Long-term trend of CO_2 and ocean acidification in the surface water of the Ulleung Basin, the East/Japan Sea inferred from the underway observational data

Jae-Yeon Kim[1], Dong-Jin Kang[2,*], Tongsup Lee[1], Kyung-Ryul Kim[3]

[1]{Department of Oceanography, Pusan National University, Busan 609-735, Korea}

[2]{Korea Institute of Ocean Science & Technology, Ansan 426-744, Korea}

[3]{School of Earth and Environment Sciences/Research Institute of Oceanography, Seoul National University, Seoul 151-747, Korea}

Correspondence to: Dong-Jin Kang (djocean@kiost.ac)

Abstract

Anthropogenic carbon is responsible for both global warming and ocean acidification. Efforts are underway to understand the role of ocean in a high CO_2 world on a global context. However, marginal seas received little attention despite their significant contribution to biogeochemical cycles. Here we report that the CO_2 increase and ocean acidification in the surface waters of the Ulleung Basin (UB) of the East/Japan Sea are much faster than the global mean, and possible causes are discussed. Fourteen observations of surface fCO_2 were made in the period from 1995 to 2009. The decadal trend of fCO_2 increment was estimated by harmonic analysis. The estimated rates of increase of fCO_2 were 1.8 μatm yr^{-1} for the atmosphere and 2.7 μatm yr^{-1} for the surface ocean. The rates exceed the global mean of 1.5 μatm yr^{-1}. The ocean acidification trend, calculated from total alkalinity and fCO_2, was estimated to be 0.03 pH units $decade^{-1}$. Surface seawater of the UB has been acidified more rapidly compared to the global mean (0.02 pH units $decade^{-1}$). Results show that, if warming strengthens the currents or advection in the marginal seas, biological pump will be enhanced. This would lead to compensation for the presumed reduction in oceanic uptake of atmospheric CO_2 in the warmer world, which warrants quantification worldwide.

1 Introduction

The IPCC (2007) announced that the anthropogenic CO_2 that has accumulated in the atmosphere since the Industrial Revolution is responsible for the enhanced greenhouse effect. The ocean is the ultimate mobile carbon storage reservoir in the Earth system. The surface ocean absorbs atmospheric CO_2 by means of both physical and biogeochemical processes. In the oceanic carbonate system, the dissolved carbon is transported to the deep ocean by the oceanic carbon pumps and eventually sequestrated in deep-sea sediments. Sabine et al. (2004) reported that about one-third of the CO_2 from the total anthropogenic CO_2 emissions is stored in the ocean. Currently, results from established oceanic time-series stations show that the trend of increasing CO_2 in surface seawater exceeds that of the atmosphere (Bates, 2001; Keeling et al., 2004) and is accelerating ocean acidification (Caldeira and Wickett, 2003; Key et al., 2004; Orr et al., 2005).

Ocean carbon uptake is commonly described as a variety of carbon pumps, which operate by thermodynamics (e.g., temperature effects on the solubility of CO_2), physical transport (e.g., mixing and advection of water masses carrying various forms of carbon), sinking of soft-tissue (e.g., carbon uptake/release and export by phytoplankton during photosynthesis/respiration), and sinking of carbonate shells (e.g., formation and dissolution of $CaCO_3$) (Volk and Hoffert, 1985). These processes are easily affected, directly or indirectly, by temperature. The world ocean's temperature from the surface to 3000 m increased by 0.06 ℃ between the mid 1950s and mid 1990s because of an enhanced greenhouse effect (Levitus et al., 2005), and previous works have shown that the trend of increasing global sea surface temperature (SST) is about 0.08 – 0.14 ℃ decade^{-1} (Nicholls et al., 1996; Molinari et al., 1997; Casey and Cornillon, 2001). Soaring SST creates stronger stratification between the surface sea (upper mixed layer) and the deep ocean. It might affect the great ocean conveyor system by weakening deep water formation. The warming will also weaken the solubility pump and the biological carbon pump through fortified stratification. Overall, a positive feedback exists between global warming and ocean carbon uptake, thus making the situation worse.

Nearly all the ocean's dynamics can be observed and studied in the East/Japan Sea (EJS) despite its relatively small dimensions (Kim and Kim, 1996; Kim et al., 2001; Kang et al., 2003). The Ulleung Basin (UB), located on the western side of the southern EJS, has complex hydrography. The bifurcation of warm current entering through the Korea Strait leaves

numerous spin-off mesoscale eddies at the surface, and beneath the surface, a number of cold currents originate from the winter overturning along the Siberian coast of the EJS. Winter mode water formation is occasionally observed (Kim et al., 1991; Seung and Kim, 1993; Talley et al., 2003). The UB supports a high productivity (>220 gC m^{-2} yr^{-1}) owing to its surface dynamics and coastal upwelling (Yamada et al., 2004; Yoo and Park, 2009). Because the UB has strong solubility pumps and biological pumps, it is an ideal laboratory for ocean carbon uptake studies. For this reason some studies were carried out in the UB (Oh et al., 1999; Choi et al., 2011, 2012). In particular, it has been noted that the role of the coastal ocean in the global carbon budget has been underestimated (Wollast, 1998; Borges et al., 2005) and under debate (Cai and Dai, 2004; Thomas et al., 2004). Therefore, the role of the carbon pump in marginal seas should be reconsidered.

To reduce the scientific uncertainty in the prediction of future climate, a better understanding of the carbon flux among various reservoirs, especially the fluxes across the air-sea interface, is required. The Takahashi climatology (Takahashi et al., 2002) utilized all of the available data for modeling but still neglected marginal seas. Here, we like to emphasize the role of the marginal sea, as exemplified by the UB, and compare it to that for the entire ocean. There is no time-series station comparable to BATS or HOT in the EJS. However, we attempted to fill the gap of oceanic CO_2 time series in the UB by interpolation using harmonic function analysis of the data from repeated measurements. Although simplified and crude, an attempt to discern the long-term trend of CO_2 uptake and acidification in a marginal sea that operates its own conveyor belt at a much faster time scale than the global ocean is here made for the first time.

2 Experimental Methods and Data Analysis

2.1 Experimental methods

Measurements of the partial pressure of CO_2 (pCO_2) in the surface water and overlying atmosphere, sea surface temperature (SST), and sea surface salinity (SSS) were carried out fourteen times in the EJS from 1995 to 2009 (Table 1). The data that were collected in the UB, the southwestern part of the EJS, were used in this study. The study area was defined as the region from 36–38 °N latitude and 130–133 °E longitude (Fig. 1).

The pCO_2 of surface seawater and the overlying atmosphere was measured by LiCor model 6252 non-dispersive infrared (NDIR) gas analyzer with a two-stage Weiss-type equilibrator. The CO_2 concentration from the NDIR was acquired every 2 seconds and, after statistical treatment for 1 minute or 2 minute, averaged data were obtained and used as the final data. Three different standard gases were used to calibrate the NDIR. Every 12 hours, a series of standard gases was analyzed for calibration; one of the standard gases was analyzed every 6 hours to check for drift of the machine. Atmospheric and surface seawater pCO_2 were analyzed every 15 minutes and 45 minutes on the hour, respectively. Surface seawater pCO_2 was analyzed by equilibrating air with surface seawater fed to the equilibrator. In-situ SST and SSS were measured using thermosalinograph (SeaBird Electronics, Model SBE-21 or SBE-45).

ΔfCO_2 is defined as the difference between fCO_2 in the surface water (fCO_2^{sea}) and overlying atmosphere (fCO_2^{atm}) ($\Delta fCO_2 \equiv fCO_2^{sea} - fCO_2^{atm}$). When we calculated ΔfCO_2, we applied average value of prior and posterior fCO_2^{atm} data to fCO_2^{sea} data, since we don't have both data at the same time and the CO_2 variation in the atmosphere is much smaller than in the seawater.

Total alkalinity (TA) was determined onboard by a potentiometric titration method using a closed cell (Millero et al., 1993). Total alkalinity was calibrated by Dickson's CRMs which are measured at every cruise. The TA data were obtained during cruises 9906, 0306, 0405, and 0410 (Table 1). Surface seawater pH values were obtained immediately after sampling by a spectrophotometric method using the indicator dye *m*-cresol purple (Clayton and Byrne, 1993). The extinction coefficients for *m*-cresol purple from Clayton and Byrne (1993) were used. Determination of surface seawater pH values was carried out during cruises 9906, 0306, 0406, and 0410 (Table 1).

2.2 Air-sea flux estimation

The CO_2 flux (mmol C m^{-2} d^{-1}) was estimated from following equation;

$$\text{Flux} = k \times s \times \Delta fCO_2$$

where k is the gas transfer velocity (cm h^{-1}), s is the solubility of CO_2 gas in seawater (mol kg^{-1} atm^{-1}; Weiss, 1974). We choose the formulas for k and the wind speed relationships used by Wanninkhof (1992). The NCEP wind speed data (http://www.cdc.noaa.gove/cdc/reanalysis)

averaged on monthly scale in 36 – 38 °N latitude and 130 – 133 °E longitude was used to estimate for flux.

3 Results and Discussion

3.1 General characteristics of fCO_2 and CO_2 flux in the UB

The secular variation of mean fCO_2^{sea} and fCO_2^{atm} of the UB from 1995 to 2009 is shown in Fig. 2. The fCO_2^{sea} ranged from 125 to 499 µatm with a mean value of 350.9 µatm (sd = 41.8), and the fCO_2^{atm} varied from 344 to 397 µatm with a mean value of 370.5 µatm (sd = 8.7). The fCO_2^{sea} was generally lower than the fCO_2^{atm} indicating that the surface seawater was undersaturated to atmospheric CO_2 in this area.

The secular trend of fCO_2 includes the long-term linear trend as well as the cyclic seasonal variation (Keeling et al., 2004). The harmonic function, which is the sum of two components defined as the one-year and half-year component, has been used primarily to reconstruct time series data. Since this analysis method was first introduced by Nojiri et al. (1999), harmonic functions have been used extensively for fCO_2 variability studies (Zeng et al., 2002; Lüger et al., 2004; Chierici et al., 2006). We applied the harmonic function analysis to fCO_2 data from the UB to estimate the decadal trend of CO_2 and to separate thermal and non-thermal effects on the fCO_2. Because harmonic function analysis is a technique for annual data, we added the long-term linear trend to the harmonic function as follows:

$$fCO_2(t) = c_0 + c_1 \times \sin(2\pi t) + c_2 \times \cos(2\pi t) + c_3 \times \sin(4\pi t) + c_4 \times \cos(4\pi t) + c_5 \times t \quad (1)$$

where t is year.

The set of six constant coefficients, c_0, c_1, c_2, c_3, c_4, and c_5,ensured a satisfactory fit of the harmonic function of Eq. (1) to the observed data (R^2 = 0.78 for seawater, R^2 = 0.96 for air), when the constant coefficients were equal to these values (Fig. 3):

c_0= -4977.46, c_1= -29.97, c_2= 0.88, c_3= -1.45, c_4= 23.41, c_5= 2.66 (for seawater),

c_0= -3308.75, c_1= 2.51, c_2= 7.22, c_3= -2.45, c_4= -1.46, c_5= 1.84 (for air).

Among the results, the c_5 value which means annual trend for fCO_2^{sea} was estimated to be 2.7 µatm yr^{-1} and that for the fCO_2^{atm} was 1.8 µatm yr^{-1}.

The secular vaiation of ΔfCO_2 is shown in Fig. 4a. Although the fCO_2^{sea} and fCO_2^{atm} varies with time, the secular variation of fCO_2 can be insignificant on the variation of ΔfCO_2, since ΔfCO_2 was calculated with fCO_2 values in the seawater and in the atmosphere at the same time. We can hardly find the long-term trend of ΔfCO_2. However, the seasonal variation of ΔfCO_2 shows that seawater is generally undersaturated with respect to the atmosphere with expection in summer (Fig. 4b) as other studies reported (Oh et al., 1999; Choi et al., 2012). Therefore, the UB serves as a sink of atmospheric CO_2, in general.

The sea-air CO_2 fluxes in 1995 and 2004 were estimated in order to evaluate the effect of the last decadal increasing trend in fCO_2. The ΔfCO_2, which estimated by The sea-air CO_2 fluxes were evaluated to be -0.95 ± 0.53 mol m^{-2} yr^{-1} for 1995 and -0.81 ± 0.49 mol m^{-2} yr^{-1} for 2004. This result shows that the UB acts as a carbon sink and its carbon sink efficiency in unit area is almost 1.7(±0.3)- fold higher than global ocean (-0.51 mol m^{-2} yr^{-1}, Takahashi et al., 2002). However the flux had been decreased about 15 % during the last decade. Choi et al. (2012) and Oh et al. (1999) reported that the annual integrated CO_2 flux in this area was -2.47±1.26 mol m^{-2} yr^{-1} and -2.2 mol m^{-2} yr^{-1}, which were larger than our results. The transiency of their observations could lead overestimation in CO_2 flux because of fewer observations (less than four times) despite of the complexity of the monthly variability and significant seasonal amplitude in fCO_2^{sea} in the UB.

3.2 Seasonal variability and controlling factors of fCO_2 in the UB

The parameters which can control the fCO_2^{sea} such as sea surface temperature (SST), mixed layer depth (MLD) based on the climatology data (de Boyer- Montégut et al., 2004), and chlorophyll-*a* concentration inferred from the SeaWiFS data between 1998 and 2007 are represented in Fig. 5. The SST reached a minimum in winter (February) and a maximum in summer (August and September) and had a mean value of 18.5 °C. The monthly variation in fCO_2^{atm} shows opposite in phase to that of SST. The monthly variation pattern of fCO_2^{atm} followed the general seasonal cycle of fCO_2 (or pCO_2) monitored in the North Hemisphere; *i.e.*, the fCO_2^{atm} reached its maximum value in spring and minimum value in summer. The fCO_2^{sea}, meanwhile, showed more complex variation than did the fCO_2^{atm}. The monthly mean of the fCO_2^{sea} in February, March, and April was lower than the average value (350.9 µatm) but that in August and December was above the average. The May–September data were

close to the average. During the warm season (June to August), relatively high fCO_2^{sea} was associated with elevated SST. During the fall and winter (October to February), monthly fCO_2^{sea} values were higher than those in spring (April and May). This difference was a result of vertical entrainment of CO_2-rich subsurface water despite the decreasing SST. Deepening of MLD (>50 m) supported high fCO_2^{sea} phenomenon in winter (Fig. 5b). Comparison of the normalized total CO_2 ($NTCO_2$) in the EJS showed the difference of ~ 100 μmol kg^{-1} in surface layer between summer and winter (Park, 1997). It also could be a result of the biological drawdown of CO_2 in March and April, when the fCO_2^{sea} attained its lowest value. During March and April, satellite-based chlorophyll-*a* concentration reached a peak value (> 1 mg m^{-3}) suggesting a possibility of biological CO_2 drawdown (Fig. 5c).

Takahashi et al. (2002) proposed a method for estimating the relative importance of the effects of biological activity and seasonal temperature change on the pCO_2 of surface seawater. In order to estimate the relative magnitude of these effects, the ratio of the thermal effect (effect of temperature change) to the non-thermal effect (effect of biological activity and vertical mixing) was adapted after the method proposed by Takahashi et al. (2002) as the following equations:

$$fCO_2^{therm} = \overline{fCO_2} \times \exp\left[0.0423 \cdot (SST - \overline{SST})\right] \quad (2)$$

$$fCO_2^{non\text{-}therm} = fCO_2 \times \exp\left[0.0423 \cdot (\overline{SST} - SST)\right] \quad (3)$$

$$\Delta fCO_2^{therm} = fCO_2^{therm}{}_{max} - fCO_2^{therm}{}_{min} \quad (4)$$

$$\Delta fCO_2^{non\text{-}therm} = fCO_2^{non\text{-}therm}{}_{max} - fCO_2^{non\text{-}therm}{}_{min} \quad (5)$$

where SST is the surface seawater temperature in °C, and the $\overline{fCO_2}$ and $\overline{SST}$ refer to the annual average of observed fCO_2 and SST values, respectively.

The dependence of fCO_2 variability in the UB on the thermal (fCO_2^{therm}) and non-thermal ($fCO_2^{non\text{-}therm}$) effects was separated by the equations above (Eq. 2 and 3). The results are shown in Figs. 5f and 5g. The fCO_2^{therm} followed the same pattern as the seasonal SST variation (Fig. 5a and 5f). The peak-to-peak amplitudes of the seasonal cycles of SST and fCO_2^{therm} (ΔfCO_2^{therm}, Eq. 4) were ~17 ℃ and 275 μatm, respectively, which means that an 1 ℃ temperature change makes a 16 μatm change in fCO_2. Takahashi et al. (1993) suggested

the thermodynamic relationship between pCO_2 and temperature ($(\partial pCO_2/\partial T)/pCO_2$ = 0.0423 ℃$^{-1}$), our data showed almost close to it (0.0462 ℃$^{-1}$). However, the ΔfCO_2^{therm} was larger than that observed at Station "P" (100 µatm, Wong and Chan, 1991) and at BATS (150 µatm, Bates, 2001), which located at the comparable latitude as the UB. Larger seasonal variation of SST (>17℃) may have caused such a difference.

On the other hand, $fCO_2^{non\text{-}therm}$ reached its highest value during the cold season and its lowest value during the warm season. The peak-to-peak amplitude was 190 µatm, which was smaller than the value from the Ross Sea (260 µatm, Sweeney 2000) but much larger than the value from BATS (115 µatm, Bates 2001). This high $fCO_2^{non\text{-}therm}$ value in winter was due to (1) relatively low biological carbon uptake during the cold season, and (2) stronger vertical mixing that pumps up high-CO_2 subsurface water. The decrease in the $fCO_2^{non\text{-}therm}$ value of more than 100 µatm in spring was attributed to carbon fixation by the spring bloom (Fig. 5e and 5c). In summer, the value decreased because of weakening of vertical mixing caused by stratification (Fig. 5b and 5e).

The seasonal variation in fCO_2 can be explained by the sum of the thermal and non-thermal effects. The ratio between the thermal effect and the non-thermal effect was estimated to be almost equal to one (ΔfCO_2^{therm} /$\Delta fCO_2^{non\text{-}therm}$ = 1.4) in the UB, which suggested that the thermal effect was almost balanced with the non-thermal effect. A similar result (0.9) was obtained at Station "P" in the eastern subarctic Pacific Ocean (50° N, 145° W) during the period 1973–1978 (Wong and Chan 1991). On the other hand, the ratio differed significantly from that found for the BATS (2.7) (Bates 2001) or the Ross Sea (0.02) (Sweeney 2000) values (Table 2).

In summary, the contribution of temperature variation to the seasonality of fCO_2 was almost equivalent to the non-thermal effect in the UB. However, the relative contribution varied with the season (Fig. 5h). Non-thermal effect contributed to the surface fCO_2 drawdown in summer, while the surface fCO_2 elevation in winter. According to Sarmiento and Gruber (2006), pCO_2 in the North Pacific is not affected by one dominant factor among SST, biological activity, and vertical mixing but is affected by their combined effect. Their explanation about the controlling factors of pCO_2 in the North Pacific could be applied to similar features of the fCO_2 in the UB.

3.3 Long-term trend of fCO_2 and ocean acidification in the UB

Long-term trend of the fCO_2 in the UB is shown in Fig. 3. The long-term atmospheric pCO_2 variation observed at Gosan station (33° 17.4′ N, 126° 9.9′ E) in Jeju Island, WMO Gloabl Atmosphere Watch Station (Cho et al., 2005) is also shown in the figure for comparison with fCO_2^{atm} measured in the UB (Fig. 3b).

The fCO_2^{atm} in the UB coincided with pCO_2 at Gosan owing to the shorter turnover time of atmospheric CO_2 than that in the ocean. The trends of increasing pCO_2 at Gosan and fCO_2 in the UB were 1.9 ppm*v* yr^{-1} and 1.8 µatm yr^{-1}, respectively. These values were slightly larger than the global mean 1.5 ppm*v* yr^{-1} (IPCC, 2007).

Since the work of Inoue et al. (1995), numerous studies have focused on the rate of CO_2 increase based on monitoring and survey data. For the sake of comparing the long-term trend of the fCO_2 in the UB with other regions, previously published data covering approximately 30 different oceanic areas are listed in Table 3. The fCO_2^{sea} in the UB had been increasing gradually at a rate of 2.7 µatm yr^{-1}, which is in the middle of the increasing rates in the mid-latitude of the Northern Hemisphere (Fig. 6). Generally, the increasing rate in most areas is close to the global average (1.5 µatm yr^{-1}; IPCC 2007), but the rate in the middle latitudes of the Northern Hemisphere is higher. We could infer from this result that human activities might influence the rate of increase of CO_2 in surface seawater.

As surface water CO_2 has been increasing, the pH of the surface seawater has been decreasing, an effect called ocean acidification. The long-term trend of pH in the UB was estimated based on total alkalinity (TA) data from this study and fCO_2^{sea}. The pH values were calculated from TA and simulated fCO_2^{sea} by CO2SYS (Lewis and Wallace, 1998). The carbonate dissociation constants (K_1 and K_2) used in these calculations was those of Mehrbach et al. (1973) as refit by Dickson and Millero (1987). We assumed the TA of surface seawater to be constant at 2266 ± 17 µeq kg^{-1}, the average value of 60 measurements during the study period, because the seasonality and secular trends were not significant. These calculated pH values were in good agreement with the measured ones (Fig. 7b). As the surface fCO_2 increased, the pH value in the surface seawater of the UB, while fluctuating seasonally, decreased at the rate of 0.03 pH units $decade^{-1}$ since 1995 (Fig. 7). The pH in the surface ocean decreased by 0.1 pH units between 1750 and 1994, which was noted as an unprecedented decline by Sabine et al. (2004).

To compare fCO_2 increasing trend and pH decreasing trend in the UB with global trend, we also plotted time series of fCO_2 and pH from Station ALOHA data during the same period. Since 1995, the pCO_2 at Station ALOHA has increased at a rate of ~1.6 μatm yr^{-1} and mixed layer pH has declined by 0.02 pH units per decade (Fig. 7). IPCC (2007) reported that the rate of decrease of pH was estimated at about 0.02 pH units $decade^{-1}$ based on station data at HOT, BATS, and ESTOC since 1980. Compared with the global trend, surface seawater of the UB has being acidified faster.

Under the assumption of constant TA and sea surface temperature, 10% increase of fCO_2 in the UB (*i.e*. 26.6 μatm increase in a decade) may reduce pH by 0.027 pH units which contributes about 87% of pH decrease in the UB during the last decade. A seawater temperature rise of 1°C may also decrease pH by 0.01 pH units at a pressure of 1 atm (Gieskes, 1969). The sea surface temperature in the EJS increased by 0.2 – 0.7 °C during the last decade (Kim et al., 2007; Yeh et al., 2010), which is a much faster warming than the global average (0.13°C $decade^{-1}$; Rayner et al., 2006). This warming contributed to a pH decrease of 0.002 – 0.007 pH units which was equivalent to 7 – 23% of the pH decline in the UB during the last decade. Therefore, the ocean acidification in the UB was mainly driven by the fCO_2 increase, while the warming effect was relatively small.

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Table 1. Information for cruises in this study. SST, fCO_2^{atm}, fCO_2^{sea}, ΔfCO_2, total alkalinity (TA), and pH are reported as averages and standard deviations. The average values are arithmetic mean of 1 minute or 2 minute averaged values in the cruise. The number of data values are listed in parentheses.

Cruise Name	Observation Period	Research Vessel	SST(℃)	SSS	fCO_2^{atm} (μatm)	fCO_2^{sea} (μatm)	ΔfCO_2 (μatm)	TA (μeq kg-1)	pH	Source
9508	6 Aug -7 Aug 1995	R/V Professor Khromov	24.1±1.5 (40)	32.6±0.3 (40)	353.0±3.1 (40)	365.4±13.6 (40)	12.4±12.8 (40)	N/A	N/A	Oh et al., 1999
9602	23 Feb - 24 Feb 1996	R/V Parvel Gordienko	9.2±1.2 (256)	34.3±0.1 (256)	362.9± 0.4 (256)	311.2±14.3 (256)	-51.7±14.4 (256)	N/A	N/A	Oh et al., 1999
9608	31 Jul - 10 Aug 1996	R/V Professor Khromov	25.6±1.0 (233)	33.4±0.7 (233)	346.8±2.6 (233)	359.7±36.3 (233)	12.9±35.7 (233)	N/A	N/A	Oh et al., 1999
9703	19 Mar – 7 Apr 1997	R/V Parvel Gordienko	12.5±1.1 (463)	34.0±0.5 (463)	364.9±3.2 (463)	266.5±12 (463)	-98.4±12.8 (463)	N/A	N/A	Oh, 1998
9906	25 Jun - 1 Jul 1999	R/V Roger Revelle	20.9±0.5 (669)	34.1±0.1 (669)	N/A	335.9±10.5 (178)	N/A	2271±10 (23)	8.08±0.02 (23)	This study
0204	12 Apr -19 Apr 2002	R/V Gargarinsky	12.8±1.4 (3857)	34.3±0.1 (3857)	375.9±8.6 (712)	280.5±41.1 (1141)	-92.8±41.2 (1142)	N/A	N/A	This study
0306	9 Jun - 14 Jun 2003	R/V Tamgu-5	19.5±0.4 (2233)	33.6±0.4 (2233)	366.9±2.3 (2233)	353.7±19.2 (1319)	-29.1±18.6 (1319)	2256±21 (21)	8.06±0.03 (29)	This study
0312	12 Dec - 22 Dec 2003	R/V Tamgu-5	16.4±0.5 (1392)	33.9±0.1 (1392)	379.7±0.9 (120)	390.1±15.3 (401)	-3.9±14.7 (401)	N/A	N/A	This study
0405	6 May - 19 May 2004	R/V Akademik Labrantiev	17.8±1.8 (5153)	34.3±0.2 (5153)	376.6±3.3 (846)	354±23.6 (3113)	-50.1±22.7 (3113)	2281±8 (8)	N/A	This study
0406	10 Jun - 11 Jun 2004	R/V Tamgu-5	19.8±0.2 (334)	34.2±0.2 (334)	368.6±0.3 (80)	336.5±7 (194)	-50.7±6.1 (194)	N/A	8.05±0.02 (21)	This study
0409	14 Sep -15 Sep 2004	R/V Tamgu-1	26.2±2.4 (806)	32.6±2.4 (806)	368.8±3.9 (64)	348.8±7.9 (245)	-26.6±8.4 (245)	N/A	N/A	This study
0410	5 Oct - 29 Oct 2004	R/V Tamgu-5	23.0±1.2 (4362)	33.4±0.2 (4362)	371.5±2.9 (563)	379.3±17.9 (2673)	-30.6±16.4 (2673)	2261±8 (8)	8.07±0.03 (8)	This study
0811	1 Nov – 7 Nov 2008	R/V Eardo	19.8±1.1 (297)	33.3±0.1 (297)	389.3±4.8 (297)	359.5±12.2 (297)	-29.8±14 (297)	N/A	N/A	This study
0907	9 Jul – 10 Jul 2009	R/V Akademik Labrantiev	21.2±0.7 (546)	33.7±0.1 (546)	375.8±4.1 (546)	397.5±22.3 (546)	21.7±21.5 (546)	N/A	N/A	This study
Average			18.5±4.23	33.9±0.54	370.5±8.7	350.9±41.8	-41.4±36.5	2266±17	8.06±0.02	

Table 2. Comparison of the contribution of thermal and non-thermal effects on the fCO_2 of surface seawater in various regions.

Station	Thermal effect (μatm)	Non-thermal effect (μatm)	Ratio (Therm/non-Therm)	location	References
UB	275	190	1.4	36-38 °N, 130-133 °E	This Study
BATS	150	55	2.7	32 ° 50' N, 64 ° 10' W	Bates, 2001; Takahashi et al., 2002
Ross Sea	5	260	0.02	76 ° 30' S, 169 °E- 177 ° W	Sweeney, 2000; Takahashi et al., 2002
Station "P"	100	115	0.9	50 ° N, 145 ° W	Wong and Chan, 1991; Takahashi et al., 2002

1 Table 3. Rate of increase of surface seawater CO_2 at various regions of the world ocean.

Region	Increasing Rate ($\mu atm\ yr^{-1}$)	Duration	Area	Reference
North Pacific	1.3±0.2	1970-2004	Whole Pacific	Takahashi et al., 2006
Central Equatorial Pacific	1.8±0.7	1979 -2001	5 °N – 5 °S	Takahashi et al., 2003
Western Equatorial Pacific	3.4±0.4	1979 -2001	5 °N – 5 °S	Takahashi et al., 2003
Western Equatorial Pacific	1.5±0.2	1985-2004	5 °S – 5 °N, 144 °E – 160 °W	Ishii et al., 2009
Western North Pacific	1.8±0.6	1984-1993	15 °N - 35 °N, 132 °E - 142 °E	Inoue et al., 1995
Western North Pacific	0.5±0.7	1984-1993	3 °N - 14 °N, 132 °E – 142 °E	Inoue et al., 1995
Western North Pacific	2.1	1995-2007	35 °N, 147.5 °E	Nojiri and Tsumori, 2007
Northwestern North Pacific	3.7	1992-1996	37.3 °N, 141.47 °E	Watai et al., 1998
Subtropical North Pacific	1.8±0.6	1996-2005	30 °N - 42 °N, 120 °E – 105 °W	Lenton et al., 2012
Central North Pacific	0.8	1995-2006	40 °N, 175 °E	Nojiri and Tsumori, 2007
Eastern North Pacific	0.9		35 °N, 140 °W	Nojiri and Tsumori, 2007
Western Subarctic North Pacific	0.6		45 °N, 155 °E	Nojiri and Tsumori, 2007
Western Subarctic North Pacific	1.6±1.7	1995-2003	42 °S – 50 °N, 150 °E – 170 °E	Lenton et al., 2012
Eastern Subarctic North Pacific	1.7		54.5 °N, 165 °W	Nojiri and Tsumori, 2007
Station "P"	1.4		50 °N, 145 °W	Nojiri and Tsumori, 2007
North Atlantic gyre	4.4	1994-2005	22 °N – 50 °N, 5 °W – 70 °W	Schuster and Watson, 2007
Eastern North Atlantic	3.6	1995-2002	36 °N – 52 °N, 10 °W – 35 °W	Lüger et al., 2004, 2006
Western North Atlantic	1.7	1995-2002	36 °N – 52 °N, 36 °W – 70 °W	Lüger et al., 2004, 2006
North Atlantic Subpolar Gyre	1.8	1982-1998	50 °N – 70 °N, 80 °W – 10 °W	Lefévre et al., 2004
Eastern Subpolar North Atlantic	3.0	1970s-1980s	50 °N – 64 °N, 32 °W – 10 °W	Omar and Olsen, 2006
Western Subpolar North Atlantic	3.0	1994-2003	53 °N – 62 °N, 20 °W – 45 °W	Corbiere et al., 2007
South Indian Ocean	1.1		20 °S	Inoue and Ishii, 2005
South Indian Ocean	1.4		40 °S	Inoue and Ishii, 2005
Southern Ocean	2.3±0.2	1995-2008	42 °S – 62 °S, Circumpol	Lenton et al., 2012
South of Australia (Sub-Antarctic Zone)	1		50 °S, 140 °E - 160 °E	Inoue and Ishii, 2005
South of Australia (Polar Frontal Zone)	1.5		55 °S, 140 °E - 160 °E	Inoue and Ishii, 2005
South of Australia (Polar Zone)	1.8		58 °S, 140 °E - 160 °E	Inoue and Ishii, 2005
ALOHA	2.5±0.3	1989-2001	22.7 °N, 158 °W	Dore et al., 2003
ALOHA	3.2±0.4	1997-2002	22.7 °N, 158 °W	Keeling et al., 2004
ALOHA	1.4±0.2	1988-1996	22.7 °N, 158 °W	Keeling et al., 2004
SEATS	4.2±3.2	1995-2004	18 °N, 116 °E	Tseng et al., 2007
ESTOC site	1.55	1995-2004	29.16 °N, 15.5 °W	Santana-Casino et al., 2007
BATS	1.67±0.28	1983-2005	31.7 °N, 64.5 °W	Bates, 2007
UB	2.7	1995-2004	36 °N - 38 °N, 130 °E - 133 °E	This study
Global Mean	1.5			IPCC, 2007

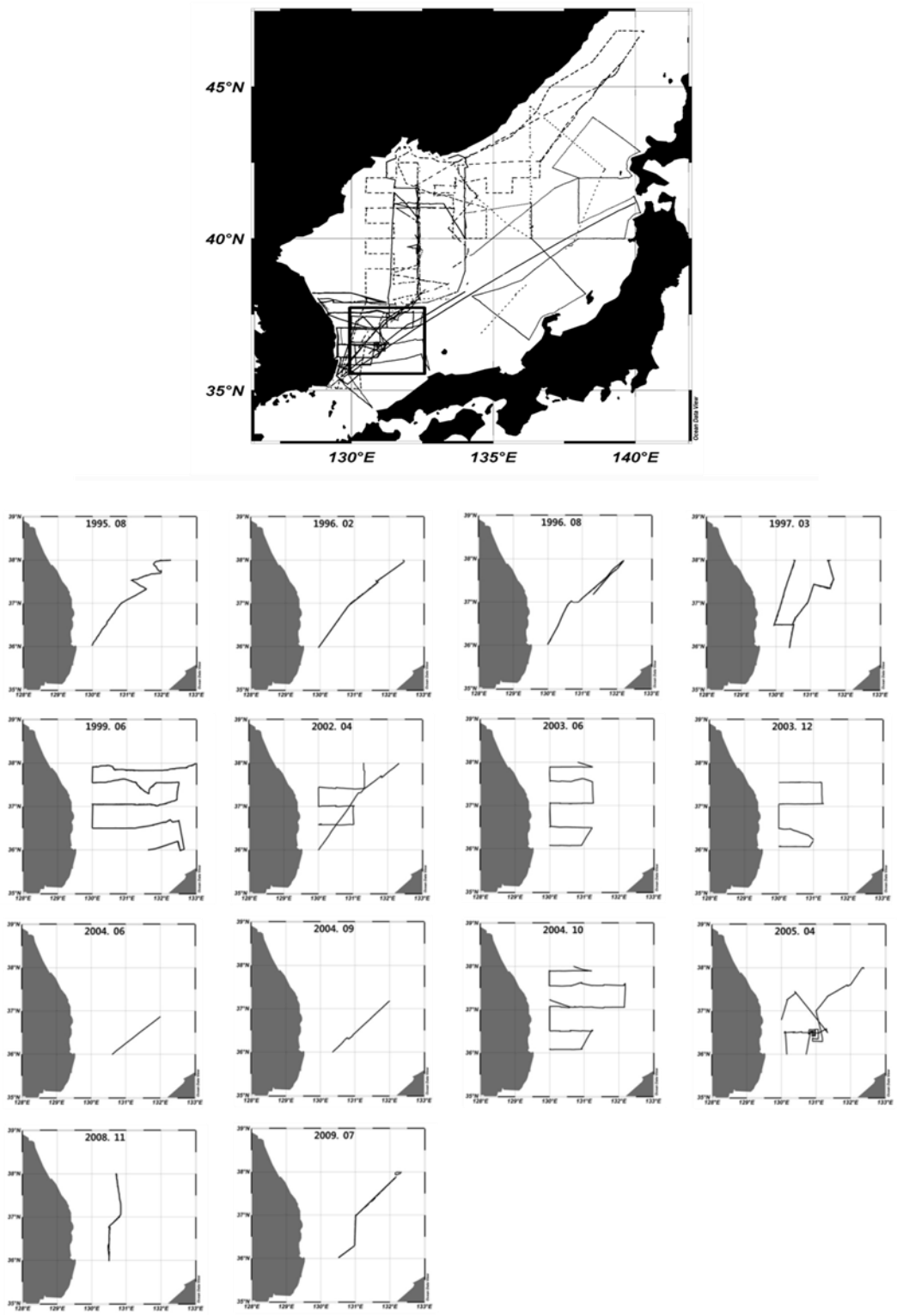

Figure 1. Maps showing the tracks of the cruises conducted between 1995 and 2009 for this study

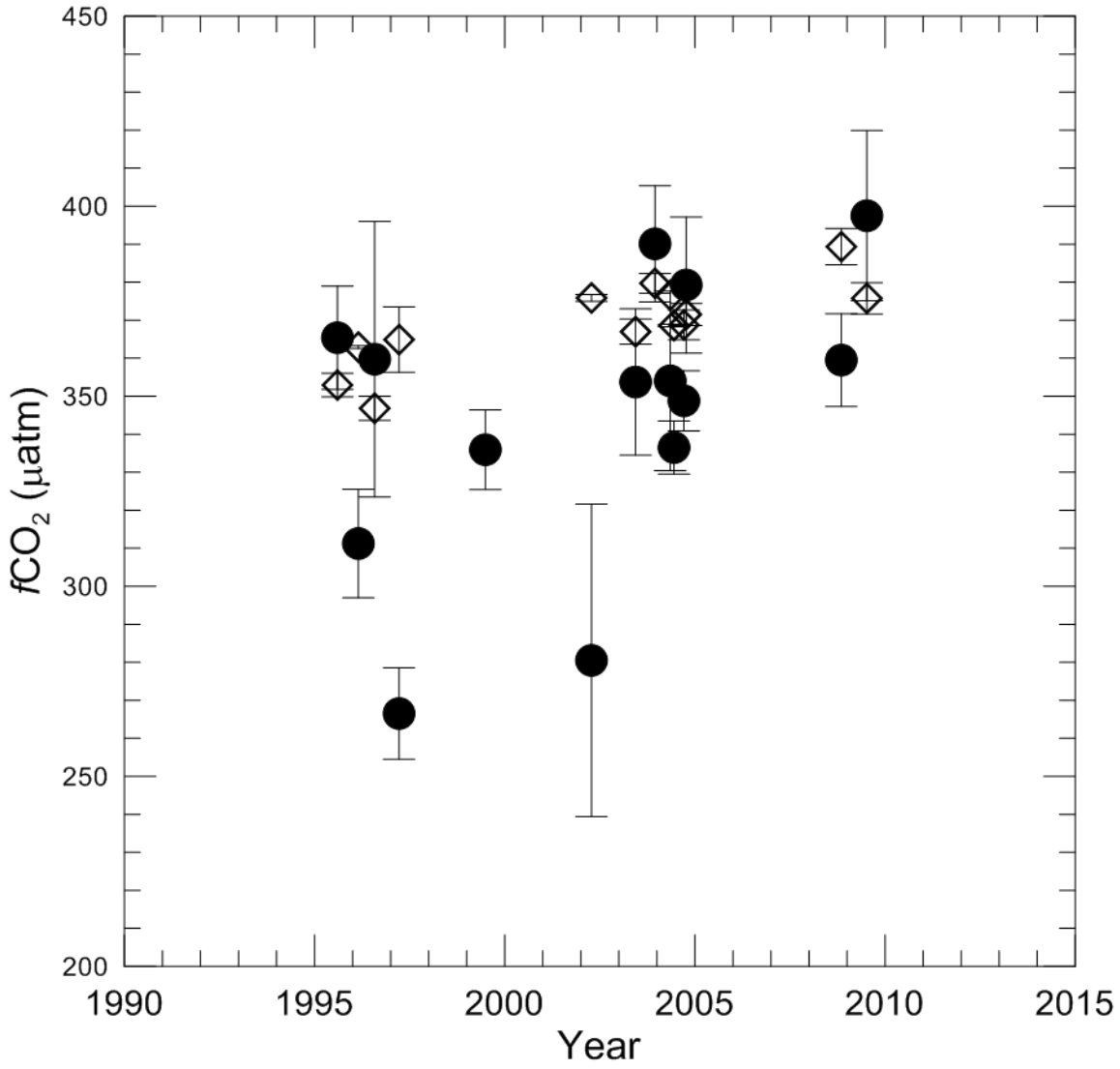

Figure 2. The secular variations in fCO_2^{sea} (filled circles) and fCO_2^{atm} (open diamonds) in the Ulleung Basin, East/Japan Sea, from 1995 to 2009. Error bars represent one standard deviation from the mean value.

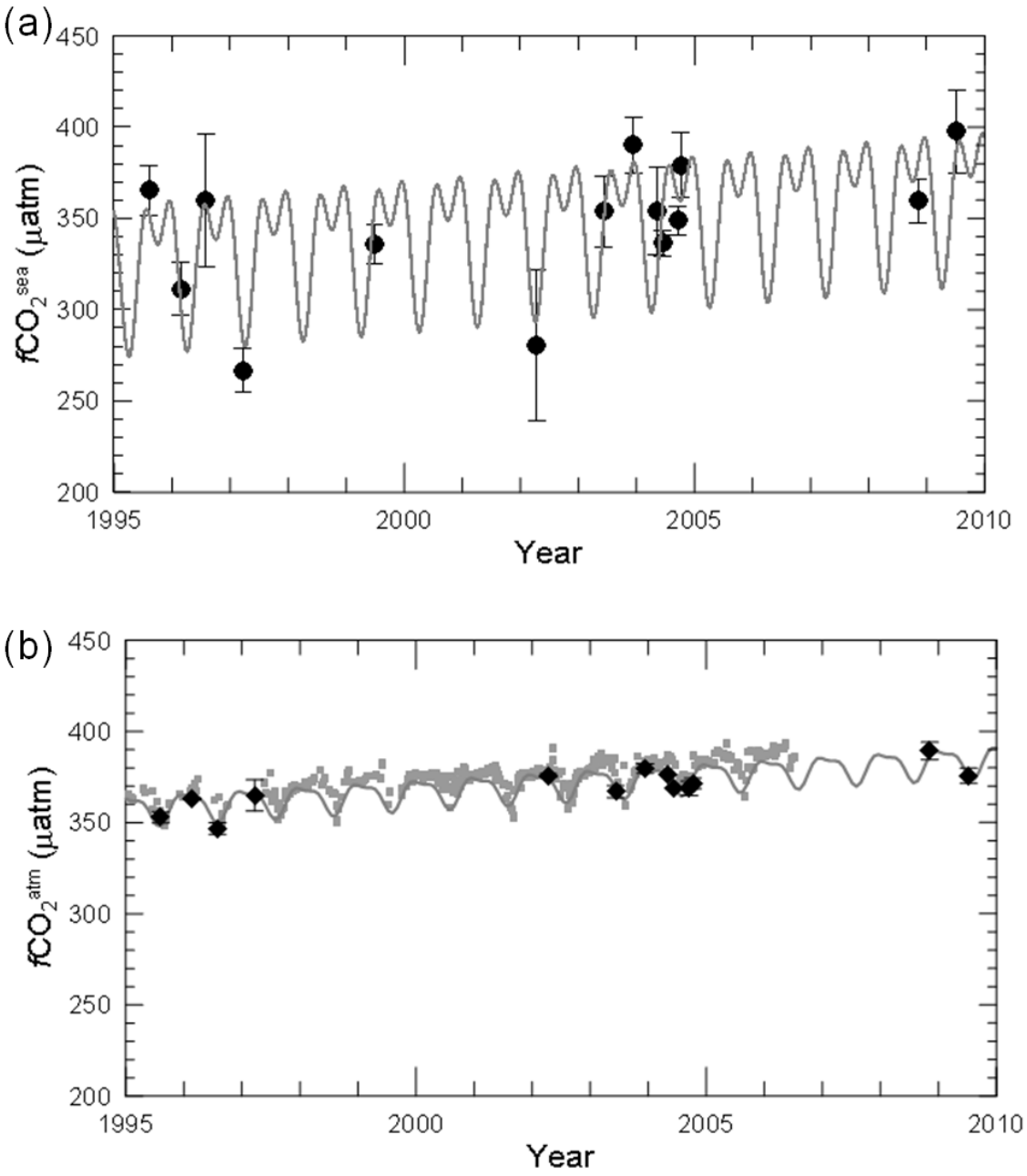

Figure 3. Long-term trend of fCO_2 at the Ulleung Basin from 1995 to 2009. (a) fCO_2^{sea} (filled circles) of surface seawater of the Ulleung Basin, (b) fCO_2^{atm} (filled diamonds) of overlying atmosphere of the Ulleung Basin. Atmospheric pCO_2 data, measured at Gosan, Jeju Island, Korea (gray dots), are also shown for comparison with fCO_2^{atm}. The gray curves represent fCO_2 fitted from a harmonic function analysis by Eq. (1).

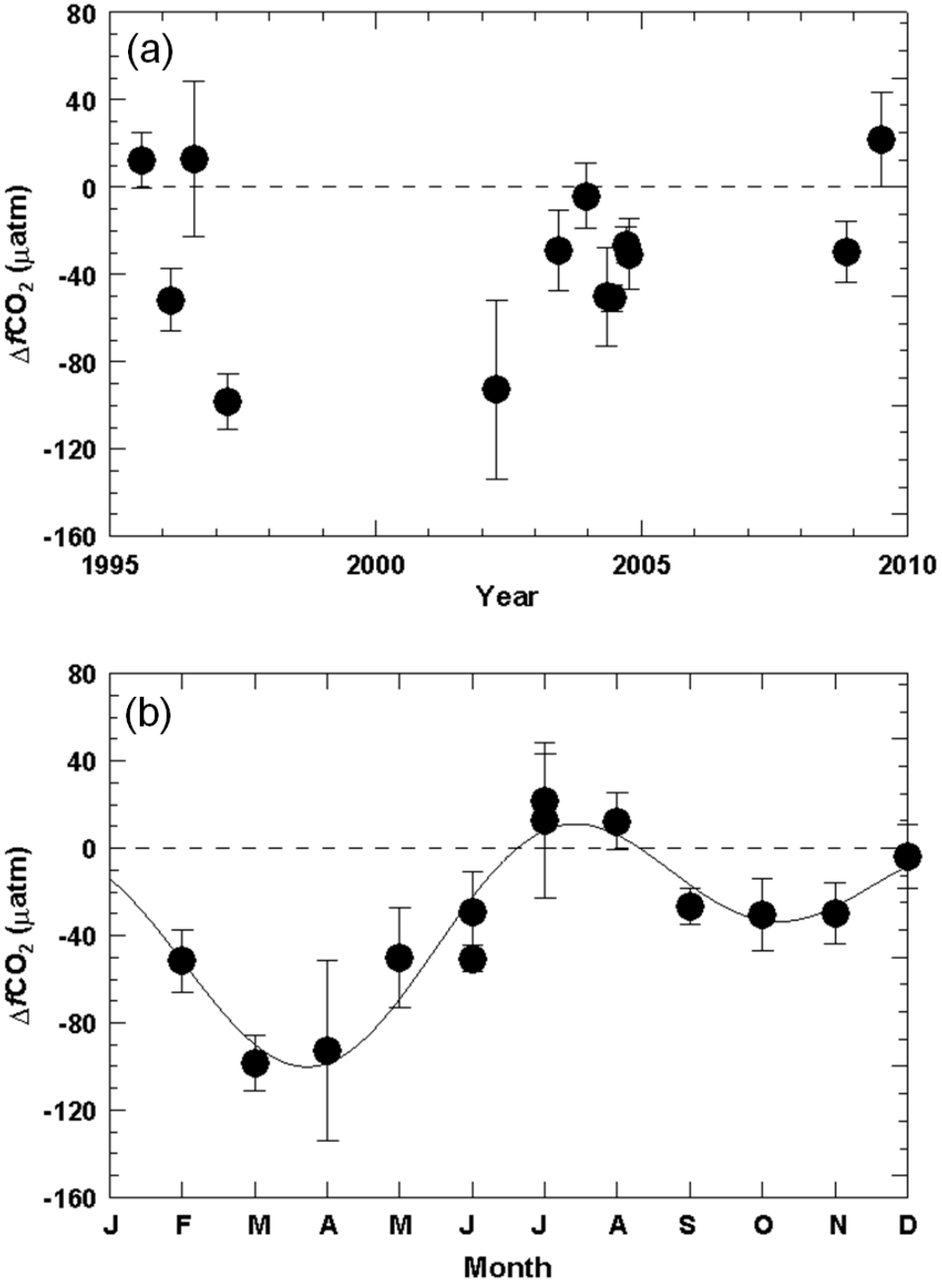

Figure 4. (a) The secular variations in ΔfCO_2 and (b) monthly variation of ΔfCO_2 in the Ulleung Basin, East/Japan Sea, from 1995 to 2009. Error bars represent one standard deviation from the mean value

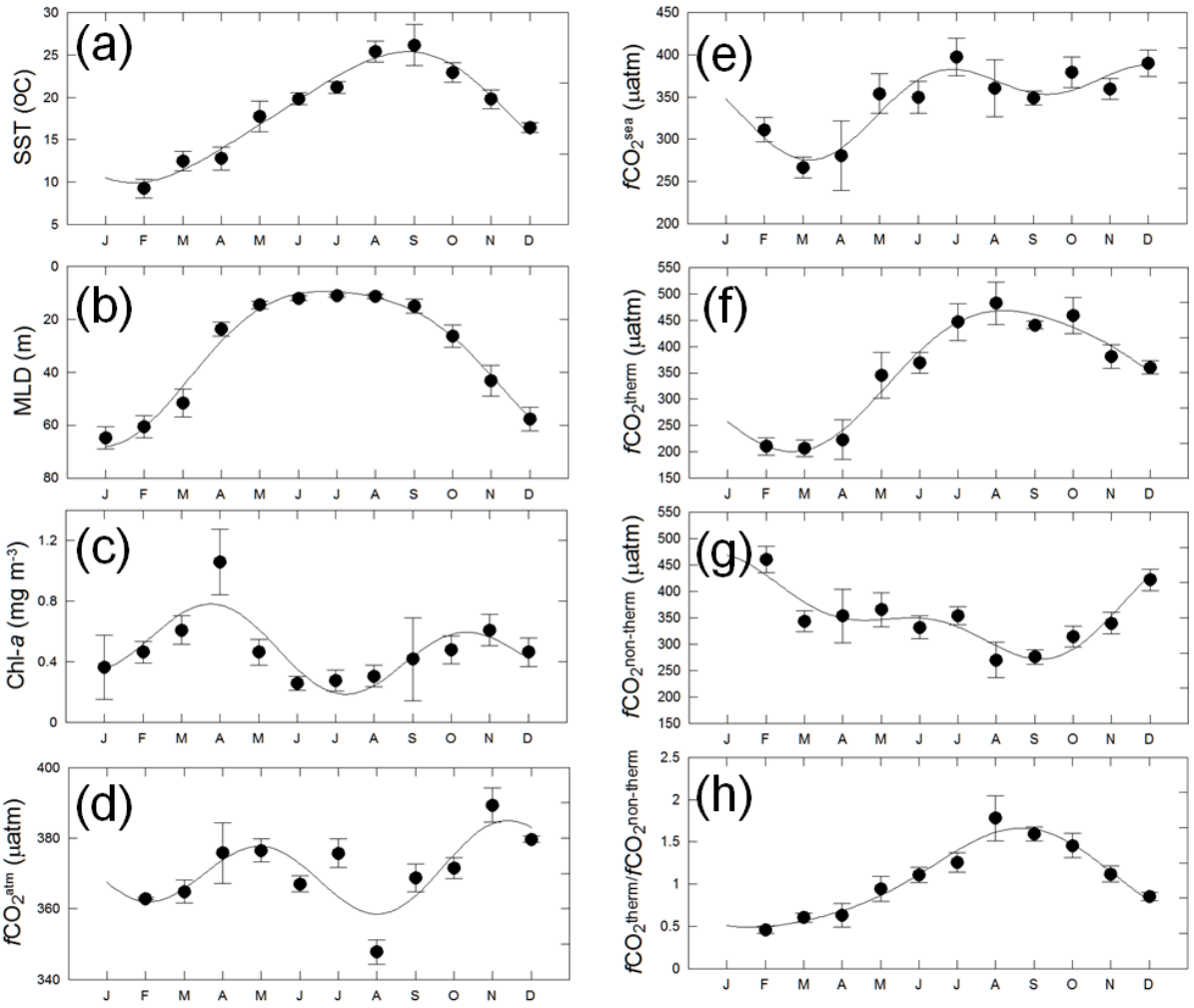

Figure 5. Monthly variations in the (a) sea surface temperature (SST), (b) mixed layer depth (MLD), (c) chlorophyll-*a* in the surface water, (d) fCO_2^{therm}, (e) $fCO_2^{non\text{-}therm}$, and (f) ratio of the thermal to non-thermal effects (fCO_2^{therm}/ $fCO_2^{non\text{-}therm}$) in the UB. The MLD was based on the climatology by de Boyer-Montégut et al. (2004). The chlorophyll-*a* was based on the SeaWiFS data between 1998 to 2007.

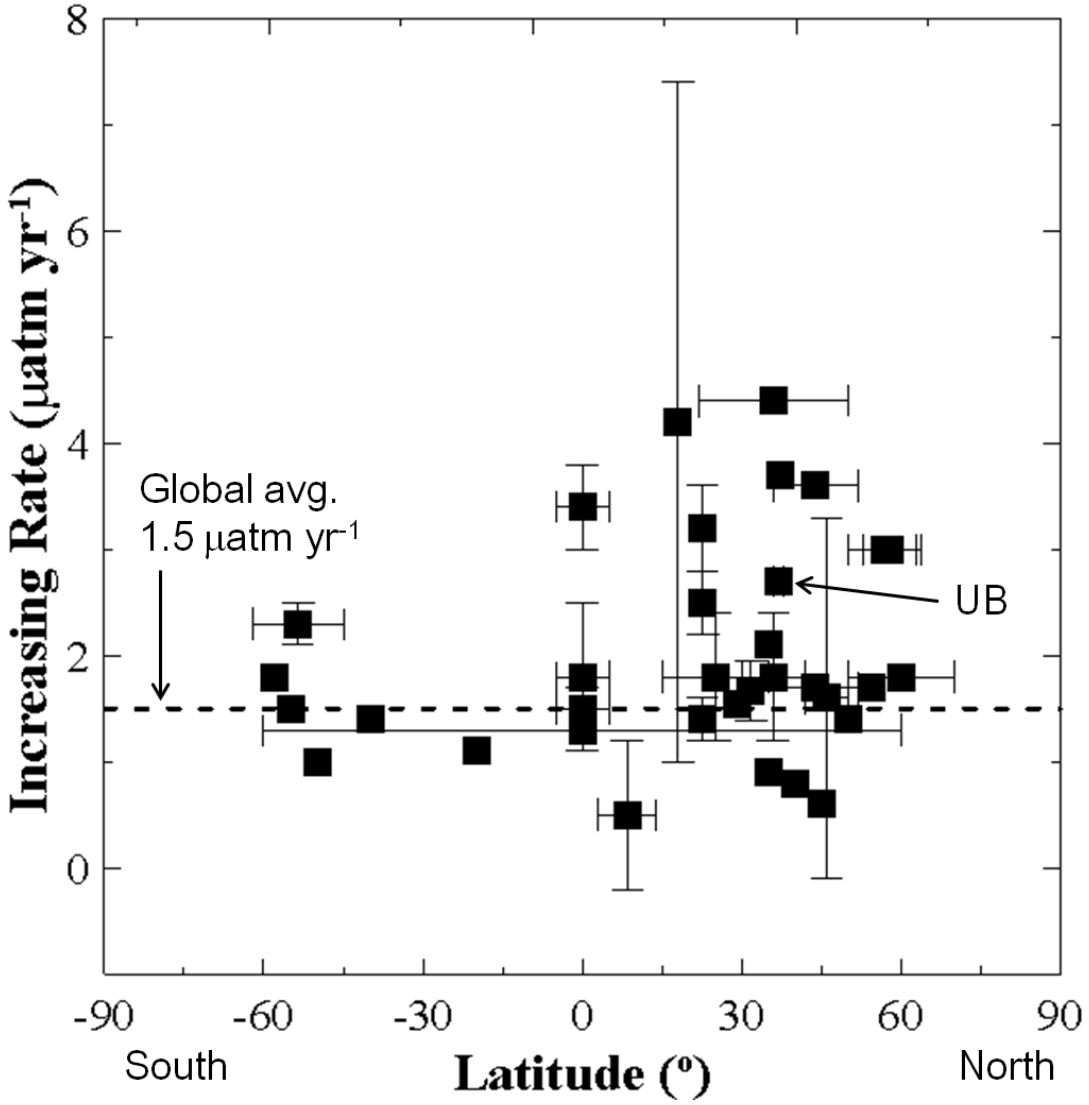

Figure 6. A plot of the rate of increase of surface seawater CO_2 versus latitude. Dashed line represents the global average of surface seawater increasing rate (1.5 μatm yr^{-1}).

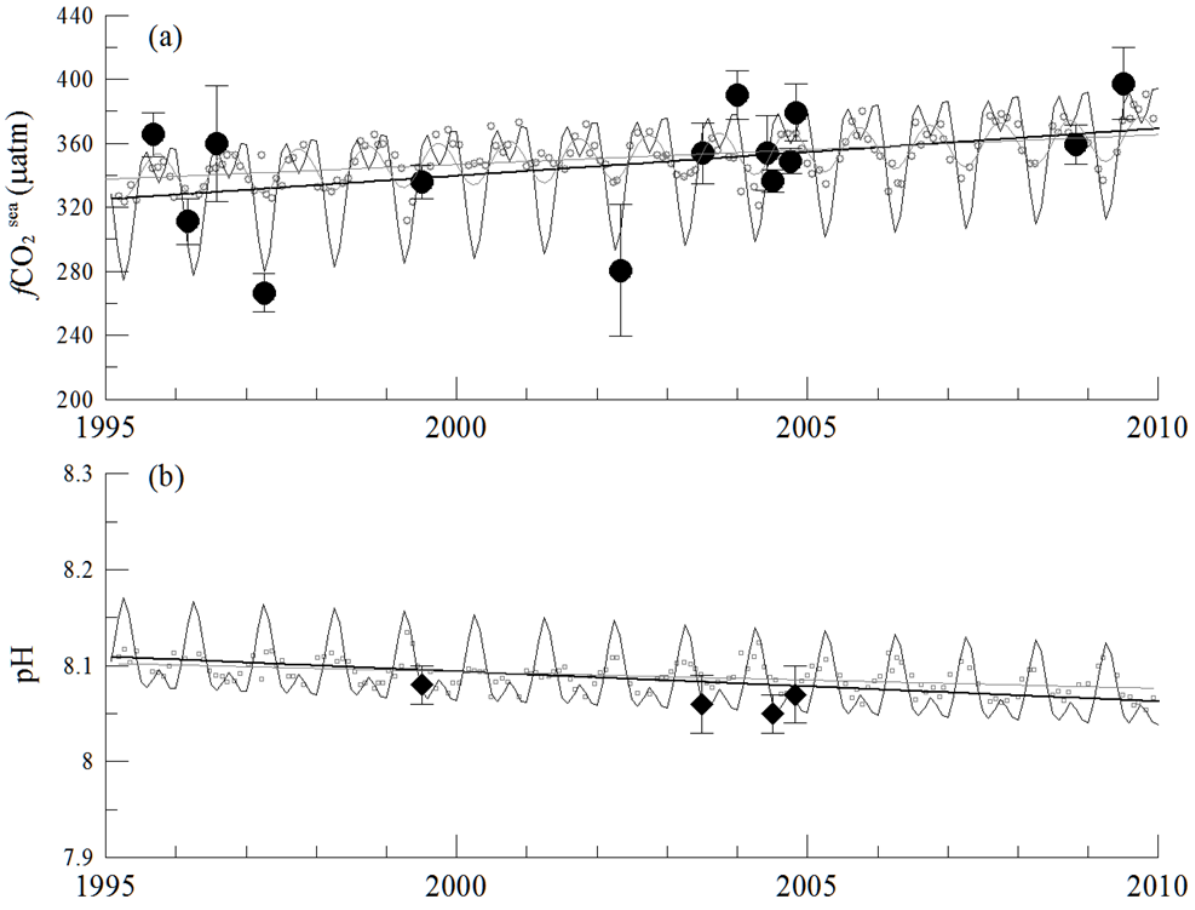

Figure 7. Secular variation of (a) fCO_2 (µatm) (black filled circles and fitting line), and (b) pH (black diamonds and fitting line) of the surface water in the UB. The time-series pCO_2 (µatm) (gray open circles) and pH (gray open squares) at Station ALOHA in the subtropical North Pacific Ocean are also shown for comparison. The pCO_2 and pH data at Station ALOHA are from D. Karl, University of Hawaii, http://hahana.soest.hawaii.edu., and are updated from Doney et al. (2009) and Dore et al. (2003).