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Variability of ΔpCO_2 in the subarctic North Pacific. A comparison of results from four expeditions

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ABSTRACT

Time-space variability of surface seawater pCO_2 is examined over the region (150°W–180°, 46°N–50°N) of the subarctic North Pacific where large meridional gradients of temperature and nutrient concentrations exist. The data were collected during four trans-Pacific expeditions in three different years (1985–1987), but within the same 30-day period of the year (August–September). Systematic measurement differences between the four data sets are estimated as <10 μatm. The inter-expedition comparison suggests that surface seawater pCO_2 in the study area is quite variable, with mean differences of up to 25 μatm and local differences up to 60 μatm. Spatial and interannual variability of surface seawater pCO_2 were found to contribute significant uncertainty to estimates of the mean ΔpCO_2 for the study area. Fluxes were calculated using ΔpCO_2 values from the four expeditions combined with gas exchange coefficients calculated from four different wind fields giving a range of -0.94 to $+4.1$ mmol CO_2 m^{-2} d^{-1}. The range of fluxes from the study area is scaled to a larger area of the North Pacific to address how this variability can translate into uncertainties in basin-wide carbon air-sea exchange fluxes.

1. Introduction

The oceanic uptake of carbon is one of the more uncertain terms in the global carbon budget and in determining the fate of fossil fuel carbon dioxide (Tans et al., 1990; Siegenthaler and Sarmiento, 1993; Sundquist, 1993). One of the methods currently employed to estimate the ocean's role in absorbing carbon is to estimate the net global flux of CO_2 from the atmosphere to the ocean by integrating local fluxes. Local fluxes of CO_2 across the air-sea interface can be calculated from the CO_2 partial pressure gradient (ΔpCO_2) across the air-sea interface combined with a gas exchange rate. Since regional data are collected in different years and seasons by various investigators, a number of assumptions are made when combining data to estimate basin-wide or global fluxes. Some of those assumptions are that the ΔpCO_2 data are intercomparable, that the time period over which the data are averaged is appropriate, that the data can be interpolated geographically and temporally, that there is no sampling bias or interannual variability, and that the parameterization of gas flux across the air-sea interface is accurate. Additionally, it is difficult to assess the uncertainty of large-scale flux estimates when the repeat measurements of CO_2 disequilib-

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ria are so few. In this study we use four data sets collected in the same region and season (over three years) by three different investigators to examine some of the assumptions and potential uncertainties in estimating large-scale CO_2 fluxes.

The intercomparability of the data sets is first examined in the methods section. Systematic analytical differences between the laboratories are found to be small, and hence the observed inter-expedition differences in ΔpCO_2 represent real oceanographic differences. In the results section, the variability in ΔpCO_2 between expeditions is described and the time-space variability of the data are examined. Spatial variability and the accuracy of data interpolations are addressed by subsampling two of the data sets at lower frequency and comparing the ship-track-mean ΔpCO_2 calculated when the data are sampled differently. The temporal variability of the data in this region is compared for time scales ranging from hours to years using high frequency data, repeat section data, and time series data from Station P. In a following section, some mechanisms for ΔpCO_2 variability are discussed. In the discussion section the findings of this study on ΔpCO_2 and CO_2 flux variability are summarized and then examined in a broader context. Local flux is estimated in eight different ways, in order to examine the sensitivity of CO_2 flux in the region to the existing ΔpCO_2 data, wind fields, and gas exchange parameterizations. The ship-track-mean flux estimates were then extrapolated to the area of the subarctic North Pacific to examine how the inter-expedition flux differences scale to regional carbon uptake differences.

The results presented here indicate that ΔpCO_2 and calculated fluxes in the subarctic North Pacific can vary significantly, even when data from the same region and season are compared. We suggest that the need for improved information about the space-time scales of oceanic pCO_2 variability is great.

2. Methods

2.1. CO_2 measurements

Measurements of p_wCO_2 in surface seawater and p_aCO_2 in the overlying atmosphere were collected from the same region of the North Pacific between 19 August and 13 September in the years 1985, 1986 and 1987. The four data sets discussed here were collected from the central subarctic Pacific between 180° and 150°W and between 46°N and 50°N (Fig. 1). The Scripps Institution of Oceanography (SIO) and Pacific Marine Environmental Laboratory (PMEL) measurements were made along a constant latitude of 47°N and 50°N, respectively, but the Lamont-Doherty Earth Observatory (LDEO) tracks follow great circle shipping routes and dip sharply to the south beyond the study region. Although this region encompasses a small portion of each expedition, this subset was chosen to minimize spatial differences.

The 1985 SIO data were collected using an automated gas chromatographic system (Weiss, 1981). Atmospheric and surface seawater measurements were each made at approximately 30-min

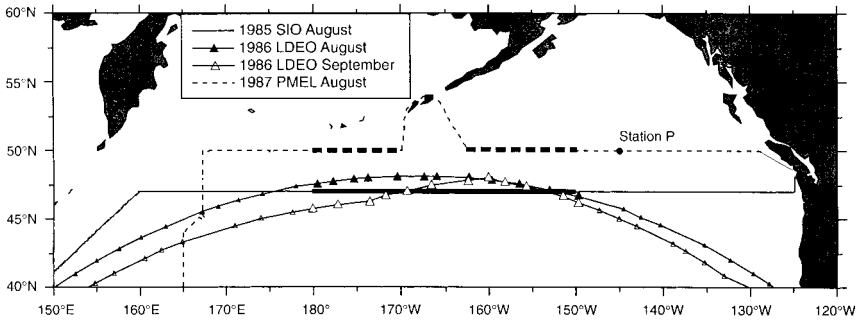

Fig. 1. Cruise tracks along which data were collected. The 1985 SIO and 1987 PMEL data were collected quasi-continuously and the sampling locations are indicated by continuous lines. The 1986 LDEO data were collected at the locations shown by the triangles, and the ship's course is indicated by the continuous line. The larger triangles indicate the station data used in this comparison. The location of Station P (50°N, 145°W) is also indicated.

intervals. Surface seawater p_wCO_2 measurements were made using a continuous flow, two-stage equilibrator which operates at low water pressure and is vented to the atmosphere. The results are reported as mole fraction of CO_2 in dry air with surface seawater values corrected to the in situ mixed layer temperature to account for the warming of seawater transiting to the equilibrator. For this study, the reported mole fraction values were converted to ΔpCO_2 using the following equation:

$$\Delta pCO_2 = (P_{total} - p_{H_2O})(x_w - x_a), \quad (1)$$

where ΔpCO_2 is the oceanic CO_2 partial pressure (p_wCO_2) minus atmospheric CO_2 partial pressure (p_aCO_2), P_{total} is the atmospheric pressure, p_{H_2O} is the partial pressure of water at mixed layer temperature, x_w is the mole fraction of CO_2 in dry gas in equilibrium with surface seawater (corrected for warming), and x_a is the mole fraction of CO_2 in dry air for the atmosphere.

The 1986 LDEO data are based on the discrete flask samples collected at selected stations (Fig. 1) and processed aboard container ships (Takahashi et al., 1991). For seawater pCO_2 measurements, approximately 500 ml of air was recirculated through a gas disperser immersed in a 4-litre seawater sample in a closed-loop gas/water equilibration system for 15 min. A 200 ml gas sample was isolated in a Pyrex flask from the equilibration system, and the temperature of equilibration and pressure of the circulating gas were recorded at the end of the equilibration process. Marine air samples were collected periodically also in 200 ml glass flasks aboard the ship. The samples were collected at stations 2 to 3° apart in longitude during two consecutive trips across the Pacific about 20 days apart in August and September, 1986. These gas samples were returned to Lamont for CO_2 analysis using a gas chromatograph similar to that developed by Weiss (1981), and the pCO_2 values thus obtained were corrected to the in situ temperature. Note that sample locations used here have been corrected from those given in Takahashi et al. (1991).

The 1987 PMEL data (Murphy et al., 1994) were collected using an equilibrator and automated analytical system similar to that used by SIO. The reported hourly values of mole fraction of CO_2 in dry air were converted to partial pressures using eq. (1), with surface seawater results corrected to in situ mixed layer temperatures. Where the ship diverted northward from 50°N, those data between 170°W and 163°W were not used in this comparison study.

2.2. *Comparison of methodologies*

Since this comparison is based on data from 3 different laboratories in 3 different years, the observed variability could result from systematic differences between the sets of observations or from real oceanographic differences between the years, or some combination of the two. Determining the comparability of p_wCO_2 data collected using different measurement techniques is not easy since the systems differ in a number of ways. However, the analyzers used to collect the data for this comparison were all gas chromatographs calibrated using CO_2-in-air gas standards measured by C. D. Keeling at SIO. The comparability of results between the three laboratories is shown by independent field comparisons, and then possible systematic differences between these four particular data sets are discussed.

An estimate of the accuracy of the atmospheric data from each expedition can be made by comparison with independent monthly mean observations made by the Geophysical Monitoring for Climatic Change Laboratory (1986, 1987, and 1988). Stations MLO (19.5°N, 155.6°W) and BRW (71.3°N, 156.6°W) were chosen for comparison because the air sampled at those locations is characteristic of remote background air and the latitudes for those stations bracket the latitude range of the observations discussed here. During August/September, the mole fraction x_a value at MLO is ~7 ppm greater than at BRW. For each expedition, mean values of x_a fall within the range of the GMCC data at these two stations (337.4–343.9 for 1985, 339.5–345.5 for 1986, and 340.4–347.7 for 1987).

Comparability of the p_wCO_2 measurements can be made by examining field data collected by these three laboratories in other locations. LDEO and SIO both measured pCO_2 during TTO/NAS expeditions in the North Atlantic. Takahashi et al. (1995) compared 137 pairs of the LDEO station data with the SIO underway pCO_2 data obtained in surface water during the 1981 TTO/NAS expeditions, and observed that their data are mutually consistent to within a mean standard

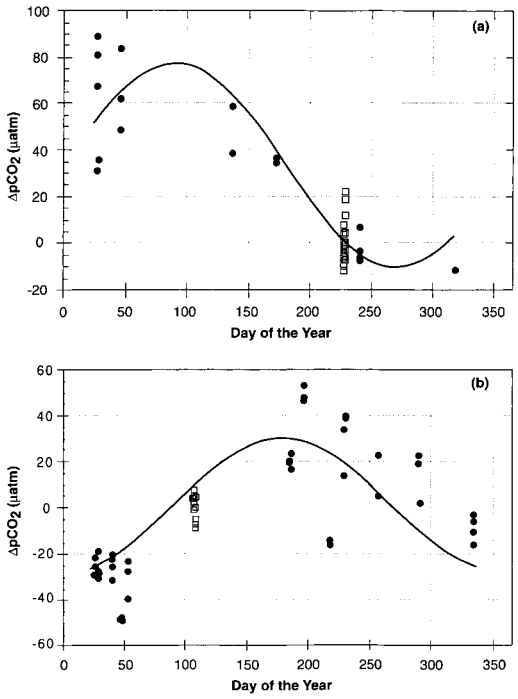

Fig. 2. Comparison of ΔpCO_2 data from one PMEL cruise (open squares) with the seasonal trend (line) defined by LDEO data (filled circles) from the same region. (a) Northwestern Pacific (48–54°N, 164–170°E) trend compared with PMEL cruise RITS/CO_2 1987. The line is a sine function fit to the data giving a rough representation of the seasonality. (b) Eastern subtropical Pacific (33–43°N, 131–138°W) trend compared with PMEL cruise RITS/CO_2, Leg 3. The line is a cosine function fit to the data giving a rough representation of the seasonality.

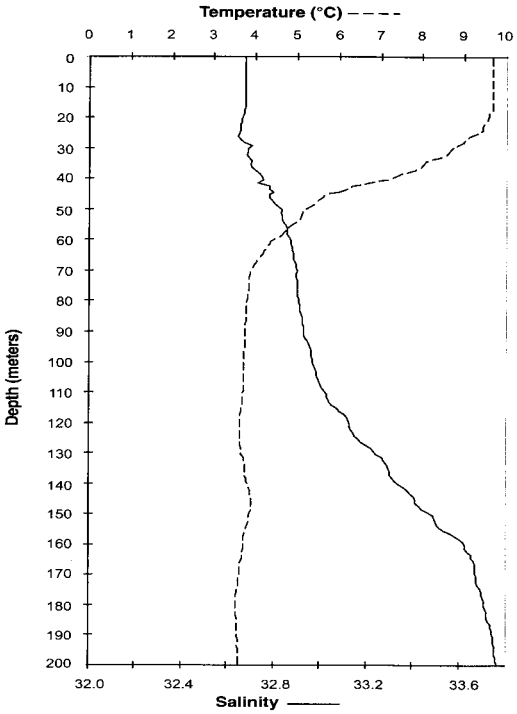

Fig. 3. Temperature and salinity versus depth at 50°N, 170°W on 20 August 1987 (Talley et al., 1988).

deviation of ± 0.7 µatm (or a RMSD of ± 7.5 µatm).

The discrete LDEO data can also be compared with quasi-continuous PMEL data in the northwestern (Fig. 2a) and eastern subtropical Pacific (Fig. 2b). Following Takahashi et al. (1991), a regional seasonal trend was defined by fitting sine and cosine curves to the data collected on multiple expeditions. The mean of the PMEL data in both cases are consistent with the trend defined by the LDEO data to within 10 µatm. These field comparisons suggest that data collected by the three laboratories are generally comparable to within 10 µatm.

Although data from these laboratories is generally comparable, it is possible that systematic differences might exist between these 4 specific data sets. For example, systematic differences could result from differences in sample depth. Samples for the 1986 LDEO data set were collected from a depth of 14–20 m, whereas samples for 1985 and 1987 were collected from ~ 5 m. Profiles of depth vs. salinity and temperature from the 1987 observations (Fig. 3), as well as Dodimead et al. (1963) and Talley et al. (1988) indicate that the mixed layer is warmer and fresher than the underlying thermocline layer. Based on the XBT data, Takahashi et al. (1991) reported that the mixed layer depth ranges from 12 to 28 m. Since some of the 1986 samples may have been drawn from below the mixed layer, they may have included cooler, saltier water with higher dissolved inorganic carbon (DIC), and hence higher p_wCO_2. Comparison of 1986 temperature and salinity data with climatological values (discussed in Subsection 3.1) suggests that LDEO 1986 temperatures are consistent with climatological values. Where LDEO 1986 salinities deviate from climatological values, they are fresher by up to 0.2.

Therefore, the observed variations cannot be attributed unequivocally to sampling depth.

The warming correction can be the largest source of uncertainty in calculating p_wCO_2 in surface waters. Seawater generally warms in transiting from the sea surface to the equilibrator where p_wCO_2 measurements are made. The measured value of p_wCO_2 can be corrected to the p_wCO_2 at in situ temperature by a number of different algorithms. For 1985 and 1987, the observations were corrected to mixed layer conditions using the effect of temperature, $\partial \ln pCO_2/\partial t$, ranging from about 4.4% $^\circ C^{-1}$ at 0°C to 3.6% $^\circ C^{-1}$ at 30°C (Weiss et al., 1982). These were computed using the apparent dissociation constants for carbonic and boric acids by Hansson (1973). The warming was always <1°C and a typical correction in p_wCO_2 was 7 μatm. For the 1986 LDEO data, the measured values were corrected to the in situ temperatures using a constant ($\partial \ln pCO_2/\partial t$) value of 4.23% $^\circ C^{-1}$ based on the direct measurements of Takahashi et al. (1993). Again, the warming was small (<1°C). To assess the potential magnitude of p_wCO_2 differences arising from differences in the warming algorithms, the 1986 data were recalculated using the Weiss et al. (1982) algorithm and the assumptions of 1 atm total pressure and 1°C warming. For the temperature range and warming magnitude for these expeditions the differences between the algorithms is small, and the differences in p_wCO_2 calculated using the two algorithms is <1 μatm.

Another difference between the data sets is that of the storage time between sampling and analysis. The 1985 and 1987 samples were analyzed using a quasi-continuous shipboard system. The 1986 gas samples were equilibrated with 4 litres of seawater within 1 h of sampling and stored for a month or longer prior to analysis. Takahashi et al. (1990) report that these gas samples can be stored stably and reliably for long periods of time.

The comparisons discussed above suggest that any systematic differences between the four data sets presented here are less than 10 μatm.

2.3. *Flux calculations*

Fluxes were calculated using the equation:

$$F = \Delta pCO_2 * E(W), \quad (2)$$

where F is CO_2 flux, E is the gas exchange coefficient, which is a function of wind speed, W. Exchange coefficients given by Liss and Merlivat (1986) and Tans et al. (1990) represent, respectively, the lower and upper boundaries between which fall most of the experimental (radon, bomb-produced and natural ^{14}C) evaluations. A more recent parameterization given by Wanninkhof (1992) is intermediate between these extremes. Fluxes were calculated using the extremes of the parameterizations to examine the potential range of fluxes estimated for the study area.

In the interest of evaluating the full range of flux possibilities, we have also used four wind speed data sets. Traditionally, climatological wind speed data have been used to estimate the mean CO_2 flux, since the p_wCO_2 data were collected over many different years. However, the wind speed in any given year can differ significantly (more than a factor of 2) from the climatological wind speed (cf., Murphy et al., 1991). We here use the Esbensen and Kushnir (1981) climatology which incorporates data collected over more than a century. Another climatology, Harrison (1989), incorporates data between 1950 and 1980 over the Pacific, and so is more representative of recent decades. Year-specific data sets from the US Navy's Fleet Numerical Oceanography Center (FNOC) and from the European Centre for Medium-Range Weather Forecasting (ECMWF) provide 2 high-frequency wind fields (6-hourly and 12-hourly winds, respectively). A more complete description of the climatologies and the FNOC wind products is given by Murphy et al. (1991). In the study region of the North Pacific, the several surface wind analyses available to us were more similar than different in their monthly mean and longer time period behavior; it will also be seen that variability in the wind is not the primary source of variability in surface carbon flux.

CO_2 fluxes were calculated for each of the 4 ΔpCO_2 data sets by combining each ΔpCO_2 with 8 different combinations of wind speed and E(W) parameterization. E(W) was calculated at each pCO_2 sampling location for each time step of the wind data for the 30-day period encompassing the pCO_2 data collection (15 days before and after). 30-day mean values of E(W) were then used to calculate local fluxes. Local fluxes were integrated to estimate a range of ship-track-mean CO_2 fluxes for the period mid-August to mid-September.

3. Results

3.1. Oceanographic environment

The study region is north of the subarctic/subtropical boundary and south of the Alaska Gyre (Favorite et al., 1976). Surface currents deduced from geostrophy (and substantiated with drift bottle data) indicate eastward flow (~ 4 cm s^{-1}) in the region 45–50°N from $\sim$180–145°W. According to the comprehensive ocean-atmosphere data set (COADS) climatological data base described by Woodruff et al. (1987), the region 45–50°N, 170°E–140°W shows a large seasonal cycle in sea surface temperature (sst), ranging from 5.3°C in March to 12.9°C in August. Large interannual variations (up to 2°C) as well as meridional (-3°C from 45°N to 50°N) and zonal gradients ($+3$°C from 170°E to 140°W) also influence temperature at a particular location (Fig. 4a). Because of the large seasonal changes, interannual variability, and strong gradients, observed sst values are strongly dependent on the sampling time and location. The 1985 47°N and 1987 50°N data sets examined here (Fig. 5a) show cooler temperatures (-2°C), than the gridded COADS climatological data near 180°W for those latitudes. August 1986 and September 1986 values agree with the climatological values (within 1°C), with the exception of 1 point at 45.8°N, 180°, which is ~ 1.5°C warmer. Observations at and west of 165°W agree with climatological values for all 4 data sets.

Climatological salinity in the region 45–50°N, 170°E–140°W shows a seasonal cycle of salinity (practical salinity scale) ranging from 33.0 in April to 32.7 in September according to the climatological data set described by Levitus et al. (1994). Meridional and zonal gradients during August/September are approximately -0.3 (from 45°N to 50°N and 170°E to 140°W). The 1985 47°N and 1987 50°N data sets (Fig. 5b) are consistent with the gridded Levitus et al. climatological data (to 0.1) at those latitudes. The LDEO 1986 data sets both show lower salinities than climatological, in places by -0.2. The salinity climatology is less smooth than for temperature, and spatial variability is also suggested by the 1985 and 1986 data sets.

Subarctic and subantarctic areas also typically show large seasonal cycles and large meridional gradients in mixed layer depth (Levitus, 1982),

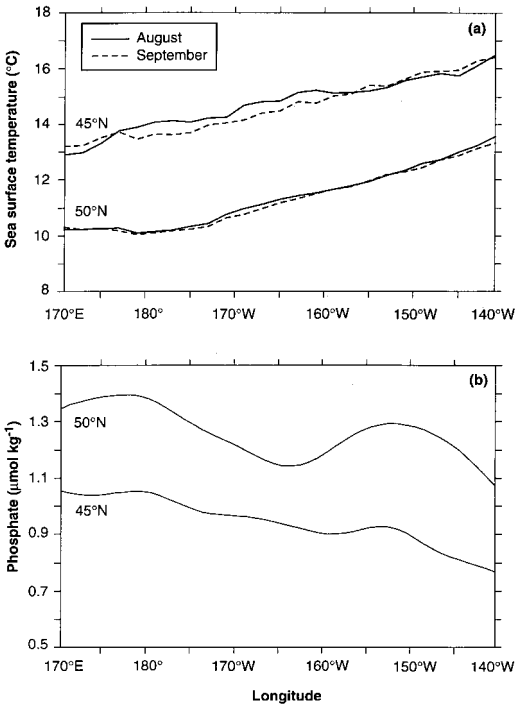

Fig. 4. Climatological data for the region 45–50°N, 170°E–140°W. (a) Sea surface temperature data for August and September along 45°N and 50°N. Data are from the comprehensive ocean-atmosphere data set (COADS) climatological data base described by Woodruff et al. (1987). (b) Annualized phosphate concentrations along 45°N and 50°N. Data are from Conkright et al. (1994).

nutrient concentrations (Conkright et al., 1994), and dissolved oxygen concentrations (Levitus and Boyer, 1994). Few nutrient data are available for these areas, but the data generally show high concentrations, a pronounced seasonal cycle, large gradients, and high variability in the subpolar regions. The gridded data of annualized phosphate concentrations (Fig. 4b) show large meridional ($+0.3$ μmol l^{-1} from 45°N to 50°N) and zonal (-0.3 μmol l^{-1} from 170°E to 140°W) gradients. Annualized phosphate concentrations are not strictly comparable with the data collected in August/September of 1985 and 1986 (Fig. 6), but the general trends are similar.

The large interannual variability in temperature, as well as zonal and meridional gradients of temperature, salinity, and nutrients are suggestive that p_wCO_2 may also show strong gradients and

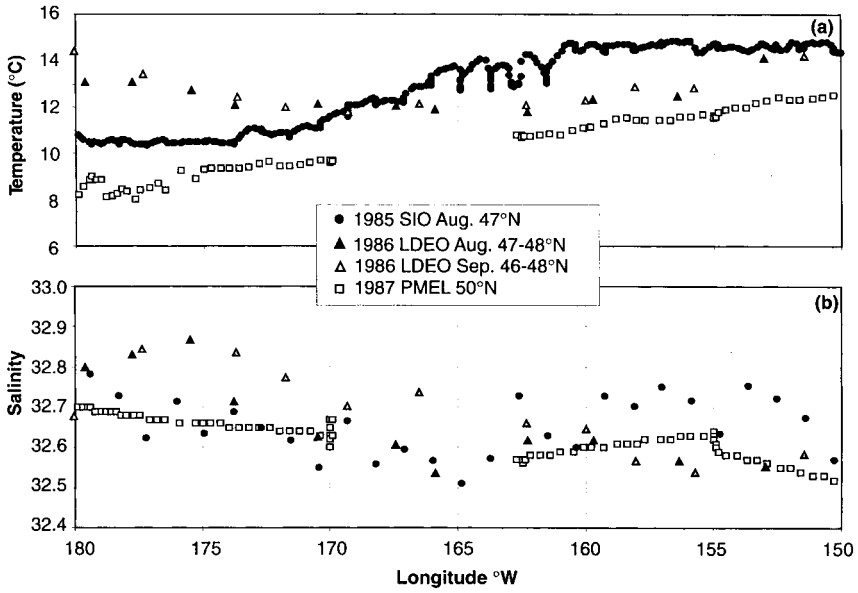

Fig. 5. Comparison of data collected on the 4 expeditions. (a) Mixed layer temperature and (b) Salinity.

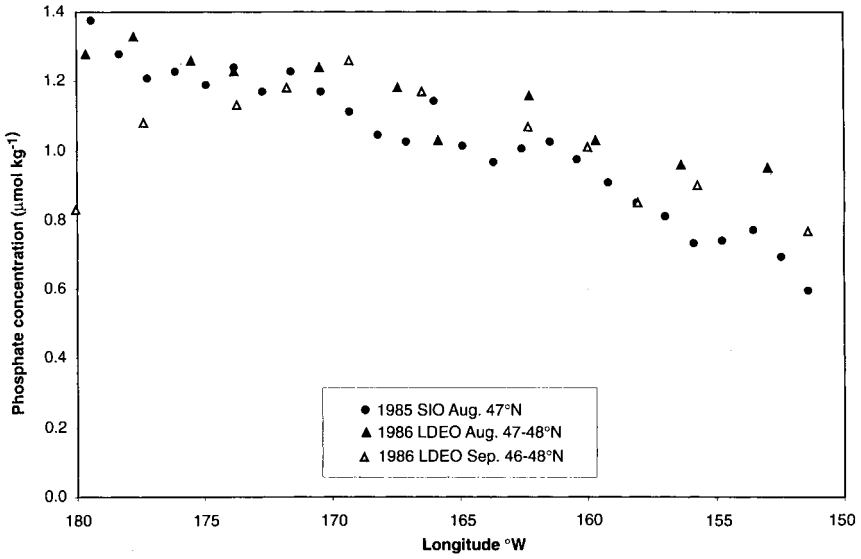

Fig. 6. Comparison of phosphate observations for the years 1985 and 1986.

interannual variations. We particularly note that the meridional gradients in sst and nutrient concentrations are amongst the steepest in the global ocean. However, the northward and westward increases in phosphate concentrations also suggests increases in DIC concentrations to the north and west. Since an increasing effect of greater DIC on seawater pCO_2 is partially counteracted by the lowering effect of colder sst, the distribution of surface water pCO_2 is expected to exhibit smaller meridional and zonal gradients than sst and the nutrient concentrations.

3.2. *Comparison of ΔpCO_2*

The August 1985 SIO ΔpCO_2 data along 47°N show near-equilibrium values with a range of −32 to +24 µatm (Table 1, Fig. 7). The highest values occur in the middle of the comparison region (near 165°W) and lowest values at the eastern end. The August 1986 data LDEO observations made between 47°N and 48°N show consistent supersaturation with respect to the atmosphere along the track ranging from 13 to 41 µatm. The September 1986 LDEO data (46°N–48°N) show mostly supersaturation, with values ranging from −23 to 14 µatm. The lowest value occurs at the western end and highest values in the middle of the comparison region. The August 1987 PMEL data obtained along 50°N show a significant zonal gradient with undersaturation to the west as low as −39 µatm and oversaturation to the east as high as +43 µatm. Taken as a whole, the composite data do not seem spatially or temporally coherent, i.e., there is no systematic east-west gradient nor is there a systematic offset between the data sets.

Significant differences are observed between the mean air-sea ΔpCO_2 values along the respective ship's tracks between 150°W and 180°W. The mean ΔpCO_2 values were obtained by integrating the area under the curves shown in Fig. 7, so that the mean values reflect an areal weighting of the

Table 1. *Inter-cruise comparison of pCO_2 data in units of µatm; data shown in Fig. 7 were integrated to obtain areally weighted ship-track-mean values*

	Ship-track-mean		Minimum ΔpCO_2	Maximum ΔpCO_2	Regional mean ΔpCO_2	Ship-track-mean ΔpCO_2 subsampled at LDEO August locations	Ship-track-mean ΔpCO_2 subsampled at LDEO September locations
	p_aCO_2	p_wCO_2					
SIO	338.4	337.6	−32	24	−0.83	+0.30	−1.0
LDEO Aug. 1986	337.5	361.2	13	41	24		
LDEO Sep. 1986	335.2	339.7	−23	14	4.6		
PMEL	335.2	335.7	−39	43	0.50	−1.6	+3.6
$\bar{x}=$	336.6	343.6					

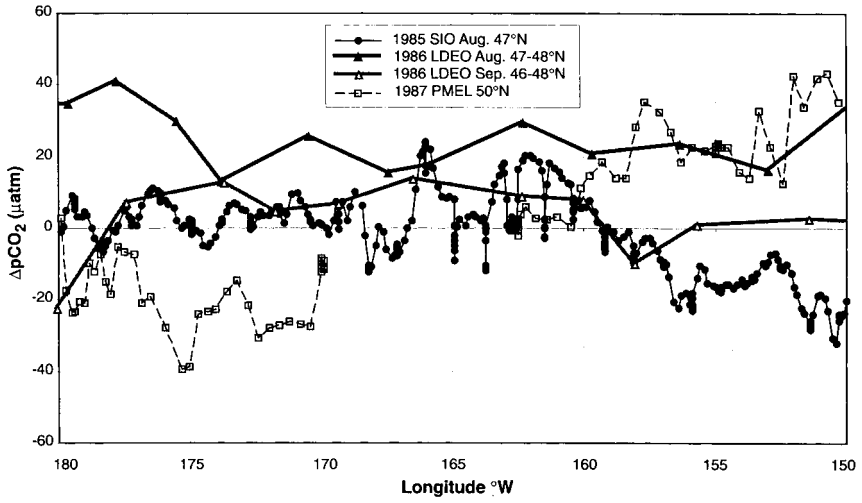

Fig. 7. Comparison of ΔpCO_2 as a function of longitude along the four cruise tracks. The region of geographically intersecting data is defined as 180° to 150°W, 46°N to 50°N.

data obtained at irregular intervals. The 1985 SIO and 1987 PMEL data give small mean values near 0, -0.83 µatm along 47°N and $+0.50$ µatm along 50°N, respectively (Table 1), indicating that waters along these tracks were either a small sink or source for atmospheric CO_2. On the other hand, the 1986 LDEO data obtained between 46°N and 48°N give larger positive values, $+24$ µatm for August and $+4.6$ µatm for September indicating that these waters were CO_2 sources.

To address pCO_2 differences resulting from interannual variations and zonal and meridional gradients in temperature between these 4 data sets, the pCO_2 values in seawater were normalized to a regional climatological sst value for August/September of 13°C (Fig. 8). To the west the normalized p_wCO_2 agree fairly well between the 4 data sets (within ± 20 µatm, with the exception of 2 points). East of 165°W, p_wCO_2 differs by as much as 80 µatm although sst for the 4 data sets agree with the COADS climatological values to within 1°C. Observed salinities for 1985 and 1987 are also consistent with climatological value; although the 1986 salinities are lower than climatological values, the normalized p_wCO_2 falls between 1985 and 1987 values. These comparisons suggest that, to the west, meridional gradients in sst and interannual differences in sst can largely account for the p_wCO_2 differences (~ 42 and 28 µatm, respectively) shown in Fig. 7. Differences in p_wCO_2 east of 165°W however cannot be attributed to spatial gradients or interannual variations in sst.

The LDEO data provide an especially interesting comparison because the measurements were made over a similar area (Fig. 1) about 18 to 25 days apart on successive trans-Pacific crossings. As shown in Fig. 5a and discussed in Subsection 3.1, August and September 1986 data show similar sst except that the September data point at 180° is warmer than the August value by ~ 1.5°C. If the pCO_2 values in seawater are normalized to a constant temperature (Fig. 8) to remove the effect of temperature differences along these two crossings, the mean September value is found to be significantly reduced (by 24 µatm) from the mean August value. On the other hand, the mean phosphate concentration observed for the September crossing is found to be smaller than that for the August crossing by about 0.15 µmol kg^{-1} (Fig. 6). Using the Redfield C/P ratio of 106, this corresponds to a photosynthetic reduction of DIC by about 15 µmol kg^{-1} and of p_wCO_2 by about 30 µatm. Therefore, the lower p_wCO_2 in September may be attributed to a net uptake of CO_2 due to photosynthesis.

Observations at the easternmost portion of the study area (50°N, 150°W) can be compared with those from the Canadian Ocean Weather Station P (50°N, 145°W) from 1973 through 1978 by

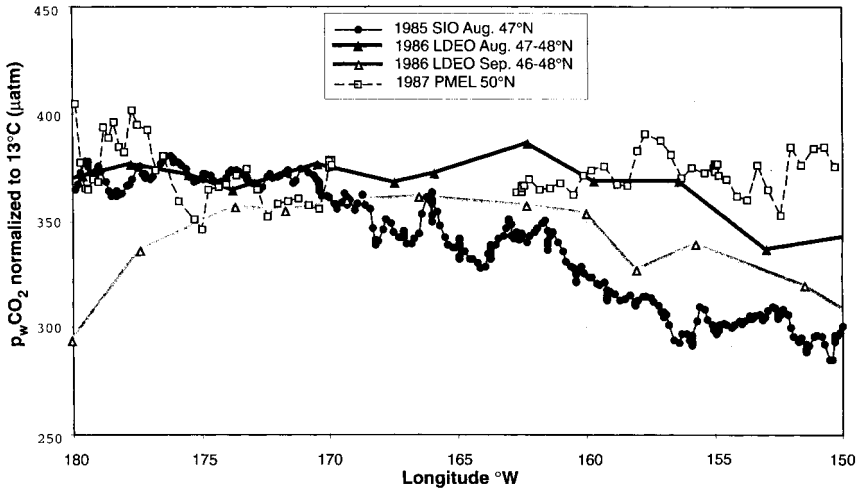

Fig. 8. Comparison of p_wCO_2 normalized to 13°C (the climatological mean temperature for the region 180°–150°W and 45°–50°N for August/September according to COADS monthly average surface marine observations).

Wong and Chan (1991). Their weekly pCO_2 data are compared in Fig. 9 with the data reported here. Their air-sea ΔpCO_2 values ranged from -23 to $+19$ µatm for the time period 15 August to 15 September. While the 1985 SIO and September 1986 LDEO data fall within this range, the August 1986 SIO and 1987 PMEL ΔpCO_2 values are 25 to 35 µatm higher than their data.

Comparisons with Station P data were also made for data which were temperature-normalized and collected at Station P and its vicinity during a 15-year period. pCO_2 data have been normalized to an annual mean temperature of 10°C and plotted against the day of a single year in Fig. 10 (after Takahashi et al., 1997). The sinusoidal seasonal trend observed in the temperature-normalized p_wCO_2 values illustrates that the total CO_2 concentration in surface waters changes seasonally, from a maximum in February due to winter upwelling to a minimum in September due to photosynthesis in strongly stratified photic waters. The magnitude of seasonal changes observed for nitrate in surface water (Parslow, 1981) is consistent with the observed seasonal variation of temperature-normalized p_wCO_2 values (Takahashi et al., 1993). The effect of seasonal changes in the total CO_2 concentration on pCO_2 partly counteracts the effect of seasonal temperature changes, thus reducing significantly the magnitude of seasonal changes in the mixed layer pCO_2. The observations of Wong and Chan (small dots in Fig. 10) scatter around the mean sinusoidal curve by about ± 25 µatm. This reflects the time variability of surface pCO_2 data including diurnal, seasonal, and interannual time scales. The spatial and temporal variations described in this study (shown in Figs. 5, 6, and discussed more fully in the next sections) are of the same magnitude as the temporal variations observed at Station P, and are consistent with the range of temperature-normalized p_wCO_2 observed during late July/early August (Fig. 10). Fig. 10 also shows that the 1973–1978 pCO_2 data are indistinguishable from the 1983–87 data of LDEO and PMEL, suggesting that the effect of

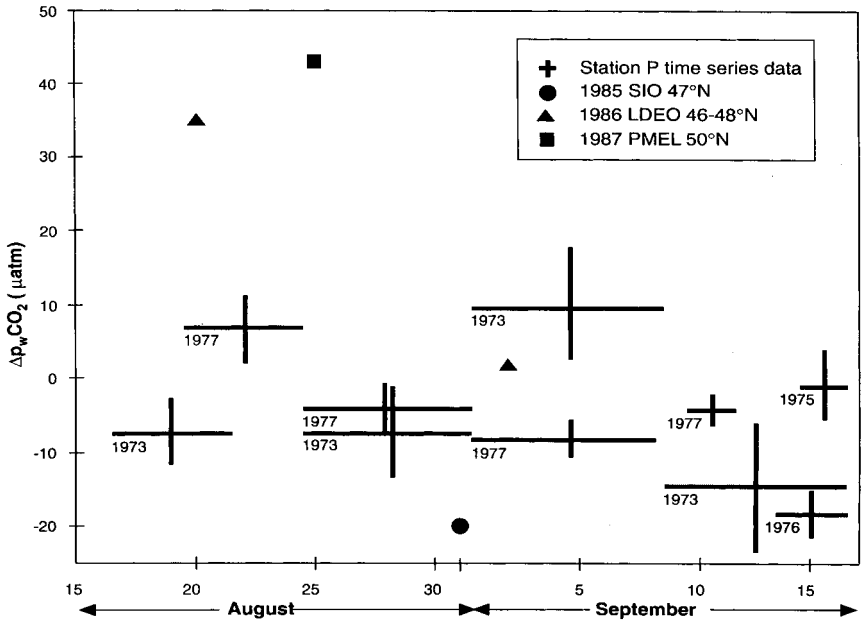

Fig. 9. Comparison of data from the easternmost extremes (150°W) of these 4 cruises with time series data from Station P (50°N, 145°W). Station P data are those reported by Wong and Chan (1991). The point of crossing indicates the mean of the combined ΔpCO_2 data, the length of the horizontal line shows the time over which the data were averaged, and the height of the vertical line indicates ± 1 sigma deviation from the mean.

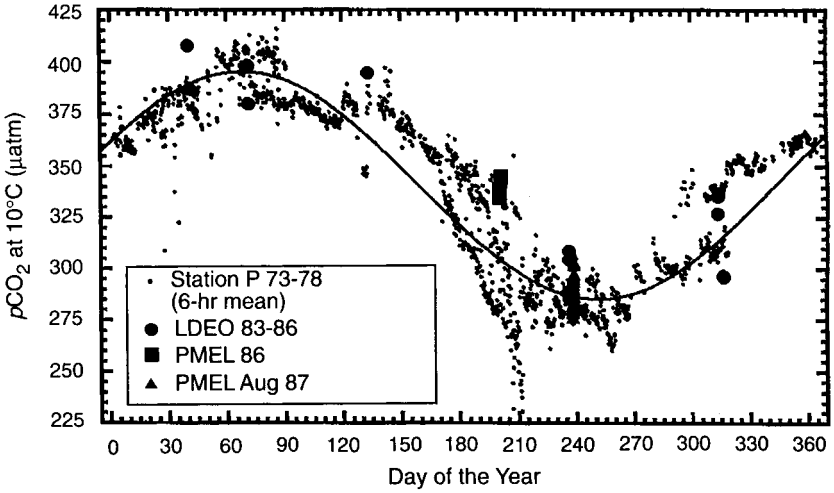

Fig. 10. Comparison of Station P data with data collected in the vicinity (48.5°N–52.5°N and 141°W–149°W) during a 15-year period. The Station P data (Wong and Chan, 1991) were collected over the period 1973–1978. The data were all normalized to an annual mean temperature of 10°C and plotted against the day of a single year. The seasonal trend is shown with a sinusoidal function (after Takahashi et al., 1997).

industrial CO_2 on seawater pCO_2 is too small to be clearly recognized above the natural noise levels at this site.

3.3. *Sampling frequency of* ΔpCO_2

Since the density of observations is quite different for the four data sets used here, it seems reasonable to ask whether differences in sampling density might affect the results discussed so far. We first note that the higher frequency sampling indicates significant variability in ΔpCO_2 over small space scales. The half-hourly SIO data show changes of up to 30 µatm over 1° of longitude. To assess the impact of sampling density on the mean ΔpCO_2 for a given set of observations we subsampled the SIO data at only the longitudes coinciding with those where LDEO data were collected. In so doing, 3% of the SIO data points are used. The mean ΔpCO_2 is increased by about 1 µatm and becomes positive (+0.30 µatm) instead of negative (−0.83 µatm) when the higher frequency data are integrated, as shown in Table 1. If subsampled at the same longitudes as the LDEO September data, mean ΔpCO_2 is decreased by 0.2 µatm to −1.0 µatm. The PMEL data show larger differences on subsampling. If sampled at the same longitudes as the LDEO August and September data, the mean ΔpCO_2 value of +0.5 µatm becomes −1.6 and +3.6 µatm, respectively. In this case, 10% of the PMEL observations were used to obtain subsampled means. Therefore, the mean values obtained on the basis of low frequency LDEO data (1 sample every 150–225 km) may be skewed by up to about 3 µatm.

3.4. *Temporal variability of* ΔpCO_2

That the seasonal variability of ΔpCO_2 is important to determining the mean ΔpCO_2 for a region has been shown using time series data (Wong and Chan, 1991; Winn et al., 1994; Bates et al., 1996) and repeat measurements over a larger region (e.g., Metzl et al. (1995) in the Indian Ocean and Takahashi et al. (1991, 1993) in the North Pacific). Estimating a mean seasonal value of ΔpCO_2 usually requires combining observations collected on several expeditions in different years. This analysis implicitly assumes that the variability of ΔpCO_2 is small over the time scale of hours to weeks and that the interannual variability is likewise small. We here examine the assumption that a single set of observations is representative of the seasonal mean value of ΔpCO_2 for a given location.

The seasonal range of mean p_wCO_2 in this region of the subarctic Pacific (45°–48.5°N,

170°–180°W) is ~50 μatm (−10 to +40) according to Takahashi et al. (1991). This assessment is based on observations from 5 LDEO expeditions including the 2 described here. This is consistent with the seasonal range at Station P (~40 μatm) observed by Wong and Chan (1991) over the years 1973 to 1978.

The variability of ΔpCO_2 on shorter time scales in this region is indicated in the Station P time series data (Wong and Chan, 1991): changes in the consecutive weekly averages of ΔpCO_2 can be as high as 25 μatm (Fig. 10). Similar variability is seen in the data presented here collected over 12 hours at Station P: SIO ΔpCO_2 values range from −11 to +0.6 μatm; PMEL values range from −17 to +4 μatm. This high frequency variability is consistent with that found in the North Atlantic (Watson et al., 1991) and in the Indian Ocean (Poisson et al., 1993).

On longer time scales, variability of p_wCO_2 assessed from limited time series data indicates that interannual differences of tens of microatmospheres are not uncommon. Interannual variability of up to 15 μatm has been reported for the ALOHA 1989–1992 data (Winn et al., 1994) and up to 30 μatm for the BATS data collected during 1991–1993 (Bates et al., 1996). In the subarctic Pacific, the interannual variability of p_wCO_2 during August/September can be estimated directly from the Station P time series data (Wong and Chan, 1991) or from the temperature-normalized data reported by Takahashi et al. (1993). In both cases, the monthly averages of p_wCO_2 from 1973–1978 vary by up to 30 μatm. Interannual variability in the equatorial Pacific driven by changes in upwelling during ENSO can be even higher (Feely at al., 1987; Inoue and Sugimura, 1992; Wong et al., 1993; Feely et al., 1995; Wanninkhof et al., 1996; Feely et al., in press). These comparisons of p_wCO_2 data on different time scales suggests that the mean seasonal variability for this region of the subarctic North Pacific is approximately 3 times larger than the high-frequency variability (hours to week) and approximately 1.5 times the interannual variability.

3.5. *Comparison of wind speed*

Wind data from the four different sources used here, calculated for the 1985 SIO sampling locations, are compared in Fig. 11. The 30-year and 150-year climatological winds are similar, varying by less than 0.5 m s^{-1}. The operational analysis winds (FNOC and ECMWF 1985 data) track one another well but are not as smooth as the climatological winds. The modeled wind speeds vary by a factor of 0.7 to 1.5 from the climatological values, but in general they tend to be higher than either of the climatological winds. Remotely sensed winds with appropriate ground-truth verification (Boutin and Etcheto, 1991; 1996) will provide consistently measured, high frequency winds which will be useful for future flux computations of this type.

Fig. 12 shows the differences in monthly averaged ECMWF wind speeds calculated for each data set's sampling locations and times. This comparison suggests that wind speed differences can be up to 30%. This difference accounts for interannual differences in wind speed as well as differences in sampling times.

3.6. *Comparison of CO_2 fluxes*

Fluxes calculated at each ΔpCO_2 sampling point were integrated to give a ship-track-mean flux for each combination of ΔpCO_2, E(W) parameterization and W for each data set. Local fluxes were integrated over the region defined by the east-west extent of the data (180° to 150°W) and 1 m north-south to give mean fluxes in units of mmol CO_2 m^{-2} day^{-1}. The fluxes calculated in this study follow the pattern of the ΔpCO_2 differences very closely. The 1985 fluxes are negative (ocean sink) and close to zero (−0.17 to 0.00 mmol CO_2 m^{-2} d^{-1}). The 1986 fluxes are larger and positive (0.38 to 4.05 mmol CO_2 m^{-2} d^{-1}) suggesting an ocean source for CO_2, but the magnitude of the fluxes for August 1986 are about 3 times higher than those of September 1986. The 1987 fluxes are both positive and negative, but the values are moderate (−0.94 to +0.25 mmol CO_2 m^{-2} d^{-1}). The range (5.0 mmol CO_2 m^{-2} d^{-1}) of the net CO_2 flux observed for this study is 4 times larger than the range (1.2 mmol CO_2 m^{-2} d^{-1}) of global mean flux (-1.3 ± 0.6 mmol CO_2 m^{-2} d^{-1}) calculated from the range of current estimates (2 ± 1 PgC yr^{-1}) for net CO_2 uptake by the global ocean.

The sign and magnitude of CO_2 fluxes across

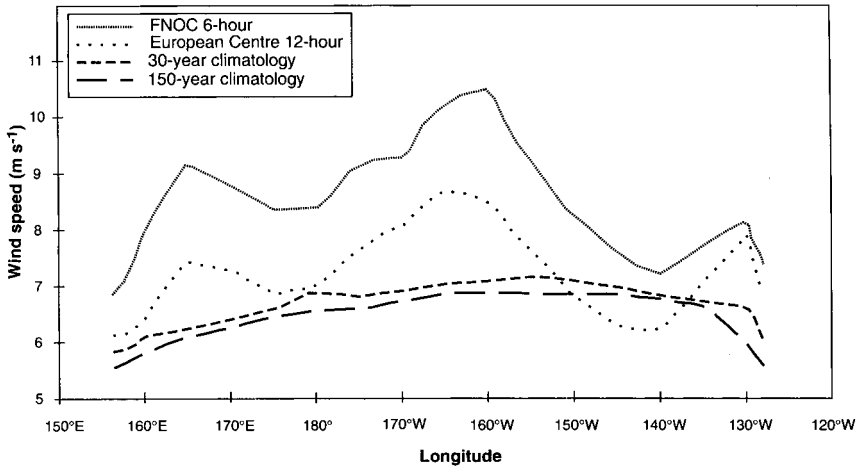

Fig. 11. Comparison of wind speeds used to calculate fluxes for the 1985 SIO data. The FNOC data are calculated from operational vector winds provided at 6-hour interval by the Fleet Numerical Oceanography Center. The European Centre wind speeds are also operational winds provided at 12-h intervals by the European Centre for Medium Range Weather Forecasting. The 30-year and 150-year climatological winds are from Harrison (1989) and Esbensen and Kushnir (1981), respectively.

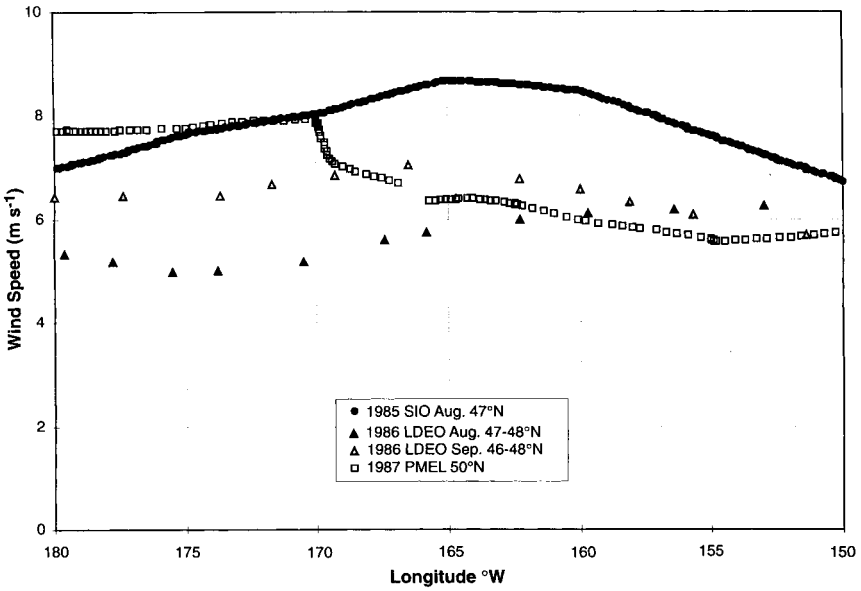

Fig. 12. Comparison of the variability in the 30-day averaged wind speeds for the four sets of ΔpCO_2 data. The winds are operational winds (blending marine observations into a meteorological model) given every 12 hours by the European Centre for Medium-Range Weather Forecasting (ECMWF).

the air-sea interface are principally determined by ΔpCO_2, and secondarily by E(W) parameterization and wind field. The uncertainty in the E(W) parameterization typically results in flux differences of the magnitude also seen here (about a factor of 2). We do not here address the more fundamental question of whether fluxes can be parameterized from wind speed. Accurate assess-

ment of fluxes will ultimately depend on developing techniques to measure CO_2 fluxes directly. Until such flux measurements can be made, we can calculate a range of fluxes and the sensitivity of the calculated flux to different parameterizations.

The effect of the wind field choice is smaller than that of the gas exchange parameterization, but can still be significant. The effect of using different wind fields to calculate fluxes is small for the 1985 data since the ship-track-mean ΔpCO_2 is so small. The effect on the 1986 September data is also small since the wind speeds from modelled data for this period were not significantly different from climatological wind speeds. The effect of wind speed differences is more pronounced for the 1986 August data, with fluxes differing by up to 60% depending on which wind field is used. In the 1987 data analysis, the flux varies from positive to negative depending on which winds are used in the calculations.

4. Mechanisms for P_wCO_2 variability

In this section we examine the individual processes of seasonal heating, biological production, wind-driven mixing and horizontal advection which can change p_wCO_2 in surface seawater on different time scales. The variability of p_wCO_2 might result from any one of several mechanisms or a combination of mechanisms.

4.1. *Seasonal heating and regional gradients*

Weiss et al. (1982) showed that seasonal heating accounted for most ($\sim$90%) of the change in p_wCO_2 in the subtropical Pacific gyres. To examine whether differences in seasonal heating might be responsible for interannual differences in p_wCO_2 in this region, the COADS sea surface temperature data (Woodruff et al., 1987) were examined. The monthly climatological data for 1946–1989 for this region indicate that the seasonal warming from May to August is 6.5°C. Seasonal warming for the years 1985, 1986 and 1987 differ by as much as 1.5°C, which translates to $\sim$23 μatm difference in p_wCO_2. Normalizing the p_wCO_2 data to one representative temperature would reduce the p_wCO_2 differences between data sets if seasonal warming alone were responsible. If interannual differences in seasonal warming were the primary cause of the ΔpCO_2 variability observed here, the temperature-normalized data would be expected to agree to within 25 μatm. Instead the picture is more complicated. Fig. 8 shows the surface water pCO_2 values normalized to the August/September climatological temperature (13°C). With the exception of two data points near the 180° meridian, temperature-normalized pCO_2 west of 163°W agree within $\pm$20 μatm. This suggests that waters in this area had similar values of alkalinity and DIC and that variations observed in the in situ pCO_2 values were due primarily to variations in water temperature. In the eastern part of the study area (between 150°W and 163°W) the agreement is less good. The 1985 SIO values observed along 50°N are greater than the 1987 PMEL values obtained along 47°N by about 70 μatm. The LDEO values obtained between these latitudes fall in between the SIO and PMEL values. This suggests a difference in the chemical composition of seawater to the east of 163°W, either because of strong meridional gradients or because of significant interannual variability in DIC. If the former, either the DIC increased northward or the alkalinity decreased northward. If the northward increase in pCO_2 is assumed to be due entirely to an increase in the DIC concentration, a northward increase of about 35 μmol kg^{-1} is expected. This is broadly consistent with the meridional gradient of DIC concentration observed for this area in 1984–1989 (Takahashi et al., 1993). If interannual variability in DIC is responsible for the variability in p_wCO_2, this variability may result from photosynthetic utilization and/or from subsurface mixing (discussed in the next two sections).

4.2. *Photosynthetic utilization*

Photosynthetic activity can significantly reduce p_wCO_2 in surface waters, and interannual differences in new production may affect the inter-expedition p_wCO_2 variations seen in the four data sets reported here. Biological productivity and chlorophyll data were not collected on these expeditions (although phosphate concentrations were measured in 1985 and 1986) so any interannual differences between 1985, 1986 and 1987 cannot be directly compared. However, net CO_2 utilization in the mixed layer can be calculated from carbon concentration data if several assump-

tions are made. The calculated value of net CO_2 utilization is consistent with measurements of primary production for the subarctic North Pacific, suggesting that variations in photosynthetic carbon utilization contribute to the observed variability in p_wCO_2.

The 1986 data are examined since DIC and phosphate were measured also (Takahashi et al., 1991). Inter-expedition differences (August–September) in pCO_2 normalized to 13°C range from 5 to 78 μatm, but the September ship-track-mean value (343.3 μatm) is 24 μatm lower than the August ship-track-mean value (367.1 μatm). Using a Revelle factor ($\partial \ln pCO_2/\partial \ln$ DIC) of 11 (Takahashi et al., 1980) and a DIC of 2050 μmol kg^{-1}, the August–September lowering of pCO_2 corresponds to a decrease of 12.5 μmol kg^{-1} in DIC concentration. Phosphate data (shown in Fig. 6) suggest that, with the exception of two points between 169°W and 166°W, the September values are on the average $\sim$0.1 μmol kg^{-1} lower than the August values. The observed decrease in pCO_2, DIC, and phosphate concentrations along the LDEO tracks are consistent with the biological utilization of CO_2 with the Redfield C/P ratio of 106 : 1. Since these LDEO measurements were made at stations occupied 18–25 days apart, the net CO_2 utilization in the mixed layer along the ship's tracks are estimated to be $\sim$160 mg C m^{-2} day^{-1}. In this calculation, a mean time period of 21.5 days and a mixed layer thickness of 23 ± 10 m observed by Takahashi et al. (1991) along the ship's tracks concurrent with the CO_2 and phosphate measurements has been used.

We compare changes in DIC inventory with primary production data collected at Station P. As discussed by Chipman et al. (1993), in vitro ^{14}C assimilation rates are not strictly comparable with net CO_2 utilization rates based on mixed layer CO_2 concentration changes. However Chipman et al. found in the North Atlantic that the mean daily reduction rate of DIC in the mixed layer over 14 days was 75% of the photosynthetic production rate (from ^{14}C assimilation rate integrated over the mixed layer and based on a 14 h incubation period). The estimate of net CO_2 utilization calculated here (160 mg C m^{-2} day^{-1}) falls within the lower range of primary production data (120–680 mg C m^{-2} day^{-1}, integrated over 20 m) collected at Station P during August 1988 and September 1987 (Welschmeyer et al., 1993). These measurements of ^{14}C assimilation were based on a 24 h incubation rate which has been estimated by Bender et al. (1992) to be 1.17 times the measured value of 14 h ^{14}C production. The calculated net CO_2 utilization rate based on changes in DIC inventory are thus consistent with ^{14}C assimilation rates integrated over the mixed layer at Station P.

The calculations thus far have suggested that variability in biological production can account for mean differences in ΔpCO_2 between data sets. Can biological activity also account for large local differences in temperature-normalized pCO_2 as high as 80 μatm? The LDEO data provide an opportunity to address this question as well. Repeat observations (18 days later) near 180° indicate a decrease in temperature-normalized ΔpCO_2 of 78 μatm and a concomitant decrease in salinity-normalized DIC of 43 μmol kg^{-1}. LDEO surface seawater pCO_2 (normalized to 13°C) at 180° in August was 371 μatm. If a Revelle factor of 11 is again assumed, the inter-expedition change in DIC calculated from the ΔpCO_2 change is 39 μmol kg^{-1}, consistent with the observed DIC decrease, but less than the value (53 ± 10 μmol kg^{-1}) calculated from the Redfield ratio and the decrease in phosphate from August to September (0.5 ± 0.1 μmol kg^{-1}). From the observed change in DIC over 18 days, a net production rate may be calculated to be 670 mg C m^{-2} day^{-1}. Since the sea-air CO_2 fluxes is relatively small, it has been neglected in this calculation. This value falls at the high end of the production range given by Welschmeyer for Station P.

These estimates of the biological role in p_wCO_2 changes are approximate because the processes of horizontal and vertical advection, gas exchange, and mixed layer depth changes have been neglected. Despite their limitations, estimates of this type do suggest that plausible levels of biological activity could produce the observed inter-expedition differences in ΔpCO_2.

4.3. *Mixing*

Variability in lateral and vertical mixing will also affect variability in p_wCO_2. Tabata (1989) however attributed most of the interannual variability of the upper ocean at Station P to variability

in vertical motions. He was able to account for 75% of the temperature variability and 80% of the salinity variability (at depths less than 1000 m) by this mechanism. Accordingly, variation of vertical mixing appears to be a major cause of variability in p_wCO_2. The importance of vertical motions to upper layer variability can also be seen in the observational and modeling results of Denman and Miyake (1973) for temperature and salinity at Ocean Station P over 6 weeks (2 to 8 times per day). The observations were compared with the results of an upper ocean model requiring routine meteorological observations as input parameters. They concluded that upper layer mixing induced by synoptic-scale weather disturbances resulted in significant temperature, salinity, and mixed layer depth fluctuations. Summer storm activity can deepen the mixed layer by as much as 40 m (from 10 to 50 m) in a day.

The importance of vertical mixing to variability in upper ocean p_wCO_2 was explicitly suggested by the 1-dimensional modeling study of Garçon et al. (1992). They modeled p_wCO_2 in the mixed layer using meteorological and hydrographic input data for the years 1971 and 1972 and found good agreement with the observations made in 1973–1978, years for which p_wCO_2 observations were made. One conclusion of their study was that episodic mixing events can induce strong short-term variability (up to 30 μatm over days) in p_wCO_2.

5. Discussion and summary

5.1. *Variability of* p_wCO_2

Mean seasonal values of ΔpCO_2 for a region are commonly determined by combining data which have been collected several months apart, in different years, and interpolated over tens of degrees in space. Such seasonal contour maps of ΔpCO_2 are necessitated by sparse data, yet there is little direct information regarding how representative is one pCO_2 data set from a particular cruise track for a larger time and space scale. The data presented here were not collected in the same location and season, but are relatively tightly constrained in time and space, and so provide an opportunity to examine the accuracy of larger scale interpolations. We note that a systematic error of 1 μatm in the annual average ΔpCO_2 results in an error of ~ 0.2 PgC yr^{-1} ($1Pg = 10^{15}g$) in estimating the net global ocean flux (Watson et al., 1991).

Comparison of 4 subarctic Pacific data sets shows that differences in the ship-track-mean ΔpCO_2 in a narrow latitudinal range (46–50°N) are as much as 25 μatm (Table 1), while ΔpCO_2 varies locally by as much as 64 μatm during the months of August and September in 1985, 86, and 87. The differences cannot be attributed to analytical or sampling frequency differences (here assessed as <10 μatm and <4 μatm, respectively) and therefore must result from real oceanographic differences. An examination of the spatial and temporal variability of ΔpCO_2 suggests that these differences can contribute significant uncertainty to regional and seasonal mean ΔpCO_2 estimates.

Regional mean estimates for late summer from sparse measurements are necessarily uncertain when the variations in higher frequency (~ 15 μatm over hours to weeks) and lower frequency (~ 30 μatm interannually) observations made during late summer are almost as high as the seasonal variation (~ 45 μatm). Spatial variability likewise contributes significant uncertainty to regional estimates of ΔpCO_2, and the subsampling study reported here (described in the results section and summarized in Table 1) shows that sampling density can affect both the magnitude and the sign of the estimated ship-track-mean ΔpCO_2.

Large spatial and temporal variability of ΔpCO_2 for some regions may contribute significantly to the uncertainty in estimating basin-wide fluxes of carbon from the atmosphere to the ocean. Polar and subpolar regions are typically undersampled and often have a summer sampling bias. Undersampling necessitates neglecting any interannual variability when combining chemical data sets, although the subarctic Pacific exhibits interannual variability in surface temperature and surface winds which are currently the subject of much study. Steep meridional gradients of temperature and nutrient concentrations in the subarctic Pacific also suggest that seasonal averages of ΔpCO_2 over large areas are more uncertain in the subpolar and polar regions than in the subtropics. Repeat observations of p_wCO_2 in the Western Pacific (Inoue et al., 1995) indicate substantially less variability (<5 μatm) at a given latitude over the region 3°N–27°N, 137°E than is shown here. Similarly, repeat seasonal observations in the cent-

ral Pacific (Wong et al., 1995) suggest smaller variability of surface seawater CO_2 concentrations in the subtropical North Pacific. These subtropical data have been averaged in time and space, and are expected to be smoother, but suggest smaller variability of p_wCO_2 in the subtropical than the subpolar regions. These findings emphasize that high-density measurements in time and space are necessary to accurately determine a mean regional and seasonal value of ΔpCO_2 in high gradient areas such as our study area in the North Pacific. We note that there are currently few systematic observations of the variability in ΔpCO_2 to help to decide how many measurements are required to determine accurate regional and seasonal mean values of CO_2 disequilibria.

5.2. *Variability of calculated CO_2 fluxes*

Ship-track-mean fluxes calculated from these 4 data sets range from -0.94 to $+4.1$ mmol CO_2 m^{-2} d^{-1}, depending on which set of ΔpCO_2, E(W) parameterization and W are used. The relative differences in fluxes calculated from these data sets result principally from differences in ΔpCO_2. We note that the range of net CO_2 fluxes calculated for this study (5.0 mmol CO_2 m^{-2} d^{-1}) is significantly larger than the range (1.2 mmol CO_2 m^{-2} d^{-1}) of global mean flux (-1.3 ± 0.6 mmol CO_2 m^{-2} d^{-1}) which corresponds to current estimates for the net CO_2 uptake for the global ocean (2 ± 1 PgC yr^{-1}).

5.3. *Broader context for flux results*

To obtain some idea of how variability of the magnitude reported here translates into uncertainties in regional carbon uptake values, we scale the range of fluxes calculated here to a larger region. We emphasize that the point of this exercise is not to predict basin-wide carbon fluxes, but to provide a broader context for the variability in ΔpCO_2 reported here.

Scaling the range of fluxes reported here to the subarctic/arctic North Pacific (40°N to 66°N) suggests some reasons for disagreements about the magnitude (and sometimes the sign) of basin-wide flux estimates. This scaling gives carbon uptake values ranging from -0.1 to $+0.3$ PgC yr^{-1}, either a small source or a small sink for carbon depending on which combination of ship-track-mean ΔpCO_2, wind field, and gas exchange parameterization is used. Whereas the fluxes calculated from the 4 data sets presented in this study are tightly constrained in time and space, they cannot be considered representative of annual fluxes for the entire subarctic/arctic region. It is interesting to note, however, that this extension gives a range of fluxes of the same magnitude as the range of estimates for the North Pacific carbon flux (-0.1 to -0.4 PgC yr^{-1}) given by three different investigators using different data sets. Takahashi et al. (1991) used 23 trans-Pacific sections to calculate that the flux of carbon from the atmosphere to the ocean in the North Pacific (20° to 57°N) is ~ 0.1 PgC yr^{-1}. Winn et al. (1994) used ALOHA time series data to suggest that the North Pacific subtropical gyre alone (15° to 40°N, 120°W to 130°E) may absorb twice that amount. And Landrum et al. (1996) estimated a range of fluxes for a 6-month period which encompasses the Takahashi et al. (1991) and Winn et al. (1994) annual estimates. They calculated that the North Pacific (15 to 54°N) acts as a sink (0.07 to 0.2 PgC) for the months February to August.

It isn't certain how widespread is variability in ΔpCO_2 of the magnitude found in this study, but it seems likely that determining a mean regional value of ΔpCO_2 from 1 set of observations may contribute significant uncertainty to basin-wide assessments of carbon uptake. At present we don't know how much information is needed to obtain an accurate regional and seasonal mean value of ΔpCO_2 — what sampling density along a given track, how much geographical coverage, or how many tracks to define a seasonal trend. Objective analyses (Landrum et al., 1996) are a first step toward assigning uncertainties to regional estimates of ΔpCO_2, but are necessarily limited by the available data. We suggest that field studies specifically designed to assess sampling questions would be useful. For the present, repeat lines in the same season provide an opportunity to address these issues. As new technology (buoy-mounted pCO_2 and wind sensors, accurate satellite wind data) becomes available it will become increasingly possible to quantitatively assess the uncertainties in our observational assessments of carbon uptake by the oceans and to design observational programs to address these uncertainties. Significant progress by several groups (Lefèvre et al., 1993; DeGrandpre, 1995; Friederich et al., 1995;

Merlivat and Brault, 1995; McNeil and Merlivat, 1996) suggests that remote CO_2 sensors will soon be available to begin assessing temporal variability at some locations.

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REFERENCES

Bates, N., Michaels, A. and Knap, A. 1996. Seasonal and interannual variability of oceanic carbon dioxide species at the U. S. JGOFS Bermuda Atlantic Time-series Study (BATS) site. *Deep-Sea Res. II* **43**, 347–383.

Bender, M., Ducklow, H., Kiddon, J., Marra, J. and Martin, J. 1992. The carbon balance during the 1989 spring bloom in the North Atlantic Ocean, 47°N, 20°W. *Deep-Sea Res.* **39**, 1707–1725.

Boutin, J. and Etcheto, J. 1991. Intrinsic error in the air-sea CO_2 exchange coefficient resulting from the use of satellite wind speeds. *Tellus* **43B**, 236–246.

Boutin, J. and Etcheto, J. 1996. Consistency of Geosat, SSM/I, and ERS-1 global surface wind speeds — Comparison with in situ data. *J. Atmos. Oceanic Technol.* **13**, 183–197.

Chipman, D. W., Marra, J. and Takahashi, T. 1993. Primary production at 47°N and 20°W in the North Atlantic Ocean: a comparison between the ^{14}C incubation method and the mixed layer carbon budget. *Deep-Sea Res.* **40**, 151–169.

Conkright, M. E., Levitus, S. and Boyer, T. P. 1994. *World Ocean Atlas* 1994, vol. 1: Nutrients. NOAA Atlas NESDIS 1, 150 pp.

DeGrandpre, M. D., Hammar, T. R., Smith, S. P. and Sayles, F. L. 1995. In situ measurements of seawater pCO_2. *Limnol. Oceanogr.* **40**, 969–975.

Denman, K. L. and Miyake, M. 1973. Upper layer modification at Ocean Station Papa: Observations and simulation. *J. Phys. Oceanogr.* **3**, 185–196.

Dodimead, A., Favorite, F. and Hirano, T. 1963. Review of oceanography of the subarctic Pacific region. *Bull. Internat. N. Pacif. Fish. Comm.* **13**, 1–195.

Esbensen, S. and Kushnir, Y. 1981. *The heat budget of the global ocean.* 29. Climatic Research Institute.

Favorite, F., Dodimead, A. and Nasu, K. 1976. Oceanography of the subarctic Pacific region. *Bull. Int. North Pac. Fish. Comm.* **33**, 1–187.

Feely, R., Gammon, R., Taft, B., Pullen, P., Waterman, L., Conway, T., Gendron, J. and Wisegarver, D. 1987. Distribution of chemical tracers in the eastern equatorial Pacific during and after the 1982–1983 El Niño/Southern Oscillation event. *J. Geophys. Res.* **92**, 6545–6558.

Feely, R., Wanninkhof, R., Cosca, C., Murphy, P., Lamb, M. and Steckley, M. 1995. CO_2 distributions in the equatorial Pacific during the 1991–1992 ENSO event. *Deep-Sea Res. II* **42**, 365–386.

Feely, R. A., Wanninkhof, R., Coyet, C., Archer, D. E. and Takahashi, T. in press. Variability of CO_2 distributions and sea-air fluxes in the central and eastern equatorial Pacific during the 1991-94 El Niño. *Deep-Sea Res. II* 000–000.

Friederich, G. E., Brewer, P. G., Herlien, R. and Chavez, F. P. 1995. Measurement of sea surface partial pressure of CO_2 from a moored buoy. *Deep-Sea Res. I* **42**, 1175–1186.

Garçon, V., Thomas, F., Wong, C. and Minster, J.-F. 1992. Gaining insight into the seasonal variability of CO_2 at Ocean Station P using an upper ocean model. *Deep-Sea Research* **39**, 921–938.

Geophysical Monitoring for Climatic Change Laboratory (GMCC, now Climate Monitoring and Diagnostics Laboratory, CMDL). 1986. *GMCC no. 14 Summary Report* 1985. National Oceanic and Atmospheric Administration, Environmental Research Laboratories, Boulder, CO.

Geophysical Monitoring for Climatic Change Laboratory (GMCC, now Climate Monitoring and Diagnostics Laboratory, CMDL). 1987. *GMCC no. 15 Summary Report 1986*. National Oceanic and Atmospheric

Administration, Environmental Research Laboratories, Boulder, CO.

Geophysical Monitoring for Climatic Change Laboratory (GMCC, now Climate Monitoring and Diagnostics Laboratory, CMDL). 1988. *GMCC no. 16 Summary Report 1987*. National Oceanic and Atmospheric Administration, Environmental Research Laboratories, Boulder, CO.

Hansson, I. 1973. A new set of acidity constants for carbonic acid and boric acid in sea water. *Deep-Sea Research* **20**, 461–478.

Harrison, D. 1989. On climatological monthly mean wind stress and wind stress curl fields over the world ocean. *J. Clim.* **2**, 57–70.

Inoue, H. and Sugimura, Y. 1992. Variations and distribution of CO_2 in and over the equatorial Pacific during the period from the 1986/88 El Niño to the 1988/89 La Niña event. *Tellus* **44B**, 1–22.

Inoue, H. Y., Matsueda, M., Ishii, K., Fushimi, M., Hirota, I. A. and Takasugi, Y. 1995. Long-term trend of the partial pressure of carbon dioxide (pCO_2) in surface waters of the western North Pacific, 1984-1993. *Tellus* **47B**, 391-413.

Landrum, L. L., Gammon, R. H., Feely, R. A., Murphy, P. P., Kelly, K. C., Cosca, C. E. and Weiss, R. F. 1996. North Pacific Ocean CO_2 disequilibrium for spring through summer, 1985–1989. *J. Geophys. Res.* **101**, 28 539–28 555.

Lefèvre, N., Ciabrini, J. P., Michard, G., Brient, B., DuChaffaut, M. and Merlivat, L. 1993. A new optical sensor for PCO_2 measurements in seawater. *Mar. Chem.* **42**, 189–198.

Levitus, S. 1982. Climatological Atlas of the World Ocean. NOAA Professional paper 13, 174 pp.

Levitus, S. and Boyer, T. P. 1994. *World Ocean Atlas* 1994, vol. 2: Oxygen, NOAA Atlas NESDIS 2, Washington, D. C., 186 pp.

Levitus, S., Burgett, T. and Boyer, T. P. 1994. *World Ocean Atlas* 1994, vol. 3: Salinity, NOAA Atlas NESDIS 3, Washington, D. C., 99 pp.

Liss, P. and Merlivat, L. 1986. Air-sea gas exchange rates: introduction and synthesis. In: *The rôle of air-sea exchange in geochemical cycling* (ed. P. Buat-Ménard). D. Reidel Publishing Company, Dordrecht, Holland, 113–127.

McNeil, C. L. and Merlivat, L. 1996. The warm oceanic surface layer: Implications for CO_2 fluxes and surface gas measurements. *Geophys. Res. Lett.* **23**, 3575–3578.

Merlivat, L. and Brault, P. 1995. CARIOCA Buoy: Carbon dioxide monitor. *Sea Technology* **36**, 23–30.

Metzl, N., Poisson, A., Louanchi, F., Brunet, C., Schauer, B. and Bres, B. 1995. Spatio-temporal distributions of air-sea fluxes of CO_2 in the Indian and Antarctic oceans. *Tellus* **47B**, 56–69.

Murphy, P., Feely, R., Gammon, R., Harrison, D., Kelly, K. and Waterman, L. 1991. Assessment of the air-sea exchange of CO_2 in the South Pacific during austral autumn. *J. Geophys. Res.* **96**, 20 455–20 465.

Murphy, P. P., Kelly, K. C., Feely, R. A. and Gammon, R. H. 1994. *Carbon dioxide concentrations in surface water and the atmosphere: PMEL cruises 1986–1989*. NOAA Tech. Memo. ERL PMEL-101 (NTIS DE95–009659), 185 pp.

Parslow, J. S. 1981. Phytoplankton-zooplankton interactions: Data analysis and modelling (with particular reference to Ocean Station P (50°N, 145°W) and controlled ecosystem experiments, PhD Dissertation, University of British Columbia, Vancouver, B. C., 400 pp.

Poisson, A., Metzl, N., Brunet, C., Schauer, B., Bres, B., Ruiz-Pino, D. and Louanchi, F. 1993. Variability of sources and sinks of CO_2 in the Western Indian and Southern Oceans during the year 1991. *J. Geophys. Res.* **98**, 22 759–22 778.

Siegenthaler, U. and Sarmiento, J. 1993. Atmospheric carbon dioxide and the ocean. *Nature* **365**, 119–125.

Sundquist, E. 1993. The global carbon dioxide budget. *Science* **259**, 934–941.

Tabata, S. 1989. Trends and long-term variability of ocean properties at Ocean Station P in the northeast Pacific Ocean. In: *Aspects of climate variability in the Pacific and the Western Americas* (ed. D. H. Peterson). AGU, Washington D. C., 113–132.

Takahashi, T., Broecker, W. S., Werner, S. R. and Bainbridge, A. E. 1980. Carbonate chemistry of the surface waters of the world oceans. In: *Isotope marine chemistry* (eds. E. D. Goldberg, Y. Horibe and K. Saruhashi). Uchida Rokakuho Publishing Co. Ltd., Tokyo, Japan, 291–326.

Takahashi, T., Goddard, J., Chipman, D. and Noonan, M. 1990. *Carbon chemistry in the confluence areas of the Brazil and Malvinas currents in the south western Atlantic Ocean* Technical Report of Research, 38 pp.

Takahashi, T., Goddard, J., Chipman, D., Sutherland, S. and Mathieu, G. 1991. *Assessment of carbon dioxide sink/source in the north Pacific Ocean: seasonal and geographic variability*, 1984–1989. Final Technical Report for contract 19X-SC428C. Lamont-Doherty Geological Observatory, 111 pp.

Takahashi, T., Olafsson, J., Goddard, J., Chipman, D. and Sutherland, S. 1993. Seasonal variations of CO_2 and nutrients in the high-latitude surface oceans: a comparative study. *Global Biogeochem. Cycles* **7**, 843–878.

Takahashi, T., Takahashi, T. T. and Sutherland, S. C. 1995. An assessment of the role of the North Atlantic as a CO_2 sink. *Phil. Trans. R. Soc. Lond. B* **348**, 143–152.

Takahashi, T., Feely, R. A., Weiss, R. F., Wanninkhof, R. H., Chipman, D. W., Sutherland, S. C. and Takahashi, T. T. 1997. Global air-sea flux of CO_2: An estimate based on measurements of sea-air pCO_2 difference: *Proc. Natl. Acad. Sci. USA* **94**, 8292–8299.

Talley, L., Martin, M., Salameh, P. and Oceanographic Data Facility 1988. *Transpacific section in the subpolar gyre*. SIO Reference 88–9, Scripps Institution of Oceanography, 245 pp.

Tans, P., Fung, I. and Takahashi, T. 1990. Observational constraints on the global atmospheric CO_2 budget. *Science* **247**, 1431–1438.

Wanninkhof, R. 1992. Relationship between wind speed and gas exchange over the ocean. *J. Geophys. Res.* **97**, 7373–7382.

Wanninkhof, R., Feely, R. A., Chen, H., Cosca, C. and Murphy, P. P. 1996. Surface water fCO_2 values in the eastern equatorial Pacific during the 1992–93 El Niño. *J. Geophys. Res.* **101**, 16 333–16 343.

Watson, A., Robinson, C., Robinson, J., Williams, P. L. B. and Fasham, M. 1991. Spatial variability in the sink for atmospheric carbon dioxide in the North Atlantic. *Nature* **350**, 50-53.

Weiss, R. 1981. Determinations of carbon dioxide and methane by dual catalyst flame ionization chromatography and nitrous oxide by electron capture chromatography. *J. Chromatogr.* **19**, 611–616.

Weiss, R. F., Jahnke, R. A. and Keeling, C. D. 1982. Seasonal effects of temperature and salinity on the partial pressure of CO_2 in seawater. *Nature* **300**, 511–513.

Welschmeyer, N. A., Strom, S., Goericke, R., DiTullio, G., Belvin, M. and Petersen, W. 1993. Primary production in the subarctic Pacific Ocean: Project SUPER. *Prog. Oceanogr.* **32**, 101–135.

Winn, C., Mackenzie, F., Carrillo, C., Sabine, C. and Karl, D. 1994. Air-sea carbon dioxide exchange in the North Pacific subtropical gyre: implications for the global carbon budget. *Global Biogeochem. Cycles* **8**, 157–163.

Wong, C. and Chan, Y.-H. 1991. Temporal variations in the partial pressure and flux of CO_2 and ocean station P in the subarctic northeast Pacific Ocean. *Tellus* **43B**, 206–223.

Wong, C., Chan, Y.-H., Page, J., Smith, G. and Bellegay, R. 1993. Changes in equatorial CO_2 flux and new production estimated from CO_2 and nutrient levels in Pacific surface waters during the 1986/87 El Niño. *Tellus* **45B**, 64–79.

Wong, C. S., Chan, Y.-H., Page, J. S., Smith, G. E., Bellegay, R. D. and Iseki, K. 1995. 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. *Tellus* **47B**, 414–430.

Woodruff, S., Slutz, R., Jenne, R. and Steurer, P. 1987. A comprehensive ocean-atmosphere data set. *Bulletin American Meteorological Society* **68**, 1239–1250.