# The Air-Sea Exchange of CO<sub>2</sub> in the East Sea (Japan Sea)

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During CREAMS expeditions,  $fCO_2$  for surface waters was measured continuously along the cruise tracks. The fCO2 in surface waters in summer varied in the range 320~440 μatm, showing moderate supersaturation with respect to atmospheric CO<sub>2</sub>. In winter, however, fCO2 showed under-saturation of CO2 in most of the area, while varying in a much wider range from 180 to 520  $\mu$ atm. Some very high  $fCO_2$  values observed in the northern East Sea (Japan Sea) appeared to be associated with the intensive convection system developed in the area. A gas-exchange model was developed for describing the annual variation of  $fCO_2$  and for estimating the annual flux of  $CO_2$  at the air-sea interface. The model incorporated annual variations in SST, the thickness of the mixed layer, gas exchange associated with wind velocity, biological activity and atmospheric concentration of CO2. The model shows that the East Sea releases CO2 into the atmosphere from June to September, and absorbs CO2 during the rest of the year, from October through May. The net annual  $CO_2$  flux at the air-sea interface was estimated to be 0.032 ( $\pm 0.012$ ) Gt-C per year from the atmosphere into the East Sea. Water column chemistry shows penetration of CO2 into the whole water column, supporting a short turnover time for deep waters in the East Sea.

#### Keywords:

- · Surface fCO<sub>2</sub> in the East (Japan) Sea,
- ·air-sea CO<sub>2</sub> flux,
- seasonal variability of fCO<sub>2</sub>,
- · solubility pump,
- · gas-exchange model.

### 1. Introduction

It is well known that the ocean plays a very important role in the overall biogeochemical cycle of CO<sub>2</sub> with its special pumping mechanisms such as solubility, biological, alkalinity and dynamic pumps (for the most recent summary, see IPCC, 1995). Thus, many studies on global changes in recent years have placed emphasis on quantifying these oceanic processes associated with CO<sub>2</sub> pumps.

The East Sea (Japan Sea) is one of the few ideal places for studying these oceanic processes on a regional scale. This is based on two facts that were confirmed during the recent CREAMS (Circulation Research of the East Asian Marginal Seas) Japan-Korea-Russia International cooperative studies on the East Sea:

- (1) The East Sea has many open ocean characteristics such as a deep convection system, large seasonal variation in the sea surface temperature (SST) and comparable biological activities.
- (2) The East Sea has undergone drastic changes in her ventilation system from the bottom water formation mode in the past to that of deep intermediate water formation at the present time (Kim and Kim, 1996).

In particular, the change in the mode of ventilation system observed in the East Sea may resemble that which may be expected to occur in open oceans in the next century associated with recent global warming (Manabe and Stouffer, 1993). Therefore, understanding the carbonate system in the East Sea could provide very important clues to changes in global carbon cycles in the next century.

During the CREAMS expeditions, studies on  $CO_2$  pumps in the East Sea were carried out. The studies included measuring parameters such as fugacity of  $CO_2$  ( $fCO_2$ ) in surface waters as well as in the atmosphere, and alkalinity and pH for the water column. In this paper, we report results mainly focused on the solubility pump operating at the airsea interface of the East Sea. A simple model is devised to describe the annual variation of  $fCO_2$  in surface waters. The net  $CO_2$  flux at the air-sea interface is also estimated from the model.

### 2. Materials and Methods

Studies on the carbonate system were carried out 4 times during the CREAMS expeditions; twice on summer cruises, 95S and 96S, on board R/V Professor Khromov, and

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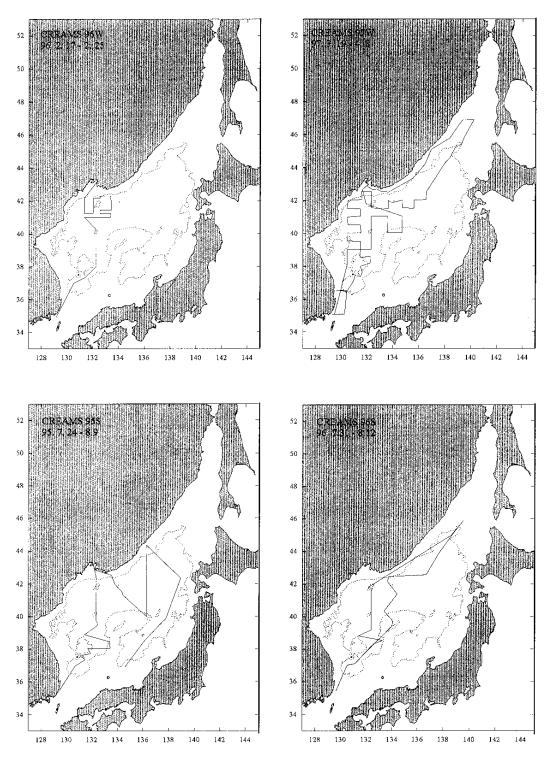


Fig. 1. Maps showing the cruise tracks of (a) CREAMS 96W in February 1996, (b) CREAMS 97W from March to April 1997, (c) CREAMS 95S from July to August 1995, and (d) CREAMS 96S expedition from July to August 1996. Dotted lines represent isopleth of 2000 m depth.

twice on winter cruises, 96W and 97W, on board R/V Parvel Gordienko. The cruise tracks of the four expeditions are shown in Fig. 1.

Partial pressures of CO<sub>2</sub> (pCO<sub>2</sub>) in surface waters and air were measured continuously along the cruise track using a non-dispersive infrared CO<sub>2</sub> analyzer (NDIR; Licor-6252). Air was taken from an air intake tower mounted at the bow through the Dekabon tube (Dekoron, 3/8 inch) and surface waters were pumped up from the cooling system of the ship engines, which was located about 5 m below the sea surface. A Weiss-type equilibrator was used for the pCO<sub>2</sub> measurement of sea waters. pCO<sub>2</sub> was measured every second and was averaged over two-minute intervals. A schematic diagram for the overall analysis system is shown in Fig. 2.

SST and salinity of sea waters entering the equilibrator were measured with a thermosalinometer (Seabird SEACAT) every ten seconds. The temperature at the initial water intake

was also measured continuously with a calibrated thermometer in order to correct for the warming effect during the flow through the tube from the intake to the equilibrator.

Along with the pCO<sub>2</sub> measurement, TA (Total Alkalinity) and pH of sea waters were determined regularly on board during the cruise. TA was determined by a potentiometric titration method (Millero *et al.*, 1993) and pH was measured spectrophotometrically (Clyton and Byrne, 1993). During the CREAMS 97W expedition, silicate and phosphate were also analyzed spectrophotometrically on board (Gordon *et al.*, 1993) along with TA and pH.

The fugacity of CO<sub>2</sub> in the moist equilibrator vapor, fCO<sub>2</sub>, was calculated according to the algorithm by Weiss and Price (1980):

$$fCO_2 = pCO_2(P^{atm} - P^{sw})exp[P^{atm}(B + 2\delta)/RT],$$

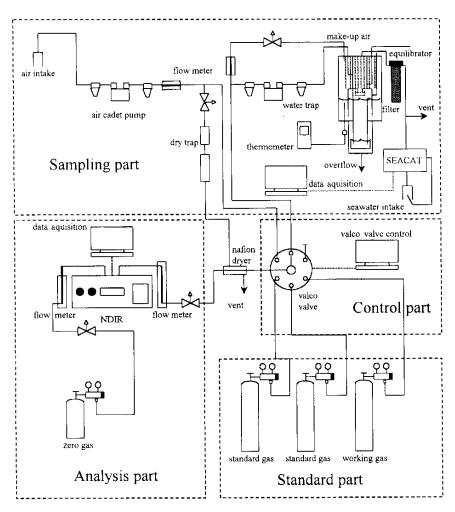


Fig. 2. Schematic diagram of CO<sub>2</sub> analysis system. pCO<sub>2</sub> is measured every second with an NDIR (non-dispersive infrared) CO<sub>2</sub> analyzer (Licor-6252). Temperature and salinity are measured every ten seconds with a thermosalinograph (SEACAT).

where pCO<sub>2</sub> is the partial pressure of CO<sub>2</sub> measured,

 $P^{\text{atm}}$  is the total barometric pressure in units of atmosphere,

 $P^{\text{sw}}$  is the saturated vapor pressure of sea water at the temperature of the measurements (Weiss and Price, 1980),

B and  $\delta$  are the virial and cross-virial coefficients for CO<sub>2</sub>, respectively (Weiss, 1974).

The *in situ* fugacity of  $CO_2$  was calculated by correcting for the warming effect during the sea water flow from the intake to the equilibrator ( $\sim 0.4$ °C) using the following equation (Weiss *et al.*, 1982):

$$\partial \ln f CO_2 / \partial t = 0.03107 - 2.785 \times 10^{-4} t - 1.839 \times 10^{-3} \ln f CO_2$$

where t is temperature in degrees Celsius.

### 3. Results

## 3.1 fCO2 variations in surface waters

The  $fCO_2$  variations in surface waters measured during the 4 cruises are shown in Fig. 3. The SST variations are also shown in the figure for comparison.

In summer,  $fCO_2$  showed a variation with a range of 320~440  $\mu$ atm, which was accompanied by a temperature variation from 15 to 28°C. The figures, especially during

CREAMS 96S, clearly show that the variation of  $fCO_2$  is quite parallel to that of SST, confirming the importance of temperature variation in controlling  $fCO_2$  in surface sea waters (Peng *et al.*, 1987; Hirota *et al.*, 1991; Inoue *et al.*, 1995). The slope of the relative change in  $fCO_2$  with respect to temperature,  $\Delta fCO_2/fCO_2/\Delta T$ , however, is  $0.022\sim0.030$  °C<sup>-1</sup>, smaller than 0.042°C<sup>-1</sup> estimated with laboratory experiment by Peng *et al.* (1987), suggesting that other effects such as gas exchange and biological consumption processes are also important in the real situation.

In winter,  $fCO_2$  showed much wider variations from ~200  $\mu$ atm to ~520  $\mu$ atm, with temperature changes from 0 to 15°C. While  $fCO_2$  values in winter were low, i.e., below atmospheric values of around 360  $\mu$ atm in most of the survey areas, some places in the northern East Sea showed high  $fCO_2$  values, producing an apparent reverse trend between  $fCO_2$  and SST (96W and early and later parts of 97W, in particular).

The process responsible for these high values can be understood by comparing  $fCO_2$  with nutrients in surface waters as shown in Fig. 4. The observed data during 97W cruise clearly show that  $fCO_2$  variation is similar to that of silicate and phosphate. The mixed-layer depths observed at several stations during the cruise were also linearly related to  $fCO_2$  values at the stations, strongly implying that these high values of  $fCO_2$  and nutrients originated from deep water

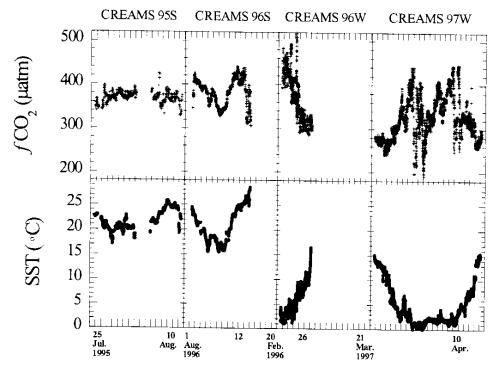


Fig. 3. Surface water fCO<sub>2</sub> and SST variations measured during 4 cruises, CREAMS 95S, CREAMS 96S, CREAMS 96W, and CREAMS 97W. Surface water fCO<sub>2</sub> varies in the range from 320 μatm to 420 μatm. The patterns of fCO<sub>2</sub> and SST variation are quite similar. SST seems to be the main parameter affecting surface water fCO<sub>2</sub>.

brought to the surface through an intensive deep convection system developed in the area in winter time (Oh, 1998). Similar results were reported in the South Atlantic and Weddell Sea areas and in the western North Pacific (Takahashi *et al.*, 1993). The possibility of utilizing  $fCO_2$  in surface waters as a surrogate for mixed-layer depths, thus locating areas of deep convection in the subpolar ocean in winter time, is further discussed elsewhere (Kim *et al.*, 1998).

### 3.2 $\Delta fCO_2$ variations in surface waters

 $\Delta f$ CO<sub>2</sub>, defined as the difference between surface water fCO<sub>2</sub> and atmospheric fCO<sub>2</sub>, (fCO<sub>2</sub><sup>sw</sup> – fCO<sub>2</sub><sup>air</sup>), represents the driving force for CO<sub>2</sub> exchanges at the air-sea interface. In Fig. 5, the distribution of  $\Delta f$ CO<sub>2</sub> is shown, revealing that

surface sea waters were, in general, supersaturated in summer with respect to atmospheric  $CO_2$  and were undersaturated in winter time. Therefore, the East Sea serves as a source of atmospheric  $CO_2$  in summer and as a sink in winter. However, notable exceptions are, of course, coastal areas with upwelling in summer, and deep convection areas in winter time.

### 4. Discussion

### 4.1 An air-sea interface gas-exchange model

A model was devised to describe the variation of  $fCO_2$  in surface waters through time and to calculate the  $CO_2$  flux at the air-sea interface. The model considered that the

# **CREAMS 97W**

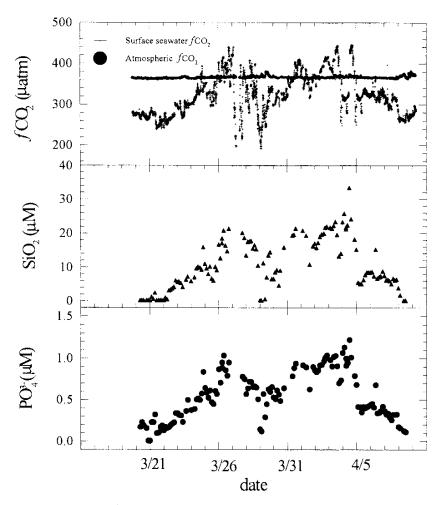


Fig. 4. Surface water fCO<sub>2</sub>, silicate, and phosphate variations against time in CREAMS 97W expedition. The trends of surface water fCO<sub>2</sub> are opposite those of SST but quite similar to those of silicate and phosphate. This means that surface water fCO<sub>2</sub> is affected by vertical mixing with deep water which is relatively rich in CO<sub>2</sub> and nutrients.

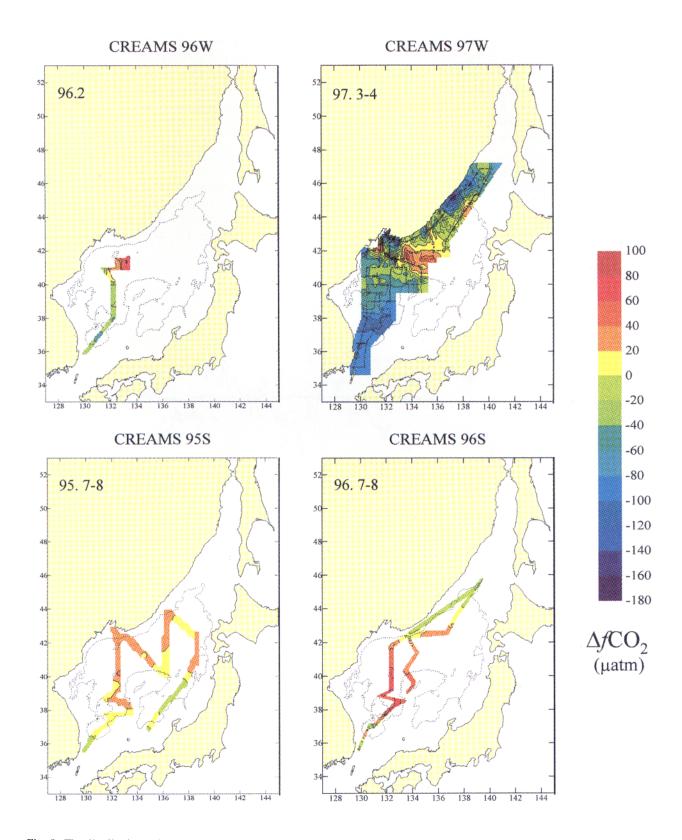


Fig. 5. The distributions of  $\Delta f$ CO<sub>2</sub> (fCO<sub>2seawater</sub> -fCO<sub>2air</sub>) in the East Sea. They reveal that surface seawaters in the East Sea are, in general, supersaturated in summer and undersaturated in winter. In some areas in northern East Sea in winter time, however, high supersaturation of CO<sub>2</sub> was also observed, which could be due to intensive vertical mixing of upper waters.

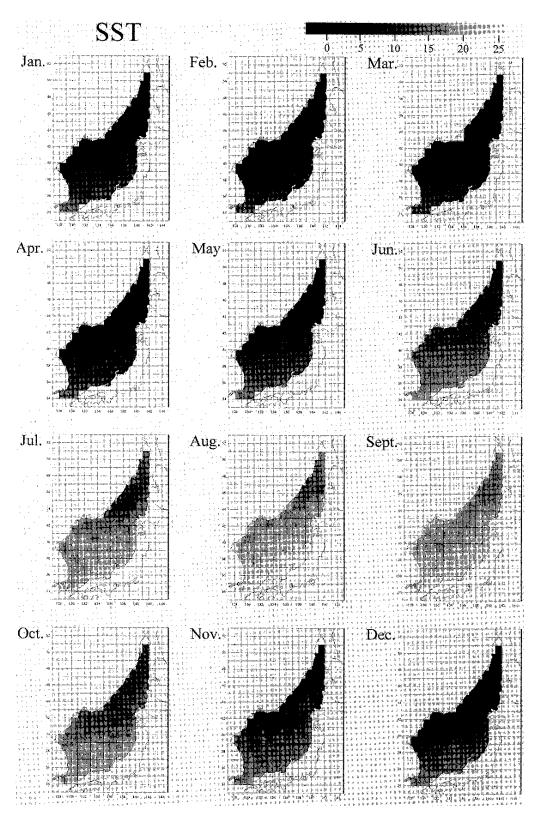


Fig. 6. Monthly mean SST in  $1^{\circ} \times 1^{\circ}$  grid in the East Sea. The East Sea can be divided into warm and cold regions with respect to SST. Even though the areas of warm and cold regions fluctuate seasonally, the boundary, which is a polar front, exists along the  $40^{\circ}$ N line except for coastal regions. The warm region represents subtropical waters whereas the cold region represents subpolar waters.

variation of surface  $fCO_2$  in time resulted from the solubility pump associated with SST and salinity variation in time and gas exchange at the air-sea interface, the biological pump, and the dynamic pump, especially the change of mixed-layer depth with time.

### 4.1.1 Model parameters

For the model calculations, data for SST, mixed-layer depth (MLD), salinity, chlorophyll-a, wind velocity, and atmospheric  $fCO_2$  were compiled for 111 grids (1° × 1° in size) as follows:

SST and salinity: The NODC (National Oceanographic Data Center) and JODC (Japan Oceanographic Data Center) databases containing data from 1932 to 1994 were used to calculate the monthly mean values for each grid.

*MLD*: MLD was defined as the depth with the maximum temperature gradient. MLD was estimated from temperature profiles obtained from the NODC CD-ROM listing data from 1932 to 1994.

Chlorophyll-a: CZCS chlorophyll-a data (Nimbus-7 satellite from 1978 to 1986) were used to estimate the characteristics of the biological pump. The biological production rate is estimated from the CZCS (Coastal Zone Color Scanner) chlorophyll concentration with an empirical CZCS chlorophyll—primary production relationship given by Smith and Baker (1977) as follows:

$$P^{\text{eu}} = 10^{[1.25 + 0.73 \log \text{Chl-}a]}$$

where  $P^{\text{cu}}$  is the average production rate (mg·m<sup>-3</sup>·d<sup>-1</sup>) in the euphotic zone and Chl-a is the CZCS chlorophyll concentration (mg·m<sup>-3</sup>).

The depth of the euphotic zone was also calculated from chlorophyll-*a* data using the empirical relationship by Smith and Baker (1978):

$$Z^{\text{eu}} = 4.6[6.9 - 6.5\log\text{Chl-}a].$$

When the integrated biological activity for the whole surface mixed layer was estimated,  $Z^{cu}$  was used instead of MLD if  $Z^{cu}$  was shallower than MLD.

Wind speed: Diurnal wind speeds for every 1° by 1° grid were calculated with the Cardone Model based on diurnal weather maps (Na et al., 1992); in model calculations actual wind data for 1993 were used. The gas transfer velocity (k<sub>w</sub>) was estimated from the diurnal wind speed as follows (Wanninkhof, 1992):

$$k_{\rm w}$$
 (m/s) =  $[2.5(0.5246 + 1.6256 \times 10^{-2}t + 4.9946 \times 10^{-4}t^2) + 0.3u^2](Sc/660)^{-1/2}$ ,

where t is SST (degrees in Celsius), u is the wind speed (m/s) at 10 m above the sea surface, and Sc is the Schmidt number.

The CO<sub>2</sub> flux at the air-sea interface was calculated as follows:

Flux = 
$$k_w \beta (fCO_2^{sw} - fCO_2^{air}) = k_w \beta (\Delta fCO_2)$$
,

where  $k_{\rm w}$  is the gas transfer velocity and  $\beta$  is the solubility of CO<sub>2</sub>.

Atmospheric fCO<sub>2</sub>: Atmospheric fCO<sub>2</sub> is spatially constant in the East Sea area. The monthly average values from 1990 to 1996, monitored at the Kosan observatory on Cheju Island (Park, M.-K., 1997), were used in the model.

*TCO*<sub>2</sub>: The depth dependence of TCO<sub>2</sub> was estimated from the average TCO<sub>2</sub> profile obtained in the Japan Basin during summer time.

The East Sea can be divided into two regions for model calculation: the northern ("cold region") shows typical characteristics for subpolar oceans, whereas the southern region ("warm region") is typical for subtropical oceans (Fig. 6; Kim and Kim, 1996). Therefore, the parameters estimated for each grid were averaged into two groups (cold region and warm region) as shown in Fig. 7, and were used for the model calculations. Except for wind velocity, all other parameters are monthly-average values and were fitted to smooth curves to give the daily variations.

### 4.1.2 Model algorithm

Figure 8 shows the algorithm for the model calculations. The procedure for model calculation is briefly summarized. It was assumed that total alkalinity (TA) is constant through time. The initial value is assumed to be 2307  $\mu$ mol kg<sup>-1</sup> which is the average value calculated from actual observations at the East Sea during the cruises (Oh, 1998). With the initial TCO<sub>2</sub>, fCO<sub>2</sub> in the air, SST, salinity, MLD and  $k_w$  as input, fCO<sub>2</sub> and pH in surface waters are calculated first and the daily CO<sub>2</sub> flux at the air-sea interface is assessed. Next, the integrated TCO<sub>2</sub> for the whole mixed layer of the day is calculated by multiplying TCO<sub>2</sub> by the mixed-layer depth (MLD);

Integrated 
$$TCO_2(d) = TCO_2(d) \times MLD(d)$$
.

Then, the integrated  $TCO_2$  for the next day, Integrated  $TCO_2(d+1)$ , is calculated by subtracting the effects of the biological pump and gas exchange between the mixed layer and atmosphere as follows:

Integrated 
$$TCO_2(d+1) = Integrated TCO_2(d) - Flux - B$$
,

where Flux represents the  $CO_2$  flux derived from gas-exchange process at the air-sea interface (mmole  $C \cdot m^{-2} \cdot d^{-1}$ ), and B is the rate of the biological pump (mmole  $C \cdot m^{-2} \cdot d^{-1}$ ) which refers to the process by which export production removes carbon from the ocean surface layer to the ocean interior (Denman *et al.*, 1995). B is assumed to be 10% of the primary production in MLD (Broecker and Peng, 1982).

These procedures are repeated for 365 days and are then started all over again for the next year until the model produces stable results, which is usually achieved at the end of three years.

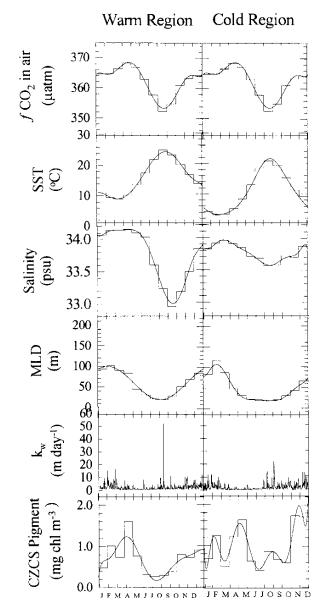


Fig. 7. Annual variation of parameters in the East Sea considered in this model. Due to differences in SST variations, the East Sea is divided into two regions (cold and warm regions) at 40°N in latitude.

### 4.1.3 Model results

Figure 9 shows the model results for warm regions and cold regions, clearly showing seasonal variation with higher values in summer and lower values in winter. The uncertainty of the model results can be estimated by running the model by varying the parameters within their own error limits (Oh, 1998). The uncertainty results mainly from the error for the rate of the biological pump. The uncertainty in  $fCO_2$  is estimated to be  $\pm 60~\mu$ atm and is drawn as dashed

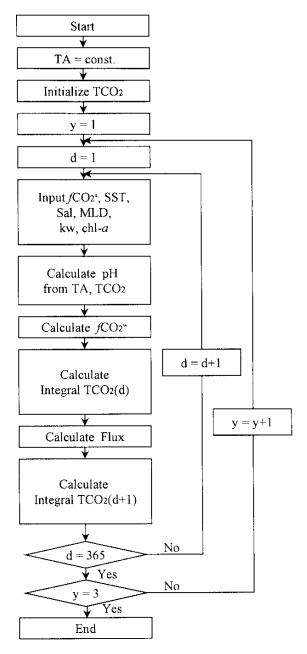


Fig. 8. The model algorithm for estimating surface water fCO<sub>2</sub> and flux at the-air sea interface through time.

curves in the figure.

The observed values during the CREAMS cruises are also shown as vertical lines in the figure. The agreement between the two sets of results is reasonably good. For the winter data, the range of anomalously high values due to the deep convection system is shown with dashed lines; this system could not be represented by the model for obvious reasons with moderate MLD in winter time.

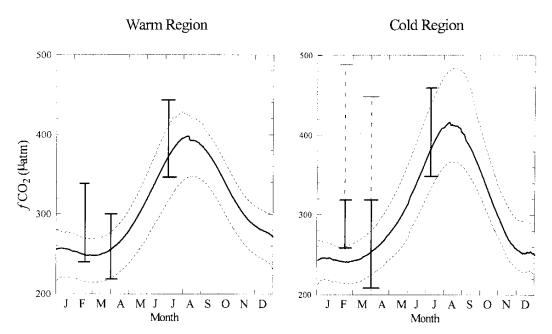


Fig. 9. The annual variation of fCO<sub>2</sub> estimated from the model in the East Sea. The values from our model are consistent with the observed ones in summer but there are some deviations in the cold region in winter.

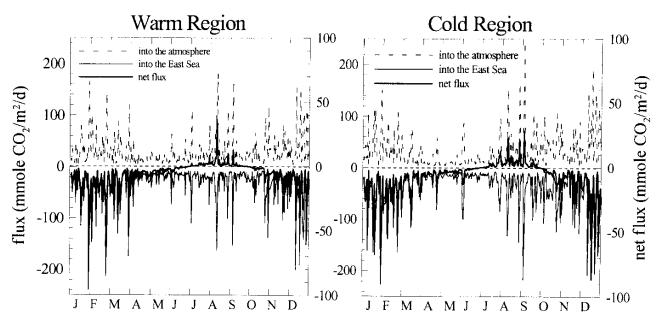


Fig. 10. The daily flux at the air-sea interface estimated from this model in the East Sea. There is a net flux from sea to air from July through September, and from air to sea from October to June. In general, gas exchanges are vigorous in winter but extremely high flux is predicted in summer due to typhoons.

# 4.2 CO2 flux at the air-sea interface

From the model calculations, the daily flux of  $CO_2$  at each region can be estimated and the results are given in Fig. 10. The net fluxes are much smaller than the real exchange

fluxes and are shown on a different scale. The CO<sub>2</sub> exchange process at the air-sea interface is more intense in winter than in summer, except when a typhoon passes through the region. The model shows that there are net fluxes from the

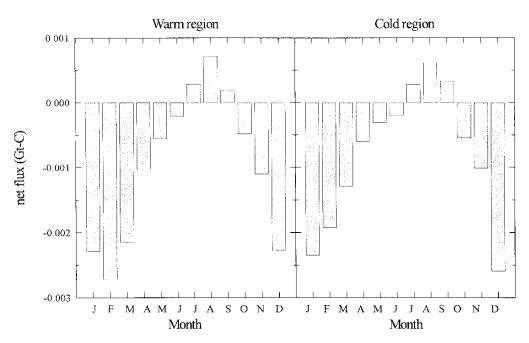


Fig. 11. Monthly integrated  $CO_2$  fluxes at the air-sea interface for the warm and cold regions in the East Sea. A negative  $CO_2$  flux means a  $CO_2$  sink to the atmosphere and a positive value is a source to the atmosphere. The East Sea behaves as a  $CO_2$  source in warmer months from July to September whereas it behaves a  $CO_2$  sink in other seasons. The net  $CO_2$  flux in the warm and cold regions is 0.014 ( $\pm 0.005$ ) Gt yr<sup>-1</sup> and 0.018 ( $\pm 0.007$ ) Gt yr<sup>-1</sup>, respectively.

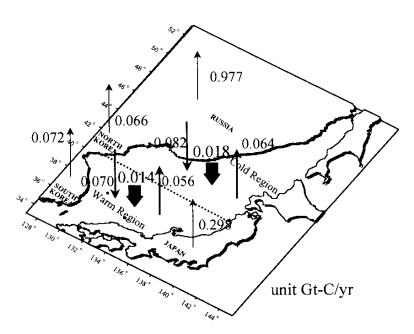


Fig. 12. The overall exchanges of CO<sub>2</sub> at the East Sea. The warm region absorbs 0.070 Gt-C and emits 0.056 Gt-C annually, resulting in a net absorption of 0.014 (± 0.005) Gt-C yr<sup>-1</sup>. In the cold region, the East Sea emits 0.064 Gt-C and absorbs 0.082 Gt-C annually, producing an annual net absorption of 0.018 (± 0.007) Gt-C yr<sup>-1</sup>. Anthropogenic emissions of CO<sub>2</sub> from nearby nations (Marland *et al.*, 1994) are also shown for comparison in the figure.

sea into the air in summer (from July to September) whereas these fluxes are from the air into the sea during the remaining period of the year (from October to May).

The monthly net flux is given in Fig. 11. The East Sea absorbs  $CO_2$  from October to May, serving as an atmospheric  $CO_2$  sink whereas it emits  $CO_2$  from June through September, working as a  $CO_2$  source. The warm region absorbs 0.070 Gt-C and emits 0.056 Gt-C annually, resulting in a net absorption of 0.014 ( $\pm 0.005$ ) Gt-C per year. In the cold region, the East Sea emits 0.064 Gt-C and absorbs 0.082 Gt-C annually, producing an annual net absorption of 0.018 ( $\pm 0.007$ ) Gt-C per year. Therefore, the East Sea appears to serve as a net sink for atmospheric  $CO_2$  and the annual net flux is 0.032 ( $\pm 0.012$ ) Gt-C per year. The overall exchanges of  $CO_2$  at the East Sea are summarized in Fig. 12. In the figure, anthropogenic emissions of  $CO_2$  from nearby nations are also shown for comparison.

#### 5. Conclusions

fCO<sub>2</sub> for surface waters was measured continuously along the cruise tracks 4 times during CREAMS expeditions. In summer, the fCO<sub>2</sub> in surface waters varied in the range 320~440  $\mu$ atm, showing moderate supersaturation with respect to atmospheric CO<sub>2</sub>. However, in winter, fCO<sub>2</sub> showed undersaturation of CO<sub>2</sub> in most of the area, while fCO<sub>2</sub> varied in a much wider range from 180 to 520  $\mu$ atm. Some very high fCO<sub>2</sub> values, observed in northern East Sea, appear to be associated with the intensive convection system developed in the area.

An air-sea gas-exchange model was developed for describing the annual variation of  $fCO_2$  and for estimating the annual flux of  $CO_2$  at the air-sea interface. The model incorporated annual variations in SST, mixed-layer depth, gas exchange associated with wind velocity, biological activity, and atmospheric concentration of  $CO_2$ . The model shows that the East Sea emits  $CO_2$  into the atmosphere from June to September, and absorbs  $CO_2$  during the rest of the year, from October through May. The net annual  $CO_2$  flux at the air-sea interface was estimated to be 0.032 ( $\pm 0.012$ ) Gt-C per year from the atmosphere into the East Sea. This amount corresponds to about 1.5% of the annual oceanic uptake of  $CO_2$  ( $2.1 \pm 0.8$  Gt-C; Tans et al., 1993), about 5 times the global average  $CO_2$  uptake rate per unit area by ocean.

Studies on the carbonate system in the water column also show penetration of CO<sub>2</sub> all the way to the bottom within entire basins, supporting a rapid turnover time in the area (Tsunogai *et al.*, 1993; Kim and Kim, 1996; Kumamoto *et al.*, 1998). The East Sea appears to have 0.25 Gt-C of excess CO<sub>2</sub> already in deep waters (Park, S. Y., 1997), indicating rapid penetration of anthropogenic CO<sub>2</sub> at the present time, consistent with the present study. Studies on constructing overall CO<sub>2</sub> cycles in the East Sea are under way at the present time.

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