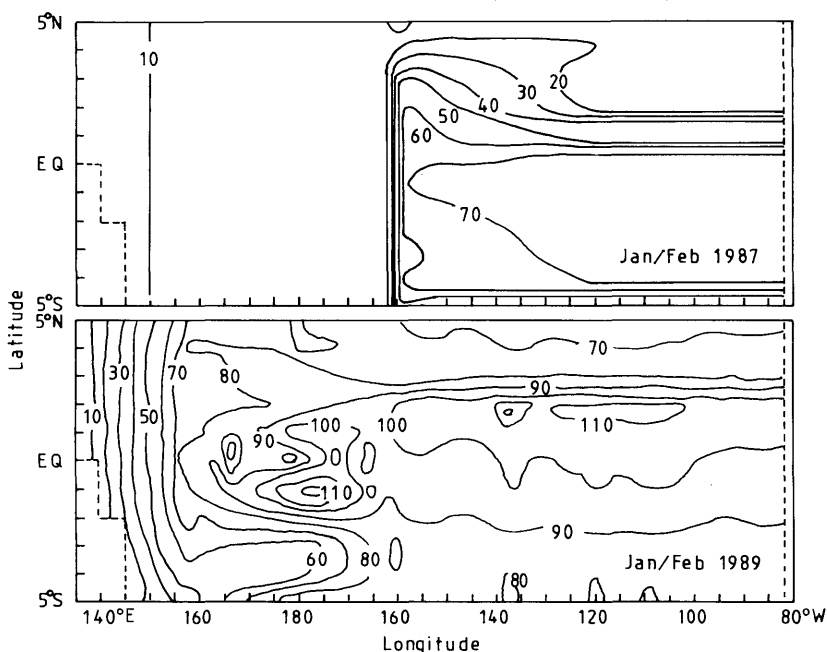


Fig. 7. Maps of estimated  $\Delta p\text{CO}_2$  in the equatorial Pacific in January and February 1987 and in January and February 1989. Area surrounded by dotted line, mostly land area, was excluded from the calculation of  $\text{CO}_2$  flux.

change in pCO<sub>2</sub> between 161°W and 160°W. Taking into account the oceanographic data, the ΔpCO<sub>2</sub> was assumed to be 5 μatm between 135°E and 159°E, and 14 μatm between 160°E and 161°W. The former is the average of ΔpCO<sub>2</sub> from 5°N to 0° along 137°E (the pCO<sub>2</sub> south of 2°N was taken from the January 1988 data), and the latter is the average between 160°E and 161°W.

The pCO<sub>2</sub> east of 159°W was not measured in January and February 1987. Extrapolations of the observed pCO<sub>2</sub> in January and February 1987 into the eastern equatorial Pacific by the relationship between the pCO<sub>2</sub> and the SST result in extraordinary high pCO<sub>2</sub>, because of the steep change in pCO<sub>2</sub> between 160.5°W and 160°W. Here, we simply use the pCO<sub>2</sub> data measured in November 1987 (~120°W), within the 1986/88 ENSO event, because of the small difference in SST

(0.5°C) between January and February 1987 and November 1987. The pCO<sub>2</sub> was then interpolated zonally with a linear function between 160°W and 120°W. East of 120°W, we assume the same north-south distribution of pCO<sub>2</sub> as along 120°W. By combining these pCO<sub>2</sub>, we obtain ΔpCO<sub>2</sub> in the equatorial Pacific in January and February 1987 (Fig. 7).

In January and February 1989, west of 160°W a linear interpolation of pCO<sub>2</sub> was made zonally between contiguous data (Fig. 2). The ΔpCO<sub>2</sub> between 135°E and 137°E was assumed to be 4 μatm, the average ΔpCO<sub>2</sub> south of 5°N along 137°E (the pCO<sub>2</sub> south of 2°N was also taken from the January 1988 data). East of 159°W, measurements of pCO<sub>2</sub> were not made during the La Niña event. We extrapolate observed pCO<sub>2</sub> in January and February 1989 into regions east of

Table 1. The CO<sub>2</sub> flux (Gt-C year<sup>-1</sup>) in the equatorial Pacific (5.5°N–5.5°S, 134.5°E–80.5°W) in Jan/Feb 1987 and Jan/Feb 1989; the CO<sub>2</sub> flux F<sub>x</sub> is calculated from eqs. (4) and (5), and F<sub>x</sub>' from eqs. (4) and (6)

| Period Jan/Feb 1987 |                                                                |                          |                                           |
|---------------------|----------------------------------------------------------------|--------------------------|-------------------------------------------|
| Region              | E (mole m <sup>-2</sup> yr <sup>-1</sup> μatm <sup>-1</sup> )  | ΔpCO <sub>2</sub> (μatm) | F <sub>x</sub> (Gt-C yr <sup>-1</sup> )   |
| 134.5°E–159.5°E     | 0.036                                                          | 8.0                      | 0.01                                      |
| 159.5°E–160.5°W     | 0.055                                                          | 14.6                     | 0.05                                      |
| 160.5°W–80.5°W      | 0.058                                                          | 46.4                     | 0.34                                      |
| 134.5°E–80.5°W      | 0.053                                                          | 31.0                     | 0.40                                      |
|                     | E' (mole m <sup>-2</sup> yr <sup>-1</sup> μatm <sup>-1</sup> ) | ΔpCO <sub>2</sub> (μatm) | F <sub>x</sub> ' (Gt-C yr <sup>-1</sup> ) |
| 134.5°E–159.5°E     | 0.016                                                          | 8.0                      | 0.005                                     |
| 159.5°E–160.5°W     | 0.026                                                          | 14.6                     | 0.03                                      |
| 160.5°W–80.5°W      | 0.027                                                          | 46.4                     | 0.16                                      |
| 134.5°E–80.5°W      | 0.025                                                          | 31.0                     | 0.20                                      |
| Period Jan/Feb 1989 |                                                                |                          |                                           |
| Region              | E (mole m <sup>-2</sup> yr <sup>-1</sup> μatm <sup>-1</sup> )  | ΔpCO <sub>2</sub> (μatm) | F <sub>x</sub> (Gt-C yr <sup>-1</sup> )   |
| 134.5°E–159.5°E     | 0.028                                                          | 36.7                     | 0.05                                      |
| 159.5°E–160.5°W     | 0.063                                                          | 81.5                     | 0.35                                      |
| 160.5°W–80.5°W      | 0.049                                                          | 90.9                     | 0.57                                      |
| 134.5°E–80.5°W      | 0.049                                                          | 78.9                     | 0.96                                      |
|                     | E' (mole m <sup>-2</sup> yr <sup>-1</sup> μatm <sup>-1</sup> ) | ΔpCO <sub>2</sub> (μatm) | F <sub>x</sub> ' (Gt-C yr <sup>-1</sup> ) |
| 134.5°E–159.5°E     | 0.012                                                          | 36.7                     | 0.02                                      |
| 159.5°E–160.5°W     | 0.030                                                          | 81.5                     | 0.16                                      |
| 160.5°W–80.5°W      | 0.022                                                          | 90.9                     | 0.26                                      |
| 134.5°E–80.5°W      | 0.023                                                          | 78.9                     | 0.44                                      |

159°W by using the relationship between the  $p\text{CO}_2$  and the SST (the SST data in each box was provided as described below). If the effect of biological activities on the  $p\text{CO}_2$  did not change considerably, this may be extrapolated well.

The map of  $\Delta p\text{CO}_2$  in January and February 1987 (Fig. 7) shows the simple pattern in the western and central equatorial Pacific, which is caused by the reduction of upwelling. In January and February 1989, the  $\Delta p\text{CO}_2$  in the central equatorial Pacific increased considerably, and shows relatively small scale  $\Delta p\text{CO}_2$  contour (Fig. 7), at least partly due to the enhanced upwelling in this area.

The monthly mean SST ( $2^\circ$  by  $2^\circ$ ) was provided by the Marine Department of JMA (MRCS, 1987a, 1987b, 1989a, 1989b), and the average SST in January and February in each box was obtained by linear interpolation to calculate the  $\text{CO}_2$  solubility ( $S_a = 34\text{‰}$ ).

We used the wind data set prepared by the Japan Meteorological Agency which has been used for the long-term weather forecasting (GANL data set, 1987, 1989). The meteorological elements such as wind speed, isobaric height, temperature etc. have been computed twice a day based on the radiosonde data. They cover a  $2.5 \times 2.5$  grid point system for 1987 and  $1.875 \times 1.875$  gridpoint system for 1989. The wind speed at each geographical position of the cruise was obtained by linear interpolation. The wind speed of GANL data set was compared with the wind speed observed on board the ship. As the height for measuring the wind speed depends on the ship, the wind speed at 10-m height was calculated by assuming a drag coefficient of  $1.5 \times 10^{-3}$ . The wind speed of GANL ( $W_{\text{GANL}}$ ) data set correlates fairly well with the observed wind speed  $W_{\text{obs}}$  ( $W_{\text{obs}} = 0.73W_{\text{GANL}} + 2.63$ ,  $r = 0.802$  for 1987;  $W_{\text{obs}} = 0.83W_{\text{GANL}} + 2.16$ ,  $r = 0.861$  for 1989). We use these linear relationships to obtain the wind speed at 10 m height above surface.

The  $\text{CO}_2$  flux was obtained twice a day, and  $\text{CO}_2$  flux of the equatorial Pacific was listed in Table 1. There is no substantial difference in gas transfer coefficient between two periods. The  $\text{CO}_2$  flux based on eqs. (6) was about half as much as that of the eq. (5) for the same  $\Delta p\text{CO}_2$  distribution (Takahashi et al., 1989, Tans et al., 1990). It is clear that net  $\text{CO}_2$  flux in January and February 1989 has increased by a factor of 2.0–2.5 compared

to that of January and February 1987 for a given relationship between wind speed and gas transfer coefficient. The  $\text{CO}_2$  flux in the region between  $160^\circ\text{E}$  and  $160^\circ\text{W}$  changes considerably due to the large variation in  $\Delta p\text{CO}_2$ . During the non-ENSO period the  $\text{CO}_2$  flux was estimated to be in the range from  $0.6 \text{ GT-C year}^{-1}$  (Smethie 1985, Feely et al., 1987) to  $0.8 \text{ GT-C year}^{-1}$  (Keeling and Revelle 1985), and during the 1982/83 ENSO event, considerably lower ( $0.02 \text{ GT-C yr}^{-1}$ ) due to the low  $\Delta p\text{CO}_2$  value. (Feely et al., 1987; Keeling and Revelle 1985). If we use the gas transfer coefficient calculated from eqs. (6),  $\text{CO}_2$  flux in January and February 1989 becomes lower than the  $\text{CO}_2$  flux during the non-ENSO period reported earlier. Here, we use the gas transfer coefficient derived from eqs. (4) and (5), which seems to be reasonable for estimating not only the  $\text{CO}_2$  flux but also the longitudinal distribution of  $p\text{CO}_2$  in the central equatorial Pacific (subsection 3.5).

In January and February 1987, the  $\text{CO}_2$  flux from the central and western equatorial Pacific decreased, but still remained positive as compared to the 1982/83 ENSO event. In January and February 1989 it increased to the level of  $1 \text{ GT-C year}^{-1}$ . If sustained throughout the year, that corresponds to about 20% of the current rate of  $\text{CO}_2$  emission from fossil fuel burning.

A high growth rate of atmospheric  $\text{CO}_2$  appeared during late 1987 and the first part of 1988, (Thoning et al., 1989a), which is the time that the SST anomalies of the eastern and central equatorial Pacific became positive, and the  $\text{CO}_2$  flux of the eastern and central equatorial Pacific are expected to be low in comparison with the non-ENSO periods. In the second half of 1988, the growth rate of atmospheric  $\text{CO}_2$  was not so large as the first half of 1988 (Keeling et al., 1989). Although sources and sinks other than the oceanic  $\text{CO}_2$  source in the equatorial Pacific play a dominant role in determining the growth rate of atmospheric  $\text{CO}_2$  between 1987 and 1989, inter-annual changes in the  $\text{CO}_2$  flux in the equatorial Pacific were large enough to disturb it.

### 3.5. Longitudinal distribution of oceanic $p\text{CO}_2$ in January and February 1989

In this section, we consider the effect on  $p\text{CO}_2$  of changes in the SST, salinity, biological activity, (Andrieu et al., 1988 and Oudot and Andrieu, 1989) and the  $\text{CO}_2$  flux during the zonal flow in the

central and western equatorial Pacific. As described above, the pCO<sub>2</sub> tends to increase with the SST decrease in the equatorial Pacific, which is opposite to the eq. (1). The salinity dependence of pCO<sub>2</sub> is expressed by eq. (3) at the condition of constant SST, pH and the dissolved inorganic carbon (DIC) concentration.

Biological activity, which is reflected in the surface water nutrient concentrations, leads to the pCO<sub>2</sub> change, and we are able to evaluate it based on the buffer factor ( $(\delta p\text{CO}_2/p\text{CO}_2)/(\delta \text{DIC}/\text{DIC})$ ) and Redfield ratio (C:N:P = 106:16:1). In the surface mixed layer (deeper than 150 m), the concentration of nutrients was nearly constant to about 60 m below the surface, and it increased gradually with depth.

The CO<sub>2</sub> exchange between the sea and the air results in changes in the DIC concentration and the corresponding change in pCO<sub>2</sub> is given by the buffer factor.

In January and February 1989, measurements of nutrient concentrations (PO<sub>4</sub><sup>3-</sup>, NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>) were made along the equator between 160° E and 160° W (JAPACS 1989). The rate and direction of seawater flow was measured continuously by using the acoustic doppler current profiler (JAPACS, 1989). Along the equator, both the pCO<sub>2</sub> and the nutrient concentration displayed a maximum at 165° W, where the meridional section of seawater temperature indicated strong upwelling. The direction of water flow at 0°, 165° W ranged from 270°

to 180° between 20 m and 100 m below the surface. At 160° W the surface water flowed toward the northwest. West of 170° W westward flow was dominant in the mixed layer along the equator and the flow rate was found to be about 0.5 m s<sup>-1</sup> (JAPACS 1989). Therefore, we calculated the pCO<sub>2</sub> change along the equator starting from 165° W to 160° E. The vertical profiles of sea water temperature and salinity indicate that the depth of the mixed layer was between 150 m and 200 m.

The calculated pCO<sub>2</sub> deviates significantly from the observed pCO<sub>2</sub> if we adopt a mixed layer depth of more than 150 m, which assumes that complete mixing in the surface mixed layer takes place within a short time. It seems to be reasonable to consider the depth of the layer in which the pCO<sub>2</sub> is affected by the CO<sub>2</sub> exchange during the zonal flow. The amount of CO<sub>2</sub> outgassing from the seawater was calculated between contiguous oceanographic stations.

Taking into account the SST, the buffer factor was assumed to be 8.5 (Sundquist, 1979; Oudot and Andrie, 1989). By changing the depth of the layer and the DIC concentration, we calculated changes in pCO<sub>2</sub> so as to minimize the square of the difference between the calculated pCO<sub>2</sub> and observed pCO<sub>2</sub>. The optimum depth, starting from 10 m with a step size of 5 m, was determined at the DIC concentration of 2040 μmol kg<sup>-1</sup> at 165° W. The optimum DIC concentration was then calculated for a given depth. We repeated this

Table 2. The pCO<sub>2</sub> distribution along the equator in Jan/Feb 1989; the buffer factor is assumed to be 8.5

| Long.                                          | SST  | Salinity | NO <sub>3</sub> <sup>-</sup> + NO <sub>2</sub> <sup>-</sup> | Fx*                                      | Fx'* | pCO <sub>2</sub> (obs) | PCO <sub>2</sub> * | PCO <sub>2</sub> '* |
|------------------------------------------------|------|----------|-------------------------------------------------------------|------------------------------------------|------|------------------------|--------------------|---------------------|
|                                                | (°C) | (‰)      | (μmol/kg)                                                   | (mole m <sup>-2</sup> yr <sup>-1</sup> ) |      | (μatm)                 | (μatm)             | (μatm)              |
| 165° W                                         | 24.8 | 34.967   | 7.7                                                         | 6.9                                      | 3.3  | 444.2                  |                    |                     |
| 170° W                                         | 25.3 | 35.353   | 7.0                                                         | 6.8                                      | 3.2  | 444.8                  | 445.7              | 445.4               |
| 175° W                                         | 25.8 | 35.133   | 6.2                                                         | 6.5                                      | 3.1  | 434.6                  | 434.9              | 434.5               |
| 180°                                           | 26.0 | 35.586   | 4.4                                                         | 5.4                                      | 2.6  | 417.7                  | 419.3              | 419.3               |
| 175° E                                         | 26.7 | 35.202   | 4.8                                                         | 4.9                                      | 2.3  | 424.8                  | 425.0              | 424.7               |
| 172.5° E                                       | 26.9 | 35.226   | 5.0                                                         | 3.6                                      | 1.6  | 422.7                  | 425.1              | 424.5               |
| 165° E                                         | 27.6 | 35.347   | 4.6                                                         | 3.0                                      | 1.3  | 430.0                  | 431.9              | 431.2               |
| 160° E                                         | 28.3 | 35.404   | 3.0                                                         | 1.5                                      | 0.6  | 428.5                  | 424.0              | 425.2               |
| Optimum depth (m)                              |      |          |                                                             |                                          |      |                        | 70                 | 30                  |
| Optimum DIC (μmol kg <sup>-1</sup> ) at 165° W |      |          |                                                             |                                          |      |                        | 2024               | 2092                |

Fx\*; CO<sub>2</sub> flux is estimated by eqs. (4) and (5).

Fx'\*; CO<sub>2</sub> flux is estimated by eqs. (4) and (6).

PCO<sub>2</sub>; the partial pressure of CO<sub>2</sub> derived from Fx.

PCO<sub>2</sub>'; the partial pressure of CO<sub>2</sub> derived from Fx'.

procedure until the square of the difference converged. In Table 2, we list the  $p\text{CO}_2$  changes along the equator between  $165^\circ\text{W}$  and  $160^\circ\text{E}$  in January and February 1989. The optimum depth was 70 m for eqs. (4) and (5), almost corresponding to the depth of constant concentration of  $\text{NO}_3^- + \text{NO}_2^-$  below the surface (60 m), and 30 m for eqs. (4) and (6). The optimum concentration of DIC was  $2024 \mu\text{mol kg}^{-1}$  for eqs. (4) and (5) and  $2092 \mu\text{mol kg}^{-1}$  for eqs. (4) and (6). The DIC concentration based on eqs. (4) and (5) is fairly close to the value reported earlier ( $2040 \mu\text{mol kg}^{-1}$  at  $0^\circ$ ,  $124.5^\circ\text{W}$  in December 1973 and  $2010 \mu\text{mol kg}^{-1}$  at  $0^\circ$ ,  $179^\circ\text{E}$  in May 1974, Broecker et al., 1982).

The effect of biological activity and the temperature play dominant role in determining the  $p\text{CO}_2$ . The  $p\text{CO}_2$  change due to the  $\text{CO}_2$  outgassing is small in comparison with the effect of biological activity and temperature change, but it clearly exists. In the present study, the  $\text{CO}_2$  exchange estimated by eqs. (4) and (5) or eqs. (4) and (6) results in a good agreement with the observed  $p\text{CO}_2$ , but both the depth of layer and the DIC concentration derived from eqs. (4) and (5) seem to be better than those of eqs. (4) and (6). This treatment is a first approximation because we did not measure the DIC concentration and buffer factor, and we treat the respective factors adiabatically. However, the present results suggest the

possibility of determining the relationship between exchange coefficient and wind speed based on the field observation.

### 3.6. Distribution of atmospheric $\text{CO}_2$ during the period from January 1987 to January 1989

At present differences between the SIO X85 mole fraction scale and ours are not yet determined. Here, we report the  $\text{CO}_2$  concentration determined by using our  $\text{CO}_2$  standard. The raw data of atmospheric  $\text{CO}_2$  concentration observed from November to December in 1987 is plotted against latitude in Fig. 8 as an example. The data was not affected by local sources and/or sinks as observed frequently at land stations, and difference in  $\text{CO}_2$  concentration between two successive measurements was usually less than 0.3 ppm, close to the natural variabilities and/or error of measurements on board the ship. Although these data include the contribution of the regional sources and sinks to the atmospheric  $\text{CO}_2$  concentration, having periodicities of a few days to several days (Thoning et al., 1989b), it is possible to extract the variation of distribution of background  $\text{CO}_2$  concentration. The average value at each degree of latitude was calculated from which the contributions of the seasonal variation and the secular variation were removed. Seasonal and long-term variation in atmospheric  $\text{CO}_2$  were estimated at every  $5^\circ$  by the linear secular

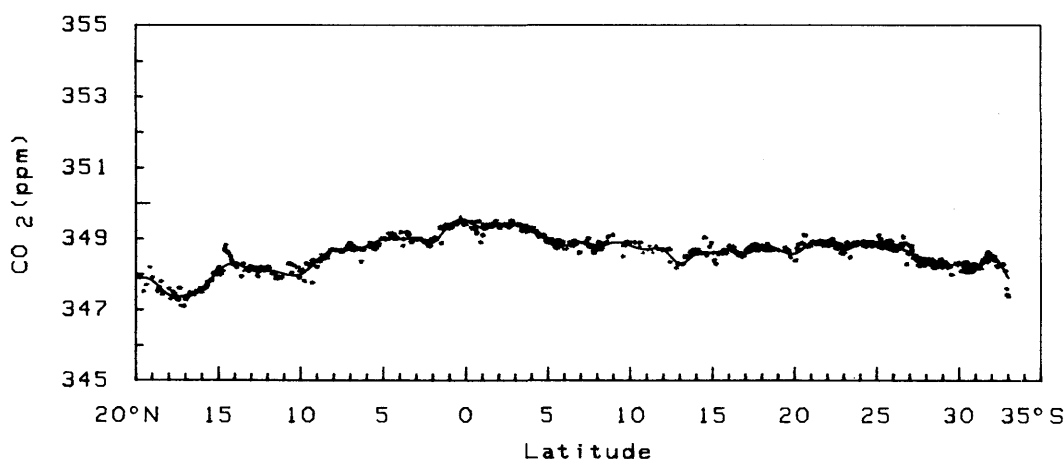


Fig. 8. The concentration of atmospheric  $\text{CO}_2$  observed during the cruise from Hawaii to Valparaiso. The smooth curves are cubic spline fits to the average of each degree of latitude. The ship left Hawaii on 28 November and arrived at Valparaiso on 6 December 1987.

trend and Fourier harmonics based on the measurements of Tanaka et al., (1987b).

$$x\text{CO}_2^{\text{NS}} = \sum_{i=1}^2 (A_i \cos(2\pi it) + B_i \sin(2\pi it)) + C + Dt, \quad (7)$$

where  $t$  is the time and  $A_i$ ,  $B_i$ ,  $C$  and  $D$  are coefficients to be determined by the least square methods.

The magnitude of correction depends on the Julian day, location and observation period, but does not usually exceed 0.5 ppm. The observational results south of 5°S, however, are not corrected for the seasonal variation, which is not well approximated by the first two Fourier harmonics (Tanaka et al., 1987b; Halter et al., 1988).

The average CO<sub>2</sub> concentration in November 30, 1987 is plotted along with the atmospheric CO<sub>2</sub> data (average in November and December 1987) of land stations (Boden et al., 1990) in Fig. 9. The north-south distribution of atmospheric CO<sub>2</sub> as measured on board the ship agrees well with the background CO<sub>2</sub> data. Key Biscayne (KEY, USA) is the land station where concentration tends to be higher than other land stations (see Fig. 7 in

Conway et al., 1988). A hump of atmospheric CO<sub>2</sub> concentration near the Hawaii islands could be caused by the land plants and anthropogenic sources. The maximum concentration of atmospheric CO<sub>2</sub> appeared near the equator, and the dip of the atmospheric CO<sub>2</sub> concentration in low latitudes of the Northern Hemisphere in this season is caused by the earlier effect of CO<sub>2</sub> uptake by land plants (Komhyr et al., 1985). The respiration of soil and land plants in boreal fall increases the CO<sub>2</sub> concentration in higher latitudes of the Northern Hemisphere (Komhyr et al., 1989).

Along 137°E, the distribution of the atmospheric CO<sub>2</sub> concentrations on January 31, 1987 shows the prominent feature that the concentration of atmospheric CO<sub>2</sub> decreased from the Japan islands southward to 15°N, and stayed nearly constant toward the equator (Fig. 10), where CO<sub>2</sub> concentration at 13°N and 14°N is nearly equal to the Guam (GMI, USA) data (Boden et al., 1990). The observed variability of atmospheric CO<sub>2</sub> at low latitude is less than that of the higher latitudes. Therefore, we consider the long-term trend of  $\delta^{13}\text{C}$  and concentration of atmospheric CO<sub>2</sub> in the fairly narrow area (between 7°N and 13°N) in Section 3.7. The high CO<sub>2</sub> concentrations from the Japan islands southward to 15°N is caused by the anthropogenic

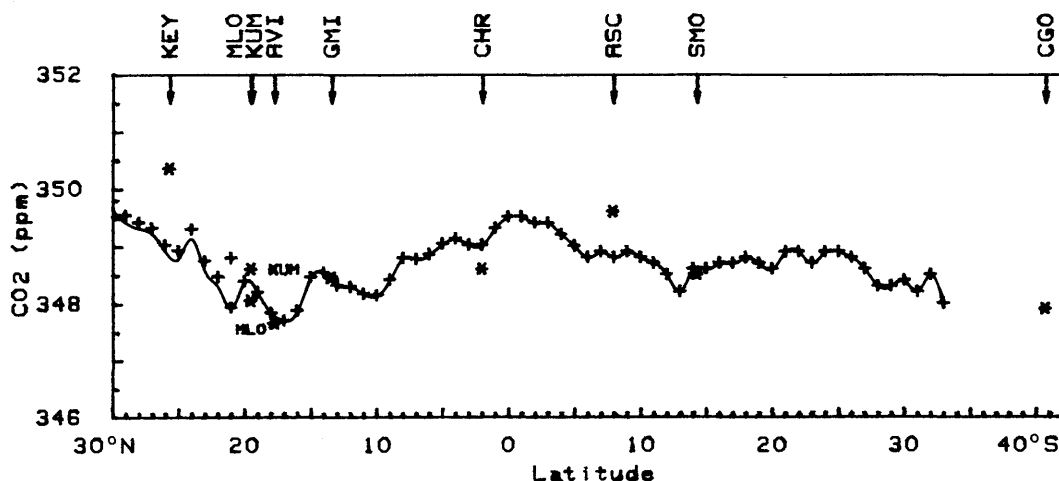


Fig. 9. The seasonally adjusted concentration of atmospheric CO<sub>2</sub> at each latitude on 30 November 1987. The CO<sub>2</sub> concentration was measured from October 31 (30°N) to December 6 (33°S). The smooth curves are cubic spline fits to the average of each degree of latitude. The site codes of land stations are those of Conway et al., (1988). ASC; Ascension Island, UK. CGO; Cape Grim, Australia. CHR; Christmas Island, Kiribati. GMI; Guam, USA. KEY; Key Biscayne, USA. KUM; Cape Kumukahi, USA. MLO; Mauna Loa, USA. SMO; American Samoa, USA.

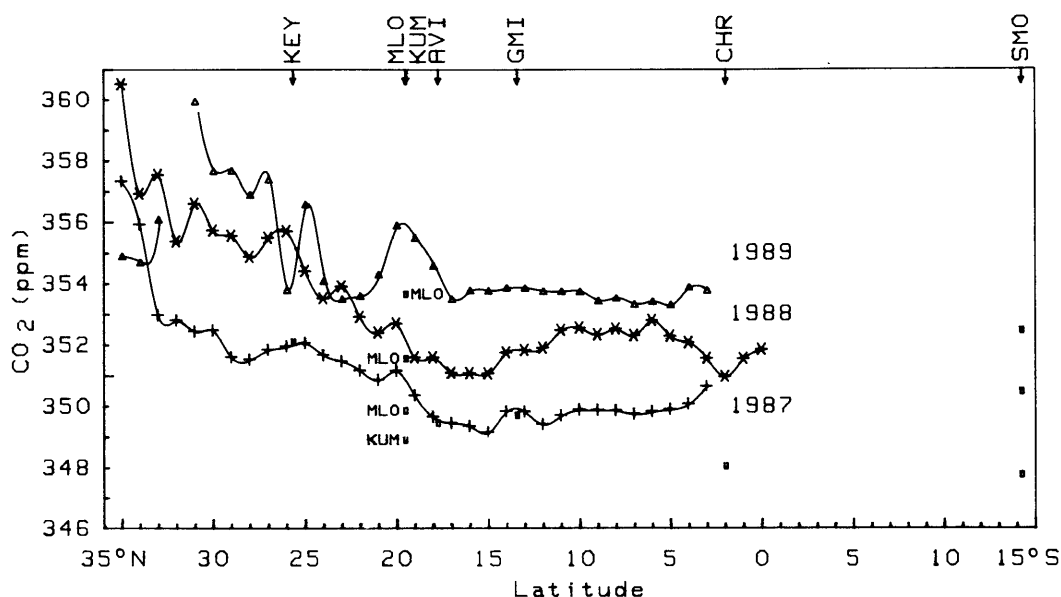


Fig. 10. The distribution of atmospheric  $\text{CO}_2$  on 31 January. The seasonal variation in  $\text{CO}_2$  concentration during the cruise was adjusted (see text). The smooth curves are cubic spline fits to the average of each degree of latitude. The concentration was measured from January 14 ( $35^\circ\text{N}$ ) to January 26 ( $3^\circ\text{N}$ ) in 1987, from January 16 ( $35^\circ\text{N}$ ) to January 30 ( $0^\circ$ ) in 1988, and January 19 ( $35^\circ\text{N}$ ) to January 30 ( $3^\circ\text{N}$ ) in 1989. The site code of the land stations is same as in Fig. 9. + : 1987, \* : 1988,  $\Delta$  : 1989.

sources and biospheric sources on land. During the boreal winter season, the strong and persistent northwest winds around the Siberian anticyclone result in a steeper gradient of the  $\text{CO}_2$  concentration in winter than is observed in the NOAA/CMDL (formerly SMCC) data (Komhyr et al., 1985). In January 1989, the distribution of the  $\text{CO}_2$  concentrations is quite different from others observed every January for the period from 1981 to 1988. The latitudinal distribution of atmospheric  $\text{CO}_2$  for 1981 to 1985 has been already reported (Inoue et al., 1987). Low concentrations around  $34^\circ\text{N}$  and  $24^\circ\text{N}$  in January 1989 are caused by the cyclone which brought air from lower latitudes. Instead of northwest wind we had prevailing south winds around  $34^\circ\text{N}$  and  $24^\circ\text{N}$ . The synoptic scale weather patterns support the present conclusion (DMA, 1989).

Differences in  $\text{CO}_2$  concentration south of  $15^\circ\text{N}$  between 1988 and 1987 were larger than those between 1989 and 1988, which are caused by the effect of regional sources in 1988 and/or high growth rate of atmospheric  $\text{CO}_2$  in 1987/88. Based

on time series analysis of  $\text{CO}_2$  data at the background stations, variations in the growth rate of atmospheric  $\text{CO}_2$  have been reported to be high in late 1987 and the first part of 1988, and sources located at northern high latitudes were suggested (Thoning et al., 1989a).

Along  $160^\circ\text{W}$ , the large increase of the  $\text{CO}_2$  concentration of atmospheric  $\text{CO}_2$  between 1989 and 1987 (4.8 ppm) is also seen in Fig. 11, and the distribution of atmospheric  $\text{CO}_2$  is fairly constant between  $20^\circ\text{N}$  and  $5^\circ\text{S}$ . A local maximum of  $\text{CO}_2$  concentration ( $\leq 1$  ppm), which exists near the equator during non-ENSO events, disappeared during the 1982/83 ENSO event (Conway et al., 1988). This is at least in part caused by the change in the  $\text{CO}_2$  flux of the equatorial marine  $\text{CO}_2$  source. As described above, the equatorial Pacific acts as a strong source during the La Niña event. Therefore, a clear maximum in the atmospheric  $\text{CO}_2$  concentration is expected near the equator in January 1989. However, only a small maximum seems to exist in the equatorial region along  $160^\circ\text{W}$  ( $\leq 0.5$  ppm) in January 1989. The reason

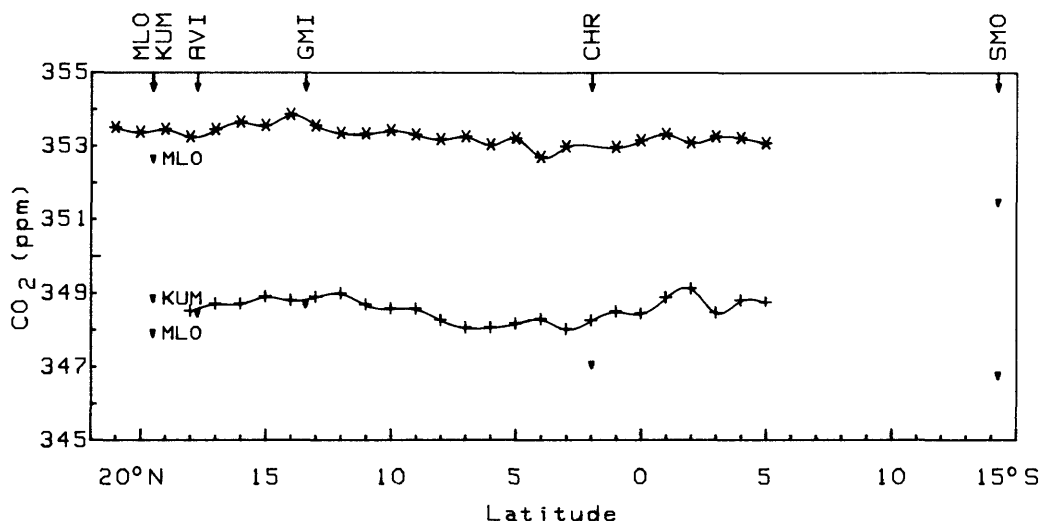


Fig. 11. The distribution of atmospheric CO<sub>2</sub> on 31 January. The seasonal variation in CO<sub>2</sub> concentration during the cruise was adjusted (see text). The smooth curves are cubic spline fits to the average of each degree of latitude. The CO<sub>2</sub> concentration was measured from February 16 (5°S) to February 22 (18°N) in 1987, and from January 13 (21°N) to January 20 (5°S) in 1989. The site code of the land station is same as in Fig. 9. + : 1987, \*: 1989.

for the small maximum is not clear at the moment, but could partly be due to the short time observation, and limited latitudinal coverage.

### 3.7. $\delta^{13}\text{C}$ of atmospheric CO<sub>2</sub> during the period from January 1989 to February 1989

We evaluate the long-term trend of  $\delta^{13}\text{C}$  of atmospheric CO<sub>2</sub>, and whether sources of oceanic CO<sub>2</sub> during the El Niño and La Niña event could affect it. When the atmospheric CO<sub>2</sub> concentration is changed by CO<sub>2</sub> exchange between the air and the sea, the rate of  $\delta^{13}\text{C}$  change with respect to the concentration in the air is much smaller than the CO<sub>2</sub> exchange between the air and the biosphere because of the small kinetic fractionation (Siegenthaler and Munnich, 1981; Inoue and Sugimura, 1985; Wanninkhof 1985).

Here, we consider the long-term trend of  $\delta^{13}\text{C}$  and the concentration of atmospheric CO<sub>2</sub> during the boreal winter season. The concentration of atmospheric CO<sub>2</sub> observed in the winter season is adjusted to January 31 for seasonal variation by Fourier harmonics (eq. (7)), and the seasonally adjusted  $\delta^{13}\text{C}$  was obtained by assuming a ratio of  $-0.05\text{‰ ppm}^{-1}$  (valid for short-term correlations, not necessarily for the long-term trend) based on observations at land stations (Keeling

et al., 1990). The atmospheric CO<sub>2</sub> concentration along 137°E on January 31, 1987 shows good agreement with the average CO<sub>2</sub> concentration between January and February in 1987 at Guam 1987 (Fig. 10), which seems to be the CO<sub>2</sub> concentration of background air. To remove the effects of regional sources and sinks from the observed data in 1988 and 1989, the concentration rise was assumed to be equal to the averages of Mauna Loa (Boden et al., 1990) and American Samoa (SMO, Tans, 1990 personal communication). The rise of the average concentration in January and February is 2.7 ppm between 1988 and 1987, and 2.0 ppm between 1989 and 1988. Differences between the background concentration and the observed concentration were attributed to regional effects, and the rate of  $\delta^{13}\text{C}$  change with concentration was taken to be  $-0.05\text{‰ ppm}^{-1}$ . The concentration and  $\delta^{13}\text{C}$  data of atmospheric CO<sub>2</sub> data between 7°N and 13°N, which is near the Guam observation site, give a negative ratio ( $-0.05\text{‰ ppm}^{-1}$ ) between 1988 ( $351.2 \pm 0.2$  ppm,  $-8.06 \pm 0.03\text{‰}$ ,  $n=6$ ) and 1987 ( $348.5 \pm 0.2$  ppm,  $-7.93 \pm 0.03\text{‰}$ ,  $n=5$ ), and a less negative ratio ( $-0.02\text{‰ ppm}^{-1}$ ) between 1989 ( $353.2 \pm 0.3$  ppm,  $-8.10 \pm 0.02\text{‰}$ ,  $n=6$ ) and 1988. The former result indicates the

enhanced CO<sub>2</sub> release by land plants and soil, and/or fossil fuel combustion. The role of the terrestrial biosphere has been emphasized by Keeling et al., (1989), who indicate lower  $\delta^{13}\text{C}$  during the El Niño event in 1987. It could be partly caused by the droughts and great fire in China observed from satellite (Levine 1990). According to the results of Mauna Loa and South Pole, the rate of  $\delta^{13}\text{C}$  change decreased in the second half of 1988, compared to that of 1987 and the first half of 1988 (Keeling et al., 1989). The decreased rate of  $\delta^{13}\text{C}$  with respect to the air CO<sub>2</sub> concentration between 1989 and 1988, which is nearly equal to the long-term trend reported earlier (Mook et al., 1983; Keeling et al., 1984; Francey 1985), implies a more important role for oceanic CO<sub>2</sub> sources in determining the atmospheric CO<sub>2</sub> concentration than for the period between 1988 and 1987. The change in the  $\delta^{13}\text{C}$  rate against concentration was at least partly due to the CO<sub>2</sub> flux (1 Gt-C yr<sup>-1</sup>) in the equatorial Pacific during the La Niña event.

#### 4. Summary

Measurements of CO<sub>2</sub> in and over the equatorial Pacific were made along with flask sampling of air during the period of the 1986/88 ENSO event to the 1988/89 La Niña event. The data on which this paper is based are available in a summarized form on floppy disks from the authors and will be made available from the Japan Climatic Data Center, JMA in the future.

The pCO<sub>2</sub> of the equatorial Pacific was low during the 1986/88 ENSO event but less dramatically than during the 1982/83 ENSO event.

The pCO<sub>2</sub> in the central equatorial Pacific in January and February 1989 was remarkably high in comparison with that of January and February 1987, which was caused by strong upwelling. Along 137°E, however, the pCO<sub>2</sub> has not varied

every boreal winter for the period from 1987 to 1989. By using the relationship between gas transfer coefficient and wind speed suggested by Takahashi et al., (1989), the equatorial Pacific CO<sub>2</sub> flux was estimated to be 0.4 Gt-C year<sup>-1</sup> in January and February 1987, and 1 Gt-C year<sup>-1</sup> in January and February 1989. The increase in CO<sub>2</sub> flux between these two period could affect the atmospheric CO<sub>2</sub> growth rate. The surface water pCO<sub>2</sub> distribution during the zonal flow along the equator was well approximated by effects on pCO<sub>2</sub> of the temperature, salinity, biological activity and the CO<sub>2</sub> flux between the sea and the air.

The growth rate of atmospheric CO<sub>2</sub> has been reported to be high in the late 1987 and the first part of 1988. The long-term trend of  $\delta^{13}\text{C}$  and the concentration of atmospheric CO<sub>2</sub> indicate the predominant role of the biosphere (droughts and fire) for the atmospheric CO<sub>2</sub> increase between 1988 and 1987, and an important role of CO<sub>2</sub> exchange between the sea and the air between 1989 and 1988, as compared to the preceding year.

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