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Geographical, seasonal and interannual variations of air-sea CO_2 exchange in the subtropical Pacific surface waters during 1983–1988

(I). Variabilities of oceanic fCO_2

By C. S. WONG[1]*, Y.-H. CHAN[2], J. S. PAGE[1], G. E. SMITH[1], R. D. BELLEGAY[1] and K. ISEKI[1,3],
[1]*Centre for Ocean Climate Chemistry, Institute of Ocean Sciences, P. O. Box 6000, Sidney, BC, Canada V8L 4B2*; [2]*Western Ecological Services, Ltd., #2-2563 Penrhyn Street, Victoria, BC, Canada V8N 1G2*;
[3]*Present address: Fisheries Oceanography Division, Seikai National Fisheries Research Institute, 49 Kokubu-machi, Nagasaki 850, Japan*

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ABSTRACT

The fugacity of CO_2 (fCO_2) in the surface seawater of the northeast (15°N–45°N) and southwest (25°S–15°S) subtropical Pacific Ocean from November 1983 to February 1988 were observed on 18 cruises on the M/V Lillooet. The northeast Pacific within 15°N–45°N was a net CO_2 sink with a mean annual ΔfCO_2 of about $-5\,\mu$atm, as the average of $-19\,\mu$atm in the boreal winter and spring and $8\,\mu$atm in the boreal summer and autumn. Our cruises also obtained the first set of austral winter and spring fCO_2 data for the southwest subtropical Pacific and thus provided a complete seasonal cycle of surface fCO_2 for this oceanic region. The southwest Pacific within 25°S–15°S, with a mean annual ΔfCO_2 of about $-20\,\mu$atm, was a CO_2 sink year round except during the austral summer of the El Niño whence ΔfCO_2 had positive values. The observed sea-surface fCO_2 showed large variability. Between the summer and the winter months, the seasonal amplitudes of fCO_2 (normalized to the climatological annual mean SST) ranged from $1\,\mu$atm to $63\,\mu$atm. The surface waters of the subtropical and the northeast Pacific also displayed annual trends in the normalized fCO_2 values. From January 1984 to January 1987, there were decreases in the seasonal means of normalized fCO_2 of 4–11 μatm yr^{-1} within 25°S–15°S, 15°N–25°N, and 25°N–35°N. These trends were within the uncertainty of the data and were not significant statistically. However, from summer 1985 to summer 1987, the normalized fCO_2 showed significant positive increase of 24 μatm yr^{-1} within 35°N–45°N. There was also an increase, though not significant statistically, of 13 μatm yr^{-1} within 25°N–35°N. The cause of the slightly larger increase in fCO_2 in northern subtropical gyre could be the anomalous intrusions of the Oyashio Current during the period 1984–1986 along the North Pacific Current, which was the source of the California Current bifurcating into the waters of 25°N–35°N and 35°N–45°N. These intrusions in late spring usually followed an intensified and eastward shift of the Aleutian Low which was especially persistent during 1977–1988.

1. Introduction

Carbon dioxide is one of the major radiatively active gases that are known to affect the Earth's climate. Since the industrial revolution, the level of atmospheric CO_2 has been increasing due mainly to the combustion of fossil fuels and from land-use changes (Keeling, 1973; Bolin, 1977; Woodwell et al., 1978; Bolin et al., 1979; Chan et al., 1980). The accumulation of CO_2 in the atmosphere around 1990s is estimated to be about 1.8 ppmv yr^{-1} (Boden et al., 1991; IPCC, 1990; 1992). This

* Corresponding author.

increase represents the annual addition of that fraction of the anthropogenic CO_2 that is not transferred to the global carbon sinks but is remained air borne. While a fraction of the anthropogenic CO_2 emission may be sequestering in the biosphere, a large part of this emission is being taken up by the ocean. The magnitude of this oceanic uptake is uncertain with disagreement (Tans et al., 1990; Quay et al., 1992; Siegenthaler and Sarmiento, 1993).

In general, the surface water of the equatorial region is a large CO_2 source. The north Atlantic Ocean and the vast oceanic areas of the southern temperate latitudes are basically CO_2 sinks (Takahashi et al., 1986; Takahashi, 1989; Tans et al., 1990). The magnitude of the flux depends on a coefficient of CO_2 air-sea exchange and the difference in the CO_2 partial pressures between the air and the overlying surface seawater (ΔpCO_2). Using a $2^\circ \times 2^\circ$ geographical distribution of seasonal ΔpCO_2 and climatological wind speed coupled to an atmospheric general circulation model, Tans et al. (1990) estimate the global annual ocean CO_2 sink of less than 1.0 Gt C yr^{-1}, in disagreement with other experimental approach based on $^{13}C/^{12}C$ (Quay et al., 1992) or modelling (Siegenthaler and Joos, 1992; Sarmiento et al., 1992). Recent reviews of the oceanic uptake of anthropogenic CO_2 (Sarmiento and Sundquist, 1992; Siegenthaler and Sarmiento, 1993) suggested that the estimate of Tans et al. neglects several important processes and that available evidence supports a mean oceanic CO_2 uptake rate of about 2 Gt yr^{-1}. However, the approach used by Tans et al. (1990) may also be improved by increasing the numbers of ΔpCO_2 and wind speed data since these two terms are known to be highly variable both spatially and temporally (Erickson, 1989; Etcheto et al., 1991; Wong et al., 1993).

Since the International Geophysical Years, the pCO_2 or ΔpCO_2 over the Pacific surface seawaters have been measured quite extensively (Keeling et al., 1965; Miyake and Sugimura, 1969; Miyake et al., 1974; Gordon et al., 1971; 1973; Kelley and Hood, 1971; Takahashi et al., 1986; Feely et al., 1987; Fushimi, 1987; Inoue et al., 1987; Inoue and Sugimura, 1988a; 1988b; Murphy et al., 1991a; 1991b; Wong and Chan, 1991; Wong et al., 1993). The global distribution of ΔpCO_2 is summarized early by Keeling (1968) and then by Takahashi et al. (1986) and Takahashi (1989). However, many areas of the Pacific Ocean are still undersampled.

The spatial and temporal variations of pCO_2 in surface seawater exert great influence on the short-term variability of the global carbon cycle. Factors that control the partial pressure of oceanic CO_2 include sea water temperature, biological activity, circulation, vertical and lateral mixing of waters. Relating these factors with ΔpCO_2 variability are important for the understanding of CO_2 air-sea exchange. During spring blooms, the drawdown of CO_2 by phytoplanktons may reach about 200 μatm within relatively short time and spatial scales (Codispoti et al., 1982; 1986; Kempe and Pegler, 1991; Watson et al., 1991). Seasonal variations of the sea-surface pCO_2 may range from about 60 μatm in the subarctic Pacific (Gordon et al., 1971) to about 20 μatm in western North Pacific and the subtropical gyres (Weiss et al., 1982; Inoue et al., 1987; Inoue and Sugimura, 1988a).

Studies on the interannual variations of pCO_2 and air-sea CO_2 flux are few (Inoue et al., 1987; Inoue and Sugimura, 1988a). One factor that exerts a large impact on the year-to-year changes in the air-sea exchange of CO_2 in the Pacific is the occurrence of El Niño (Feely et al., 1987; Fushimi, 1987; Inoue and Sugimura, 1988b; Wong et al., 1993). However, the El Niño effects in the eastern or central tropical Pacific differ from those observed in the western Pacific (Fushimi, 1987; Inoue and Sugimura, 1988b).

The secular increase in total dissolved inorganic carbon (DIC) or partial pressure of CO_2 (pCO_2) in surface seawater (Broecker et al., 1979; Keeling, 1983; Takahashi et al., 1983) or change in isotopic $^{13}C/^{12}C$ ratio (Quay et al., 1992) can be considered a direct proof of the oceanic uptake of fossil fuel carbon. For DIC in surface seawater, the increase is predicted to be about 0.7 μmol CO_2 kg^{-1} but is also not easy to measure in practice because of the limitation of CO_2 methodology and the natural variability of DIC. The secular increase of pCO_2 follows loosely the rise in atmospheric CO_2 at a rate of about 1.5 ± 0.5 μatm yr^{-1} (Takahashi et al., 1983; Roos and Gravenhorst, 1984; Inoue and Sugimura, 1988a). One of the causes of the uncertainty of this estimate is the changes in methods for pCO_2 measurements made over decades. Repeat measurements in the oceanic regions using the same technique, as done in this

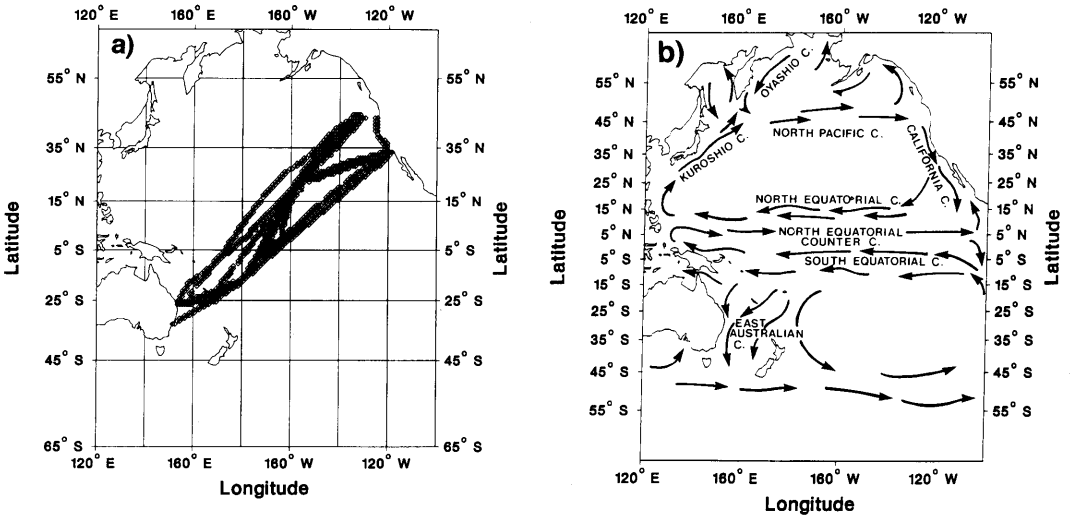

Fig. 1. (a) Cruise tracks of M/V Lillooet. (b) Major surface current systems of the Pacific.

Table 1. *The M/V Lillooet Cruises*

Cruise No.	Dates	Notes
452S	30 Nov 1983–21 Dec 1983	GC data for 28°S–7°N, 22–34°N; route 1
453N	8 Jan 1984–24 Jan 1984	no GC data north of 12°S; route 3
458S	29 Jun 1984–25 Jul 1984	GC data within 21°S–24°N only; route 1
463N	10 Jan 1985–26 Jan 1985	route 2
465N	12 Mar 1985–29 Mar 1985	route 2
469N	12 Aug 1985–30 Aug 1985	route 2
473N	16 Jan 1986– 2 Feb 1986	no GC data south of 20°S; route 4
475N	23 Mar 1986–13 Apr 1986	route 2
479N	1 Aug 1986–19 Aug 1986	GC data for 21°S–2°N, 22–31°N; route 5
481N	11 Oct 1986–28 Oct 1986	no GC data north of 26°N; route 2
483N	14 Dec 1986– 2 Jan 1987	route 2
485N	21 Feb 1987–12 Mar 1987	route 2
487N	1 May 1987–26 May 1987	route 3
489N	21 Jul 1987–10 Aug 1987	route 2
491N	20 Sep 1987– 8 Oct 1987	route 5
493N	26 Nov 1987–11 Dec 1987	route 6
494S	31 Dec 1987–12 Jan 1988	route 1
495N	4 Feb 1988–20 Feb 1988	route 6

route 1: Vancouver–Oakland–Sydney
route 2: Brisbane–Noumea–Suva–Honolulu–Vancouver
route 3: Brisbane–Noumea–Suva–Long Beach–Vancouver
route 4: Brisbane–Vancouver
route 5: Brisbane–Noumea–Suva–Honolulu–Oakland–Vancouver
route 6: Brisbane–Honolulu–Oakland–Vancouver

program, are necessary for determining the long-term trend of pCO_2 in surface seawater.

In 1983, the Centre for Ocean Climate Chemistry at the Institute of Ocean Sciences initiated a ship-of-opportunity program as part of the Oceanic CO_2 Monitoring Project to study the carbon system of the Pacific Ocean (Wong et al., 1984; 1993). Various properties of ocean surface waters along the ship routes (Fig. 1) were sampled periodically for several years. The partial pressures of CO_2 and nutrient levels in surface seawater and net CO_2 fluxes near the central tropical Pacific within 15°S–15°N before and during the 1986/1987 El Niño were reported earlier (Wong et al., 1993). This report presents the CO_2 fugacity (fCO_2) measurements during the 18 cruises on the M/V Lillooet between November 1983 and February 1988. The oceanic areas discussed in this report cover the zones of southwest subtropical Pacific within 25°S–15°S and northeast Pacific within 15°N–45°N (Table 1).

2. Method

The ship-of-opportunity program was carried out on board the German container-carrier M/V Lillooet which sailed between Canada and Australia six times a year. The three south-bound cruises sailed from Vancouver to Long Beach or Oakland, USA, then crossed the equator at about 170°W–160°W, and arrived finally in Brisbane or Sydney, Australia. For the 15 north-bound cruises, the route normally started from Brisbane, Australia, and crossed the equator at about 170°W–180°W. Except for a few cruises, the north-bound route included a stop at Honolulu. Four of the cruises also visited Oakland before heading north. Several of the north-bound cruises also included other ports-of-visit at Noumea, New Caledonia, and Sauva, Fiji. Two of the cruises, 453N and 487N, developed engine problems early on and sailed directly from Suva to Long Beach, California, before heading to Vancouver.

Details of the procedure for sampling and analyzing equilibrium CO_2 in surface seawater were described fully in Wong et al. (1993). In the following, we give only a description of the methodology together with other details relevant to this report.

2.1. CO_2 in atmosphere and surface seawater

The CO_2 sampling system installed on the ship was designed to measure both the contents of CO_2 in the atmosphere and in a dried aliquot of a gas phase equilibrated with surface seawater. A bellows-diaphragm pump delivered seawater from a sea chest ~5 m below sea level in the engine room at a rate of 20 l min^{-1} with a diversion through a seawater loop to a shower-type pCO_2 equilibrator (Keeling et al., 1965). Volumes of seawater and enclosed air in the equilibrator were 2 l and 1 l respectively. The seawater was being circulated continuously in a loop of stainless steel tubing. All pipes were insulated to keep the temperature difference between the intake and the equilibrator to within ~1°C under field condition for most of the time. Temperatures of seawater at the intake and in the equilibrator were recorded and their difference was used to correct for the temperature effect on the fugacity of CO_2 in seawater.

The mole fractions of CO_2 were measured, after catalytic conversion to methane in pure hydrogen carrier gas, by gas chromatography with a flame-ionization detector (Weiss, 1981). A series of automated solenoid valves provided time-sequence sampling of CO_2-in-air from a single gas standard, the equilibrated air, and the marine air. Each set of analyses included two 1 ml samples of the standard gas bracketing a sample of each of the seawater and atmospheric air. The difference in the two standard-gas peak areas was used to adjust the respective peak area of the samples proportionately. Total time for the 4 measurements, signal integration and data logging were about 17 min in the first 3 cruises, but adjusted to about 27 min in later cruises. The working standards were prepared with near-ambient concentration of CO_2 which were calibrated in our laboratory with WMO primary standards obtained from the Scripps Institution of Oceanography (SIO). Additional flask samples of marine air were also collected daily in quadruplicate for comparison purpose. A shorebased nondispersive infrared analyzer was used to measure the mole fractions of atmospheric CO_2 in these samples.

The position of the ship was relayed from a satellite navigation system directly to the microcomputer attached to the analyzing system. The printout from the computer included the analysis

readings of CO_2 in the various sources, position of the ship, air temperature, and seawater temperatures both at the intake and in the equilibrator. Additional meteorological records for the same Marsden Squares covering the cruise tracks were obtained from the National Climatic Data Center of NOAA, USA. The records included mean sea level barometric pressure, wind speed, and wet-bulb and dry-bulb temperatures.

2.2. *Computations and data analyses*

The equilibrium fCO_2 of seawater in the dataset displayed fairly large variability. After checking with the cruise logs, a few of the extreme outliers were deleted from the dataset before further analysis. All sample measurements were averaged daily. Individual values of those oceanographic or climatic variables required for fCO_2 or flux calculations were estimated by cubic spline interpolation. To analyze for time variations, the weighted daily averages were used to compute the monthly or seasonal means. The boreal winter (or austral summer) included January and February of the current year plus December of the preceding year.

2.2.1. Fugacity of CO_2 in air. For the first three cruises, the levels of atmospheric CO_2 were interpolated from results of the flask samples collected during the cruises. For the other cruises, in addition to our flask CO_2 data, we also relied on the data of atmospheric CO_2 level from the network of GMCC stations operated by NOAA, USA, as done for the case of equatorial Pacific data set (Wong et al., 1993). For each sampling cycle, the atmospheric CO_2 level was estimated through two interpolation steps. The mole fractions of atmospheric CO_2 on the days when the ship crossed the same latitudes as those Pacific stations at Cape Grim (Tasmania), Cape Matulala (American Samoa), Christmas Island, Cape Kumukahi (Hawaii), Sand Island (Midway Islands), and Cape Meares (Oregon, USA) were first interpolated from the respective CO_2 time series (Boden et al., 1991). Thus, for each cruise, we obtained a set of values representing the spatio-temporal distribution of atmospheric CO_2 levels. The appropriate data were interpolated to obtain the mole fraction of atmospheric CO_2 for each GC sampling cycle at a given time and latitude. Each interpolated value of CO_2 concentration was converted to atmospheric CO_2 fugacity (in μatm) using the equations of Weiss (1974) as in Wong and Chan (1991). The CO_2 fugacity, assuming moisture-saturated at the air-sea interface, was computed by using the recorded barometric pressure and the saturated vapour pressure at sea surface temperature. The differences between fugacity and partial pressure, which depend on ambient pressure and gas composition (UNESCO, 1991), were less than $\pm 0.4\%$ of ambient pCO_2 observed during our cruises, i.e., about ± 1.5 μatm at 380 μatm level to ± 0.8 μatm at 190 μatm.

2.2.2. Fugacity of CO_2 in equilibrium with surface seawater. As in atmospheric CO_2, the level of CO_2 in equilibrium with surface seawater was also expressed as CO_2 fugacity (fCO_2). To compute the seawater fCO_2 (in μatm), the mole fraction of CO_2 equilibrated with seawater at in situ temperature was converted to CO_2 fugacity assuming saturated vapour pressure in the equilibrator (Wanninkhof and Thoning, 1993). Any change in the value of fCO_2 due to temperature difference after sampling at the intake was corrected by an empirical equation relating the dependency of fCO_2 temperature coefficient (PT, in $\%\ °C^{-1}$) on salinity (S). This equation, which is expressed as,

$$PT = 4.272 + 5.244 \times 10^{-2}\, S - 2.524 \times 10^{-3}\, S^2, \quad (1)$$

was fitted from the experimental data obtained in our laboratory (Wong, unpubl. data). Since the seawater sample was collected at ~ 5 m below sea surface, the recorded bulk SST might be slightly higher than the actual water temperature at the air-sea interface (Hasse, 1971). A correction for this "thermal skin effect" (Robertson and Watson, 1992) in calculating air-sea fluxes of CO_2 from our samples will be discussed in Part II of this report.

2.2.3. Normalized fugacity of CO_2. In comparing the long-term variations of fCO_2 observed during our cruises, we adjusted the in situ fCO_2 values to remove the effect of temperature. Eq. (1) was used to adjust for the different temperature effects on fCO_2 in various oceanic zones by normalizing the values of daily mean fCO_2 to the climatological annual mean SST for each zone. The climatological means, which were estimated from the dataset compiled by Esbenson and Kushnir (1981), were 25.5°C, 24.6°C, 20.0°C and 15.3°C for zones 25°S–15°S, 15°N–25°N,

25°N–35°N and 35°N–45°N respectively. The normalized daily means were averaged over a given season to obtain the seasonal mean.

3. Results and discussion

For clarity in presenting the results, the southwest subtropical Pacific and the northeast Pacific regions are divided into four zones: 25°S–15°S within about 157°E–175°E, 15°N–25°N within 170°W–130°W, 25°N–35°N within 165°W–125°W and 35°N–45°N within 150°W–130°W. Large portion of the cruise tracks lie within the north Pacific subtropical gyre. The six cruises that sailed between Honolulu and Oakland or Long Beach also traverse the coastal waters of the California Current within the region 28°N–32°N and 125°W–120°W (Table 1, Fig. 1). Data from these coastal waters collected occasionally

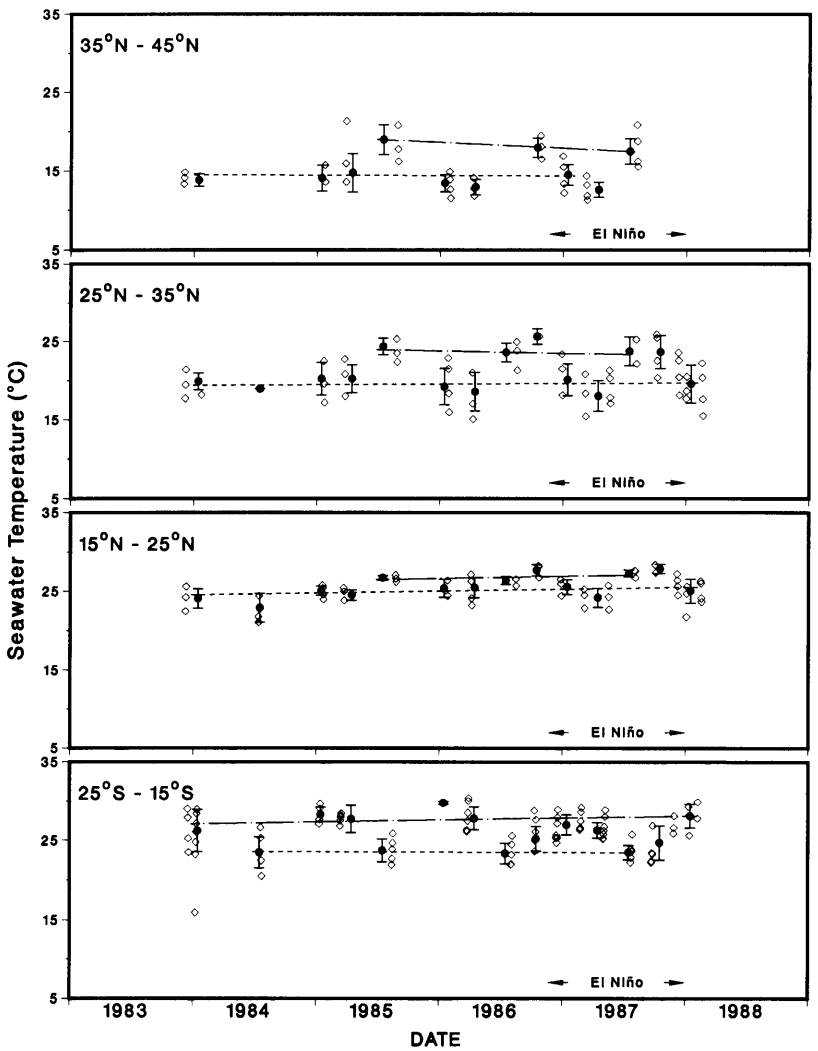

Fig. 2. Sea-surface temperature of the southwestern and northeastern subtropical Pacific observed during the cruises. The open diamonds are daily means and the solid circles are seasonal means with the error bar representing $\pm 1\sigma$. The boreal winter (austral summer) includes January and February of the current year and December of the preceding year.

between Vancouver and Oakland or Long Beach were not used in this study. Only results of open-ocean data were discussed below. Figs. 2, 3 give the zonal averages of SST and salinity in surface seawaters for all the cruises. Results from the equatorial region have been discussed in a previous report (Wong et al., 1993). Fig. 4 shows the seasonal variation in fCO_2 within the latitudinal zones in seasonal groups. The average differences in CO_2 fugacities in equilibrium with surface seawater and in the overlying air [$\Delta fCO_2 = fCO_2(\text{sw}) - fCO_2(\text{air})$] is similarly displayed in Fig. 5. All dates and times are given in Universal Time Coordinated (UTC).

3.1. *SST and salinity*

The SST during 1984–1988 sampled within 25°S–15°S averaged about 27.1 ± 2.2°C in the austral summer but lowered to about 23.5 ± 1.5°C in austral winter (Fig. 2). In the northern

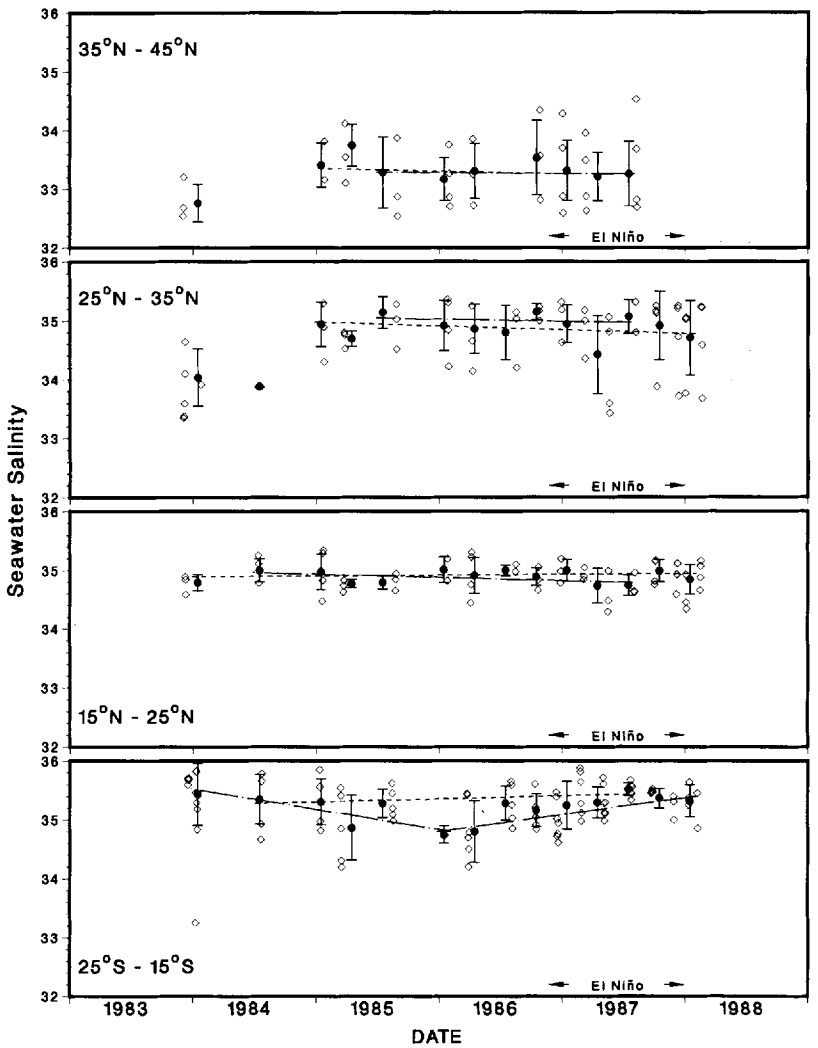

Fig. 3. Salinity of surface seawater from the southwestern and northeastern subtropical Pacific observed during the cruises. The open diamonds are daily means and the solid circles are seasonal means with the error bar representing $\pm 1\sigma$. The boreal winter (austral summer) includes January and February of the current year and December of the preceding year.

region, the SST during boreal summer decrease from averages of 25.3 ± 3.3°C within 15°N–25°N to 23.5 ± 2.1°C within 25°N–35°N and to 18.2 ± 2.0°C within 35°N–45°N (Fig. 2). Within 25°S–15°S and 15°N–25°N, the summer SST observed during the 1986/1987 El Niño period were 1–2°C warmer than the SST observed during the non-El Niño years. However in the northern waters within 25°N–45°N, there was a slight decrease of summer SST from 1985 to 1987. Averages of winter SST for the northern zones, i.e., 15°N–45°N, lowered from 24.9 ± 1.4°C to 14.3 ± 2.8°C northward. The summer SST averages for both latitudinal zones at 25°S–15°S and 15°N–25°N were within the same range, but the winter SST within 25°S–15°S averaged about 1°C lower than those observed within 15°N–25°N. The SST observed in winter, as well as those observed in spring and autumn, were not noticeably different before or during the El Niño.

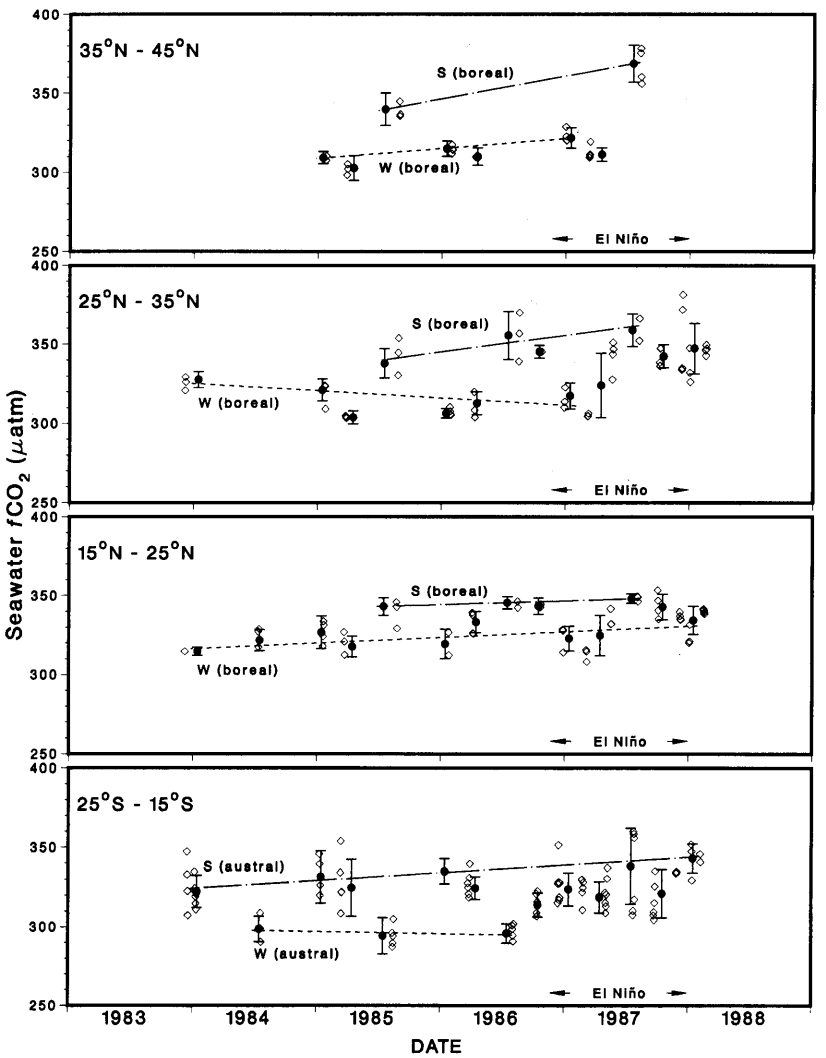

Fig. 4. Averages of fCO_2 in surface seawater observed at the southwestern and northeastern subtropical Pacific. The open diamonds are daily means and the solid circles are seasonal means with the error bar representing $\pm 1\sigma$. Annual trends for both the summer and the winter means are also shown. The boreal winter (austral summer) includes January and February of the current year and December of the preceding year.

During 1984–1988, the averaged surface salinity within 25°S–15°S showed hardly a seasonal variation with a summer mean of 35.32 ± 0.45 and a winter mean of 35.35 ± 0.31. Slightly lower values of salinity were observed during the austral autumn of 1985 and 1986 (Fig. 3). No seasonal variation in salinity was observed within 15°N–25°N. However, the seasonal averages of salinity in this zone, i.e., 34.87 ± 0.19 in summer and 34.88 ± 0.24 in winter, were slightly fresher than those from 25°S–15°S (Fig. 3). In the northeast Pacific, the surface salinity gradually decreased northward to a summer mean of 33.27 ± 0.58 and a winter mean of 33.14 ± 0.47 within 35°N–45°N. Salinity within 25°N–45°N observed during each cruise decreased poleward by as much as 2 units as the ship crossed different water masses from the subtropical gyre to the California Current. In these two northern zones, the overall annual trends showed a slight decrease in salinity from the 1985 winter to the 1988 winter (Fig. 3).

Although the observed salinity from our cruises agreed generally with the distribution of surface salinity of the Pacific (Pickard and Emery, 1982), abnormally low values of salinity were observed in northern waters during the 1983/1984 winter (Fig. 3). The SST from this region during summer 1984 was also colder than normal. In fact, there was a large pool of cold water between 40°N to 50°N across the Pacific in the whole year of 1984 (NOAA, 1984). The low SST and low salinity also supported the observation of an anomalous southward intrusion of the less saline Oyashio Current to the North Pacific Current in 1984 (Sekine, 1988). This condition in late spring commonly occurred following an intensified and eastward shift of the Aleutian low in the previous winter half year and stronger southeastward wind stress by the Siberian high in the western North Pacific. Associated with the increase in surface wind stress was a southward shift of the subarctic circulation and an increased biological activities in the central North Pacific (Emery and Hamilton, 1985; Venrick et al., 1987; Trenberth and Hurrell, 1994).

3.2. *Sea surface fCO_2*

In the southern subtropical gyre 25°S–15°S , there were large differences between the averages of fCO_2 (Fig. 4) observed during the austral winter (about 296 ± 8 μatm for 1984–1986) and those during the austral summer (about 327 ± 13 μatm). In the 1987 austral winter during the El Niño, the fCO_2 increased to an average of 338 μatm, about 42 μatm over the 1984–1986 mean. These ranges of values from the southern zone were slightly less than the averages obtained from 15°N–25°N, i.e., 340 ± 12 μatm in the boreal summer and 328 ± 11 μatm in the boreal winter (Fig. 4). In boreal summer 1984, the fCO_2 value within 15°N–25°N was about 15 μatm lower than the 1985–1987 mean. The averages of fCO_2 within 15°–25° north and south of the equator during the 1986/1987 El Niño were higher than those observed before the El Niño; more so in summer than in winter. This was in contrast to the condition within the equatorial region where the fCO_2 values were reduced significantly during the El Niño (Wong et al., 1993).

In the northern waters within 25°N–45°N, there were more fCO_2 data in winter than in summer. The mean summer fCO_2 within 25°N–35°N and 35°N–45°N were respectively 351 ± 15 μatm and 354 ± 18 μatm. In winter, the mean fCO_2 within 25°N–35°N and 35°N–45°N were 332 ± 20 μatm and 316 ± 7 μatm respectively (Fig. 4). The higher mean winter value from 25°N–35°N reflected the inclusion of high fCO_2 (averaged 347 μatm) observed in the 1988 winter.

Distribution of CO_2 in tropical Pacific Ocean was also studied by Inoue and Sugimura (1992). A small section of their cruises also crossed our tracks in February 1987 and 1988. In February 1987, they recorded an average pCO_2 of 350 μatm within the equatorial belt of 5°S–5°N along 180° (Inoue and Sugimura, 1992). This value was slightly lower than the averages of 356–367 μatm from our cruise 485N in February and March 1987 within the same latitudinal belt but a few degrees to the east (Wong et al., 1993). The contour map of ΔpCO_2 by Keeling (1968) showed a gradient of about +15 μatm along the equatorial Pacific from 180° to 170°W. Thus, the value from Inoue and Sugimura (1992) would be comparable to our results if their pCO_2 value is converted to fCO_2 (about 1–2 μatm less) as well as adjusted for the geographical variation. In February 1988, the pCO_2 at 15°S and 154°E recorded by Inoue and Sugimura (1992) was about 350 μatm whereas the average fCO_2 at 15°S latitude observed during

our February 1988 cruise was slightly lower at 339 μatm.

3.3. ΔfCO_2

The ΔfCO_2 [fCO_2(sw) − fCO_2(air)] value typically reflects the state of CO_2 disequilibrium of surface seawater. A positive ΔfCO_2 signifies that the surface seawater is a potential for CO_2 evasion; conversely, a negative ΔfCO_2 value indicates a potential for CO_2 invasion. For the southwest subtropical Pacific covered by our cruises, the ΔpCO_2 map of Tans et al. (1990) shows a range of −40 to −20 μatm during the January to April period for the same general geographical area. No data are shown for the July through October period (Tans et al., 1990). Murphy et al. (1991a, b) provided ΔfCO_2 from this oceanic area for the austral autumn. Our data set provides the first austral winter/spring measurements of ΔfCO_2, and together with data from the austral summer/autumn seasons, the complete annual cycle of ΔfCO_2 in the southwest subtropical Pacific. In austral winter and spring, this oceanic region within 25°S–15°S was basically a CO_2 sink with averaged ΔfCO_2 of -33 ± 20 μatm and -22 ± 14 μatm respectively (Fig. 5). During austral summer and autumn, the magnitude of ΔfCO_2 values were smaller and averaged about -9 ± 14 μatm and -18 ± 13 μatm respectively. Low positive values of ΔfCO_2 were obtained in 4 of the 11 cruises that were sampling during the warming seasons with the cruises in January and February 1988 recorded positive ΔfCO_2 of about 5 ± 12 μatm and 6 ± 5 μatm respectively. A slightly higher ΔpCO_2 value of about 10 μatm was observed by Inoue and Sugimura (1992) in February 1988 at 15°S. Based on all our data, the annual mean ΔfCO_2 for this oceanic region was about −20 μatm.

Recently, Murphy et al. (1991b) studied the CO_2 air-sea exchange during austral autumn in the southern Pacific ocean and also provided a ΔpCO_2 contour map for the study area. Their results of sea surface CO_2 data for the southwest subtropical Pacific observed during May 1987 could be used to corroborate our results from Cruise 487N in early May 1987. The contours for this oceanic area given by Murphy et al. (1991b) were from −10 to −30 μatm, whereas the latitudinal averages of ΔfCO_2 from Cruise 487N were between −5 and −31 μatm. Our results for austral summer and autumn, as well as those for southwestern Pacific from Murphy et al. (1991b), agree fairly well with the ΔpCO_2 contour of −30 to 10 μatm provided by Miyake et al. (1974). These values suggested that this region of the southwest Pacific is probably a lesser CO_2 sink as estimated by Tans et al. (1990).

In the northeast subtropical Pacific during boreal winter and spring before the mature phase of the 1986/1987 El Niño, the zonal averages of our shipboard ΔfCO_2 values were -17 ± 8 μatm within 15°N–25°N, -26 ± 6 μatm within 25°N–35°N, and -32 ± 5 μatm within 35°N–45°N (Fig. 5). The ranges of ΔfCO_2 showed less negative values during the 1987/1988 winter after the mature phase of the El Niño: -9 ± 6 μatm within 15°N–25°N and -3 ± 14 μatm within 25°N–35°N. The overall average of −19 μatm for northern Pacific waters represented a larger CO_2 sink than that suggested by the ΔpCO_2 values shown on the map of Tans et al. (1990). Their ΔpCO_2 values for the northeastern Pacific region within 35°N–45°N were only between −10 to −20 μatm, although their January–April values for the same oceanic area within 15°N–35°N were about −20 to −40 μatm, implying only 50% of the strength of the northern Pacific oceanic sink indicated by our 1984–1988 measurements of -32 ± 5 μatm.

During boreal summer/autumn at the northeast subtropical Pacific, the values of ΔfCO_2 from our cruises were mostly positive, i.e., 7 ± 5 μatm for 15°N–25°N during 1985–1987, 8 ± 9 μatm for 25°N–35°N, and 17 ± 11 μatm for 35°N–45°N (Fig. 5). One exception was the averaged ΔfCO_2 of −19 μatm within 25°N–35°N for the 1984 summer. The overall average ΔfCO_2 for the northern zones was about 8 μatm. Most of these summer averages fell within the ranges of values appeared in the ΔpCO_2 map for the July–October period given by Tans et al. (1990): about 10–40 μatm within 15°N–35°N and 0–20 μatm within 35°N–45°N.

Takahashi (1989) gave estimates of mean annual ΔfCO_2 for the Pacific north of 10°N. His values were 9 μatm for the oceanic area within 10°N–40°N east of the dateline, and 4 μatm for the area within 40°N–60°N east of the dateline. Tans et al. (1990) also provided mean values for the Pacific north of 15°N: −11 μatm for January to April, 14 μatm for July to October, and an

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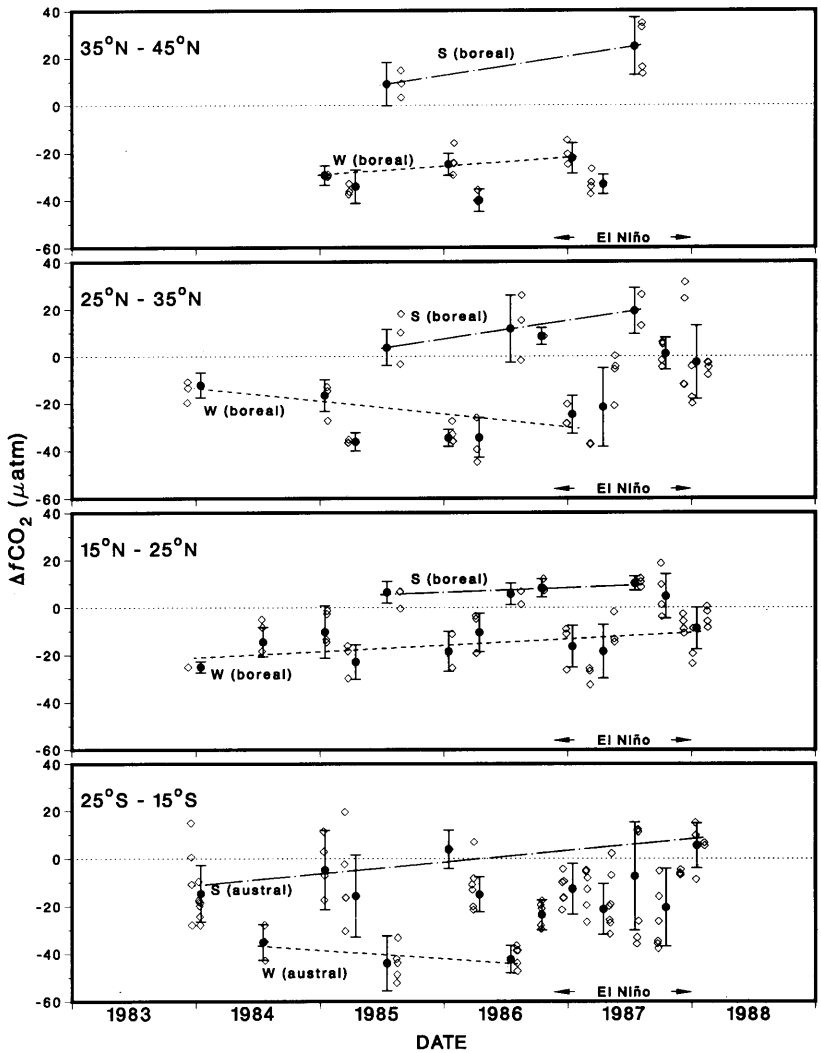

Fig. 5. Averages of ΔfCO_2 [$fCO_{2(sw)} - fCO_{2(air)}$] estimated for the southwestern and northeastern subtropical Pacific. The open diamonds are daily means and the solid circles are seasonal means with the error bar representing $\pm 1\sigma$. Annual trends for both the summer and the winter means are also shown. The boreal winter (austral summer) includes January and February of the current year and December of the preceding year.

annual mean of 2 μatm. Takahashi et al. (1993) obtained an annual mean of almost zero for 30° N–38° N as shown in his Fig. 10 on seasonal change in the eastern subtropical Pacific. The ΔfCO_2 results from our cruises for the northeast Pacific 15° N–45° N averaged about -19 μatm for winter and spring and about 8 μatm for summer and autumn, giving an annual mean ΔfCO_2 of about -5 μatm. These estimates, which differed by about 14 μatm as compared with the average of 9 μatm given by Takahashi (1989), suggested that the northeast Pacific is a relatively weak CO_2 sink annually. Note that our averages did not cover the eastern North Pacific area south of 28° N and east

of 154°W, and we also did not extrapolated ΔfCO_2 values into this area as was done by Tans et al. (1990).

3.4. *Seasonal variation of sea-surface fCO_2*

Studies on the diurnal and daily variations of fCO_2 in surface seawater might help in the understanding of the interaction between marine environment and its biological components (Oudot and Andrié, 1989) However, our data sets focused on relatively large-scale changes in both time and space rather than on short-term variations. In several occasions, our cruises sampled the same oceanic regions during the warming and the cooling seasons of the year, and thus, supplied the necessary data to study the seasonal and annual variations of fCO_2 in the subtropical Pacific. In general, the summer averages were higher than those values obtained in winter and reflected the effect of seasonal heating of surface seawater. The amplitudes between winter/spring and summer/autumn seasons unadjusted for temperature effects lied mostly within 20–50 μatm (Fig. 4).

The seasonal amplitudes (summer/winter) of the surface fCO_2 in the subtropical gyre within 15°N–25°N from winter 1984 to winter 1988 averaged about 21 μatm (Fig. 4). Weiss et al. (1982) estimated that the average seasonal amplitude of fCO_2 in surface seawater of the tropical arm of the north Pacific subtropical gyre was about 20 μatm. They concluded that this seasonal variation of surface fCO_2 in the subtropical gyre, with small air-sea CO_2 exchange and low biological production, could be adequately explained by the seasonal changes in SST and salinity. Our result was in general agreement with that of Weiss et al. (1982). However, similar analyses of the fCO_2 values normalized at the mean SST for this latitudinal zone still showed some residual variations (Fig. 6). Furthermore, the amplitude from summer 1983 to winter 1984 was about 31 μatm, slightly over two times the averaged standard error of 14.5 μatm for the normalized fCO_2 estimates (Fig. 6). Since the SST-normalized values presumably were affected equally by temperature, the variations showed in the normalized values might represent the residual effects from other factors, e.g., air-sea exchange, upwelling or lateral mixing as well as biological production and respiration. Our results, especially those of summer 1985, for this zone in the north Pacific subtropical gyre suggested that the effects from factors other than temperature could not be totally ignored.

Similar analyses of the SST-normalized fCO_2 for the southern Pacific within 25°S–15°S also suggested that the relatively large changes in fCO_2 during the 1986/1987 El Niño were not caused by the increased SST alone. The residual variation of the SST-normalized fCO_2 resulted in estimated seasonal amplitudes of about 43 μatm and 63 μatm in 1987–1988 (Fig. 6). These values were 1.5–2 times the averaged standard error of 27 μatm of the normalized fCO_2 data. For the northern zones within 25°N–35°N, the average amplitude of normalized fCO_2 was about 40 μatm between 1985 and 1986, and about 50 μatm between summer 1987 and winter 1988; well over the averaged standard error of 30 μatm of the normalized fCO_2 data. These analyses indicated that, in certain seasons, the variations of fCO_2 in the subtropical Pacific could not be explained by changes in SST alone.

3.5. *Interannual variations of sea surface fCO_2*

In our monitoring programs, the sampling from about the same oceanic regions periodically from December 1983 to February 1988 allow us to study the interannual variation of fCO_2 in surface seawater of the subtropical Pacific. A component of the variation in sea surface fCO_2 in response to the annual changes in atmospheric CO_2 could be estimated by the difference between the annual trends of the fCO_2 (Fig. 4) and ΔfCO_2 (Fig. 5) data. This component of the interannual variabilities might also be removed from the annual increase in SST-normalized fCO_2 data (Fig. 6) to assess the responses of surface fCO_2 to other factors such as changes in lateral transport, upwelling or biological activities.

Fig. 4 also showed the changes in the summer and winter seasonal averages of sea surface fCO_2. In the southwest Pacific (25°S–15°S), the austral winter fCO_2 decreased at an average rate of 1.4 μatm yr^{-1} from 1984 to 1986 but the austral summer fCO_2 increased at about 5 μatm yr^{-1} from 1984 to 1988. Similarly, the annual increases for the seasonal averages of ΔfCO_2 in austral winter and summer were respectively -3.6 μatm yr^{-1} and 4.9 μatm yr^{-1} (Fig. 5). In Fig. 6, the averages of the normalized fCO_2 from austral summer showed an annual increase of about 9.6 μatm yr^{-1}.

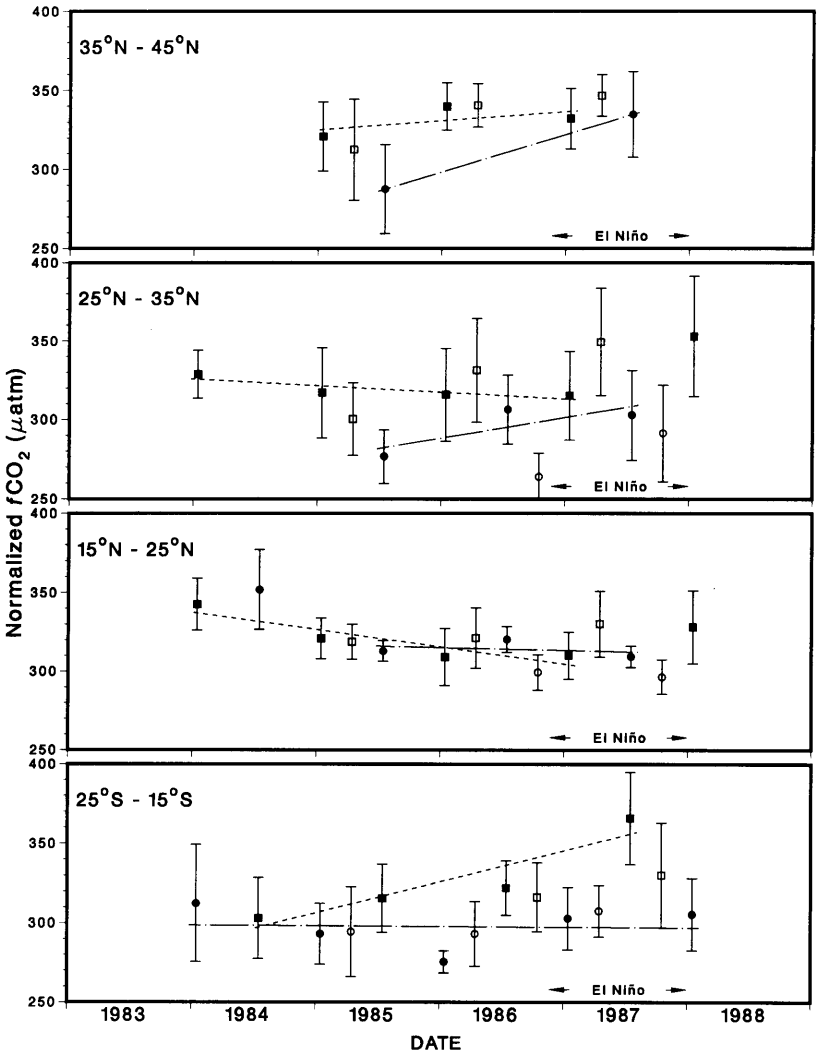

Fig. 6. Seasonal averages of fCO_2 normalized to the climatological mean SST of the respective zone. The mean SST used in the computation was 25.5°C for 25°S–15°S, 24.6°C for 15°N–25°N, 20.0°C for 25°–35°N, and 15.3°C for 35°N–45°N. Different markers are used for the four seasons: winter, solid square; spring, open square; summer, solid circle; and autumn, open circle. The boreal winter (austral summer) includes January and February of the current year and December of the preceding year. The error bars represent $\pm 1\sigma$. Trend lines for both the summer and the winter seasonal means are also given.

Out of which, about 2.2 μatm yr^{-1}, which was as the difference between annual increases in fCO_2 and ΔfCO_2, represented the response of surface seawater to the annual increase in the atmospheric CO_2 level. The averages of normalized fCO_2 from austral winter, which decreased initially from 1984 to 1986 and then increased thereafter to 1988, showed an overall decrease of 0.4 μatm yr^{-1}. However, with an average standard error of about 27 μatm for the SST-normalized fCO_2 data, both the small winter increase and summer decrease were considered not significant.

In the north subtropical Pacific at 15°N–25°N, the seasonal averaged fCO_2 increased at about

3.6 μatm yr^{-1} in boreal winter from 1984 to 1988 and about 2.5 μatm yr^{-1} in boreal summer from 1985 to 1987. For the seasonal ΔfCO_2, the annual trends were 2.6 μatm yr^{-1} and 1.8 μatm yr^{-1} for the winter and summer averages respectively. The annual increases attributed to responses to atmospheric CO_2 were relatively small, about 1 μatm yr^{-1} and 0.6 μatm yr^{-1} respectively for the winter and summer averaged ΔfCO_2. When the seasonal averages of SST-normalized fCO_2 were considered, the annual trend was -4 μatm yr^{-1} for the winters of 1984–1988, and -1.8 μatm yr^{-1} for the summers of 1985–1987. These annnual trends for the seasonal averages of SST-normalized fCO_2 were also within the uncertainty of the data.

The summer averaged fCO_2 and ΔfCO_2 within 25°N–35°N increased at 10.5 μatm yr^{-1} and 7.8 μatm yr^{-1} respectively. For the winter averages in this region, the trends decreased from 1984 to 1986 and then increased thereafter to 1988. Annual trends for averaged fCO_2 and ΔfCO_2 for 1984–1986 were respectively -10.7 μatm yr^{-1} and -11.2 μatm yr^{-1}, and for 1986–1988, 20.5 μatm yr^{-1} and 16.0 μatm yr^{-1} respectively. Within 35°N–45°N, the winter averaged fCO_2 and ΔfCO_2 for 1985–1987 increased respectively at 6.2 μatm yr^{-1} and 3.6 μatm yr^{-1}. Only the 1985 and 1987 summer averages of both fCO_2 and ΔfCO_2 were available from this zone. The annual increases for fCO_2 and ΔfCO_2 were 14 μatm yr^{-1} and 8 μatm yr^{-1}, respectively.

The general trend of the normalized fCO_2 within 25°N–35°N (Fig. 6) for the summer 1985–1987 averages was 13 μatm yr^{-1}, and for the 1984–1987 winter averages, -4.2 μatm yr^{-1}. Within 35°N–45°N, both the summer and winter trends showed increases of 24 μatm yr^{-1} and 6 μatm yr^{-1} respectively. Although the summer increase in the normalized fCO_2 within 25°N–35°N as well as the winter increase within 35°N–45°N were all within the $1\sigma(\sim 20$ μatm yr$^{-1})$ of their seasonal mean estimates, the fact that the summer increases were observed within the two northern zones suggested that the surface fCO_2 may indeed have increased in this region from 1984 to 1987. The cause of this increase in the northern Pacific was probably the anomalously cold water along the North Pacific Current during 1984–1986.

The previous analyses of the SST-normalized fCO_2 suggested that the increase in surface fCO_2 within 25°N–45°N of the Pacific from 1984–1988 could not be explained entirely as a result of the increases in SST and oceanic CO_2 uptake. Other changes responsible for supplying CO_2 to this oceanic region might be involved. Our study period happened to be in the last half of the decade when the Aleutian low for winter time from 1977 to 1988 were deeper by 3 mb than normal (Trenberth and Hurrell, 1994). Associated with this substantial variation were large-scale changes in wind stress curl and the corresponding Sverdrup transport in the ocean. The southward intrusion of the high-CO_2 Oyashio Current to the North Pacific (Sekine, 1988) might occurred several times over this period and could cause the increase in fCO_2 in the region.

The above estimates of the year-to-year increase, which relied entirely on sea surface fCO_2 measurements obtained within a few years, are not necessarily indicative of the secular increase of fCO_2 in surface seawater. Changes in fCO_2 are known to be strongly influenced by decadal or even longer term atmosphere-ocean variations in the Pacific. To estimate the secular increase of sea surface fCO_2, other historical data, e.g., $\Delta^{14}C$ and ^{13}C records of banded coral or other tracers (Van Scoy and Druffel, 1993; Druffel and Griffin, 1993), sampled from the same general oceanic areas are required to assess the contribution from various sources.

4. Conclusions

This report presents measurements of the sea-surface fCO_2 in the northeast (15°N–45°N) and southwest (25°S–15°S) subtropical Pacific from November 1983 to February 1988. For the southwest Pacific within 25°S–15°S, the austral winter/spring fCO_2 data composed the first such data set for this oceanic region, and with the fCO_2 data from the other seasons, provided the full annual cycle of sea-surface fCO_2 for the same region. The observed sea-surface fCO_2 exhibited relatively large geographical and seasonal variations. Our results showed that the seasonal variations of the fCO_2 in these areas of about 20–50 μatm were strongly influenced by the 1977–1988 atmosphere-ocean variations in the North Pacific and by the 1986–1987 El Niño.

Between December 1983 and February 1988, the northeast subtropical Pacific was a small CO_2 sink during boreal winter and spring with ΔfCO_2 of about $-19\,\mu$atm. In the boreal summer and autumn seasons, this oceanic region became a CO_2 source with an average ΔfCO_2 of $8\,\mu$atm. Hence, during our study period, the northeast Pacific was a relatively weak CO_2 sink annually with a mean annual ΔfCO_2 of about $-5\,\mu$atm. This value differed by about $14\,\mu$atm as compared with the mean of $9\,\mu$atm given by Takahashi (1989) for the Pacific within 10°N–40°N east of the dateline. For the southwest subtropical Pacific, the region was basically a CO_2 sink year round with an annual mean ΔfCO_2 of about $-20\,\mu$atm, except during the summer months of the El Niño period whence ΔfCO_2 was positive. Our results for the southwest Pacific, as well as those from Miyake et al. (1974) and Murphy et al. (1991b), suggested that this oceanic region is probably a lesser CO_2 sink as estimated by Tans et al. (1990).

Variabilities of sea-surface fCO_2 in response to annual increase in atmospheric CO_2 level and to changes in SST were estimated by analyzing the various trends of in situ fCO_2, ΔfCO_2, and the fCO_2 values normalized to the climatological mean SST. After removing the response of fCO_2 to changes in atmospheric CO_2 from the estimated SST-normalized fCO_2, the annual trends of the residuals were computed. Except for the summer averages within 35°N–45°N, the annual variations of the normalized fCO_2 were mainly within the uncertainty of the data.

In the northern waters within 35°N–45°N, the normalized fCO_2 showed a significant increase of about $24\,\mu$atm yr^{-1} among summer means and only a small increase of $6\,\mu$atm yr^{-1} among the winter means. The summer increase of normalized fCO_2 was also displayed in surface waters further south with 25°N–35°N but at a lower rate of $13\,\mu$atm yr^{-1}. Although this rate was not statistically significant, the fact that the summer increases were observed within the two northern zones from 25°N to 45°N suggested that the surface fCO_2 in the northern Pacific may have increased during 1984 to 1987. The increase probably reflected the intrusions of the high-CO_2 Oyashio Current to the central North Pacific region following some episodes of winter intensification and eastward shift of the Aleutian low from 1977 to 1988.

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