Biogeosciences, 11, 2443–2454, 2014
www.biogeosciences.net/11/2443/2014/
doi:10.5194/bg-11-2443-2014

Biogeosciences Open Access

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

J.-Y. Kim[1], D.-J. Kang[2,3], T. Lee[1], and K.-R. Kim[4,5]

[1]Department of Oceanography, Pusan National University, Busan 609-735, South Korea
[2]Oceanographic Measurement and Instrument Calibration Service Center, Korea Institute of Ocean Science & Technology, Ansan 426-744, South Korea
[3]Marine Environmental System Science, Korea University of Science & Technology (UST), Daejeon 305-350, South Korea
[4]GIST college, Gwangju Institute of Science and Technology, Gwangju 500-712, South Korea
[5]Research Institute of Oceanography, Seoul National University, Seoul 151-742, South Korea

Correspondence to: D.-J. Kang (djocean@kiost.ac)

Received: 5 April 2013 – Published in Biogeosciences Discuss.: 14 June 2013
Revised: 26 February 2014 – Accepted: 12 March 2014 – Published: 5 May 2014

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 the CO_2 increase and ocean acidification in the surface waters of the Ulleung Basin (UB) of the East/Japan Sea, and possible causes are discussed. Fourteen observations of surface fCO_2 were made in the period from 1995 to 2009. The contribution of temperature variation to the seasonality of fCO_2 was almost equivalent to the non-thermal effect in the UB. However, the difference of relative contribution with the season makes two seasonal peaks of fCO_2 in the surface water of the UB. Non-thermal effect contributed to the surface fCO_2 drawdown in summer, whereas the surface fCO_2 elevation in winter. The decadal trend of fCO_2 increment was estimated by harmonic analysis. The estimated rates of increase of fCO_2 were $1.8 \pm 0.4\,\mu atm\,yr^{-1}$ for the atmosphere and $2.7 \pm 1.1\,\mu atm\,yr^{-1}$ for the surface water. The ocean acidification trend, calculated from total alkalinity and fCO_2, was estimated to be -0.03 ± 0.02 pH units decade^{-1}. These rates seem to be higher than observations at most other ocean time-series sites during the same period of time. Sustained observations are required to understand more accurate trend in this area.

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 °C 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 °C 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., 2009) 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 a 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 timescale 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 14 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 s and, after statistical treatment for 1 min or 2 min, averaged data were obtained and used as the final data. Three different standard gases were used to calibrate the NDIR. Every 12 h, a series of standard gases was analyzed for calibration; one of the standard gases was analyzed every 6 h to check for drift of the machine. Atmospheric and surface seawater pCO_2 were analyzed every 15 min and 45 min 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 an average value of prior and posterior fCO_2^{atm} data to fCO_2^{sea} data, since we do not 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 on board by a potentiometric titration method using a closed cell (Millero et al., 1993). Total alkalinity was calibrated by certified reference materials provided by A. Dickson of Scripps Institute of Oceanography, 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). pH values were calculated at in situ sea surface temperature.

2.2 Air–sea flux estimation

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

$$\text{Flux} = k \times s \times \Delta f,$$

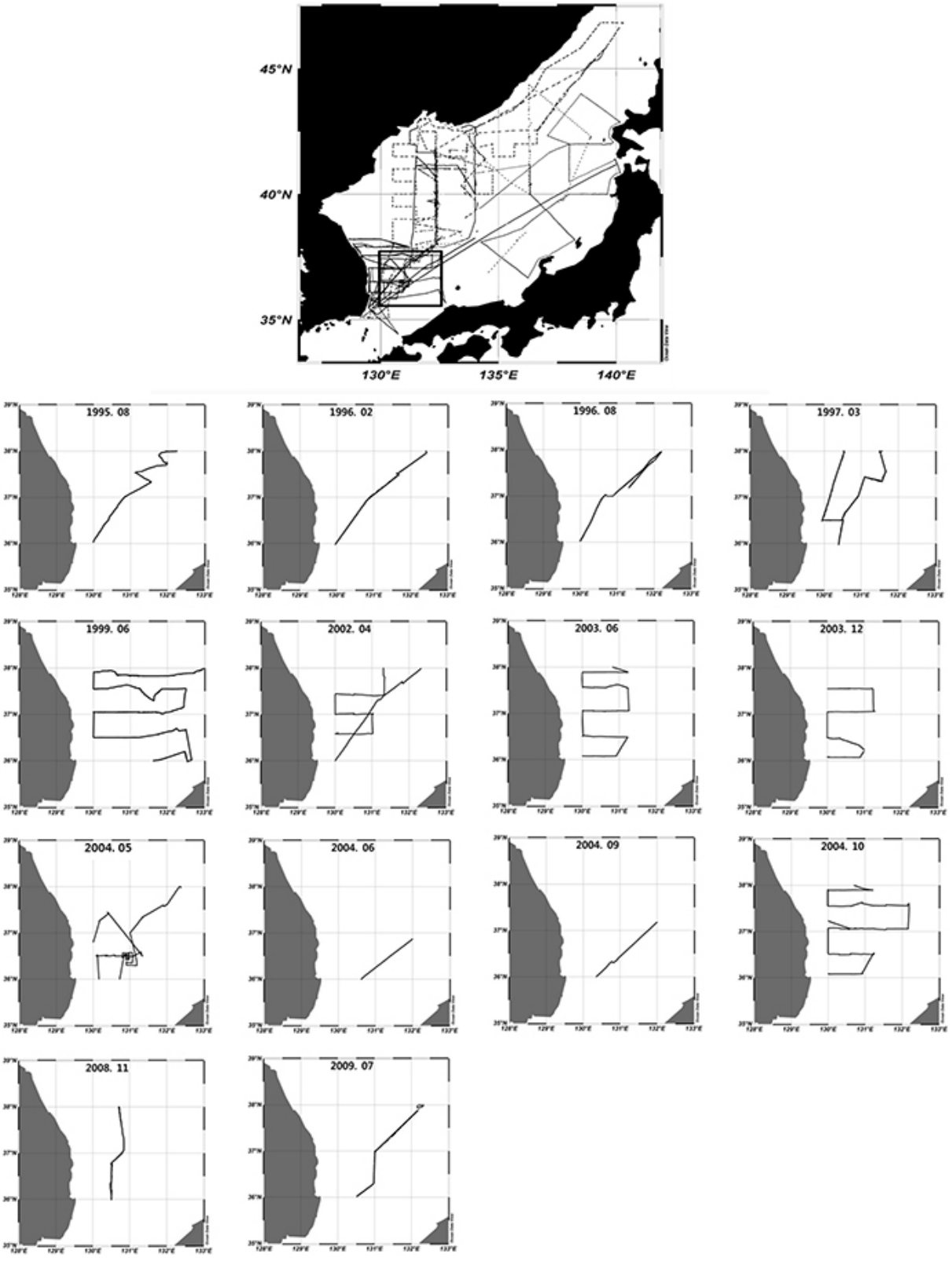

Fig. 1. Maps showing the tracks of the cruises conducted between 1995 and 2009 for this study.

Table 1. Information for cruises in this study: SST, SSS, 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 or 2 min averaged values in the cruise. The number of data values IS listed in parentheses.

Cruise name	Observation period	Research vessel	SST (°C)	SSS	fCO_2^{atm} (µatm)	fCO_2^{sea} (µatm)	ΔfCO_2 (µatm)	TA (µeq kg^{-1})	pH	Source
9508	6–7 Aug 1995	RV *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	23–24 Feb 1996	RV *Pavel 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	RV *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	RV *Pavel 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	RV *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	12–19 Apr 2002	RV *Gagarinsky*	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	9–14 Jun 2003	RV *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	12–22 Dec 2003	RV *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	6–19 May 2004	RV *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	10–11 Jun 2004	RV *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	14–15 Sep 2004	RV *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	5–29 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	1–7 Nov 2008	RV *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	9–10 Jul 2009	RV *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	

where k is the gas transfer velocity (cm h^{-1}), and s is the solubility of CO_2 gas in seawater (mol kg^{-1} atm^{-1}; Weiss, 1974). We choose the long-term scale formulas for k and the wind speed relationships used by Wanninkhof (1992). The NCEP wind speed data (http://www.cdc.noaa.gov/cdc/reanalysis) averaged on monthly scale in 36–38° N latitude and 130–133° E longitude were 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 1-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 (RMSD = 17.22 µatm, $R^2 = 0.78$ for seawater, RMSD = 2.02 µatm, $R^2 = 0.96$ for air; Fig. S1 in the Supplement), 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).

Uncertainties for the annual trend were estimated by the equation $\pm[\sigma^2/(\Sigma(X_i^2) - N(X_{mean})^2)]^{1/2}$, where $\sigma^2 =$

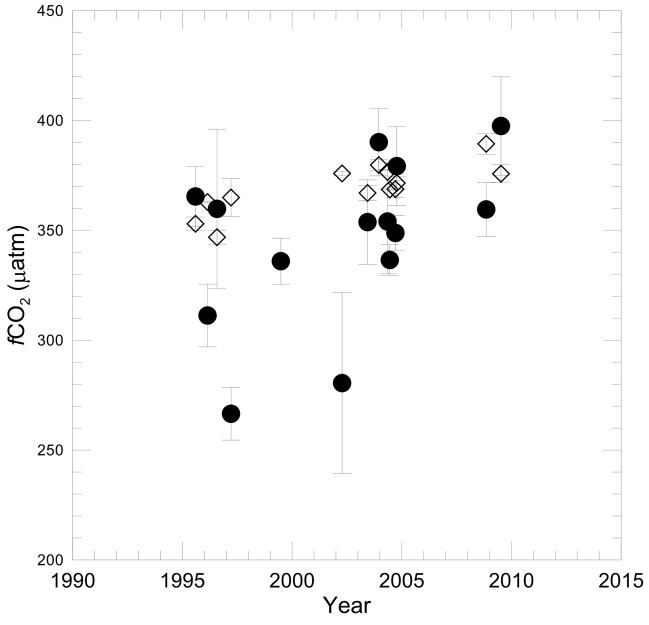

Fig. 2. The secular variations in fCO_2^{sea} (filled circles) and fCO_2^{atm} (open diamonds) in the Ulleung Basin, East/Japan Sea, from 1995 to 2009. Error bars represent one standard deviation from the mean value.

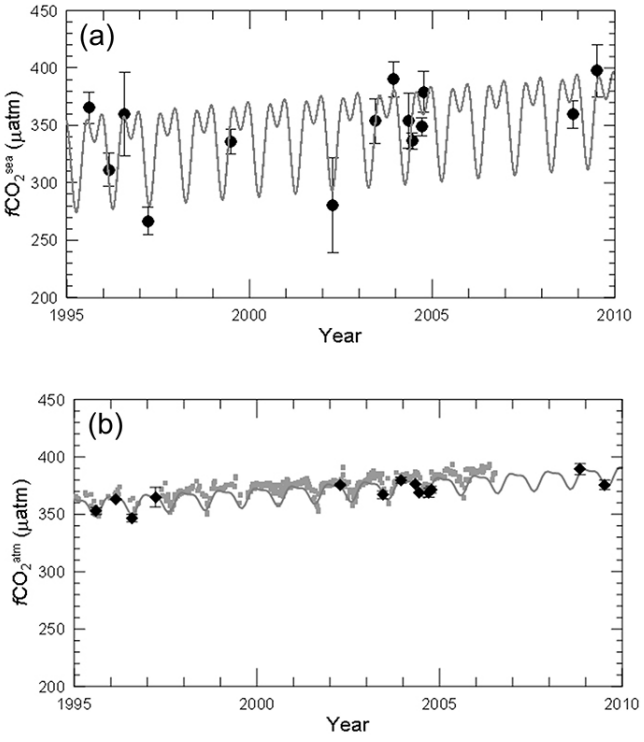

Fig. 3. Long-term trend of fCO_2 at the Ulleung Basin from 1995 to 2009. **(a)** fCO_2^{sea} (filled circles) of surface seawater of the Ulleung Basin, **(b)** fCO_2^{atm} (filled diamonds) of overlying atmosphere of the Ulleung Basin. Atmospheric pCO_2 data, measured at Gosan, Jeju Island, South Korea (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).

$[(\Sigma(Y_i\text{-}Y_i^{'})^2)/(N-2)]$ is the variance around the fitted Eq. (1), and Y is fCO_2, X is year, and N is number of data (Takahashi et al., 2009). Among the results, the c_5 value, which means annual trend for fCO_2^{sea}, was estimated to be 2.7 ± 1.1 µatm yr^{-1} and that for the fCO_2^{atm} was 1.8 ± 0.4 µatm yr^{-1} (see Fig. S1 in the Supplement).

The secular variation of ΔfCO_2 is shown in Fig. 4a. Although the fCO_2^{sea} and fCO_2^{atm} vary 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 exception 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 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 $1.7(\pm 0.3)$-fold higher than global ocean (-0.51 mol m^{-2} yr^{-1}, Takahashi et al., 2002). 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 to 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 that 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; that is, 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

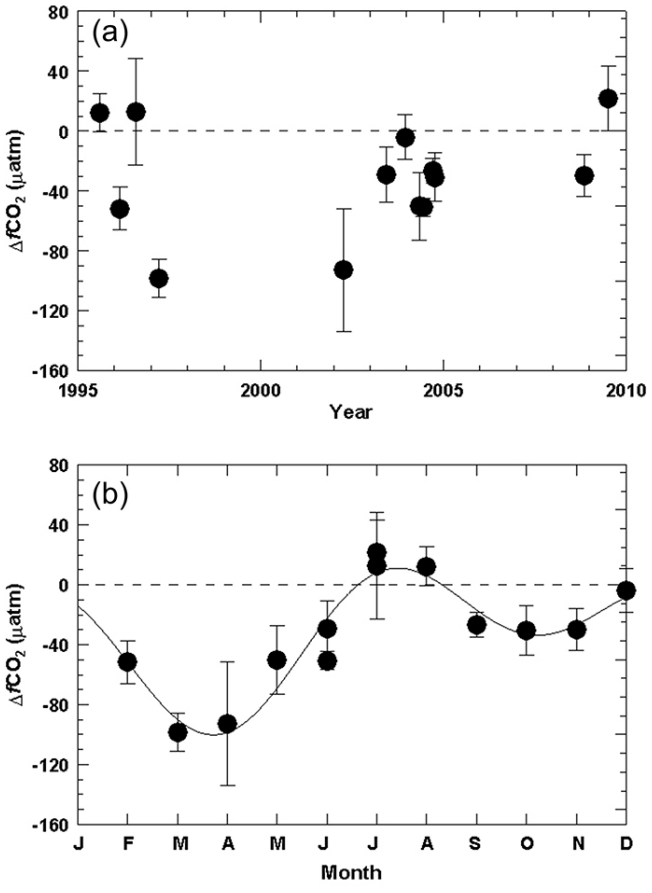

Fig. 4. (a) The secular variations in ΔfCO_2 and **(b)** monthly variation of ΔfCO_2 in the Ulleung Basin, East/Japan Sea, from 1995 to 2009. Error bars represent one standard deviation from the mean value.

of the fCO_2^{sea} in February–June and September–November was lower than the fCO_2^{air}, but that in July and August was above the fCO_2^{air} (Fig. 4b). The fCO_2^{sea} in December data appeared close to the fCO_2^{air}. 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 $\sim 100\,\mu mol\,kg^{-1}$ 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).

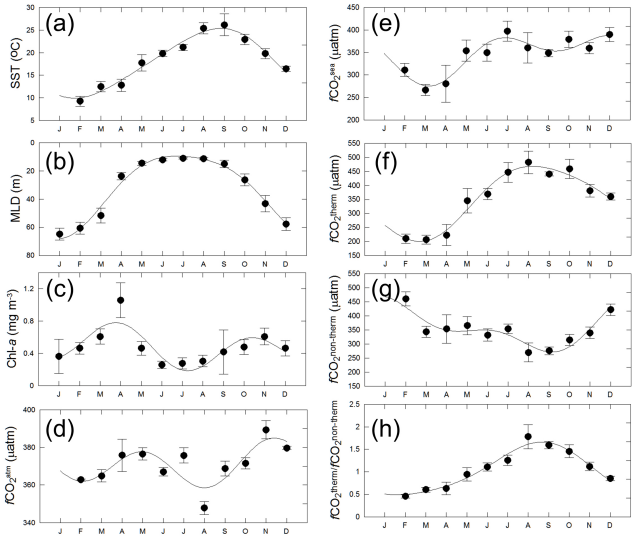

Fig. 5. Monthly variations in the **(a)** sea surface temperature (SST), **(b)** mixed layer depth (MLD), **(c)** chlorophyll *a* in the surface water, **(d)** fCO_2^{atm}, **(e)** fCO_2^{sea}, **(f)** fCO_2^{therm}, **(g)** $fCO_2^{non-therm}$, and **(h)** ratio of the thermal to non-thermal effects ($fCO_2^{therm}/fCO_2^{non-therm}$) 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 and 2007.

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:

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

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

$$\Delta fCO_2{}^{\text{therm}} = fCO_{2\,\max}{}^{\text{therm}} - fCO_{2\,\min}{}^{\text{therm}} \quad (4)$$

$$\Delta fCO_2{}^{\text{non-therm}} = fCO_{2\,\max}{}^{\text{non-therm}} - fCO_{2\,\min}{}^{\text{non-therm}} \quad (5)$$

where SST is the surface seawater temperature in °C, and the $\overline{fCO_2}$ and $\overline{\text{SST}}$ 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 ($fCO_2^{non-therm}$) effects was separated by the equations above (Eqs. 2 and 3). The results

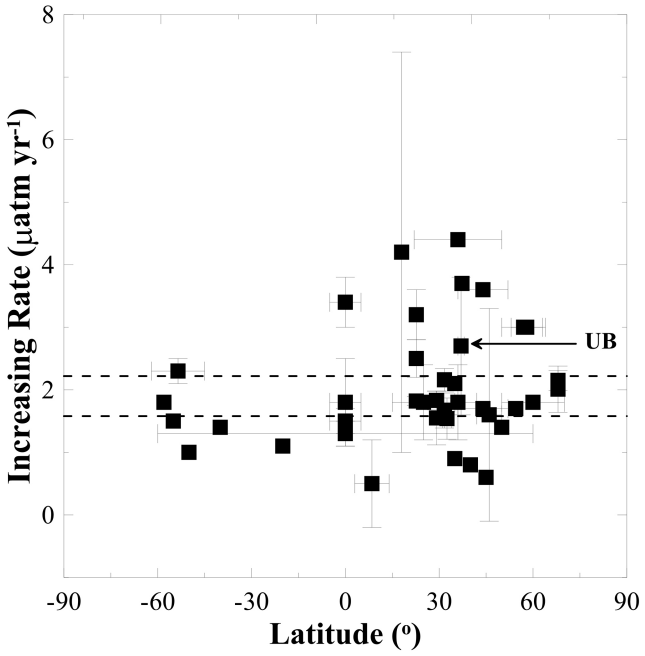

Fig. 6. A plot of the rate of increase of surface seawater CO_2 versus latitude. Dashed lines represent the range of long-term trend at major ocean time-series stations (Rhein et al., 2013).

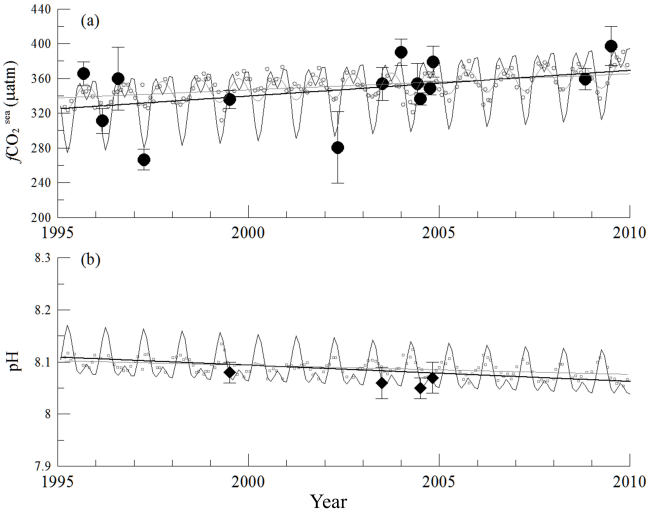

Fig. 7. Secular variation of **(a)** fCO_2 (μatm) (black filled circles and fitting line), and **(b)** pH (black diamonds and fitting line) of the surface water in the UB. The time-series pCO_2 (μatm) (gray open circles) and pH (gray open squares) at Station ALOHA in the subtropical North Pacific Ocean 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).

are shown in Fig. 5f and g. The fCO_2^{therm} followed the same pattern as the seasonal SST variation (Fig. 5a and f). The peak-to-peak amplitudes of the seasonal cycles of SST and fCO_2^{therm} ($\Delta fCO_2^{\text{therm}}$, Eq. 4) were $\sim 17\,^{\circ}$C and 275 μatm, respectively, which means that an $1\,^{\circ}$C temperature change makes a 16 μatm change in fCO_2. However, the $\Delta fCO_2^{\text{therm}}$ was larger than that observed at Station "P" (100 μatm; Wong and Chan, 1991) and at BATS (150 μatm, Bates, 2001), which are located at the comparable latitude as the UB. Larger seasonal variation of SST ($> 17\,^{\circ}$C) may have caused such a difference.

On the other hand, $fCO_2^{\text{non-therm}}$ 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 $fCO_2^{\text{non-therm}}$ 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 $fCO_2^{\text{non-therm}}$ value of more than 100 μatm in spring was attributed to carbon fixation by the spring bloom (Fig. 5e and c). In summer, the value decreased because of weakening of vertical mixing caused by stratification (Fig. 5b and e).

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 ($\Delta fCO_2^{\text{therm}}$ $/\Delta fCO_2^{\text{non-therm}} = 1.4$) 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) on Jeju Island, WMO Global Atmosphere Watch Station (Cho et al., 2005), is also

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)

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^{atm} in the UB were 1.9 ppm yr^{-1} and 1.8 µatm yr^{-1}, respectively. These values are comparable to the several ocean time-series data (between 1.88 ± 0.02 µatm yr^{-1} and 1.92 ± 0.01 µatm yr^{-1}) during same period (Rhein et al., 2013).

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^{sea} in the UB with other regions, previously published data covering more than 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 ± 1.1 µatm yr^{-1}, which is in the middle of the observed range of increasing rates in the mid-latitude of the Northern Hemisphere (Fig. 6). Although it has been reported for some regions of the oceans that the rate of fCO_2 change is dependent on the period of observations (Ishii et al., 2009; McKinley et al., 2011; Midorikawa et al., 2012), the increasing rate in most areas is close to the long-term trend at major ocean time-series stations (between 1.58 ± 0.07 µatm yr^{-1} and 2.22 ± 0.11 µatm yr^{-1}; Rhein et al., 2013). However the rate in the mid-latitudes of the Northern Hemisphere is higher. Compared to observations from the same period of time, the increasing rate of fCO_2 in the UB that we found appears to be higher than most (Table 3) and warrants further investigation.

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 were 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, secular trends, and the relationships between total alkalinity with salinity and temperature (Lee et al., 2006) were not significant (see Fig. S2 in the Supplement). 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, has changed at the rate of -0.03 ± 0.02 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 other ocean time-series stations, 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 ~ 2.16 µatm yr^{-1}, and mixed layer pH has declined by ~ 0.02 pH units per decade (Fig. 7). Rhein et al. (2013) reported that the increasing rates of fCO_2 in the seawater were in range of 1.58 ± 0.07 µatm yr^{-1} and 2.22 ± 0.11 µatm yr^{-1}, and the rates of decrease of pH were estimated between 0.015 ± 0.001 pH units decade^{-1} and 0.022 ± 0.004 pH units decade^{-1} based on long-term ocean time-series station data in the North Atlantic and North Pacific oceans. Compared with those data, the trend of CO_2 and ocean acidification in the surface seawater of the UB seems to be slightly faster as the faster warming trend than the global one in this area (Kim et al, 2007; Yeh et al., 2010).

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 contributed 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 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.

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

Region	Increasing Rate (μatm yr^{-1})	Period	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°–35°N, 132°–142° E	Inoue et al. (1995)
Western North Pacific	0.5 ± 0.7	1984–1993	3°–14°N, 132°–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°–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)
Coast of western North Pacific	1.54 ± 0.33	1994–2008	31 °–34° N, 136°–140° E	Ishii et al. (2011)
North Atlantic gyre	4.4	1994–2005	22°–50° N, 5°–70° W	Schuster and Watson (2007)
Eastern North Atlantic	3.6	1995–2002	36°–52° N, 10°–35° W	Lüger et al. (2004, 2006)
Western North Atlantic	1.7	1995–2002	36°–52° N, 36°–70° W	Lüger et al. (2004, 2006)
North Atlantic Subpolar Gyre	1.8	1982–1998	50° N–70° N, 80°–10° W	Lefévre et al. (2004)
Eastern Subpolar North Atlantic	3.0	1970s–1980s	50°–64° N, 32°–10° W	Omar and Olsen (2006)
Western Subpolar North Atlantic	3.0	1994–2003	53°–62° N, 20°–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°–62° S, circumpolar	Lenton et al. (2012)
South of Australia (Sub-Antarctic Zone)	1		50° S, 140°–160° E	Inoue and Ishii (2005)
South of Australia (Polar Frontal Zone)	1.5		55° S, 140°–160° E	Inoue and Ishii (2005)
South of Australia (Polar Zone)	1.8		58° S, 140°–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	1.82 ± 0.07	1988–2009	22.7° N, 158° W	Rhein et al. (2013)
SEATS	4.2 ± 3.2	1999-2003	18° N, 116° E	Tseng et al. (2007)
ESTOC	1.55	1995–2004	29.16° N, 15.5° W	Santana-Casino et al. (2007)
ESTOC	1.83 ± 0.15	1995–2009	29.16° N, 15.5° W	Rhein et al. (2013)
BATS	1.67 ± 0.28	1983–2005	31.7° N, 64.5° W	Bates (2007)
BATS	2.16 ± 0.18	1995–2009	31.7° N, 64.5° W	Rhein et al. (2013)
IS	2.15 ± 0.16	1985–2006	68.0° N, 12.67° W	Olafsson et al. (2009)
IS	2.01 ± 0.37	1985–2009	68.0° N, 12.67° W	Rhein et al. (2013)
UB	2.7 ± 1.1	1995–2009	36°–38° N, 130°– 133° E	This study

Supplementary material related to this article is available online at http://www.biogeosciences.net/11/2443/2014/bg-11-2443-2014-supplement.pdf.

Acknowledgements. The authors appreciate the editor and reviewers for invaluable and fruitful comments and suggestions. We also would like to express sincere appreciation to captains and crews of RVs *Roger Revelle*, *Professor Gagarinsky*, *Akademik Lavrentiev*, *Tamgu-1*, *Tamgu-5*, and *Eardo*. This research was a part of the project titled "East Asian Seas Time series-I (EAST-I)", funded by the Ministry of Oceans and Fisheries, South Korea (PM57520).

Edited by: K.-K. Liu

References

Bates, N.: Interannual variability of oceanic CO_2 and biogeochemical properties in the Western North Atlantic subtropical gyre, Dee-Sea Res. II, 48, 1507–1528, 2001.

Bates, N.: Interannual variability of the oceanic CO_2 sink in the subtropical gyre of the North Atlantic Ocean over the last 2 decades, J. Geophys. Res., 112, C09013, doi:10.1029/2006JC003759, 2007.

Borges, A. V., Delille, B., and Frankignoulle, M.: Budgeting sinks and sources of CO_2 in the coastal ocean: Diversity of ecosystems counts, Geophys. Res. Lett., 32, L14601, doi:10.1029/2005GL023053, 2005.

Cai, W.-J. and Dai, M.: Comment on "Enhanced Open Ocean Storage of CO_2 from Shelf Sea Pumping", Science, 306, 1477, doi:10.1126/science.1102132, 2004.

Caldeira, K. and Wickett, M. E.: Anthropogenic carbon and ocean pH, Nature, 425, 365 pp., 2003.

Casey, K. S. and Cornillon, P.: Global and regional sea surface temperature trends, J. Climate, 14, 3801–3818, 2001.

Chierici, M., Fransson, A., and Nojiri, Y.: Biogeochemical processes as drivers of surface fCO_2 in contrasting provinces in the subarctic North Pacific Ocean, Glob. Biogeochem. Cy., 20, GB1009. doi:10.1029/2004GB002356, 2006.

Cho, C., Oh, S.-N., Park, K.- J., Choi, B. –C., Park, M.–K., and Kim, K.-R.: The distribution characteristics of atmospheric CO_2 in the Korean Peninsula and the expectations on its annual growth trend by the year 2015, Asia-Pacific J. Atm. Sci., 41, 371–385, 2005.

Choi, S. H., Kim, D., Shim, J. H., and Min, H. S.: The spatial distribution of surface fCO_2 in the Southwestern East Sea/Japan Sea during summer 2005, Ocean. Sci. J., 46, 13–21, 2011.

Choi, S. H., Kim, D., Shim, J. H., Kim, K.-H., and Min, H. S.: Seasonal variations of surface fCO_2 and sea-air fluxes in the Ulleung Basin of the East/Japan Sea, Terr. Atmos. Ocean. Sci., 23, 343–353, 2012.

Clayton, T. D. and Byrne, R. H.: Spectrophotometric seawater pH measurements: Total hydrogen ion concentration scale calibration of m-cresol purple and at sea results, Deep-Sea Res., 40, 2115–2129, 1993.

Corbiere, A., Metzl, N., Reverdin, G., Brunet, C., and Takahashi, T.: Interannaul and decadal variability of the oceanic carbon sink in the North Atlantic subpolar gyre, Tellus, 59B, 168–178, 2007.

de Boyer-Montégut, C., Madec, G., Fisher, A. S., Lazar, A., and Iudicone, D.: Mixed layer depth over the global ocean: An examination of profile data and a profile-based climatology, J. Geophys. Res., 109, C12003, doi:10.1029/2004JC002378, 2004.

Dickson, A. G. and Millero, F. J.: A comparison of the equilibrium constants for the dissociation of carbonic acid in seawater media, Deep-Sea Res., 34, 1733–1743, 1987.

Doney, S., Fabry, V. J., Feely, R. A., and Kleypas, J. A.: Ocean acidification: the other CO_2 problem, Ann. Rev. Mar. Sci., 1, 169–192, 2009.

Dore, J. E., Lukas, R., Sadler, D. W., and Karl, D. M.: Climate driven changes to the atmospheric CO_2 sink in the subtropical North Pacific Ocean, Nature, 424, 754–757, 2003.

Gieskes, J. M.: Effect of temperature on the pH of seawater, Limnol. Oceanogr., 14, 679–685, 1969.

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

Inoue, H. Y. and Ishii, M.: Variations and trends of CO_2 in the surface seawater in the Southern Ocean south of Australia between 1969 and 2002, Tellus B, 57, 58–69, 2005.

Intergovernmental Panel on Climate Change (IPCC): Climate change 2007: The physical science basis, Cambridge University Press, 996 pp., 2007.

Ishii, M., Inoue, H., Midorikawa, T., Saito, S., Tokieda, T., Sasano, D., Nakadate, A., Nemoto, K., Metzl, N, Wong, C, S, and Feely, R. A.: Spatial variability and decadal trend of the oceanic CO_2 in the western equatorial Pacific warm/fresh water, Deep-Sea Res. II, 56. 591–606, 2009.

Ishii, M., Kosugi, N., Sasano, D., Saito, S., Midorikawa, T., and Inoue, H. Y.: Ocean acidification off the south coast of Japan: A result from time series observations of CO_2 parameters from 1994 to 2008, J. Geophys. Res. Oceans, 116, C06022, doi:10.1029/2010JC006831, 2011.

Kang, D.-J., Park, S., Kim, Y.-G., Kim, K., and Kim, K.-R.: A Moving-Boundary Box Model (MBBM) for Oceans in Change: An Application to the East/Japan Sea, Geophys. Res. Lett. 30, 1299, doi:10.1029/2002GL016486, 2003.

Keeling, C. D., Brix, H., and Gruber, N.: Seasonal and long-term dynamics of the upper ocean carbon cycle at Station ALOHA near Hawaii, Glob. Biogeochem. Cy., 18, GB4006, doi:10.1029/2004GB002227, 2004.

Key, R. M., Kozyr, A., Sabine, C. L., Lee, K., Wanninkhof, R., Bullister, J., Feely, R. A., Millero, F., Mordy, C., and Peng, T. –H.: A global ocean carbon climatology: Results from GLODAP, Glob. Biogeochem. Cy., 18, GB4031, doi:10.1029/2004GB002247, 2004.

Kim, K., Kim, K.-R., Chung, J. Y., Yoo, H. S., and Park, S. G.: Characteristics of physical properties in the Ulleung Basin, J. Korean Soc. Oceanogr., 26, 83–100, 1991.

Kim, K., Kim, K. -R., Min, D., Volkov, Y., Yoon, J. –H., and Takematsu, M.: Warming and Structural Changes in the East Sea (Japan Sea): A Clue to the future Changes in Global Oceans?, Geophys. Res. Lett., 28, 3293–3296, 2001.

Kim, K.-R. and Kim, K.: What is happening in the East Sea (Japan Sea)?: Recent chemical observation during CREAMS 93–96, J. Korean Soc. Oceanogr., 31, 164–172, 1996.

Kim, S., Zhang, C.-I., Kim, J.-Y., Oh, J.-H., Kang, S., and Lee, J. B.: Climate variability and its effects on Major Fisheries in Korea, Ocean Sci. J., 42, 179–192, 2007.

Lee, K., Tong, L. T., Millero, F. J., Sabine, C. L., Dickson, A. G., Goyet, C., Park, G.-H., Wanninkof, R., Feely, R. A., and Key, R. M.: Global relationships of total alkalinity with salinity and temperature in surface waters of the world's oceans, Geophys. Res. Lett., 33, L19605, doi:10.1029/2006GL027207, 2006.

Lefévre, N., Watson, A. J., Olsen, A., Rios, A. F., and Pérez, F. F.: A decrease in the sink for atmospheric CO_2 in the North Atlantic, Geophys. Res. Lett., 31, L07306, doi:10.1029/2003GL018957, 2004.

Lenton, A., Metzl, N., Takahashi, T., Kuchinke, M., Matear, R. J., Roy, T., Sutherland, S. C., and Tilbrook, B.: The observed evolution of oceanic pCO_2 and its drivers over the last two decades, Glob. Biogeochem. Cy., 26, GB2021, doi:10.1029/2011GB004095, 2012.

Lewis, E. and Wallace, D. W. R.: CO2SYS—Program developed for the CO_2 system calculations, Carbon Dioxide Information Analysis Center, Oak Ridge, p. 31, 1998.

Levitus, S., Antonov, J., and Boyer, T.: Warming of the world ocean, 1955–2003, Geophys. Res. Lett., 32, L02604, doi:10.1029/2004GL021592, 2005.

Lüger, H., Wallace, D.W. R., Körtzinger, A., and Nojiri, Y.: The pCO_2 variability in the midlatitude North Atlantic Ocean during a full annual cycle, Glob. Biogeochem. Cy., 18, GB3023, doi:10.1029/2003GB002200, 2004.

Lüger, H., Wanninkhof, R., Wallace, D. W. R., and Körtzinger, A.: CO_2 fluxes in the subtropical and subarctic North Atlantic based on measurements from a volunteer observing ship, J. Geophys. Res., 111, C06024, doi:10.1029/2005JC003101, 2006.

McKinley, G. A., Fay, A. R., Takahashi, T., and Metzl, N.: Convergence of atmospheric and North Atlantic carbon dioxide trends on multidecadal timescales, Nat. Geosci., 4, 606–610, doi:10.1038/ngeo1193, 2011.

Mehrbach, C., Culberson, C. H., Hawley, J. E., and Pytkowicz, R. M.: Measurement of the apparent dissociation constants of carbonic acid in seawater at atmospheric pressure, Limnol. Oceanogr., 18, 897–907, 1973.

Midorikawa T., Ishii, M., Kosugi, N., Sasano, D., Nakano, T., Saito, S., Sakamoto, N., Nakano, H., and Inoue, H. Y.: Recent deceleration of oceanic pCO_2 increase in the western North Pacific in winter, Geophys. Res. Lett., 39, L12601, doiL10.1029/2012GL051665, 2012.

Millero, F.J., Zhang, J.-Z., Lee, K., and Campbell, D. M.: Titration alkalinity of seawater. Mar. Chem., 44, 153–165, 1993.

Molinari, R. L., Mayer, D. A., Fester, J. F., and Bezdek, H. F.: Multiyear variability in the near-surface temperature structure of the midlatitude western North Atlantic Ocean, J. Geophys. Res., 102, 3267–3278, 1997.

Nicholls, N., Grusz, V. V., Jouzel, J., Karl, T., Ogallo, L., and Parker, D.: Observed climate variability and change. In: Houghton, J. and IPCC (Eds.), Climate Change 1995: The Science of Climate Change, Cambridge University Press, Cambridge, 133–192, 1996.

Nojiri, Y., Fujinuma, Y., Zeng, J., and Wong, C. S.: Monitoring of pCO_2 with complete seasonal coverage utilizing a cargo ship M/S Skaugran between Japan and Canada/US, Proceedings of the Second International Symposium of CO_2 in the Oceans, Tsukuba, Japan, 17–23, 1999.

Nojiri, Y. and Tsumori, H.: Ocean Carbon Source and Sinks Regional View: North Pacific: 'Data analysis of 12 years of monthly observation by the 3 VOS', Changing Times: An International Ocean Biogeochemical Time-Series Workshop, IOC/UNESCO, Paris, France, 2007.

Oh, D.-C.: A study on the characteristics of fCO_2 distributions and CO_2 flux at the air-sea interface in the seas around Korea, M. S. thesis, Seoul National University, 1998.

Oh, D.-C., Park, M.-K., Choi, S.-H., Kang, D.-J., Park, S. Y., Hwang, J. S., Andreev, A., Hong, G. H., and Kim, K.-R.: The air-sea exchange of CO_2 in the East Sea (Japan Sea), J. Oceanogr., 55, 157–169, 1999.

Olafsson, J., Olafsdottir, S. R., Benoit-Cattin, A., Danielsen, M., Arnarson, T. S., and Takahashi, T.: Rate of Iceland Sea acidification from time series measurements, Biogeosciences, 6, 2661–2668, 2009,
http://www.biogeosciences.net/6/2661/2009/.

Omar, A. M. and Olsen, A.: Reconstructing the time history of the air-sea CO_2 disequilibrium and its rate of change in the eastern subpolar North Atlantic, 1972–1989, Geophys. Res. Lett., 33, L04602, doi:10.1029/2005GL025425, 2006.

Orr, J. C., Fabry, V. J., Aumont, O., Bopp, L., Doney, S. C., Feely, R. A., Ganadesikan, A., Gruber, N., Ishida, A., Joos, F., Key, R. M., Lindsay, K., Maier-Reimer, E., Matear, R., Monfray, P., Mouchet, A., Najjar, R. G., Plattner, G. –K., Rodgers, K. B., Sabine, C. L., Sarmiento, J. L., Schlitzer, R., Slater, R. D., Totterdell, I. J., Weirig, M. –F., Yamanaka, Y., and Yool, A.: Anthropogenic ocean acidification over the twenty-first century and its impact on calcifying organisms, Nature, 437, 681–686, 2005.

Park, S. Y.: A study on the carbon cycle in the deep waters of the East Sea, M.S. thesis, Seoul National University, 1997.

Rayner, N. A., Brohan, P., Parker, D. E., Folland, C. K., Kennedy, J. J., Vanicek, M., Ansell, T. J., and Tett, S. F. B.: Improved analyses of changes and uncertainties in sea surface temperature measured in situ since the Mid-Nineteenth Century: The HadSST2 Dataset, J. Climate, 19, 446–469, 2006.

Rhein, M., Rintoul, S.R., Aoki, S., Campos, E., Chambers, D., Feely, R.A., Gulev, S., Johnson, G.C., Josey, S.A., Kostianoy, A. , Mauritzen, C., Roemmich, D., Talley, L.D., and Wang, F.: Observations: Ocean. In: Stocker, T.F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P.M. (Eds.), Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge, 255–315, 2013.

Sabine, C. L., Feely, R. A., Gruber, N., Key, R. M., Lee, K., Bullister, J. L., and Wanninkhof, R.: The ocean sink for anthropogenic CO_2, Science, 305, 367–371, 2004.

Santana-Casiano, J. M., González-Dávila, M. , Rueda, M.-J., Llinás, O., and González-Dávila, E.-F.: The interannual variability of oceanic CO_2 parameters in the northeast Atlantic subtropical gyre at the ESTOC site, Glob. Biogeochem. Cy., 21, GB1015, doi:10.1029/2006GB002788, 2007.

Sarmiento, J. L. and Gruber, N.: Ocean biogeochemical dynamics, Princeton University Press, Princeton, 2006.

Schuster, U. and Watson, A. J.: A variable and decreasing sink for atmospheric CO_2 in the North Atlantic, J. Geophys. Res., 112, C11006, doi:10.1029/2006JC003941, 2007.

Seung, Y. H. and Kim, K.: A numerical modeling of the East Sea circulation, J. Korean Soc. Oceanogr., 28, 292–304, 1993.

Sweeney, C.: Biogeochemical regimes and meso-scale variability in the Ross Sea, Antarctica, Ph. D. Thesis, Columbia University, 2000.

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

Takahashi, T., Sutherland, S. C., Sweeney, C., Poisson, A., Metzl, N., Tilbrook, B., Bates, N., Wanninkhof, R., Feely, R. A., Sabine, C., Olafsson, J., and Nojiri, Y.: Global sea– air CO_2 flux based on climatological surface ocean pCO_2, and seasonal biological and temperature effects, Deep-Sea Res. II, 49, 1601–1622, 2002.

Takahashi, T., Sutherland, S. C., Feely, R. A., and Cosca, C. E.: Decadal variation of the surface water pCO_2 in the western and central equatorial Pacific, Science, 203, 852–856, 2003.

Takahashi, T., Sutherland, S. C., Feely, R. A., and Wanninkhof, R.: Decadal change of the surface water pCO_2 in the North Pacific: a synthesis of 35 years of observations, J. Geophys. Res., 111, C07S05, doi:10.1029/2005JC003074, 2006.

Takahashi, T., Sutherland, S. C., Wanninkhof, R., Sweeney, C., Feely, R. A., Chipman, D. W., Hales, B., Friederich, G., Chavez, F., Sabine, C., Watson, A., Bakker, D. C. E., Schuster, U., Metzl, N., Inoue, H. Y., Ishii, M., Midorikawa, T., Nojiri, Y., Körtzinger, A., Steinhoff, T., Hoppema, M., Olafsson, J., Arnarson, T. S., Tilbrook, B., Johannessen, T., Olsen, A., Bellerby, R., Wong, C. S., Delille, B., Bates, N. R., and de Barr, H. J. W.: Climatological mean and decadal change in surface ocean pCO_2, and net sea-air

CO_2 flux over the global oceans, Deep-Sea Res. II, 56, 554–577, 2009.

Talley, L. D., Lovanov, V., Ponomarev, V., Salyuk, A., Tishchenko, P., Zhabin, I., and Riser, S.: Deep convection and brine rejection in the Japan Sea, Geophys. Res. Lett., 30, 4, doi:10.1029/2002GL016451, 2003.

Thomas, H., Bozec, Y., Elkalay, K., and de Baar, H. J. W.: Enhanced open ocean storage of CO_2 from shelf sea pumping, Science, 304, 1005–1008, 2004.

Tseng, C.-M., Wong, G. T. F., Chou, W.-C., Lee, B.-S., Sheu, D.-D, and Liu, K.-K: Temporal variations in the carbonate system in the upper layer at the SEATS station, Deep-Sea Res. II, 54, 1448–1468, 2007.

Volk, T. and Hoffert, M. I.: Ocean carbon pumps: analysis of relative strengths and efficiencies in ocean-driven atmospheric CO_2 changes, in: The Carbon Cycle and Atmospheric CO_2: Natural Variations Archean to Present, edited by: Sundquist, E. T. and Broecker, W. S., Geophysical Monograph 32, American Geophysical Union, Washington, DC, 99–110, 1985.

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

Watai, T., Kikuchi, M., and Nakazawa, T.: Temporal variations of surface oceanic and atmospheric CO_2 fugacity and total dissolved inorganic carbon in the northwestern North Pacific, J. Oceanogr., 54, 323–336, 1998.

Weiss, R. F.: Carbon dioxide in water and seawater: the solubility of a non-ideal gas, Mar. Chem., 2, 203–205, 1974.

Wollast, R.: Evaluation and comparison of the global carbon cycle in the coastal zone and in the open ocean, in: The Global Coastal Ocean, edited by: Brink, K. H. and Robinson, A. R., John Wiley & Sons, 213–252, 1998.

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

Yamada, K., Ishizaka, J., Yoo, S., Kim, H. C., and Chiba, S.: Seasonal and interannual variability of sea surface chlorophyll a concentration in the Japan/East (JES), Prog. Oceanogr., 61, 193–211, 2004.

Yeh, S. -W., Park, Y.-G., Min, H., Choi, C.-H., and Lee, J.-H.: Analysis of characteristics in the sea surface temperature variability in the East/Japan Sea, Prog. Oceanogr., 85, 213–223, 2010.

Yoo, S. and Park, J.: Why is the southwest the most productive region of the East Sea/Sea of Japan?, J. Mar. sys., 78, 301–315, 2009.

Zeng, J., Nojiri, Y., Murphy, P. P., Wong, C. S., and Fujinuma, Y.: A comparison of pCO_2 distributions in the northern North Pacific using results from a commercial vessel in 1995–1999, Deep-Sea Res. II, 49, 5303–5315, 2002.