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Spatial variability and decadal trend of the oceanic CO₂ in the western equatorial Pacific warm/fresh water

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Abstract

Data of the partial pressure of carbon dioxide in surface seawater ($p\text{CO}_{2\text{sw}}$) collected in the western equatorial Pacific (144°E – 160°W, 5°S – 5°N) since the 1980's have been used to determine the spatial variability and decadal trends in $p\text{CO}_{2\text{sw}}$. A total of 109 cruises since 1983, including 30 cruises since 1990 with total dissolved inorganic carbon (TCO_2) measurements, are synthesized for this zone. The western equatorial Pacific warm/fresh surface water where $T \geq 29.0$ °C and $S \leq 34.8$ is nitrate-depleted and it is moderately supersaturated with CO_2 ($0 < \Delta p\text{CO}_2 (= p\text{CO}_{2\text{sw}} - p\text{CO}_{2\text{air}}) / \mu\text{atm} < 40$). A slight CO_2 undersaturation ($-20 < \Delta p\text{CO}_2 / \mu\text{atm} < 0$) was also observed on many cruises where thin, low density layers ($\sigma_t < 21.4$) capped barrier layers. The undersaturation may result from net biological uptake of CO_2 due to nitrogen fixation. Excluding the data in such a extremely light waters, we determined changes in the linear growth rate of $p\text{CO}_{2\text{sw}}$ of $+0.3 \pm 1.3 \mu\text{atm year}^{-1}$ for 1985 - 1990, $+2.2 \pm 0.7 \mu\text{atm year}^{-1}$ for 1990 - 1999, and $-0.2 \pm 1.0 \mu\text{atm year}^{-1}$ for 1999 – 2004. The rate of increase was $+1.5 \pm 0.2 \mu\text{atm year}^{-1}$ for the entire period (1985 - 2004). The variation in the rate of increase of $p\text{CO}_{2\text{sw}}$ is fairly consistent with the change in the rate of increase of salinity-normalized TCO_2 from $+2.1 \pm 0.4 \mu\text{mol kg}^{-1} \text{ year}^{-1}$ during 1992 - 1999 to $+0.4 \pm 0.9 \mu\text{mol kg}^{-1} \text{ year}^{-1}$ during 1999 - 2004. These changes are anti-correlated with the decadal variation in the geostrophic mass transport from subtropics of both hemispheres into the equatorial zone in the Pacific.

1. Introduction

The oceans play an important role in regulating the increase of carbon dioxide (CO₂) in the atmosphere through their ability to take up and release CO₂. The oceans have already absorbed 118 ± 19 PgC as CO₂ for the period from 1800 to 1994, about 50 % of CO₂ emitted by the fossil fuel burning during the same period (Sabine et al., 2004a) and are continuing to absorb CO₂ at an average rate of 1.4 to 1.8 PgC per year (Gloor et al., 2003; Gurney et al., 2004; Takahashi et al., 2002; 2008, this issue). Such a crucial function of the oceans should be monitored by long-term measurements of the CO₂ chemistry in the oceans. Moreover, observing long-term trends and interannual to decadal variability of CO₂ uptake are subjects of high priority in understanding the global carbon cycle and its changes associated with climate change and the atmospheric CO₂ increase.

Inoue et al. (1995) and Midorikawa et al. (2005) used data of regular winter-time observations collected since the early 1980's to show that the partial pressure of CO₂ in surface water ($p\text{CO}_{2\text{sw}}$) in the western North Pacific along 137°E meridian has increased at a similar rate to the partial pressure of CO₂ in the overlying atmosphere ($p\text{CO}_{2\text{air}}$) over the entire subtropical zone. The increase in the surface CO₂ as well as its variability in the subtropics has also been documented on the basis of the monthly or more frequent observations in the North Atlantic near Bermuda (Bates et al., 1996; Gruber et al., 2002) and in the central North Pacific near Hawaii (Dore et al., 2003; Keeling et al., 2004). At the latter site, a change in the rate of CO₂ increase has been ascribed to changes in the

circulation and regional pattern of precipitation that are possibly linked to each other. Inoue et al. (1999) also found that the rate of increase of $p\text{CO}_2\text{sw}$ in the western South Pacific is similar to that in the atmosphere. Recently Takahashi et al. (2006; 2008, this issue) concluded that the mean rate of increase of $p\text{CO}_2\text{sw}$ for the open ocean areas in the North Pacific is indistinguishable from the mean rate of increase of $p\text{CO}_2\text{air}$.

For the oceans at high latitude, it is particularly difficult to identify long-term trends in the oceanic CO_2 since the variability is much larger than in the subtropics. Nevertheless, several studies of the North Atlantic subpolar gyre show that $p\text{CO}_2\text{sw}$ is increasing faster than in the atmosphere (Lefevre et al., 2004; Omar and Olsen, 2006; Corbiere et al., 2007; Schuster and Watson, 2007; Takahashi et al., 2006; 2008, this issue). The processes controlling these changes include warming, changes in circulation and/or biological productivity, but their relative importance has not yet been clearly quantified. On the other hand, Inoue and Ishii (2005) analyzed the trend of $p\text{CO}_2\text{sw}$ in the Southern Ocean to the south of Australia in austral summer since late 1960s. They found similar rates of increase to $p\text{CO}_2\text{air}$ in the zones to the south of the Sub-Antarctic Front (SAF), but slower rates of increase to the north of SAF due to deep convection in winter. Metzl (2008, this issue) and Takahashi et al. (2008, this issue) also documented the increase of $p\text{CO}_2\text{sw}$ in other regions of the Southern Ocean. The rates of $p\text{CO}_2\text{sw}$ increase they reported are comparable to or larger than the rate of $p\text{CO}_2\text{air}$ increase.

The equatorial Pacific is also a zone that has attracted considerable attention in terms of the long-term trend as well as interannual variability in the oceanic CO₂ (Takahashi et al., 2003; Feely et al., 2006).

The surface layer of the equatorial Pacific consists of two major domains. In the eastern zone, upwelling and turbulent mixing driven by the easterly trade winds bring cold, saline and nutrient-CO₂-rich water of the Equatorial Undercurrent (EUC) to the surface. Accordingly, sea surface salinity (SSS) is higher ($S > 35$) and sea surface temperature (SST) is lower ($22 < T/^{\circ}\text{C} < 29$). There are appreciable amounts of macronutrients such as nitrate, phosphate and silicic acid, and chlorophyll concentrations exceed 0.3 mg m^{-3} in the surface layer (Le Borgne et al., 2002a; Matsumoto et al., 2004). Since total dissolved inorganic carbon (TCO₂) is high but total alkalinity (TA) remains relatively low, $p\text{CO}_{2\text{sw}}$ is high and thus CO₂ efflux to the atmosphere often exceeds $10 \text{ mmol m}^{-2} \text{ day}^{-1}$ (e.g., Ishii et al., 2004; Feely et al., 2006). Water upwelled near the equator diverges poleward and advects westward as a South Equatorial Current (SEC) while raising SST due to solar heat influx and reducing the concentrations of nutrients and TCO₂ due to biological production and CO₂ emission to the atmosphere (Sabine et al., 2004b). In the western zone, SSS is low ($S < 34.8$) and SST is high ($T > 29.5 \text{ }^{\circ}\text{C}$) because of high annual precipitation (You, 1998) and a large solar heat influx. As a result of the stratification thus attained, nitrate is completely depleted and chlorophyll levels are low ($< 0.1 \text{ mg m}^{-3}$). Since the difference in $p\text{CO}_{2\text{sw}}$ and $p\text{CO}_{2\text{air}}$ is small, the net air-sea

CO₂ flux is also small (< 3 mmol m⁻² day⁻¹; e.g., Ishii and Inoue, 1995). The narrow boundary region between these cold/saline water to the east and warm/fresh water to the west is usually recognized by the steep zonal gradient of SSS and *p*CO_{2sw} rather than SST (e.g., Inoue et al., 1996; Le Borgne et al., 2002b). However, the relationship between SSS-SST and between SSS-TCO₂ in the boundary region change with time, reflecting that the freshwater flux and heat flux are not fully coupled to each other and that the warm/fresh and cold/saline waters are mixing in this boundary region (Ishii et al., 2004).

The longitude of the boundary region between the warmer/fresher water to the west and cooler/more saline water to the east strongly fluctuates because of the El Niño Southern Oscillation (ENSO). As a result, the CO₂ flux from the entire equatorial Pacific varies significantly. The equatorial Pacific is the largest CO₂ source of the ocean with the annual mean of the net CO₂ efflux estimated to reach ca. +0.4 PgC (Cosca, et al., 2003; Feely et al., 2004 ; Ishii et al., 2004). In the long-term, the trend of the distribution of cooler/more saline water in the equatorial Pacific potentially exerts a significant effect on the net budget of the air-sea CO₂ exchange for the global oceans and their role in absorbing the anthropogenically emitted CO₂. If the rate of increase of *p*CO_{2sw} significantly differs from that in the atmosphere, the trend of *p*CO_{2sw} can also exert a significant effect on the net air-sea CO₂ exchange for the ocean.

Feely et al. (1999) showed that *p*CO_{2sw} is increasing at a similar rate to that of atmosphere in

the cold/saline upwelling region. They used data from 1961 (Keeling et al., 1965) as a benchmark and corrected for the spatial and interannual variations of $p\text{CO}_{2\text{sw}}$ using an empirical $p\text{CO}_{2\text{sw}} - \text{SST}$ relationship. Takahashi et al. (2003) examined the change in the rate of increase of $p\text{CO}_{2\text{sw}}$ around 1990 in the central ($170^\circ\text{W} - 120^\circ\text{W}$, $5^\circ\text{S} - 5^\circ\text{N}$) and western regions ($135^\circ\text{E} - 175^\circ\text{E}$, $5^\circ\text{S} - 5^\circ\text{N}$). They estimated that $p\text{CO}_{2\text{sw}}$, calculated to a constant temperature of 28°C to correct for solubility changes, has increased dramatically in the central region from $-0.3 \pm 1.7 \mu\text{atm year}^{-1}$ for the period 1979 to 1990 to $+3.2 \pm 0.6 \mu\text{atm year}^{-1}$ for the period 1990 to 2001. In the western region they reported a rate of change from $-1.0 \pm 0.3 \mu\text{atm year}^{-1}$ for 1979 to 1990 increasing to $+3.0 \pm 0.2 \mu\text{atm year}^{-1}$ for 1990 to 2001. They ascribe these decadal scale changes to the increased upwelling due to the regime shift referred to as the Pacific Decadal Oscillation (Hare and Mantua, 2000). Feely et al. (2006) also examined $p\text{CO}_{2\text{sw}}$ increase in the same geographical regions as Takahashi (2003) using a longer record extending to 2004. They detected that temperature-normalized $p\text{CO}_{2\text{sw}}$ decreased before 1990 and increased at a similar rate to $p\text{CO}_{2\text{air}}$ after 1990. Furthermore, Inoue et al. (2006) analyzed the trend of $p\text{CO}_{2\text{sw}}$ in the western equatorial Pacific ($140^\circ\text{E} - 160^\circ\text{W}$) using a multi-linear regression method based on SST, SSS and concentration of nitrate. They found that $p\text{CO}_{2\text{sw}}$ is increasing at a rate of $+1.3 \pm 0.3 \mu\text{atm year}^{-1}$ in the western warm/fresh water and $+1.4 \pm 0.5 \mu\text{atm year}^{-1}$ in the upwelling zone for the period from 1987 to 2003.

In the present study we expand on the previous work to describe the change in the oceanic

CO₂ in the western equatorial Pacific. We emphasize the time-trend of $p\text{CO}_2\text{sw}$ since the mid 1980's in the spatially fluctuating warm/fresh water, rather than in a fixed geographical zone. The trend of surface TCO₂ since the early 1990's is also analyzed to validate and attribute the $p\text{CO}_2\text{sw}$ trend. Large spatial variability that hinders precise time-trend analysis was reduced by using salinity data to distinguish between the warm/fresh water and cold/saline water that is influenced more by equatorial upwelling. We often observed mesoscale TCO₂ drawdown, possibly the result of biological activity associated with nitrogen fixation in the warm/fresh surface water above barrier layers. Since it is also a source of pronounced spatial variability and not intrinsic to the time-trend that relates to the anthropogenic CO₂ accumulation and circulation change, its effect was removed.

In the following sections we used concurrent measurements of $p\text{CO}_2\text{sw}$ and TCO₂ along the equator to describe zonal variations of surface CO₂ system and discuss their controlling factors. Then time-trends of $p\text{CO}_2\text{sw}$ and TCO₂ in the western equatorial Pacific including those at better-sampled sites and in the broad area of warm/fresh water over the last two decades are discussed.

2. Data sources

We use data of $p\text{CO}_2\text{sw}$ in the western equatorial Pacific collected during 104 cruises spanning the years from 1983 to 2004, and TCO₂ data collected during 30 cruises from 1990 to 2004 (Figure 1

and Table 1) for the equatorial zone (5°S – 5°N) between 144°E and 160°W. The longitude boundaries were chosen because the variation of $p\text{CO}_{2\text{sw}}$ to the west of 143°E is affected by the coastal biological activity in the New Guinea Coastal Current, while the western equatorial Pacific warm/fresh water spreads eastward across the 160°W meridian during strong ENSO warm periods (Inoue et al., 2001). Most $p\text{CO}_{2\text{sw}}$ data were acquired along meridians until 1993 and cruises with more zonal coverage and surface TCO_2 measurements occurred after that.

Underway measurements of $p\text{CO}_{2\text{sw}}$ and TCO_2 were made using shipboard pumping systems that continuously pumped seawater from the ship's sea chest. For $p\text{CO}_{2\text{sw}}$ measurements, gas equilibration methods with shower-head type equilibrators equipped with a non-dispersive infrared gas analyzer or gas chromatograph detection were used. For the total of 55 cruises by the RVs Natsushima, Ryofu Maru, Hakuho Maru, Hakurei Maru, Kaiyo, Mirai, Keifu Maru, analytical systems were similar design (JMA/MRI-system) (Inoue, 2000). Comparability of data obtained by this system with those obtained by other systems has been examined during an intercomparison exercise made in the North Atlantic on board the RV Meteor (Körtzinger et al., 1999; 2000). The data of $p\text{CO}_{2\text{sw}}$ from JMA/MRI-system were in good agreement ($< \pm 2 \mu\text{atm}$) with those from a system that has been used in FLUPAC cruise of the RV L' Atalante (Poisson et al., 1993) (see Table 1) for a wide range of $p\text{CO}_{2\text{sw}}$. Analytical systems of the type described by Wanninkhof and Thoning (1993), Wong et al. (1993), Sabine and Key (1997) and Feely et al (1998) were used for the

other 48 cruises and various authors also reported a measurement precision typically better than ± 2 μatm .

All reference standards used onboard underwent pre- and post-cruise calibration against standards traceable to the primary standards of the World Meteorological Organization. Partial pressure of CO_2 ($p\text{CO}_2$) was calculated from the dry mole fraction of CO_2 ($x\text{CO}_2$) measured in the equilibrator headspace using the saturated-water vapor pressure (p_{sw}) equation of Weiss and Price (1980) and the atmospheric pressure (P):

$$p\text{CO}_2 = x\text{CO}_2 \cdot (P - p_{\text{sw}}) \quad (1)$$

The $p\text{CO}_2$ values were corrected for warming between the seawater intake and equilibrator after Copin-Montégut (1988; 1989). For those dataset that report fugacity of CO_2 in surface water ($f\text{CO}_{2\text{sw}}$) but not $x\text{CO}_2$ in the equilibrator headspace, $p\text{CO}_{2\text{sw}}$ was calculated from $f\text{CO}_{2\text{sw}}$ and fugacity coefficient (Dickson et al., 2007). All $p\text{CO}_2$ values were then recalculated to values at $P = 1$ atm.

The method of TCO_2 measurements has been described in Ishii et al. (1998) and were made coulometrically (Johnson et al., 1985). Precision of TCO_2 analysis within a single cruise is typically better than $\pm 2 \mu\text{mol kg}^{-1}$. Certified Reference Material, provided by Dr A. G. Dickson (http://www.mpl.ucsd.edu/people/adickson/CO2_QC/index.html), was analyzed routinely and indicated a precision of measurements similar to the level of $\pm 2 \mu\text{mol kg}^{-1}$.

In the following section, total alkalinity (TA) was computed from $p\text{CO}_{2\text{sw}}$, TCO_2 , temperature and salinity using the dissociation constants of carbonic acid given by Lueker et al. (2000).

3. Spatial variability

In spite of the high SST ($T > 29$ °C), surface seawater in the western equatorial Pacific warm/fresh water is usually moderately supersaturated with respect to atmospheric CO_2 [$0 < \Delta p\text{CO}_2 / \mu\text{atm} < +40$; $\Delta p\text{CO}_2 = p\text{CO}_{2\text{sw}} - p\text{CO}_{2\text{air}}$]. Concurrent measurements of $p\text{CO}_{2\text{sw}}$, TCO_2 , SST and SSS that have been made in 25 cruises in the western equatorial Pacific (Table 1) showed that the moderate spatial variability of $p\text{CO}_{2\text{sw}}$ in the warm/fresh water is influenced by the fluctuations of SST and SSS as well as that of NTCO_2 . The modest oversaturations at such high surface temperature appear to result from both low salinity ($S < 34.8$) and salinity-normalized TCO_2 ($\text{NTCO}_2 = \text{TCO}_2 \cdot S_n / S$; about $1950 \mu\text{mol kg}^{-1}$ at $S_n = 35$).

A typical case in which the fluctuations of SST and SSS were playing a major role in the variation of $p\text{CO}_{2\text{sw}}$ is that in the 1997/98 strong ENSO warm period we observed in December 1997 during the RV Kaiyo's 9714 cruise to the west of 180° and in February 1998 during the RV Mirai's 9801 cruise to the east of 179°E (Fig.2). In this period, $p\text{CO}_{2\text{sw}}$ decreased to the east from 377 ± 4 (1s) μatm between 144°E and 155°E to 354 ± 3 μatm between 175°W and 165°W , whereas spatial variations of NTCO_2 ($1948 \pm 3 \mu\text{mol kg}^{-1}$ at $S = 35$) and salinity-normalized TA

(NTA) ($2294 \pm 2 \mu\text{mol kg}^{-1}$ at $S = 35$) calculated were small over these regions. Apart from the small increase of $p\text{CO}_{2\text{sw}}$ due to the small increase of NTCO_2 (*ca.* $+4 \mu\text{mol kg}^{-1}$ on the average) between 175°E and 180° in December, the eastward decrease of $p\text{CO}_{2\text{sw}}$ was mainly attributable to the variation of SST and SSS that also decreased to the east by about 0.7°C and 0.46 , respectively ($29.9 \pm 0.2^\circ\text{C}$ to $29.2 \pm 0.2^\circ\text{C}$ and 34.66 ± 0.10 to 34.20 ± 0.08).

We also identified significant spatial variability of NTCO_2 of about 10 or $20 \mu\text{mol kg}^{-1}$ and a resultant $p\text{CO}_{2\text{sw}}$ variability in all other cruises in which we surveyed a broad expanse of warm/fresh waters. For example, in January 2003, surface NTCO_2 showed little spatial variation over the region between 145°E and 179°E ($1949.0 \pm 1.9 \mu\text{mol kg}^{-1}$), but was significantly lower in the region between 179°E and 173°W ($1940.1 \pm 2.5 \mu\text{mol kg}^{-1}$) (Fig.3). Consequently, $p\text{CO}_{2\text{sw}}$ decreased to the level slightly below that of $p\text{CO}_{2\text{air}}$ in the more easterly region. The region of low NTCO_2 and $p\text{CO}_{2\text{sw}}$ was located in the eastern limb of the warm/fresh water, just to the west of upwelling region where SSS is higher ($S > 34.8$). Another noticeable example of NTCO_2 drawdown in the warm/fresh water was observed in January 2002. A drawdown of surface NTCO_2 was seen broadly between 152°E and 170°E , and surface water between 160°E and 170°E was in near equilibrium with atmospheric CO_2 .

We found low NTCO_2 and a resultant low $p\text{CO}_{2\text{sw}}$ in surface waters with extremely low density ($\sigma_t < 21.4$) due to high-temperature and low-salinity. The low density surface waters are

associated with the formation of the thick barrier layers (Fig.4). The barrier layer forms in the warm water under a fresh surface water. In the barrier layer, the temperature is homogeneous, but salinity increases with depth, thus forming a halocline within the surface isothermal layer (Ando and McPhaden, 1997). It is formed either by the input of heavy rainfall due to the strong atmospheric convection in the western equatorial Pacific, or by the subduction of the cold/saline water of the central equatorial Pacific below the warm/fresh surface waters of the western equatorial Pacific. This insulates the surface shallow and warm mixed-layer from cooler and relatively nutrient and CO₂ rich water below the thermocline (Lukas and Lindstrom, 1991). The barrier layer induces a high near-surface temperature anomaly, and a high correlation between the barrier layer thickness and the near-surface temperature is observed in the western equatorial Pacific (Fujii and Kamachi, 2003). In January 2003, thick barrier layers with a low salinity cap were found between 178°E and 173°W at depths between 50m and 100m (Fig.4a); in January 2002, it was observed between 152°E and 172°E between 30m and 100m (Fig.4b).

Drawdown of NTCO₂ in the extremely light ($\sigma_t < 21.4$) and stratified surface waters within and above these barrier layers is accompanied by the drawdown of phosphate (Fig.5). Nitrate is completely depleted within the isothermal layer, suggesting net CO₂ uptake associated with nitrogen fixation, may be responsible for the drawdown of NTCO₂ above thick barrier layers. Midorikawa et al. (2006) also demonstrated that, in the far western equatorial zone at 137°E (3°N - 6°N), the TCO₂ drawdown above barrier layers is responsible for the negative TCO₂ anomaly observed in the ENSO cold periods when thick barrier layers tend to form at that region. Thus, in addition to the

fluctuations of SST and SSS, TCO_2 drawdown due probably to biological CO_2 uptake above thick barrier layer is a factor that is often affecting the spatial variation in $p\text{CO}_{2\text{sw}}$ in the western equatorial Pacific warm/fresh waters.

The thermocline/nitracline in the western equatorial Pacific becomes shallower in the ENSO warm periods and shoaling raises the rate of new production dramatically below 40m (Turk et al., 2001). The same shoaling may affect the CO_2 chemistry in the warm/fresh water in the western equatorial Pacific since TCO_2 also changes markedly across the thermocline/nitracline (see Fig. 5). However, no significant correlation has been found between surface NTCO_2 and thermocline/nitracline depth, and it is unclear whether these processes significantly change the surface CO_2 chemistry.

4. Time-trends

4.1. Interannual variability in the western equatorial Pacific

The $p\text{CO}_{2\text{sw}}$ in the equatorial Pacific shows large variability in space and time associated with the ENSO warm and cold events (e.g., Inoue and Sugimura, 1992; Feely et al., 1999; Feely et al., 2002; Ishii et al., 2004). The variability in the net CO_2 flux from the entire equatorial Pacific is considered to be the major origin of the interannual variability in the budget of oceanic sink for anthropogenic CO_2 (Le Quéré et al., 2000; Obata and Kitamura, 2002; Peylin et al., 2005; Park et al., 2006).

A large CO₂ efflux from the surface ocean during the ENSO cold periods results from the spread of cold/saline CO₂-rich surface water to the west. Even at 165°E in the western equatorial Pacific, surface water exhibited high supersaturation with respect to CO₂ ($\Delta p\text{CO}_2 > 70 \mu\text{atm}$) during the cold periods in 1989, 1995-1996, 1998 – 2000 and 2000-2001. In these periods, NTCO₂ at $S = 35$ was higher than $1970 \mu\text{mol kg}^{-1}$, SSS was higher than 35.0, and SST was lower than 28.5 °C (Fig.6). For normal and warm periods, the 165°E site typically showed NTCO₂ at $S = 35$ reduced to below $1960 \mu\text{mol kg}^{-1}$, SSS below 34.8, and SST above 29.0 °C. Consequently, $\Delta p\text{CO}_2$ fell below +40 μatm and even dropped to near 0 μatm when surface water was extremely light ($\sigma_t < 21.4$) due to warm ($T > 30.0$ °C) or fresh ($S < 34.2$) conditions described in section 3.

Surface waters were more frequently and more strongly affected by the equatorial upwelling in the central region at 180°, compared to 165°E (Fig.7). The range of variation in SST was large ($26.0 < T / ^\circ\text{C} < 30.5$). NTCO₂ at $S = 35$ often exceeded $2000 \mu\text{mol kg}^{-1}$ and SSS exceeded 35.0 in cold periods. For warm periods in 1987, 1994, 1997-1998, 2002-2004, warm/fresh water spread eastward across 180° and SSS and NTCO₂ dropped to below 34.8 and $1960 \mu\text{mol kg}^{-1}$, respectively, and $\Delta p\text{CO}_2$ fell below +30 μatm . Extremely light surface waters in which $\Delta p\text{CO}_2$ was close to +0 μatm have also been observed in the warm periods in late 1994 and in early 2003. It appears that the range of variation in the hydrographic and CO₂ properties in the warm/fresh water observed at 165°E and 180° are similar. This is also the case for all the warm/fresh waters observed periodically in the entire

zone of investigation.

The variability of $p\text{CO}_{2\text{sw}}$ and NTCO_2 in the equatorial Pacific (135°E - 95°W, 10°S - 5°N) can be approximated using multiple regression equations as a function of SST, SSS and the year of observation. These equations have been revised on the basis of the same data set as described in Ishii et al. (2004) (Nakadate and Ishii, 2007). We applied different equations for warm/fresh waters and for cold/saline waters to approximate NTCO_2 . Equation 2 applies to the west of 150°E and for the region where $S \leq 34.8$ to the east of 150°E that represent warm/fresh waters. Equation 3 is for other regions where $S > 34.8$ to the east of 150°E that represents cold/saline waters in the divergence zone.

Warm/fresh water:

$$\text{ntco}_2 = -0.118 - 0.776 t + 0.021 s + 0.107 t^2 - 0.003 ts - 0.008 s^2 + 0.012 y \quad (2)$$

Cold/saline water :

$$\begin{aligned} \text{ntco}_2 = & 0.378 - 0.790 t + 0.164 s - 0.136 t^2 + 0.305 ts - 0.457 s^2 + 0.016 t^3 \\ & - 0.190 t^2s + 0.319 ts^2 - 0.007 s^3 + 0.014 y \end{aligned} \quad (3)$$

$$\text{NTCO}_2 / \mu\text{mol kg}^{-1} = \text{ntco}_2 \cdot 50 + 2000 \quad (4)$$

where $t = (\text{SST}/^\circ\text{C} - 25) / 3$, $s = (\text{SSS} - 35) / 0.5$ and $y = \text{year} - 2000.0$.

Calculated NTA at $S = 35$ is also empirically expressed as Eq(5) and (6) for all the zone:

$$\text{nta} = 6.046 - 0.105 t + 0.016 t^2 \quad (5)$$

$$\text{NTA} / \mu\text{mol kg}^{-1} = \text{nta} \cdot 50 + 2000 \quad (6)$$

The regression errors ($\pm 1\sigma$) are $\pm 6.3 \mu\text{mol kg}^{-1}$ for the NTCO_2 in the warm/fresh water, $\pm 6.8 \mu\text{mol}$

kg^{-1} for the NTCO_2 in the cold/saline water, and $\pm 5.1 \mu\text{mol kg}^{-1}$ for NTA. The above equations are used with the time – space fields of SST and SSS obtained by objective analysis (Ishii et al. 2005) and in situ measurements (Delcroix et al., 2001), respectively, and those obtained by data-assimilation (Usui et al., 2006). The dissociation constants of carbonic acid (Lueker et al. 2000) and solubility of CO_2 in seawater (Weiss, 1974) were used with the NTA, NTCO_2 , SST and SSS value to compute the fields of $p\text{CO}_{2\text{sw}}$.

4.2. Decadal trend in the warm/fresh water

In order to analyze the time-trend of $p\text{CO}_{2\text{sw}}$ and TCO_2 in the nitrate-depleted warm/fresh waters, we first removed the immediate effect of equatorial upwelling that supplies nutrient- CO_2 -rich water to the surface by focusing on data with $T \geq 29.0 \text{ }^\circ\text{C}$ and $S \leq 34.8$. The trends of $p\text{CO}_{2\text{sw}}$ at in-situ temperature and salinity, $p\text{CO}_{2\text{sw}}$ normalized at $T = 29.5 \text{ }^\circ\text{C}$ and $S = 34.5$, TCO_2 normalized at $S = 34.5$ (instead of $S = 35$ in the previous sections), SST, SSS, and the density in these warm/fresh waters are shown in Fig. 8. For most cruises, both extremely light water with $\sigma_t < 21.4$ and moderately light water with $\sigma_t \geq 21.4$ were observed. The $p\text{CO}_{2\text{sw}}$ at in-situ T and S , T - S -normalized $p\text{CO}_{2\text{sw}}$, and NTCO_2 in the extremely light waters were significantly lower than those in the moderately light waters.

Next, we eliminated the data for those extremely light waters with $\sigma_t < 21.4$ (green dots). We

consider the drawdown of CO₂ most likely due to biological activity in the extremely light surface waters, to represent noise which complicates the interpretation of surface CO₂ time-trend in terms of anthropogenic CO₂ accumulation and ocean circulation change. The remaining data are unevenly distributed in space and time and to minimize the effect of small scale variability, we have analyzed the time trends using the mean value for each cruise (Fig. 8; blue open square).

The linear regression analysis for the trends of $p\text{CO}_{2\text{sw}}$ and normalized $p\text{CO}_{2\text{sw}}$ revealed an increase at a mean rate of $+1.4 \pm 0.2$ and $+1.2 \pm 0.2 \mu\text{atm year}^{-1}$, respectively, over two decades from 1985 to 2004 (Table 2, a). The increase of surface CO₂ in the warm/fresh water is also seen in the trend of NTCO₂ that is increasing at a rate of $+0.8 \pm 0.3 \mu\text{mol kg}^{-1} \text{ year}^{-1}$ in the period from 1992 to 2004 (Table 3, a). The increase rate of normalized $p\text{CO}_{2\text{sw}}$ of $+1.2 \pm 0.2 \mu\text{atm year}^{-1}$ is slightly below that of $p\text{CO}_{2\text{air}}$ measured concurrently on-board during most cruises of $+1.52 \pm 0.03 \mu\text{atm year}^{-1}$ and an increase of $+1.56 \pm 0.02 \mu\text{atm year}^{-1}$ calculated from atmospheric data collected during the same period at Christmas Island (1.70°N, 157.17°W) in the equatorial Pacific (Tans and Conway, 2005). The slower rate of increase in the surface waters relative to the atmosphere is in part due to low normalized $p\text{CO}_{2\text{sw}}$ ($< 360 \mu\text{atm}$) and NTCO₂ ($< 1915 \mu\text{mol kg}^{-1}$) observed in the several cruises after mid-1990's to the west of 166°E and south of 1°S. The region where low surface CO₂ was observed is the region where South Equatorial Counter Current (SECC) flows to the east (Fine et al., 1994). We believe this region is likely to reduce the rates of increase

due to the influence of coastal water transported from off of New Guinea by the SECC, like the coastal influence in the region to the west of 143°E.

By eliminating the data in this region as a third step, we obtained the linear rate of increase of normalized $p\text{CO}_{2\text{sw}}$ and NTCO_2 of $+1.4 \pm 0.2 \mu\text{atm year}^{-1}$ and $+1.3 \pm 0.3 \mu\text{mol kg}^{-1} \text{ year}^{-1}$, respectively. These rates of increase are in good agreement with those in the further confined equatorial zone between 1°S and 1°N from 144°E to 160°W; $+1.4 \pm 0.3 \mu\text{atm year}^{-1}$ and $+1.3 \pm 0.3 \mu\text{mol kg}^{-1} \text{ year}^{-1}$. In the following sections, we discuss the trend of surface CO_2 in the warm/fresh waters in the zone of 144°E – 160°W and 5°S – 5°N, but excluding the low CO_2 region of 144°E – 166°E and 5°S – 1°S.

4.3. Sensitivity of the trend to the threshold values delineating the warm/fresh waters

Large spatial variations in $p\text{CO}_{2\text{sw}}$ and normalized $p\text{CO}_{2\text{sw}}$ of over $30 \mu\text{atm}$ are observed for some cruises, *e.g.*, in January 1994 in the region between 145°E and 176°W. A sharp peak is also apparent in September – November 1994. This peak consists of data from 3 cruises conducted by different organizations over the region between 160°E to 170°W. All these data were collected in the warm/fresh waters where SST and SSS are close to the threshold values delineating the warm/fresh waters, $T = 29 \text{ }^\circ\text{C}$ and $S = 34.8$. The occasional large spatial variation and a sharp peak of surface CO_2 suggest the selection criteria ($T \geq 29 \text{ }^\circ\text{C}$, $S \leq 34.8$, $\sigma_t \geq 21.4$) do not completely eliminate the

influence of equatorial upwelling of nutrient and CO₂-rich water, or eliminate the presumed effect of nitrogen fixation that reduces surface CO₂. Therefore, it is important to examine if the rate of increase of surface CO₂ is sensitive to the choice of threshold values delineating the warm/fresh waters from nutrient and CO₂ rich upwelled waters and from extremely light ($\sigma_t < 21.4$) surface waters within and above barrier layers.

First, we changed the criteria to $T \geq 29.5$ °C and $S \leq 34.65$ while the threshold for density remains $\sigma_t \geq 21.4$ in order to focus on the warmer/fresher waters and exclude more water potentially influenced by equatorial upwelling (Table 2 and 3; d). With this change, the total number of data points reduced from 5,691 in 72 cruises to 1,302 in 49 cruises for $p\text{CO}_{2\text{sw}}$. For TCO_2 , the total number of data points reduced from 878 in 21 cruises to 214 in 13 cruises. A sharp peak in $p\text{CO}_{2\text{sw}}$ in late 1994 also disappeared. As a result, the linear trend of $p\text{CO}_{2\text{sw}}$ and normalized $p\text{CO}_{2\text{sw}}$ for 1985 to 2004 decreased from $+1.5 \pm 0.2 \mu\text{atm year}^{-1}$ to $+1.3 \pm 0.3 \mu\text{atm year}^{-1}$, and $+1.4 \pm 0.2 \mu\text{atm year}^{-1}$ to $+1.2 \pm 0.2 \mu\text{atm year}^{-1}$, respectively. The linear trend of NTCO_2 for 1992 to 2004 also decreased from $+1.3 \pm 0.3 \mu\text{mol kg}^{-1} \text{ year}^{-1}$ to $0.8 \pm 0.4 \mu\text{mol kg}^{-1} \text{ year}^{-1}$. The “student’s t -test” indicates that these changes are significant at 95% confidence intervals.

Secondly, we changed the criteria to $\sigma_t \geq 21.5$ while keeping the threshold for temperature and salinity at $T \geq 29.0$ °C and $S \leq 34.8$ to exclude more light waters that could be influenced by TCO_2 drawdown above the barrier layer (Table 2 and 3; e). With this change, the total number of data

points reduced from 5,691 in 72 cruises to 4,076 in 61 cruises for $p\text{CO}_{2\text{sw}}$, and 878 in 21 cruises to 576 in 15 cruises for TCO_2 . With this new threshold for density, the linear trend of $p\text{CO}_{2\text{sw}}$ and normalized $p\text{CO}_{2\text{sw}}$ for 1985-2004 did not change, but that of NTCO_2 for 1992-2004 decreased from $+1.3 \pm 0.3 \mu\text{mol kg}^{-1} \text{ year}^{-1}$ to $0.7 \pm 0.3 \mu\text{mol kg}^{-1} \text{ year}^{-1}$. The “student’s t-test” indicates that change in the rate of increase of NTCO_2 is significant at the 95% confidence level.

It is also worth examining how rates of increase change when no threshold is assumed for density, i.e., rates of increase are determined by allowing large spatial variability (Table 2 and 3; f.). Under this criterion, a much larger number of data are available; 17,035 data points in 85 cruises for $p\text{CO}_{2\text{sw}}$ and 2,534 data points in 28 cruises for TCO_2 . In spite of the larger variability in space (Fig.9), the rate of increase of $p\text{CO}_{2\text{sw}}$ and that of normalized $p\text{CO}_{2\text{sw}}$ did not change ($+1.5 \pm 0.2 \mu\text{atm year}^{-1}$ and $+1.4 \pm 0.2 \mu\text{atm year}^{-1}$). By contrast, NTCO_2 showed a significant decrease from $+1.3 \pm 0.3$ to $+0.7 \pm 0.2 \mu\text{mol kg}^{-1} \text{ year}^{-1}$.

These sensitivity tests show the rates of increase vary between $+1.3$ and $+1.6 \mu\text{atm year}^{-1}$ for $p\text{CO}_{2\text{sw}}$, between $+1.2$ and $+1.4 \mu\text{atm year}^{-1}$ for normalized $p\text{CO}_{2\text{sw}}$, and $+0.7$ and $+1.3 \mu\text{mol kg}^{-1} \text{ year}^{-1}$ for NTCO_2 depending on the choice of criteria that delineate the warm/fresh waters. The greater uncertainty in the rate of increase for NTCO_2 compared to $p\text{CO}_{2\text{sw}}$ appears in part due to a much shorter record and fewer data.

4.4. Decadal variability in the rates of increase

The rate of change in $p\text{CO}_{2\text{sw}}$ and normalized $p\text{CO}_{2\text{sw}}$ appear to have increased around 1990 (Table 1; Takahashi et al. 2003 and Feely et al. 2006). A broad peak was also observed to occur around 2000 in $p\text{CO}_{2\text{sw}}$, normalized $p\text{CO}_{2\text{sw}}$ and NTCO_2 (Figure 9). It is likely that the increase in $p\text{CO}_{2\text{sw}}$ in the western equatorial Pacific warm/fresh water slowed in late 1990s and that surface water is now changing toward a state of under-saturation with respect to atmospheric CO_2 (Fig.9). These changes are apparently coincident with the decadal change in the transport convergence of the shallow meridional overturning circulation into the equatorial zone of the Pacific reported by McPhaden and Zhang (2002; 2004).

The rate of increase of $p\text{CO}_{2\text{sw}}$ and normalized $p\text{CO}_{2\text{sw}}$ during 1985-1990 were estimated as ranging between -0.1 and $+1.3 \mu\text{atm year}^{-1}$ and between $+0.3$ and $+1.7 \mu\text{atm year}^{-1}$ depending on the criteria for warm/fresh water chosen (Table 2; b, d, e and f). The rate of increase of normalized $p\text{CO}_{2\text{sw}}$ for the pre-1990 period is larger in this study than estimates from Takahashi et al (2003) ($-1.0 \pm 0.3 \mu\text{atm year}^{-1}$) and Feely et al. (2006) ($-0.93 \pm 0.83 \mu\text{atm year}^{-1}$). These authors used all data from west of 175°E without applying selection criteria to exclude the influence of the equatorial upwelling and low density surface layers. The results from our study are more likely to be representative of changes in the warm and low salinity pool of water that typically occupies the western equatorial Pacific.

For the period 1990-1999, the rate of increase of $p\text{CO}_2\text{sw}$ ($+2.0$ to $+2.2 \mu\text{atm year}^{-1}$) and of normalized $p\text{CO}_2\text{sw}$ ($+2.0$ to $+2.1 \mu\text{atm year}^{-1}$) were significantly larger than in pre-1990 period. The results for our study are consistent with estimates from Takahashi et al.(2003) ($+3.4 \pm 0.4 \mu\text{atm year}^{-1}$ and $+3.0 \pm 0.2 \mu\text{atm year}^{-1}$) and Feely et al.(2006) ($+2.3 \pm 0.5 \mu\text{atm year}^{-1}$ and $+1.6 \pm 0.6 \mu\text{atm year}^{-1}$) for the post-1990 period in that the increase of $p\text{CO}_2\text{sw}$ and normalized $p\text{CO}_2\text{sw}$ were faster than that of the atmospheric $p\text{CO}_2$ ($+1.5 \mu\text{atm year}^{-1}$). The rate of increase of NTCO_2 , $+1.4$ to $+2.1 \mu\text{mol kg}^{-1} \text{ year}^{-1}$, during 1992-1999 was also large (Table 3; b, d, e and f). However, decreases to values from -0.3 to $-0.1 \mu\text{atm year}^{-1}$ for $p\text{CO}_2\text{sw}$, -0.5 to $+0.0 \mu\text{atm year}^{-1}$ for normalized $p\text{CO}_2\text{sw}$ and -1.3 to $+0.4 \mu\text{mol kg}^{-1} \text{ year}^{-1}$ for NTCO_2 occurred after 1999. Since the period of records after 1999 is only 6 years, the rates of increase depend on the selection of criteria, but are distinctly slower than the rates for the preceding period from 1990 to 1999. The decadal variability indicates that rate of increase will be influenced by the period of record, which may in part explain the slightly slower rate of increase of $p\text{CO}_2\text{sw}$ and normalized $p\text{CO}_2\text{sw}$ than that of $p\text{CO}_2\text{air}$ for the entire period we investigate.

5. Discussion

Atmospheric $p\text{CO}_2$ increased at a rate of $+1.56 \mu\text{atm year}^{-1}$ (or $+0.43 \% \text{ year}^{-1}$) on average during the last two decades (Tans, 2008; WDCGG, 2008). Since the air-to-sea gross CO_2 flux is

proportional to $p\text{CO}_2^{\text{air}}$, it implies that the gross CO_2 flux from the atmosphere to the surface ocean would have increased at $+0.43 \text{ \% year}^{-1}$ given no other changes. In the western equatorial Pacific, the monthly mean surface wind speed from NCEP-NCAR reanalysis (Kistler et al., 2001) is low ($4.0 \pm 1.1 \text{ m sec}^{-1}$ in 1990s) and the gas exchange coefficient is no more than $0.05 \text{ mmol m}^{-2} \text{ day}^{-1} \mu\text{atm}^{-1}$ when computed using the piston velocity of CO_2 based on long-term averaged wind speeds (Wanninkhof, 1992). Therefore, in a simple two box model consisting of the atmosphere and ocean surface mixed-layer with a depth of 60m, a $+1.56 \mu\text{atm year}^{-1}$ increase in $p\text{CO}_2^{\text{air}}$ would result in the increase in $p\text{CO}_2^{\text{sw}}$ and TCO_2 at a rate no more than $+0.9 \mu\text{atm year}^{-1}$ and $+0.5 \mu\text{mol kg}^{-1} \text{ year}^{-1}$, respectively. These rates are significantly smaller than those evaluated in this study (Table 2 and 3), and suggest that transport of anthropogenic CO_2 within the ocean is also playing an important role for the oceanic CO_2 increase in the warm/fresh waters in the western equatorial Pacific.

Upper water of the equatorial Pacific are considered to have its origin in the subduction regions in the subtropics and subtropical – sub(ant)arctic transition zones. These waters are transported to the equatorial zone via the subtropical shallow meridional overturning circulation. The pathways consist of subduction, equatorward advection in the subtropical gyre, branching off into the equatorward low-latitude western boundary currents or interior ocean meridional geostrophic transport, converging into the EUC that flows eastward across the equatorial Pacific, and supplying the cold/saline nutrient and CO_2 -rich water to the surface layer of the equatorial Pacific. The

upwelled nutrients support biological drawdown of CO₂ and the surface waters freshen and warm as they are advected westward and poleward in a shallow Ekman layer (McCreary and Lu, 1994; Fine et al., 1994, Rodgers et al., 2003). Therefore, it is expected that any CO₂ increase in the waters that subduct in the subtropics and subtropical – sub(ant)arctic transition zones and changes in the shallow meridional overturning circulation would affect the oceanic CO₂ increase in the equatorial Pacific. McPhaden and Zhang (2002; 2004) reported that equatorward geostrophic transport convergence across 9°S and 9°N has slowed down from $20.5 \pm 1.6 \text{ Sv}$ ($\times 10^6 \text{ m}^3 \text{ s}^{-1}$) during 1980 – 1989 to $14.0 \pm 1.5 \text{ Sv}$ during 1990 – 1999 or $13.4 \pm 1.6 \text{ Sv}$ during 1992 – 1998, but rebounded to $24.1 \pm 1.8 \text{ Sv}$ during 1998-2003. The transport changes accompany the positive and negative SST anomalies in the eastern equatorial Pacific. Izumo (2005) showed that mass transports of equatorward convergence, EUC, upwelling, surface divergence, and SEC strongly covaried on the interannual time scale of ENSO and correlated with the eastern equatorial Pacific SST. The higher rates of increase of $p\text{CO}_{2\text{sw}}$, normalized $p\text{CO}_{2\text{sw}}$ and NTCO_2 presented in this study are anti-correlated with the decadal variation of the equatorward geostrophic transport described in McPhaden and Zhang (2002, 2004). Greater surface CO₂ rates of increase occurred during the slow transport period of the 1990s. Low CO₂ rates of increase occurred before 1990 and after 1999 during periods of greater transport. The 5-year mean SST in the western equatorial Pacific also shows a positive anomaly during 1988-1997 and a negative anomaly during 1998 - 2002 (Fig.10), consistent with a westward spread of the

cool/saline waters in the mid-1990s.

We have no compelling explanations at present for this decadal scale anti-correlation between the rate of increase of surface CO₂ in the western equatorial Pacific warm/fresh waters and the equatorward geostrophic mass transport. It is expected that the slowdown of the meridional circulation would reduce the transport of anthropogenic CO₂ from the subduction regions to the equatorial zone. Thus, the rate of CO₂ increase in the EUC and in the branches of the subtropical shallow meridional overturning cell might be expected to slowdown. However, since a water mass would receive more rain of organic matter, the slowdown of the circulation may result in more subsurface remineralization and higher CO₂ and nutrient concentrations, in the water transported and upwelled in the equatorial zone.

Another possibility relates to the net community production and CO₂ outflux to the atmosphere in the divergence zone to the east of the warm/fresh waters. LeBorgne et al. (2002b) reported that marked zonal variations have not been seen in the divergence zone for the biogeochemical parameters such as the integrated chl.*a*, phytoplankton characteristics, plankton biomass, ¹⁴C uptake rate, and sinking flux of particulate organic carbon. They also found no strong indications of ENSO-related variability on these biogeochemical parameters within the divergence zone. On the other hand, it is well known that the ENSO-related variability has a significant effect on $\Delta p\text{CO}_2$ and CO₂ outflux in the equatorial divergence zone with larger $\Delta p\text{CO}_2$ and larger CO₂

outflux during the ENSO cold period and vice versa (Inoue et al., 1992; Feely et al., 2002). Therefore, during the slow-transport decade of the 1990's, the ENSO warm events occur more frequently, and upwelled water will lose less CO₂ to the atmosphere during the poleward divergence and westward advection in the SEC. This could result in a larger CO₂ content in the surface water of the western equatorial Pacific after complete nitrate depletion through biological uptake. In contrast, during the period of greater transport, ENSO cold events occur more frequently and upwelled water loses more CO₂ to the atmosphere resulting in a smaller CO₂ content in the surface water of the western equatorial Pacific. However, this hypothesis is not supported for the interannual time-scale where correlation between normalized $p\text{CO}_{2\text{sw}}$ or NTCO_2 in the warm/fresh water and westward spread of the cold/saline water is not seen.

The present study has revealed significant decadal variability in $\Delta p\text{CO}_2$, i.e., the state of CO₂ supersaturation, in the western equatorial Pacific warm/fresh water pool. The study shows the importance of continued monitoring of surface CO₂ in the equatorial Pacific in order to understand the decadal and longer-term variability. The air-sea CO₂ exchange in the western equatorial Pacific is strongly influenced by the shallow meridional overturning circulation and continued monitoring of CO₂ parameters in the EUC and the pycnocline of the North and South Pacific are important to determine the drivers of decadal and longer-term change in $p\text{CO}_{2\text{sw}}$ throughout the equatorial Pacific. Investigations of the variability of the oceanic CO₂ parameters in space and time in the

branches of the shallow meridional overturning circulation would help to elucidate the links between the changes in the meridional overturning circulation and CO₂ efflux from the equatorial Pacific, and to evaluate the role of the North and South Pacific in naturally sequestering the anthropogenic CO₂ from the atmosphere.

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Figure captions

- Fig.1 Time-longitude distribution of (a) $p\text{CO}_{2\text{sw}}$ data and (b) surface TCO_2 data collected in the western equatorial Pacific ($144^\circ\text{E} - 160^\circ\text{W}$, $5^\circ\text{S} - 5^\circ\text{N}$) that are superposed on the time-longitude fields of $5^\circ\text{N} - 5^\circ\text{S}$ averaged $p\text{CO}_{2\text{sw}}$ (μatm) and NTCO_2 ($\mu\text{mol kg}^{-1}$) respectively, as derived from the empirical method described in Section 4.1.
- Fig.2 Longitudinal distribution of (a) $p\text{CO}_{2\text{sw}}$ and $p\text{CO}_{2\text{air}}$, (b) temperature, (c) salinity, (d) NTCO_2 at $S = 35$, (e) calculated NTA at $S = 35$, and (f) density in surface water of the western equatorial Pacific in December 1997 and in January-February 1998 observed during the RV Kaiyo's KY9714 cruise and RV Mirai's MR9801 cruise.
- Fig.3 Longitudinal distribution of (a) $p\text{CO}_{2\text{sw}}$ and $p\text{CO}_{2\text{air}}$, (b) temperature, (c) salinity, (d) NTCO_2 at $S = 35$, (e) calculated NTA at $S = 35$, and (f) density in surface water of the western equatorial Pacific observed in January 2003 during RV Mirai's MR02K6 cruise (red) and in January 2002 during RV Mirai's MR02K1 cruise (blue).
- Fig. 4 Vertical section of temperature ($^\circ\text{C}$) (contour lines) superimposed on vertical section of salinity along the equator in the western Pacific (a) in January 2003 during R/V Mirai MR02K6 cruise and (b) in January 2002 during the MR02K1 cruise. Barrier layers found at each CTD or XCTD stations located at 1° intervals in longitude are indicated by plus symbols. Criteria of vertical gradient of temperature and density to determine isothermal layer and mixed layer is $< -0.05^\circ\text{C m}^{-1}$ and $< 0.01 \text{ kg m}^{-4}$, respectively.
- Fig. 5 CTD/sampler cast showing vertical profiles of (a) temperature, salinity, density and a thick barrier layer, and (b) phosphate, nitrate, NTCO_2 and their deficits above the isothermal layer. The cast was made at 179.31°E 0.01°S on 21 January 2003, during the R/V Mirai MR02K6 cruise. The mixed layer depth (MLD), isothermal layer depth (ILD) and the barrier layer (BL) are indicated.
- Fig. 6 Surface water variability in (a) $p\text{CO}_{2\text{sw}}$, (b) NTCO_2 at $S = 35$, (c) temperature, and (d) salinity for the period from 1987 to 2004 in the $2^\circ \times 1^\circ$ (longitude \times latitude) pixel centered at 165°E 0° . The symbol "+" shows data in the extremely light waters ($\sigma_t < 21.4$) that have been potentially affected by the local biological activity. The symbol "□" shows data in the upwelling zone ($T / ^\circ\text{C} < 29.0$ or $S > 34.8$), and "●" in the warm/fresh waters. In (a), the $p\text{CO}_{2\text{air}}$ observed on board (\circ) and from CO_2 concentration data at Christmas Island, Republic of Kiribati at 1.70°N , 157.17°W (thin line) (Tans and Conway, 2005) are shown. Open horizontal bars with "C" and closed bars with "W" indicate ENSO cold and warm periods (JMA, 2006), respectively. Thin broken lines in (a) and (b) correspond to the variability of $p\text{CO}_{2\text{sw}}$ and NTCO_2 , respectively, derived from empirical relationships with SST and SSS (Nakadate and Ishii, 2007). Thin broken lines in (c) and (d) are COBE monthly mean SST (Ishii et al., 2005) and

gridded monthly mean SSS (Delcroix et al., 2001) for 1983-1992 and monthly mean SST and SSS reproduced by ocean data assimilation “MOVE” (Usui et al., 2006) for 1993 - 2005.

Fig.7 Same as Fig.6, but centered at $180^\circ, 0^\circ$.

Fig.8 Trends of (a) in situ $p\text{CO}_{2\text{sw}}$, (b) temperature-salinity-normalized $p\text{CO}_{2\text{sw}}$, (c) NTCO_2 at $S = 34.5$, (d) SST, (e) SSS, and (f) density in the western equatorial Pacific warm/fresh water with $\text{SSS} \leq 34.8$ and $\text{SST} \geq 29.0^\circ\text{C}$ for the zone of $144^\circ\text{E} - 160^\circ\text{W}$ and $5^\circ\text{S} - 5^\circ\text{N}$. Green dots and open triangles indicate data in the extremely light waters with $\sigma_t < 21.4$ and their average for each cruise. Blue dots and open squares indicate data in the moderately light waters with $\sigma_t \geq 21.4$ and their average in each cruise. In (a) and (b), $p\text{CO}_{2\text{air}}$ from CO_2 concentration at Christmas Island, Republic of Kiribati, at $157.17^\circ\text{W}, 1.70^\circ\text{N}$ (thin line) (Tans and Conway, 2005) are shown.

Fig.9 Same as Fig.8, but for the moderately light warm/fresh waters ($\text{SSS} \leq 34.8$, $\text{SST} \geq 29.0^\circ\text{C}$ and $\sigma_t \geq 21.4$) for the zone of $144^\circ\text{E} - 160^\circ\text{W}$ and $5^\circ\text{S} - 5^\circ\text{N}$ except for $144^\circ\text{E} - 166^\circ\text{E}$ and $5^\circ\text{S} - 1^\circ\text{S}$.

Fig.10 Monthly mean SST anomaly (thin line) and its 5-year running mean (thick line) for the Nino.4 region ($160^\circ\text{E} - 150^\circ\text{W}, 5^\circ\text{S} - 5^\circ\text{N}$). Data were taken from Japan Meteorological Agency (<http://www.data.jma.go.jp/gmd/cpd/data/elnino/index/nino4anm.html>).

Table 1

List of cruises with concurrent measurements of temperature, salinity, $p\text{CO}_{2\text{sw}}$ and/or TCO_2 in surface water in the western equatorial Pacific (144°E – 160°W, 5°S – 5°N).

Ship	Cruise Name	Date of cruise (mm/dd/yy – mm/dd/yy)	Latitude	Longitude	$p\text{CO}_{2\text{sw}}$	TCO_2
Lillooet	Lill452s	12/13/83 - 12/14/83	5°S - 0°	166°W - 160°W	uw*	-
Lillooet	Lill458s	07/17/84 - 07/18/84	5°S - 2°N	167°W - 160°W	uw	-
Lillooet	Lill463n	01/15/85 - 01/16/85	5°S - 5°N	178°W - 170°W	uw	-
Lillooet	Lill465n	03/19/85 - 03/21/85	5°S - 5°N	172°W - 166°W	uw	-
Lillooet	Lill469n	08/19/85 - 08/21/85	5°S - 5°N	173°W - 168°W	uw	-
Lillooet	Lill473n	01/19/86 - 01/21/86	5°S - 5°N	173°E - 180°	uw	-
Lillooet	Lill475n	04/01/86 - 04/02/86	5°S - 5°N	173°W - 167°W	uw	-
Lillooet	Lill479n	08/09/86 - 08/10/86	5°S - 2°N	173°W - 169°W	uw	-
Lillooet	Lill481n	10/19/86 - 10/19/86	0° - 5°N	169°W - 166°W	uw	-
Lillooet	Lill483n	12/22/86 - 12/24/86	5°S - 5°N	173°W - 166°W	uw	-
Natsushima	Na8701	02/01/87 - 02/19/87	5°S - 5°N	160°E - 160°W	uw	-
Lillooet	Lill485n	03/01/87 - 03/03/87	5°S - 5°N	171°W - 166°W	uw	-
Akademik Korolev	SAGA_II	05/27/87 - 05/29/87	5°S - 5°S	160°E	uw	-
Ryofu Maru	RY8706	06/20/87 - 06/23/87	4°S - 5°N	155°E	uw	-
Oceanographer	TEW-3	07/19/87 - 07/26/87	5°S - 5°N	165°E	uw	-
Lillooet	Lill489n	07/29/87 - 08/01/87	5°S - 5°N	173°W - 163°W	uw	-
Lillooet	Lill491n	09/28/87 - 09/30/87	5°S - 5°N	173°W - 167°W	uw	-
Lillooet	Lill494s	01/06/88 - 01/08/88	5°S - 3°N	169°W - 160°W	uw	-
Hakuho Maru	KH88-1	02/01/88 - 02/04/88	5°S - 5°N	151°E - 153°E	uw	-
Lillooet	Lill495n	02/08/88 - 02/11/88	5°S - 3°N	175°E - 174°W	uw	-
Hakuho Maru	KH88-1	02/29/88 - 03/01/88	5°S - 0°	169°E - 170°E	uw	-
Oceanographer	RITS/CO2-88	04/27/88 - 05/03/88	5°S - 5°N	171°W - 170°W	uw	-
Oceanographer	EPOCS-88	05/12/88 - 05/18/88	5°S - 0°N	170°W - 160°W	uw	-
Natsushima	Na8901	01/18/89 - 02/05/89	0°	160°E - 160°W	uw	-
Natsushima	Na9001	01/15/90 - 02/18/90	5°S - 0°N	147°E - 160°W	uw	-
Malcom Baldrige	CGC-90	04/07/90 - 04/12/90	5°S - 5°N	170°W	-	dis [†]
Ryofu Maru	RY9006	06/27/90 - 06/29/90	4°S - 3°N	155°E	uw	-
Hakuho Maru	KH90-2	09/13/90 - 10/06/90	5°S - 5°N	180° - 160°W	uw	-
Hakuho Maru	KH90-3	11/04/90 - 11/17/90	5°S - 5°N	160°E - 179°E	uw	-
Natsushima	Na9101	01/17/91 - 02/27/91	5°S - 5°N	147°E - 160°W	uw	-

Table 1 (continued)

Ship	Cruise Name	Date of cruise (mm/dd/yy – mm/dd/yy)	Latitude	Longitude	$p\text{CO}_2\text{sw}$	TCO_2
Ryofu Maru	RY9106	06/18/91 - 06/21/91	4°S - 5°N	155°E	uw	-
Malcom Baldrige	MB1992_170	04/06/92 - 04/11/92	5°S - 5°N	170°W	uw	dis
Ryofu Maru	RY9206	06/21/92 - 06/24/92	4°S - 5°N	155°E	uw	-
Discoverer	DI1992_170	08/21/92 - 08/23/92	5°S - 5°N	170°W	uw	-
Hakurei Maru	NOPACCS_92	08/31/92 - 09/21/92	5°S - 5°N	172°E - 175°E	uw	-
John V. Vickers	WOCE_P13	10/12/92 - 10/16/92	5°S - 5°N	164°E - 165°E	-	dis
Discoverer	DI1993_170	03/08/93 - 03/13/93	5°S - 2°N	170°W - 167°W	uw	-
Hakurei Maru	NOPACCS_93-1	04/22/93 - 04/24/93	0°S - 5°N	174°E - 175°E	uw	-
Thomas Thompson	WOCE_P14N	08/08/93 - 08/22/93	4°S - 4°N	179°E	-	dis
Hakurei Maru	NOPACCS_93-2	09/01/93 - 09/21/93	5°S - 5°N	175°E	uw	-
Thomas Thompson	WOCE_P10	10/12/93 - 10/20/93	5°S - 5°N	145°E - 148°E	uw	dis
Kaiyo	9307	01/06/94 - 02/01/94	4°S - 5°N	144°E - 165°W	uw	uw
Hakurei Maru	NOPACCS_94-1	04/26/94 - 05/17/94	5°S - 5°N	175°E	uw	-
Hakurei Maru	NOPACCS_94-2	09/15/94 - 09/26/94	4°S - 5°N	158°E - 175°E	uw	-
L'Atalante	FLUPAC	09/27/94 - 10/17/94	5°S - 5°N	165°E - 160°W	uw	dis
John P. Tully	WOCE_P15N	10/27/94 - 11/02/94	4°S - 5°N	169°W	-	dis
Discoverer	DI1994_170	11/05/94 - 11/09/94	5°S - 5°N	170°W	uw	-
Discoverer	DI1994_180	11/15/94 - 11/18/94	5°S - 5°N	180°	uw	-
Kaiyo	9406	11/19/94 - 12/13/94	0° - 5°N	146°E - 170°E	uw	uw
Hakuho Maru	KH94-4_leg-1	11/27/94 - 11/29/94	5°S - 5°N	151°E - 154°E	uw	uw
Hakuho Maru	KH94-4_leg-4	02/06/95 - 02/07/95	5°S - 5°N	147°E - 154°E	uw	uw
Hakurei Maru	NOPACCS_95-1	04/21/95 - 05/27/95	5°S - 5°N	144°E - 164°E	uw	-
Malcom Baldrige	MB1995_08B	12/02/95 - 12/14/95	5°S - 5°N	180° - 170°W	uw	-
Malcom Baldrige	MB1995_08C	12/22/95 - 12/24/95	5°S	170°W - 160°W	uw	-
Kaiyo	9511	12/24/95 - 01/19/96	0°N - 5°N	145°E - 165°W	uw	uw
Kaiyo	9601	01/24/95 - 02/15/96	3°S - 5°N	144°E - 169°E	uw	
Ryofu Maru	RF9601	02/10/96 - 02/19/96	4°S - 5°N	144°E - 165°E	uw	-
Discoverer	CGC-96	03/04/96 - 03/06/96	5°S - 0°	169°W	-	dis
Hakurei Maru	NOPACCS_96-1	04/19/96 - 06/05/96	5°S - 5°N	151°E - 165°E	uw	-
Ka'imimoana	Ka1996_03	06/26/96 - 07/13/96	5°S - 5°N	164°E - 180°	uw	-
Ka'imimoana	Ka1996_04	07/25/96 - 08/01/96	5°S - 5°N	170°W	uw	-
Ryofu Maru	RF9611	11/04/96 - 11/18/96	5°S - 5°N	145°E - 165°E	uw	-
Ka'imimoana	Ka1996_07	11/28/96 - 12/02/96	5°S - 5°N	170°W	uw	-

Table 1 (continued)

Ship	Cruise Name	Date of cruise (mm/dd/yy – mm/dd/yy)	Latitude	Longitude	$p\text{CO}_2\text{sw}$	TCO_2
Ka'imimoana	Ka1997_03	05/20/97 - 05/27/97	5°S - 5°N	170°W	uw	-
Ka'imimoana	Ka1997_04	06/11/97 - 06/26/97	5°S - 5°N	164°E - 180°	uw	-
Ryofu Maru	RF9709	10/11/97 - 10/21/97	2°S - 5°N	145°E - 165°E	uw	-
Ka'imimoana	Ka1997_07	11/21/97 - 12/10/97	5°S - 5°N	170°W	uw	-
Kaiyo	9714	12/10/97 - 12/24/97	0° - 5°N	145°E - 180°	uw	uw
Kaiyo	9801	01/05/98 - 01/24/98	5°S - 5°N	144°E - 165°E	uw	uw
Mirai	9801	01/31/98 - 02/07/98	0° - 5°N	176°E - 161°W	uw	dis
Ka'imimoana	Ka1998_03	06/20/98 - 06/26/98	5°S - 5°N	170°W	uw	-
Ka'imimoana	Ka1998_04	07/09/98 - 07/24/98	5°S - 5°N	164°E - 180°	uw	-
Ryofu Maru	RF9809	10/15/98 - 10/25/98	3°S - 5°N	147°E - 165°E	uw	-
Ka'imimoana	Ka1998_07	11/04/98 - 11/08/98	5°S - 1°S	170°W - 160°W	uw	-
Ka'imimoana	Ka1998_08	11/30/98 - 12/02/98	2°N - 5°N	179°W - 175°W	uw	-
Mirai	MR98K2	01/03/99 - 01/17/99	0° - 5°N	144°E - 167°W	uw	dis
Kaiyo	9901	01/26/99 - 02/02/99	0° - 5°N	144°E - 169°E	uw	uw
Mirai	MR99K1	02/15/99 - 03/21/99	0° - 5°N	147°E - 165°E	uw	uw
Ka'imimoana	Ka1999_03	07/16/99 - 07/26/99	5°S - 5°N	176°E - 170°W	uw	-
Mirai	MR99K6	10/28/99 - 11/13/99	5°S - 5°N	144°E - 156°E	uw	uw
Kaiyo	9909	11/16/99 - 11/21/99	1°S - 5°N	144°E - 150°E	uw	-
Mirai	MR99K7	11/27/99 - 12/11/99	0° - 5°N	144°E - 167°W	uw	uw
Ka'imimoana	Ka2000_03	06/29/00 - 07/05/00	5°S - 5°N	170°W	uw	-
Ka'imimoana	Ka2000_04	07/20/00 - 08/04/00	5°S - 5°N	164°E - 180°	uw	-
Ryofu Maru	RF0009	10/15/00 - 10/26/00	3°S - 5°N	144°E - 165°E	uw	-
Ka'imimoana	Ka2000_06	10/29/00 - 11/08/00	5°S - 5°N	177°E - 170°W	uw	-
Ka'imimoana	Ka2000_08	11/18/00 - 12/02/00	5°S - 5°N	164°E - 180°	uw	-
Mirai	MR00K8	01/11/01 - 01/30/01	0° - 5°N	145°E - 160°W	uw	uw
Keifu_Maru	KS0101	02/10/01 - 02/11/01	0° - 5°N	165°E	uw	-
Mirai	MR01K1	02/23/01 - 03/15/01	5°S - 5°N	147°E - 156°E	uw	uw
Ryofu Maru	RF0110	11/04/01 - 11/15/01	5°S - 5°N	147°E - 165°E	uw	-
Hakuho Maru	KH01-3	12/07/01 - 12/10/01	5°S - 5°N	165°W - 160°W	uw	uw
Mirai	MR02K1	01/22/02 - 02/07/02	0° - 5°N	145°E - 160°W	uw	uw
Keifu_Maru	KS0201	02/08/02 - 02/16/02	3°S - 5°N	147°E - 165°E	uw	-
Mirai	MR02K2	03/02/02 - 03/20/02	5°S - 5°N	145°E - 156°E	uw	uw

Table 1 (continued)

Ship	Cruise Name	Date of cruise (mm/dd/yy – mm/dd/yy)	Latitude	Longitude	$p\text{CO}_2\text{sw}$	TCO_2
Ka'imimoana	Ka2002_03	06/18/02 - 06/23/02	5°S - 5°N	170°W	uw	-
Keifu_Maru	KS0206	07/04/02 - 07/08/02	0°S - 5°N	144°E	uw	-
Ka'imimoana	Ka2002_07	10/15/02 - 10/19/02	4°S - 5°N	160°W	uw	-
Ryofu Maru	RF0210	10/27/02 - 11/11/02	5°S - 5°N	147°E - 176°E	uw	-
Mirai	MR02K6	12/18/02 - 01/28/03	5°S - 5°N	145°E - 160°W	uw	uw
Keifu_Maru	KS0301	02/08/03 - 02/18/03	3°S - 5°N	147°E - 165°E	uw	-
Keifu_Maru	KS0306	07/19/03 - 08/03/03	5°S - 5°N	145°E - 165°E	uw	dis
Ka'imimoana	Ka2003_07	11/05/03 - 11/09/03	5°S - 5°N	170°W	uw	-
Ka'imimoana	Ka2003_08	11/20/03 - 12/04/03	5°S - 5°N	165°E - 180°	uw	-
Ryofu Maru	RF0401	01/30/04 - 02/22/04	5°S - 5°N	144°E - 165°E	uw	dis
Keifu_Maru	KS0401	02/15/04 - 02/18/04	5°S - 5°N	144°E - 165°E	uw	-
Keifu_Maru	KS0406	07/01/04 - 07/25/04	5°S - 5°N	145°E - 165°E	uw	dis
Ka'imimoana	Ka2004_03	07/09/04 - 07/13/04	2°S - 5°N	170°W	uw	-
Ka'imimoana	Ka2004_04	07/23/04 - 08/10/04	5°S - 5°N	165°E - 180°	uw	-

* 'uw' refers to underway measurement.

† 'dis' refers to measurement with discrete bottle sampling

Table 2

Linear increase rate of $p\text{CO}_2\text{sw}$ and normalized $p\text{CO}_2\text{sw}^*$ in the warm/fresh waters of the western equatorial Pacific at various regions and hydrographic conditions

No.	Area		Conditions				Linear increase rate of $p\text{CO}_2$ (in $\mu\text{atm yr}^{-1}$)			
	Latitude	Longitude	Temp.	Salinity	Density		1985-2004	1985-1990	1990-1999	1999-2004
a.	5°S-5°N	144°E- 160°W	≥ 29.0	≤ 34.8	≥ 21.4	$p\text{CO}_2\text{sw}$	$+1.4 \pm 0.2$ (75)	$+0.1 \pm 1.1$ (15)	$+2.4 \pm 0.6$ (41)	-0.5 ± 1.0 (31)
						$n\text{-}p\text{CO}_2\text{sw}$	$+1.2 \pm 0.2$	$+0.1 \pm 1.1$	$+2.3 \pm 0.6$	-0.7 ± 1.0
b.	5°S-5°N	144°E- 160°W [†]	≥ 29.0	≤ 34.8	≥ 21.4	$p\text{CO}_2\text{sw}$	$+1.5 \pm 0.2$ (72)	$+0.3 \pm 1.3$ (13)	$+2.2 \pm 0.7$ (39)	-0.2 ± 1.0 (29)
						$n\text{-}p\text{CO}_2\text{sw}$	$+1.4 \pm 0.2$	$+0.5 \pm 1.2$	$+2.1 \pm 0.6$	$+0.0 \pm 1.0$
c.	1°S-1°N	144°E- 160°W	≥ 29.0	≤ 34.8	≥ 21.4	$p\text{CO}_2\text{sw}$	$+1.6 \pm 0.3$ (46)	$+3.1 \pm 1.5$ (6)	$+2.1 \pm 0.8$ (28)	-1.2 ± 1.3 (16)
						$n\text{-}p\text{CO}_2\text{sw}$	$+1.4 \pm 0.3$	$+1.7 \pm 1.2$	$+2.2 \pm 0.7$	-0.6 ± 1.3
d.	5°S-5°N	144°E- 160°W [†]	≥ 29.5	≤ 34.65	≥ 21.4	$p\text{CO}_2\text{sw}$	$+1.3 \pm 0.3$ (49)	-0.1 ± 2.1 (8)	$+2.0 \pm 0.7$ (27)	-0.3 ± 0.9 (23)
						$n\text{-}p\text{CO}_2\text{sw}$	$+1.2 \pm 0.2$	$+0.3 \pm 1.6$	$+2.0 \pm 0.7$	-0.5 ± 0.9
e.	5°S-5°N	144°E- 160°W [†]	≥ 29.0	≤ 34.8	≥ 21.5	$p\text{CO}_2\text{sw}$	$+1.5 \pm 0.2$ (61)	$+1.3 \pm 1.4$ (11)	$+2.1 \pm 0.7$ (33)	-0.3 ± 1.1 (25)
						$n\text{-}p\text{CO}_2\text{sw}$	$+1.4 \pm 0.2$	$+1.6 \pm 1.3$	$+2.0 \pm 0.6$	-0.4 ± 1.1
f.	5°S-5°N	144°E- 160°W [†]	≥ 29.0	≤ 34.8	not defined	$p\text{CO}_2\text{sw}$	$+1.5 \pm 0.2$ (85)	$+0.9 \pm 1.3$ (19)	$+2.0 \pm 0.5$ (45)	-0.1 ± 1.0 (31)
						$n\text{-}p\text{CO}_2\text{sw}$	$+1.4 \pm 0.2$	$+0.7 \pm 1.1$	$+2.0 \pm 0.5$	-0.2 ± 1.1
g.	Atmospheric $p\text{CO}_2$ at Christmas Is.					$p\text{CO}_2\text{air}$	$+1.63 \pm 0.01$	$+1.74 \pm 0.08$	$+1.54 \pm 0.04$	$+1.77 \pm 0.08$

* Normalized at $t=29.5^\circ\text{C}$ and $S=34.5$. Number of cruises in parenthesis.

[†] Data from the south-western limb of the zone ($> 1^\circ\text{S}$ and $< 166^\circ\text{E}$) were not included in the analysis.

Table 3

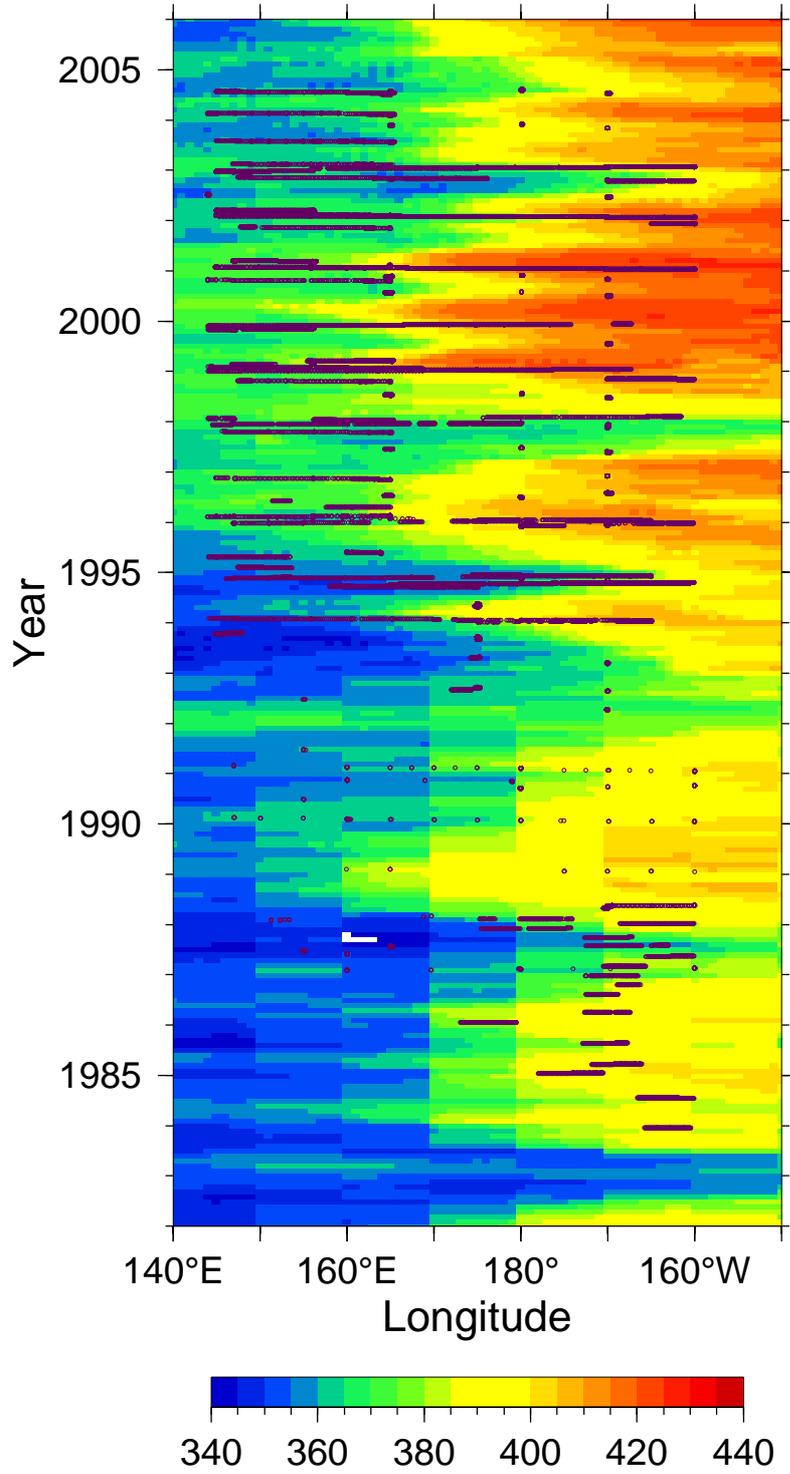
Linear rate of increase of NTCO₂ in the warm/fresh waters of the western equatorial Pacific at various regions and conditions

No.	Area		Conditions			Linear increase rate of NTCO ₂ * (in $\mu\text{mol kg}^{-1} \text{yr}^{-1}$)		
	Latitude	Longitude	Temp.	Salinity	Density	1992-2004	1992-1999	1999-2004
a.	5°S - 5°N	144°E - 160°W	≥ 29.0	≤ 34.8	≥ 21.4	+0.8 \pm 0.3 (22)	+2.1 \pm 0.4 (11)	-1.2 \pm 0.9 (10)
b.	5°S - 5°N	144°E - 160°W [†]	≥ 29.0	≤ 34.8	≥ 21.4	+1.3 \pm 0.3 (21)	+2.1 \pm 0.4 (15)	+0.4 \pm 0.9 (9)
c.	1°S - 1°N	144°E - 160°W	≥ 29.0	≤ 34.8	≥ 21.4	+1.3 \pm 0.3 (18)	+2.3 \pm 0.3 (13)	-0.3 \pm 1.6 (8)
d.	5°S - 5°N	144°E - 160°W [†]	≥ 29.5	≤ 34.65	≥ 21.4	+0.8 \pm 0.4 (13)	+2.0 \pm 0.6 (8)	-1.3 \pm 1.4 (7)
e.	5°S - 5°N	144°E - 160°W [†]	≥ 29.0	≤ 34.8	≥ 21.5	+0.7 \pm 0.3 (15)	+1.4 \pm 0.6 (10)	+0.3 \pm 0.9 (8)
f.	5°S - 5°N	144°E - 160°W [†]	≥ 29.0	≤ 34.8		+0.7 \pm 0.2 (28)	+1.5 \pm 0.4 (20)	-0.2 \pm 0.6 (12)

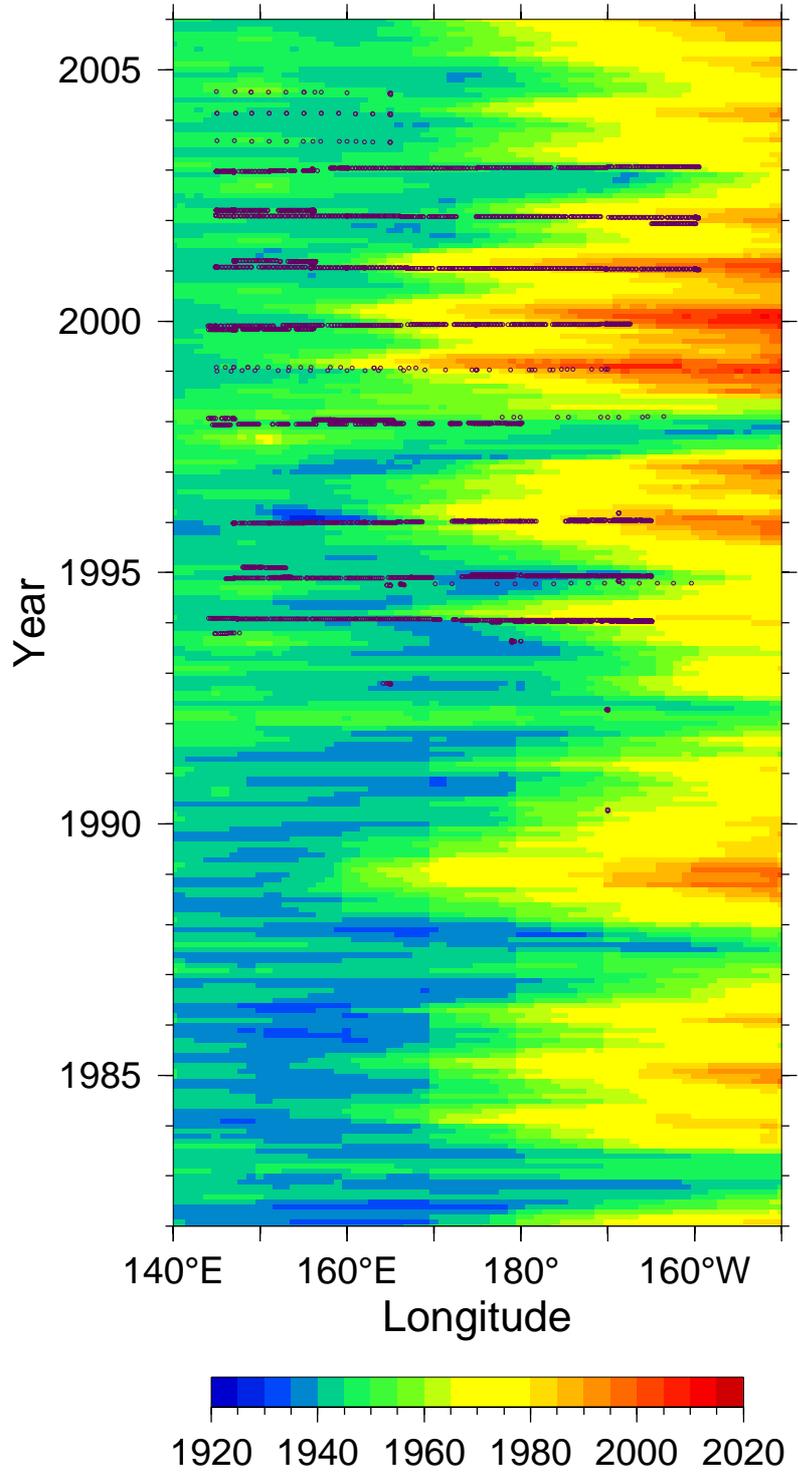
*Normalized at S=34.5. Number of cruises in parenthesis.

[†]Data from the south-western limb of the zone ($> 1^\circ\text{S}$ and $< 166^\circ\text{E}$) were not included in the analysis.

(a) pCO₂sw



(b) NTCO2



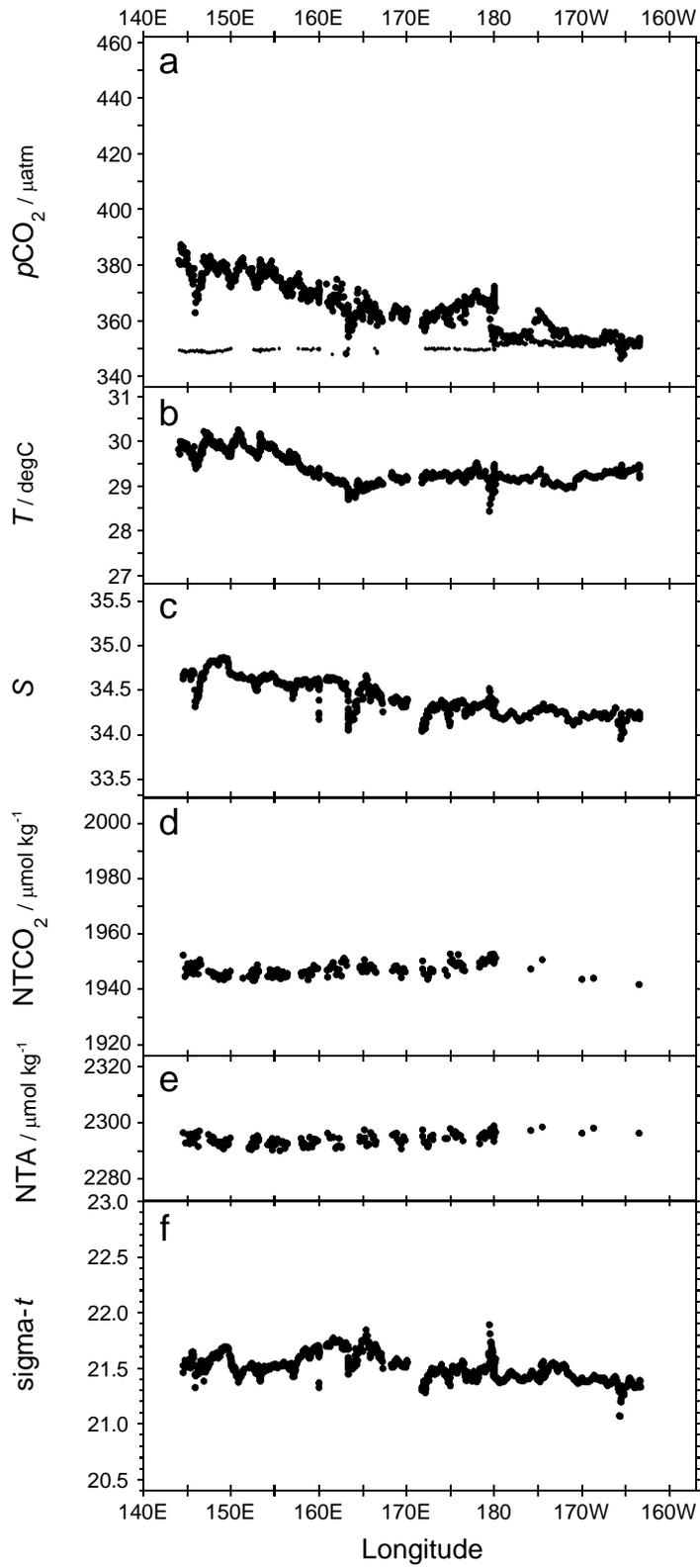


Fig. 2 Ishii et al.

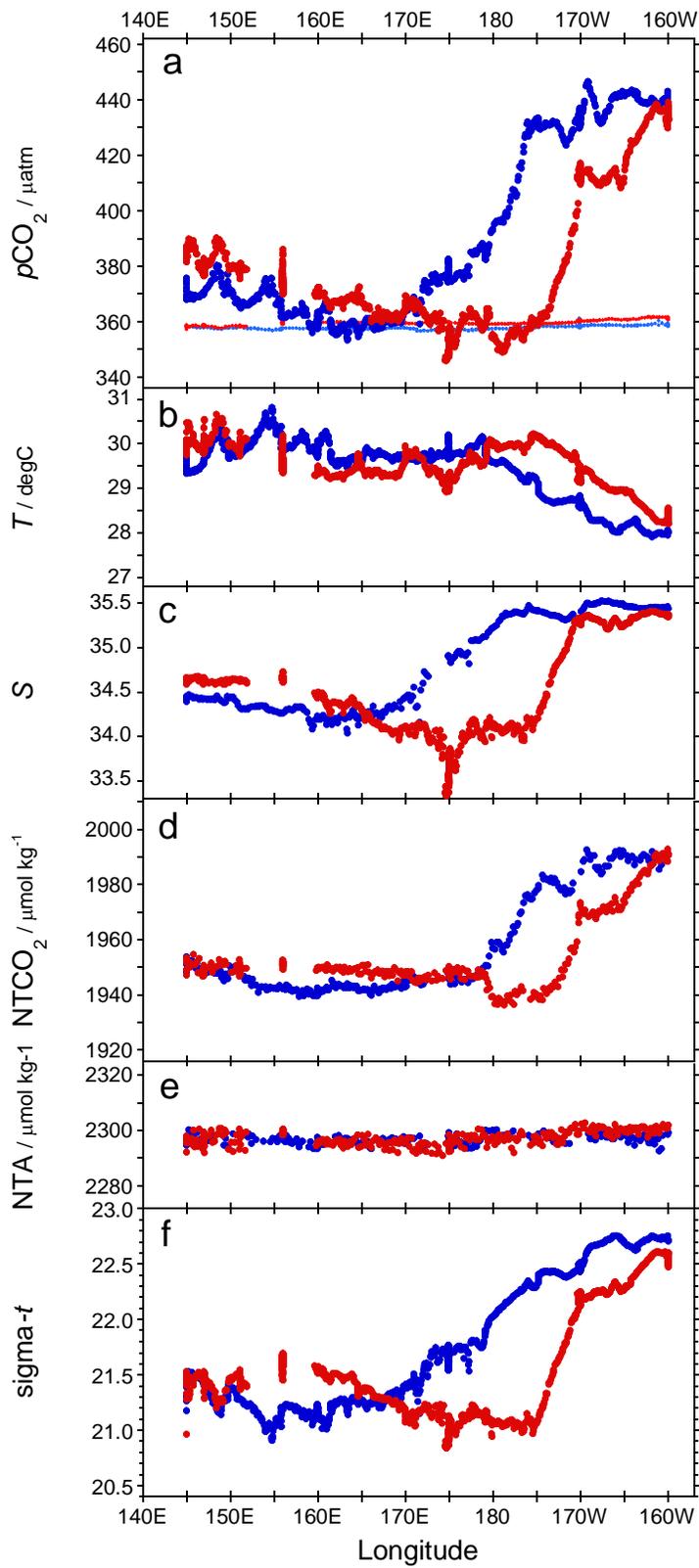
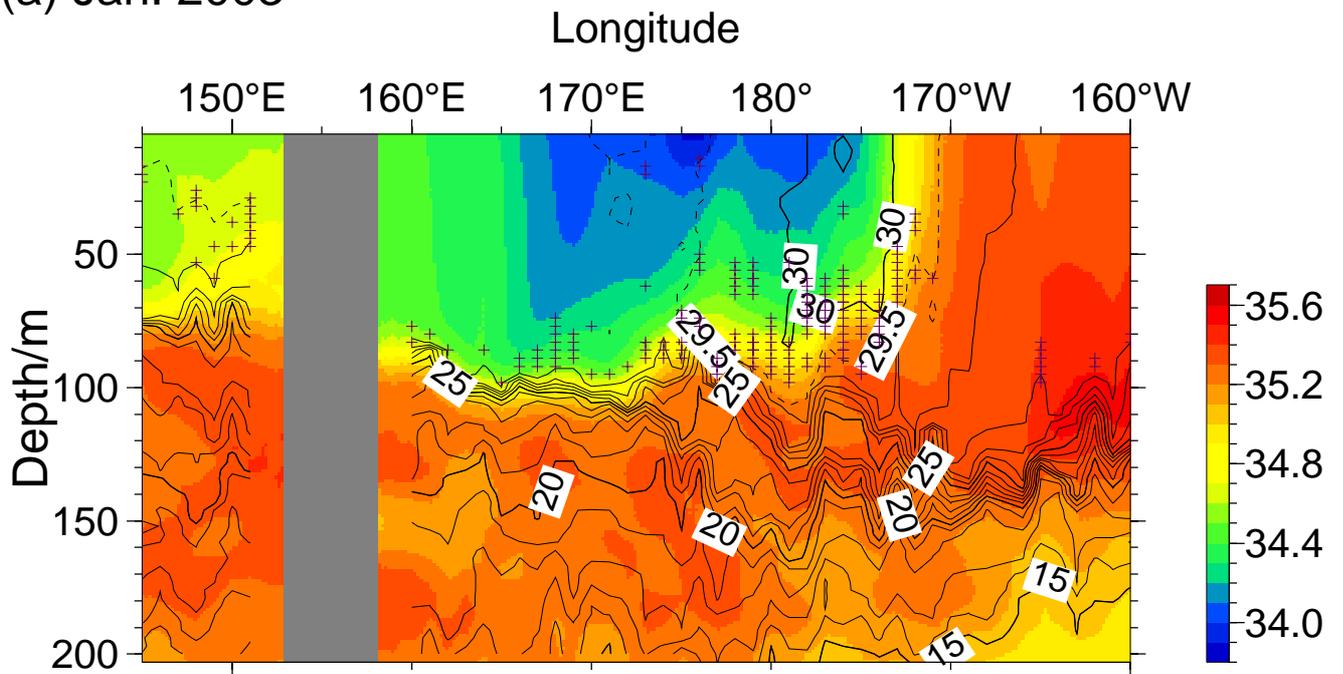
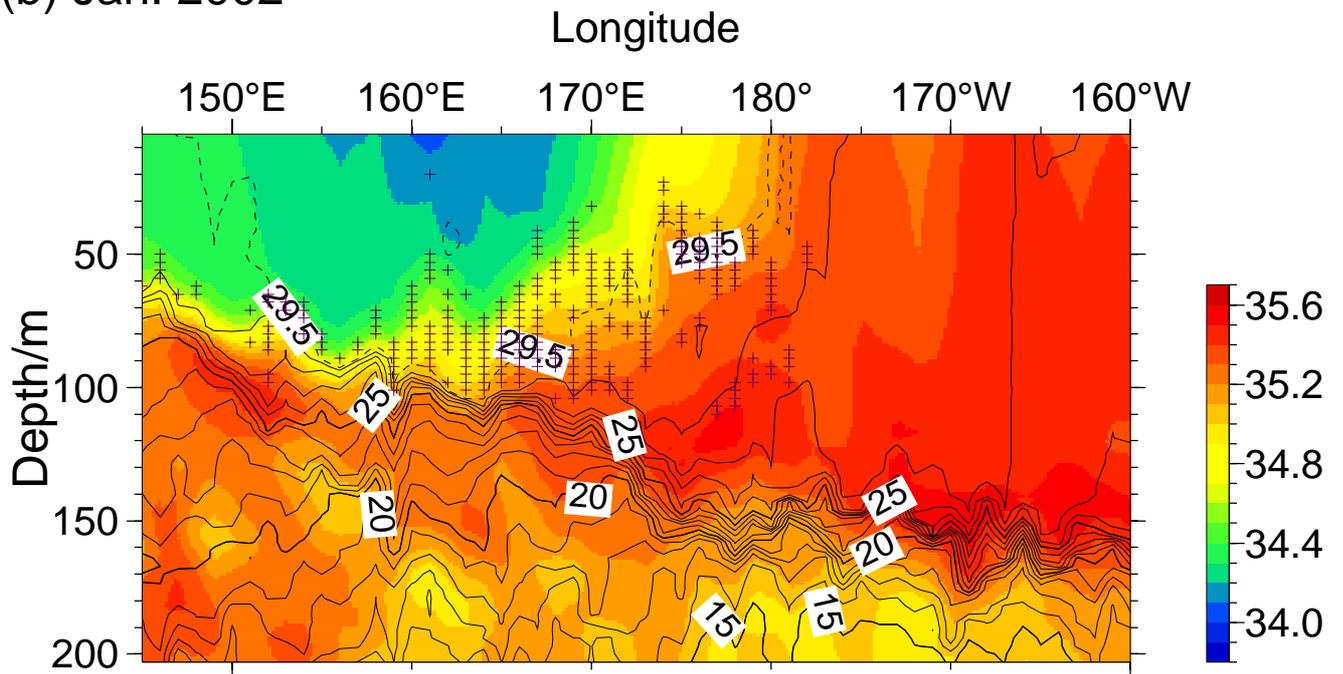


Fig. 3 Ishii et al.

(a) Jan. 2003



(b) Jan. 2002



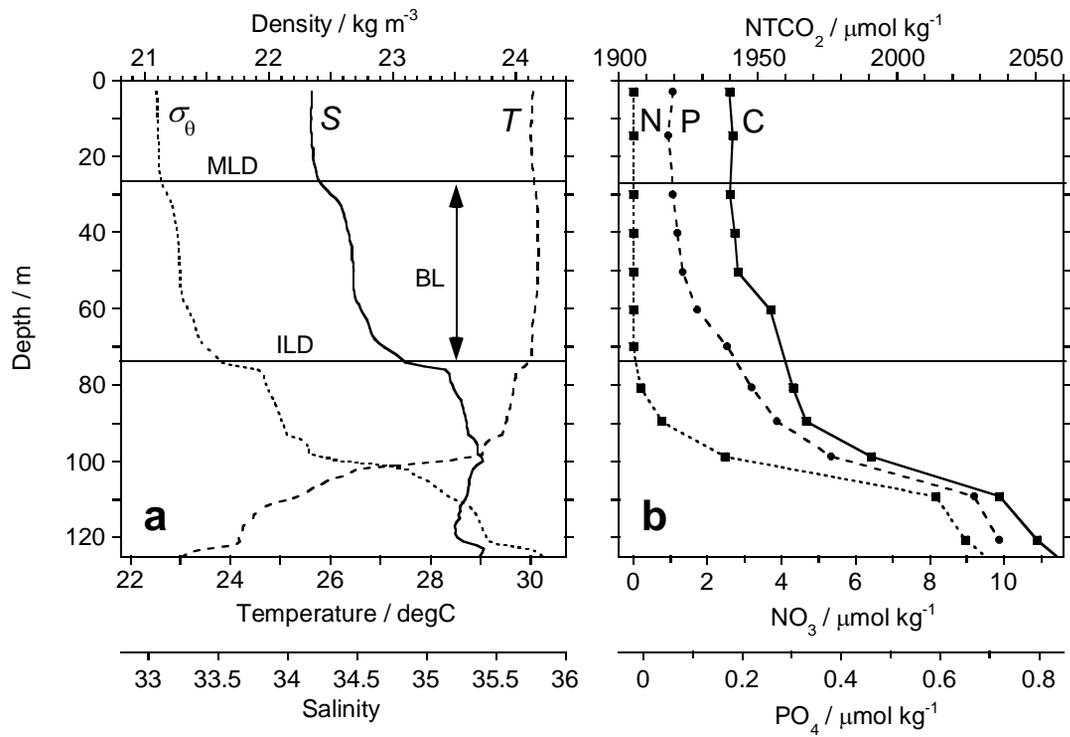


Fig. 5 Ishii et al.

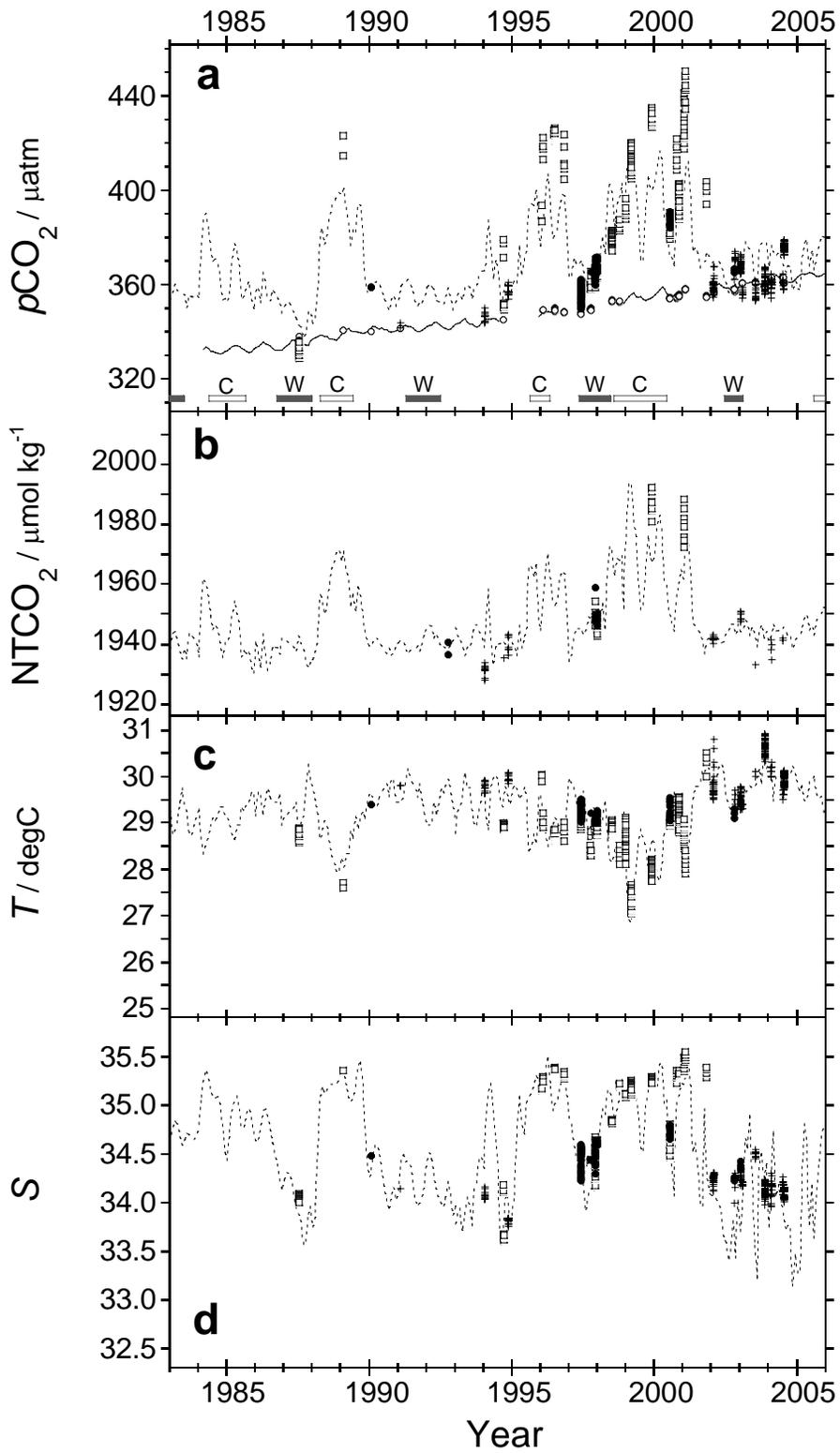


Fig. 6 Ishii et al.

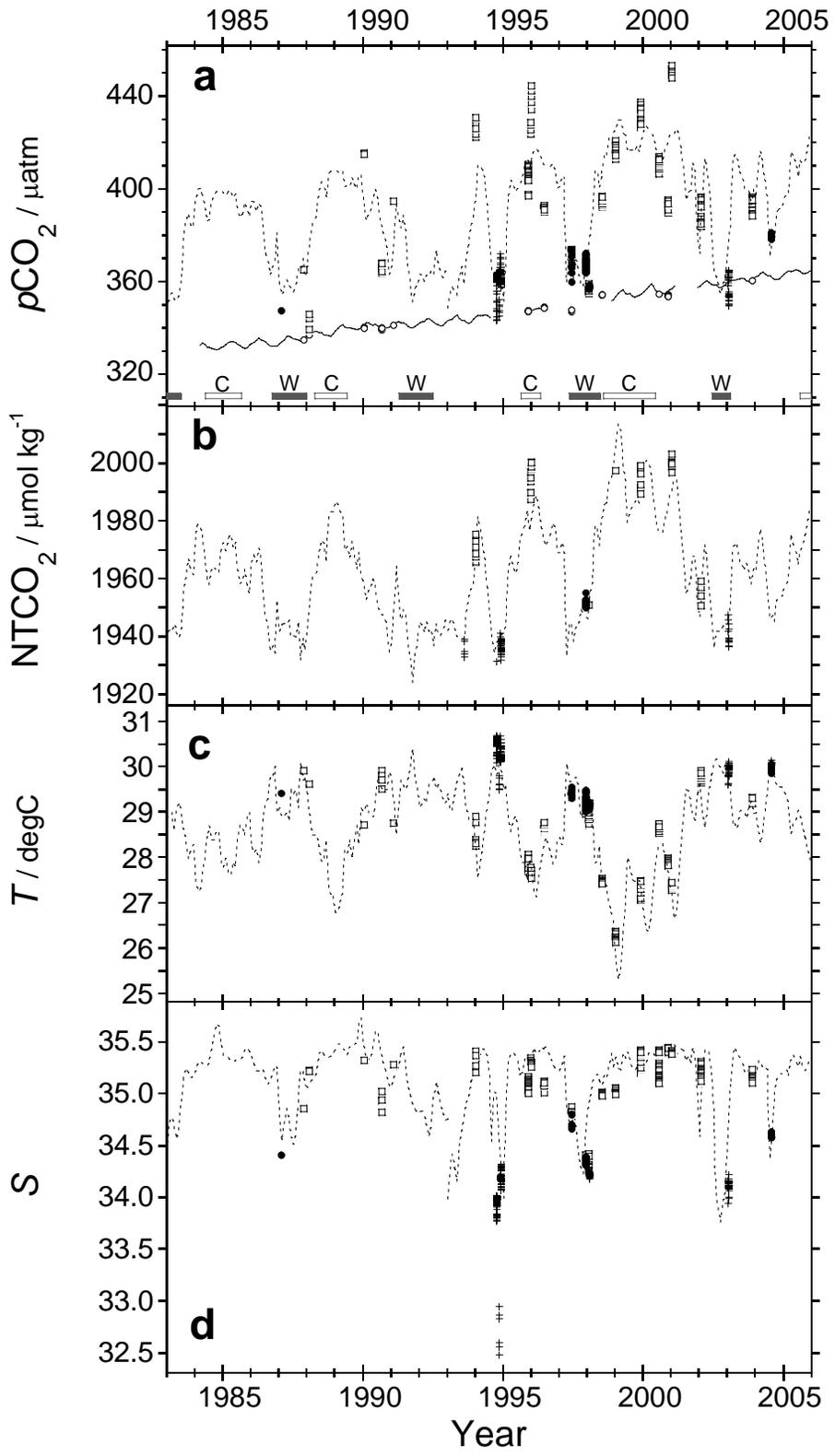


Fig. 7 Ishii et al.

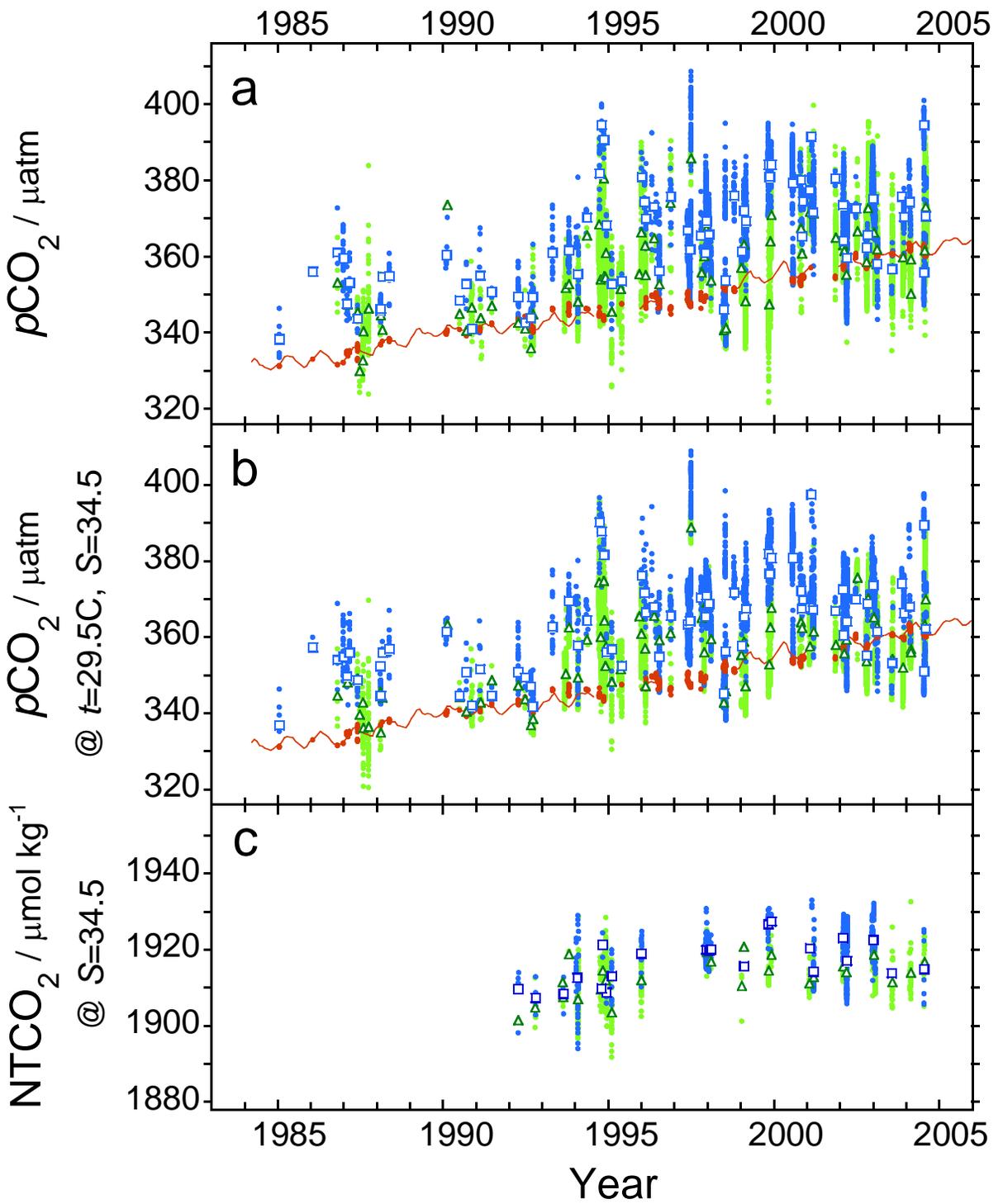


Fig.8 Ishii et al

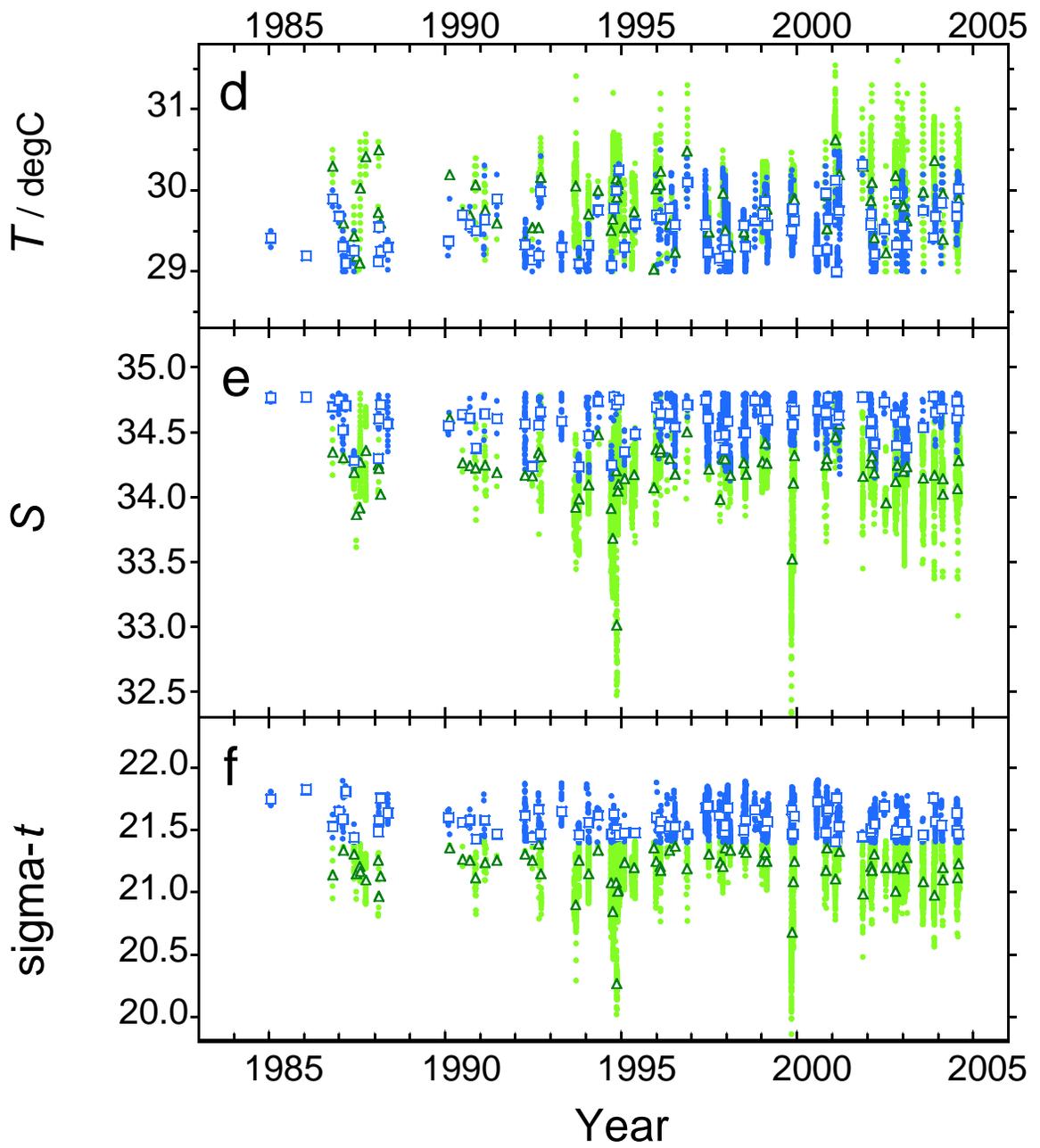


Fig.8 (continued) Ishii et al

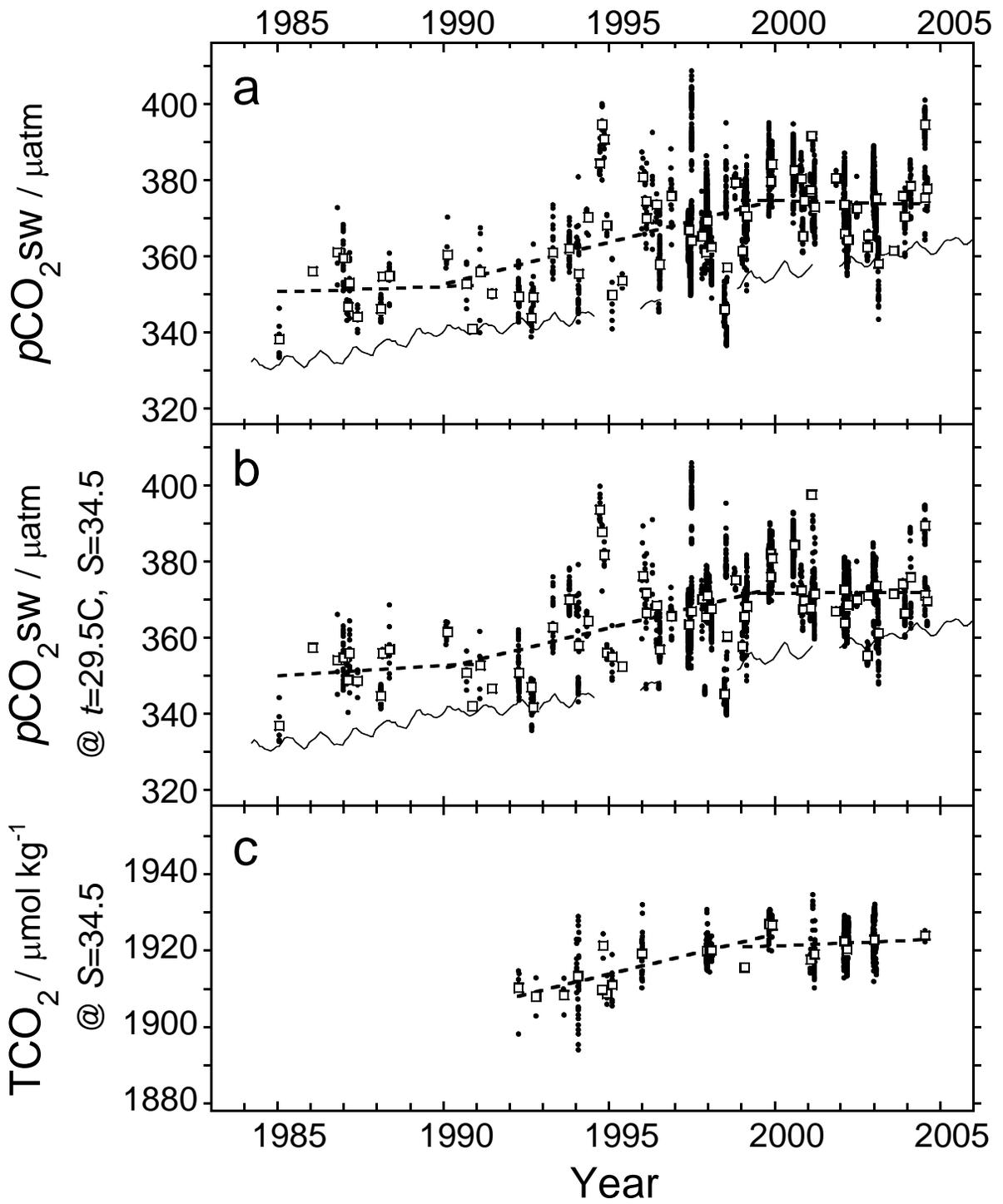


Fig.9 Ishii et al

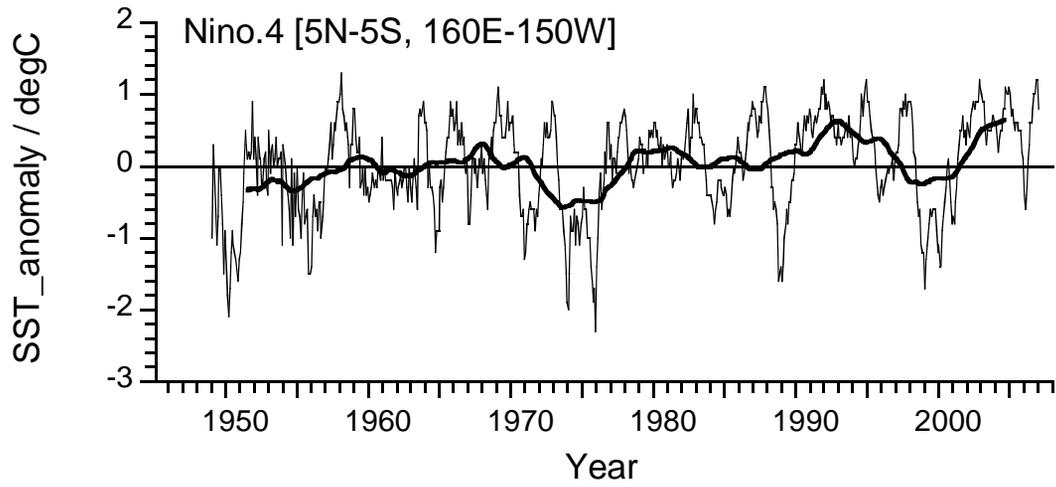


Fig.10 Ishii et al