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# **Continuous observations of atmospheric and oceanic CO<sub>2</sub> using a moored buoy in the East China Sea: Variations during the passage of typhoons**

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## Abstract

An automatic measuring system for the partial pressure,  $p\text{CO}_2$ , of atmospheric and oceanic carbon dioxide was developed. The system was mounted on a moored buoy for routine observation of maritime meteorology in the East China Sea.  $\text{CO}_2$  observations were conducted from 29 June 1997 to 6 January 1998. During the observation period, the atmospheric  $p\text{CO}_2$  showed little variation (341 to 365  $\mu\text{atm}$ ), whereas  $p\text{CO}_2$  in the surface water varied significantly (308 to 408  $\mu\text{atm}$ ). In the summer,  $p\text{CO}_2$  was higher in the surface water than in the overlying atmosphere, implying that this area was a source for atmospheric  $\text{CO}_2$ , though it became a sink after late September. Time series data clearly exhibited significant short-term variations in the oceanic  $p\text{CO}_2$ , *i.e.*, sudden variations during the passage of typhoons, and diurnal variations driven by the diurnal variations in the sea-surface temperature under calm conditions. The effects of typhoons on ocean–atmosphere  $\text{CO}_2$  exchange at the surface could differ, depending on the relative position of the mooring site with respect to the center of the moving typhoons. These differences result from the different contributions of sea-surface cooling, entrainment, and upwelling. The efflux enhanced by three typhoons accounted for 60% of the efflux of  $\text{CO}_2$  in the warm season. It is suggested that typhoons have a significant impact on the carbon cycle in the western subtropical North Pacific.

*Keywords:* Moored buoy;  $\text{CO}_2$  flux; Typhoon; Entrainment; Upwelling; Sea-surface cooling.

*Regional index terms:* North Pacific; Subtropical region; East China Sea.

## 1. Introduction

Understanding the spatial and temporal variations of oceanic carbon dioxide ( $\text{CO}_2$ ) is urgently important in order to more precisely estimate the future evolution of atmospheric  $\text{CO}_2$  concentrations in response to a given industrial energy policy. However, compared with the well-known distribution of the partial pressure of  $\text{CO}_2$  in the atmosphere ( $p\text{CO}_2^{\text{air}}$ ), the distribution of  $p\text{CO}_2$  in surface waters ( $p\text{CO}_2^{\text{sea}}$ ) is poorly understood because  $p\text{CO}_2^{\text{sea}}$  data have traditionally been collected on board research vessels or ships of opportunity on a limited temporal and spatial scale. Although the climatological monthly distribution of  $p\text{CO}_2^{\text{sea}}$  in the global surface waters and subsequent global  $\text{CO}_2$  fluxes have been estimated based on a compilation of shipboard ocean  $\text{CO}_2$  data (e.g., Takahashi et al., 2002), the uncertainties in the global estimates are large, but can be reduced by using improved data sets. The interannual variability is not known. Continuous  $p\text{CO}_2^{\text{sea}}$  measurements in selected key areas over long periods would be beneficial for identifying the temporal characteristics representative of those areas. A  $p\text{CO}_2$  measuring system that can be mounted on buoys is required. It should have sufficient stability and sensitivity to determine the temporal variability, and to collect continuous measurements over a period of several months to a year.

A few autonomous  $p\text{CO}_2$  measurement systems have been developed that can be used for continuous measurements with a moored buoy (e.g., Friederich et al., 1995; DeGrandpre et al., 1995) or a drifting buoy (Merlivat and Brault, 1995). A high-resolution time series of  $p\text{CO}_2^{\text{sea}}$  revealed variations on many scales in the subtropical North Atlantic (Bates et al., 2000), the equatorial Atlantic (Bakker et al., 2001), the Mediterranean Sea (Hood and Merlivat, 2001; Copin-Montégut et al., 2004), the equatorial Pacific (Chavez et al., 1999), and the subtropical North Pacific (Friederich et al., 2002). These measurements suggest that it is possible to estimate the full annual air-sea  $\text{CO}_2$  flux from monthly sampling, and to estimate

$p\text{CO}_2^{\text{sea}}$  and the air-sea flux based on the relationship between  $p\text{CO}_2^{\text{sea}}$  and sea-surface temperature (SST) (Hood and Merlivat, 2001). Moreover, mooring-based measurements have also captured the variability missed between monthly ship visits: e.g., the detection of large weekly variations in  $\text{CO}_2$  flux, driven by changes in wind speed, during a transition period from the El Niño to La Niña events (Chavez et al., 1999); the unusually high  $\text{CO}_2$  efflux from the central California upwelling zone during the La Niña event (Friederich et al., 2002); and the onset of phytoplanktonic bloom induced by rapid stabilization of the surface layer (Copin-Montégut et al., 2004).

Hood et al. (2001) observed a sharp decrease in SST and increases in  $p\text{CO}_2^{\text{sea}}$  and nitrate concentration during a storm with wind speeds reaching 16 to 17  $\text{m s}^{-1}$ , followed by a decrease in  $p\text{CO}_2^{\text{sea}}$  and nitrate concentration and a concomitant increase in fluorescence. The  $\text{CO}_2$  flux during the storm doubled due to the elevated wind speed. These results suggest the entrainment of nutrients and the resultant biological production derived from short-term wind-induced mixing events. The effect of heavy storms, such as tropical cyclones, on the carbon cycle in the upper ocean is not well understood, compared with their effect on the upper thermal and physical structure. Bates et al. (1998a) indicated that hurricanes exert an important influence on ocean–atmosphere  $\text{CO}_2$  exchange over subtropical oceans, based on shipboard observations made before and after the passage of hurricanes. Autonomous measurement platforms can be useful for monitoring surface properties related to the carbon cycle and for the understanding of the effects of tropical storms. Progress has been made recently using  $\text{O}_2$  and  $\text{N}_2$  measurements taken during hurricanes (D'Asaro and McNeil, 2007).

We developed a new automatic measuring system for  $p\text{CO}_2^{\text{air}}$  and  $p\text{CO}_2^{\text{sea}}$  that was mounted on a moored buoy operated by the Japan Meteorological Agency (JMA) for routine observations of maritime meteorology. In the present paper, we report the development of

this system and observations made in the East China Sea. In particular, the effects of three typhoons that passed near the buoy on the variations in  $p\text{CO}_2^{\text{sea}}$  are discussed.

## 2. Measurements and apparatus

The  $\text{CO}_2$  measuring system was mounted on a 10-meter-diameter moored buoy in the East China Sea ( $28^\circ 10' \text{N}$ ,  $126^\circ 20' \text{E}$ ; depth, 136m; Fig. 1) from June 29, 1997, through January 6, 1998. A  $\text{CO}_2$  gas analyzer, standard gas bottles (48 liter), batteries, a controller, and recording and transmission systems were housed inside the buoy body. A non-dispersive infrared (NDIR) analyzer (LI-6262, LI-COR Inc.) was used as a  $\text{CO}_2$  gas analyzer with no modification because of its good stability, tolerance to vibration, low power demand, and manageable size, as reported by Friederich et al. (1995). An air-sea equilibrator was installed in the equilibrator installing hole, penetrating the bottom of the buoy and becoming linked with the sea surface (Fig. 1). The lower compartment of the equilibrator was located below the mean waterline (Fig. 2) in order to ensure minimal deviation of the water temperature from the ambient temperature as well as a minimal power demand for the pumping up of seawater. The seawater intake port was covered with a copper mesh. In order for the system to operate without maintenance for a long period, as well as for rapid equilibration, we adapted a cascade equilibrator, in which a number of steps are provided to splash the seawater as it flows down. The seawater splash quickens equilibration between the seawater and the air circulating in a looped line consisting of the equilibrator and a buffer mounted outside of the buoy to compensate for the pulsating air flow (Fig. 3). The equilibration time response of the system was examined by comparison with ship measurements taken with a shower-head equilibrator on board R/V *Ryofu-Maru* of JMA during a cruise from the subarctic to equatorial region ( $48^\circ \text{N}$  to  $2^\circ \text{S}$  along  $165^\circ \text{E}$ ) that lasted for 39 days (October to November) in 1996. An insignificant difference ( $p = 0.18$ ) between the ship and buoy systems for an

equilibration time of 30 minutes was observed in the  $p\text{CO}_2^{\text{sea}}$  measurements for the same water, pumped from the ship's bottom.

The  $\text{CO}_2$  measuring system was powered by 77 add-water-type air batteries (Matsushita Battery Industrial Co.; 1.3 V, 2000 Ah). The maximum power supplied by the set of batteries was 12V at 5.2A. The batteries can provide sufficient energy for measurements to be made eight times per day for one year. Power management, switching circuit control, and data acquisition in the  $\text{CO}_2$  measuring system were all accomplished by a CPU 50 controller (Kimoto Engineering Ltd.).

A series of  $\text{CO}_2$  measurements in air, seawater and standard gasses were made every three hours. A sample of air was pumped from an air intake installed on the mast (7 m high) for meteorological measurements. Seawater was pumped up from the bottom of the buoy at a depth of about 1.0 m below the surface. All of this water flow was introduced into an air-sea equilibrator. The background air for atmospheric measurements or seawater-equilibrated air for seawater measurements was dried using a Perma Pure Dryer<sup>TM</sup> tube (Perma Pure, Ltd.) and was then introduced into an NDIR analyzer for measurements of the molar fraction of  $\text{CO}_2$  in the sample air. All NDIR output voltages were measured under ambient pressure while stopping the air stream temporarily, averaged over 10 sec and stored on a memory card. These were calibrated every three hours using four  $\text{CO}_2$ -in-air working standard gases with nominal mixing ratios of 270, 330, 360, and 405 ppm. A quadratic function was used for fitting the NDIR output voltages to the certified concentrations. These gases underwent pre- and post-observation calibration against standards certified by the World Meteorological Organization (WMO). In this paper, we report  $\text{CO}_2$  concentrations based on the 1995 WMO scale. The  $\text{CO}_2$  mole fraction was converted to  $p\text{CO}_2$  based on an equation from Weiss and Price (1980). The values of  $p\text{CO}_2^{\text{air}}$  and  $p\text{CO}_2^{\text{sea}}$  were normalized to a

sea-level pressure (SLP) of 1 atm in order to represent the variations in the carbonate system. Seawater temperatures, both at the bottom of the buoy and in the equilibrator, were recorded as well. Their difference was  $0.03 \pm 0.14$  °C during the whole period and corrected for the temperature effect on  $p\text{CO}_2$  using an equation from Weiss et al. (1982). The measurements with the temperature difference of larger than 0.5 °C were only 13 in 1525 measurements.

### 3. Results and discussion

#### 3.1. Comparison with shipboard measurements

The  $\text{CO}_2$  measurements were performed in the East China Sea from June 29, 1997, through January 6, 1998 (for 191 days). The buoy was located approximately 250 km northwest of Okinawa Island in about 136 m of water ( $28^\circ 10' \text{N}$ ,  $126^\circ 20' \text{E}$ ; Fig. 1). The mooring site was located to the west of the main stream of the Kuroshio. The intercomparison of  $\text{CO}_2$  measurements between the buoy and shipboard were made during July 15 to 17, 1997. The ship  $p\text{CO}_2$  data were collected on board R/V *Ryofu-Maru* of JMA in the vicinity ( $<0.5$  mile) of the buoy. The hourly shipboard data were generated with an NDIR analyzer (model 880, Beckman), coupled to a shower-head equilibrator in line with the seawater intake at 4 m depth, as reported earlier (Inoue and Sugimura, 1992; Midorikawa et al., 2006). During the comparison, no time lag was observed in the temporal variations of the two  $p\text{CO}_2$  data sets. The offset of the buoy measurements from the ship measurements was  $0.2 \pm 1.0$   $\mu\text{atm}$  for  $p\text{CO}_2^{\text{air}}$  and  $2.2 \pm 1.6$   $\mu\text{atm}$  for  $p\text{CO}_2^{\text{sea}}$ . The standard deviation of the offset for  $p\text{CO}_2^{\text{sea}}$  was comparable to that for the  $p\text{CO}_2^{\text{sea}}$  measurements on board R/V *Ryofu-Maru* reported earlier by Inoue and Sugimura (1992), although possible sources of this offset may be the different surface properties of water at different depths or a different efficiency of equilibration due to the use of different types of equilibrators in the respective

measurements.

### 3.2. Comparison with $p\text{CO}_2^{\text{air}}$ measurements on land

The  $p\text{CO}_2^{\text{air}}$  data were compared with those from continuous atmospheric measurements made at the Yonagunijima Weather Station (24.47°N, 123.02°E) by JMA (Watanabe et al., 2000) on Yonaguni Island, located approximately 500 km southwest of the buoy site. The offset between the buoy and land measurements of  $p\text{CO}_2^{\text{air}}$  for the overall study period was  $0.6 \pm 2.5$  ppm for hourly data and  $0.5 \pm 1.6$  ppm for the 24-hr moving average. The data also indicated no temporal change in the offset between the two  $p\text{CO}_2^{\text{air}}$  measurements over the whole period.

### 3.3. Seasonal variations

The  $p\text{CO}_2^{\text{sea}}$  in the surface water was highly variable on different time scales, ranging from daily to seasonal (Fig. 4). On a seasonal time scale,  $p\text{CO}_2^{\text{sea}}$  exhibited a large variation, with a maximum value of 408  $\mu\text{atm}$  in late August and a minimum value of 308  $\mu\text{atm}$  in early January. The  $p\text{CO}_2^{\text{sea}}$  was lower than  $p\text{CO}_2^{\text{air}}$  from June 29 through early July, indicating that this area was a sink for atmospheric  $\text{CO}_2$  during this period. The  $p\text{CO}_2^{\text{sea}}$  increased with increasing water temperature and exceeded  $p\text{CO}_2^{\text{air}}$  in mid July. This area was primarily a source from mid July through mid September, when the water temperature exceeded 27.3°C. In late September,  $p\text{CO}_2^{\text{sea}}$  decreased with decreasing water temperature and fell below  $p\text{CO}_2^{\text{air}}$ , indicating that this area became a sink once again. The  $p\text{CO}_2^{\text{sea}}$ , normalized to a temperature of 25°C ( $n\text{-}p\text{CO}_{2, T=25}$ ), was confined almost completely within a narrow range of  $314 \pm 6.7$   $\mu\text{atm}$  from early July through mid October except for typhoon events, indicating that the high  $p\text{CO}_2^{\text{sea}}$  in summer was due to the high temperature. Temporal changes in  $p\text{CO}_2^{\text{sea}}$  during this

period appear to be primarily attributable to variations in water temperature. From November through January,  $n\text{-}p\text{CO}_{2,T=25}$  increased gradually. It has been suggested that dissolved inorganic carbon (DIC) supplied from the lower layer through enhanced vertical mixing would contribute to this increase in  $p\text{CO}_2^{\text{sea}}$ . However, because the effects of decreasing temperature on  $p\text{CO}_2^{\text{sea}}$  exceeded those of increasing DIC concentrations,  $p\text{CO}_2^{\text{sea}}$  could decrease during this period.

### 3.4. Diurnal variations

On July 29 to 31, August 23 to 25, and August 31 to September 2, a clear diurnal  $p\text{CO}_2^{\text{sea}}$  signal was observed, with the largest observed diurnal variation of 56  $\mu\text{atm}$ , reaching a maximum at 15 to 18 o'clock and a minimum at 6 o'clock. During these days, the weather was fine and wind speeds were very low ( $< 3 \text{ m s}^{-1}$ ; mostly  $< 1 \text{ m s}^{-1}$  around the maximum  $p\text{CO}_2^{\text{sea}}$ ). Surface stratification developed, and the prominent surface warming and cooling patterns that are characteristic of the diurnal thermal cycle due to the intense solar heat flux were observed. The times of maximum and minimum diurnal changes in water temperature coincided with those for  $p\text{CO}_2^{\text{sea}}$ , whereas the  $n\text{-}p\text{CO}_{2,T=25}$  showed little diurnal variation during these days. A linear regression of  $p\text{CO}_2^{\text{sea}}$  against water temperature for two to four consecutive days gave a relationship of  $d(\ln p\text{CO}_2)/dT = 4.11 \pm 0.27$  to  $4.38 \pm 0.39 \text{ \% } ^\circ\text{C}^{-1}$ , which coincides well with the relationship of  $4.23 \text{ \% } ^\circ\text{C}^{-1}$  from Takahashi et al. (1993), although the relationship in the buoy observations was more dispersive due to superimposed noise. These results indicate that diurnal changes in  $p\text{CO}_2^{\text{sea}}$  occurred in response to the diurnal thermal cycle. The observed  $p\text{CO}_2^{\text{sea}}$  cycle can be explained in terms of the temporal evolution of the warm surface layer. However, the  $\text{CO}_2$  flux during this period was small due to the very weak wind, as described below.

### 3.5. Effects of typhoons on $p\text{CO}_2^{\text{sea}}$ variations

The buoy data revealed significant variations in  $p\text{CO}_2^{\text{sea}}$  on a time scale of several days to a week. The most pronounced temporal changes in  $p\text{CO}_2^{\text{sea}}$  were associated with the passage of typhoons. Three typhoons passed close to the buoy in August and September, 1997 (Fig. 5). The first typhoon, Typhoon Tina (T9711), passed directly over the buoy site on August 7 to 8. The second one, Typhoon Winnie (T9713), passed 300 km southwest (on the left side) of the buoy, and the third, Typhoon Oliwa (T9719), passed 300 km east (on the right side) of the buoy. During the passage of T9711, T9713, and T9719, respective wind speed maxima of 29.5, 30.0 and 20.6  $\text{m s}^{-1}$  (Figs. 4 and 6 to 8) and wave-height maxima of 10.9, 11.1, and 8.6 m were observed. During these periods,  $\text{CO}_2$  observations were conducted appropriately without any break, demonstrating the high performance of the new  $p\text{CO}_2$  measurement system under severe conditions.

The three typhoons had different influences on the surface distributions of temperature and  $p\text{CO}_2^{\text{sea}}$  at the buoy site as follows. In the case of T9713, the temperatures at depths of 1 m, 50 m, and 100 m approached each other (Fig. 7c), indicating that the mixed layer at the buoy site had deepened due to strong vertical turbulent mixing, since the relative position of the buoy from the center of the moving typhoon was on the right side of its moving direction (Wada, 2005). In the case of T9711, a converging between the temperatures at 1 m and 50 m was observed before the time of minimum SLP (Fig. 6). This was also due to vertical turbulent mixing caused by strong wind. After the typhoon center had passed, however, temperatures decreased concurrently at all three depths (1 m,  $-1.5^\circ\text{C}$ ; 50 m,  $-6.3^\circ\text{C}$ ; 100 m,  $-4.2^\circ\text{C}$ ), which cannot be explained by enhanced surface mixing alone. The salient decreases

in temperatures at depths of 50 m and 100m suggest that cool water was transported from the lower layer by an upwelling event. In fact, the cyclonic wind of the typhoon produced a divergent current in the upper ocean and, subsequently, the mixed layer in the vicinity of typhoon center tended to become shallow by Ekman pumping since the shear-induced vertical turbulent mixing became negligibly small (Wada et al., 2008). Then, a cyclonic eddy occurred behind the typhoon, inducing upwelling (e.g., Wada, 2002). Sea-surface cooling was relatively small because the mixed layer was so thick that sufficient cool water did not reach the surface. Nonetheless, it is worth noting that the upwelling event played an important role in the ocean response to T9711, which is different from T9713. In T9719, which was relatively weak and passed on the right side of the buoy site, only small changes in temperature at 1 m and 50 m were observed (Fig. 8c). Since the typhoon center was far from the buoy and since T9719 was smaller than T9713, only small changes were caused by vertical turbulent mixing with no effect from near-inertial currents or anticyclonic eddy (Wada and Usui, 2007).

In both T9711 and T9713, changes in  $p\text{CO}_2^{\text{sea}}$  were small before the minimum SLP, and just after the closest approach of the typhoon,  $p\text{CO}_2^{\text{sea}}$  decreased rapidly to the same level as  $p\text{CO}_2^{\text{air}}$  within 9 hr (Figs. 6 and 7).  $p\text{CO}_2^{\text{sea}}$  normalized to a temperature of 29°C ( $n\text{-}p\text{CO}_{2, T=29}$ ) increased from about the time when the wind speed increased to over  $15 \text{ m s}^{-1}$  until about the time when the wind speed fell below  $20 \text{ m s}^{-1}$ , synchronized approximately with the decrease in SST, indicating a DIC supply due to vertical turbulent mixing corresponding to entrainment at the mixed-layer base. Then,  $n\text{-}p\text{CO}_{2, T=29}$  decreased gradually from about the time when the wind speed fell below  $15 \text{ m s}^{-1}$ .  $n\text{-}p\text{CO}_{2, T=29}$  continued to increase for a longer time because of the large size of T9713, whereas the  $n\text{-}p\text{CO}_{2, T=29}$  increase in T9711 was more rapid and its magnitude was larger under the conditions of the upwelling event. It is

considered that  $p\text{CO}_2^{\text{sea}}$  remained almost constant because the effects of DIC entrainment on  $p\text{CO}_2^{\text{sea}}$  were counteracted by those of sea surface cooling and  $\text{CO}_2$  efflux to air during the period of predominance of vertical turbulent mixing during the first half of each typhoon passage. During the period of rapid  $p\text{CO}_2^{\text{sea}}$  decrease,  $n\text{-}p\text{CO}_{2, \text{T}=29}$  exhibited insignificant change, indicating a dominant contribution of decreasing SST to the  $p\text{CO}_2^{\text{sea}}$  decrease during this period. After  $p\text{CO}_2^{\text{sea}}$  fell to the same level as  $p\text{CO}_2^{\text{air}}$ , subsequent gradual decreases in  $n\text{-}p\text{CO}_{2, \text{T}=29}$  in both typhoons suggest a possible occurrence of biological DIC consumption, since the contribution of air-sea  $\text{CO}_2$  flux was estimated to be almost zero.

In T9719,  $p\text{CO}_2^{\text{sea}}$  increased from the same level as  $p\text{CO}_2^{\text{air}}$  to a higher one before the minimum SLP (Figs. 8e and f).  $n\text{-}p\text{CO}_{2, \text{T}=29}$  also exhibited a small increase during the passage of the typhoon. The  $p\text{CO}_2^{\text{sea}}$ , which was in equilibration with the air before the passage, could have been elevated by a supply of DIC from the lower column resulting from vertical turbulent mixing, which would exceed the contribution of a small SST decrease. A distinct  $p\text{CO}_2^{\text{sea}}$  decrease corresponding to the SST drop just after the closest approach of the typhoon was not observed. The effects of T9719 on the variations of  $p\text{CO}_2^{\text{sea}}$  and SST were relatively small due to weak vertical turbulent mixing, since this typhoon passed on the right side of the buoy.

In our observations,  $n\text{-}p\text{CO}_{2, \text{T}=29}$  increased in all three cases (T9711, +34; T9713, +20; T9719, +15  $\mu\text{atm}$ ), indicating the increases in DIC concentration due to enhanced vertical turbulent mixing. In addition, an upwelling event could enhance the increases in DIC concentration in T9711, in combination with vertical turbulent mixing. These features are different from those reported for tropical cyclones in the Sargasso Sea, where temperature was the dominant control on  $p\text{CO}_2^{\text{sea}}$  variability and no DIC change was observed (Bates et al., 1998b). It is considered that the intrusion of DIC-rich subsurface Kuroshio waters along the

continental shelf floor associated with the typhoon passage (Chen et al., 2003) could have a significant influence on the surface carbonate system in the East China Sea, as compared with the thick DIC-poor surface waters in the subtropical North Atlantic (Brix et al., 2004).

### 3.6. Variations in air-sea CO<sub>2</sub> fluxes

The air-sea fluxes of CO<sub>2</sub> ( $F$ ) were calculated using the gas-transfer coefficient ( $E(W)$ ) from Wanninkhof (1992) for the short term, based on the difference ( $\Delta p\text{CO}_2$ ) between  $p\text{CO}_2^{\text{sea}}$  and  $p\text{CO}_2^{\text{air}}$  and the wind speed, observed every three hours on the buoy, as follows:

$$F = E(W) \cdot (p\text{CO}_2^{\text{sea}} - p\text{CO}_2^{\text{air}}) = E(W) \cdot \Delta p\text{CO}_2. \quad (1)$$

The three-hourly fluxes ranged from -2.8 to 0.8 mmol C m<sup>-2</sup> per 3 hr (Fig. 4g) except for higher fluxes (max. 4.9 mmol C m<sup>-2</sup> per 3 hr in T9711) associated with typhoon winds. The fluxes were negative during the initial two weeks and after late September, indicating absorption of CO<sub>2</sub> into the ocean during these periods, whereas they were positive during mid July through mid September, indicating evolution from the ocean. This area took up 296 mmol m<sup>-2</sup> of CO<sub>2</sub> from the atmosphere as the net flux for the overall observation period of 191 days. The area acted totally as a sink during the period.

Little flux was estimated for days when a high  $p\text{CO}_2^{\text{sea}}$  was observed, corresponding to a diurnal thermal cycle caused by low wind speeds. In contrast, three typhoons had a significant impact on the CO<sub>2</sub> flux in this area. It is suggested that strong winds and a DIC supply from the lower column that occurs during the passage of a typhoon increased the efflux CO<sub>2</sub> from ocean to atmosphere, despite the decreases in  $p\text{CO}_2^{\text{sea}}$  due to sea-surface cooling. The fluxes associated with three typhoons were estimated to be 27 mmol C m<sup>-2</sup> for T9711, 37 mmol C m<sup>-2</sup> for T9713 and 26 mmol C m<sup>-2</sup> for T9719. The greatest flux was estimated for

the strong and large T9713, but not for T9711, which passed directly over the mooring site with the lowest SLP and highest  $n\text{-}p\text{CO}_2$ ,  $T=29$ .

On the basis of Eq. 1, we examined how the variations in the gas-transfer coefficient,  $p\text{CO}_2^{\text{sea}}$ , and  $p\text{CO}_2^{\text{air}}$  affected the  $\text{CO}_2$  flux during each typhoon event. The differential of the  $\text{CO}_2$  flux,  $F$ , with respect to time can be written as

$$\frac{\partial F}{\partial t} = \frac{\partial E(W)}{\partial t} \cdot \Delta p\text{CO}_2 + \frac{\partial p\text{CO}_2^{\text{sea}}}{\partial t} \cdot E(W) - \frac{\partial p\text{CO}_2^{\text{air}}}{\partial t} \cdot E(W), \quad (2)$$

where the first term of the right side of Eq. 2 represents the changes in  $\text{CO}_2$  flux by those in the gas-transfer coefficient, the second term by those in  $p\text{CO}_2^{\text{sea}}$  and the third term by those in  $p\text{CO}_2^{\text{air}}$ . These three terms were approximated by the central difference. The calculated values for the respective terms and their sum are shown in Fig. 9. For both T9711 and T9713, the increase in  $\text{CO}_2$  efflux before the minimum SLP was mainly caused by the increase in wind speed, and the rapid decrease in efflux after the minimum SLP by the decrease in  $p\text{CO}_2^{\text{sea}}$ . During the passage of T9719,  $p\text{CO}_2^{\text{sea}}$  was a major factor controlling the  $\text{CO}_2$  flux, though the maximum wind speed reached to the level of  $20 \text{ m s}^{-1}$ . As can be easily seen,  $p\text{CO}_2^{\text{air}}$  had only a minor effect on the variations in flux for all typhoon events.

In T9711 and T9713,  $p\text{CO}_2^{\text{sea}}$  was not the main driver for the temporal changes in  $\text{CO}_2$  flux before the minimum SLP, but it is noteworthy that  $p\text{CO}_2^{\text{sea}}$  contributed to the enhancement of the efflux through holding  $\Delta p\text{CO}_2$  positive. Small variations in  $p\text{CO}_2^{\text{sea}}$  before the minimum SLP were due to the balance between anti-correlated SST and DIC variations; the decrease in  $p\text{CO}_2^{\text{sea}}$  due to cooling and the  $\text{CO}_2$  efflux to air were compensated for by the supply of DIC from the lower layer, as mentioned in Section 3.5, leading to a persistently positive  $\Delta p\text{CO}_2$  and consequent enhancement of the efflux. The retention time of a positive  $\Delta p\text{CO}_2$  with a significant  $\text{CO}_2$  efflux (more than  $0.3 \text{ mmol C m}^{-2}$  per 3 hr) was 36 hr in T9711, 66 hr in T9713 and 54 hr in T9719. In T9713 and T9719, which passed over an area away

from the mooring site, sustained vertical turbulent mixing continued to supply DIC from the lower layer for a longer time and enhanced the efflux, irrespective of a relatively small  $n\text{-}p\text{CO}_2$ ,  $T=29$  increase, compared with those in T9711. In the intense T9711 and T9713, just after the closest approach of the typhoon,  $p\text{CO}_2^{\text{sea}}$  decreased to the same level as  $p\text{CO}_2^{\text{air}}$  with persistently high levels of  $n\text{-}p\text{CO}_2$ ,  $T=29$  (Figs. 6 and 7), leading to the discontinuance of efflux  $\text{CO}_2$ . The discontinuance of efflux  $\text{CO}_2$  was not triggered by the decrease in DIC concentration due to  $\text{CO}_2$  evolution to the air but rather by the decrease in  $p\text{CO}_2^{\text{sea}}$  due to the SST drop. The impact of precipitation on the SST drop is unclear, but it is likely that the precipitation also contributed to sea-surface cooling and the consequent discontinuance of  $\text{CO}_2$  evolution.

In conclusion, the efflux of  $\text{CO}_2$  in the warm season was enhanced by the passage of typhoons. In the present study, the total flux ( $90 \text{ mmol C m}^{-2}$ ) for three typhoons accounted for 60% of the efflux of  $\text{CO}_2$  from the ocean during the period from mid July through mid September, when this area acted as a source for atmospheric  $\text{CO}_2$ . This ratio is higher than that in the Sargasso Sea (44%; Bates et al., 1998a), based on shipboard observations before and after the passage of hurricanes. At the same time, the three typhoons decreased the net flux of  $\text{CO}_2$  into the ocean by 30% during the observation period.

### *3.7. Contribution of typhoons in the western subtropical North Pacific*

Based on best-track data for 1961 to 2004 taken from the Regional Specialized Meteorological Center (RSMC) Tokyo – Typhoon Center ([http://www.jma.go.jp/jma/jma-eng/jma-center/rsmc-hp-pub-eg/RSMC\\_HP.htm](http://www.jma.go.jp/jma/jma-eng/jma-center/rsmc-hp-pub-eg/RSMC_HP.htm)), the frequency of typhoons was investigated for each  $5^\circ \times 5^\circ$  box in the western subtropical North Pacific (Table 1). The size of the boxes was determined from the typical range of sea-surface

cooling by the passage of typhoons, which expands to several hundred kilometers (Wada et al., 2008). An average of 3.6 typhoons per year passed through the box centered in the vicinity of the buoy site, which was slightly higher than three typhoons observed around the buoy. The averages of  $37.9 \text{ m s}^{-1}$  for maximum wind speed and 3.6 days per year for duration were higher and longer than those observed on the buoy, respectively. Elsewhere, the average number of typhoons was 24.8 in the region of  $10$  to  $30^\circ\text{N}$ ,  $120$  to  $160^\circ\text{E}$  in a year. The average duration of 3.8 days per year for each box of  $5^\circ \times 5^\circ$  was comparable, while the average of  $30.9 \text{ m s}^{-1}$  for maximum wind speed was relatively low, compared with that around the buoy site. The higher wind speed around the buoy site was attributable to the frequent passage of intense typhoons, which developed up to the mature stage (Wada and Usui, 2007).

Based on the climatological flux by Takahashi et al. (2002), the efflux of  $\text{CO}_2$  from the ocean in the warm season around the buoy site ( $249 \text{ mmol C m}^{-2}$ ) is higher than that averaged in the western subtropical North Pacific ( $132 - 176 \text{ mmol C m}^{-2}$  for  $10$  to  $30^\circ\text{N}$ ,  $120$  to  $160^\circ\text{E}$ ; Table 1), corresponding to relatively higher  $\Delta p\text{CO}_2$  and wind speed around the buoy site. Considering the differences in wind speed and duration between the buoy site and the western subtropical North Pacific, the contribution of typhoons to the air-sea  $\text{CO}_2$  flux in the western subtropical North Pacific was estimated (Table 1) and compared with the climatological flux from Takahashi et al. (2002). The  $\text{CO}_2$  efflux ( $121 \text{ mmol C m}^{-2}$ ) enhanced by typhoons accounted for  $69 - 92\%$  (average,  $76\%$ ) of the summer efflux in the western subtropical North Pacific. These estimates are higher than the contribution ( $20 - 54\%$ ) of tropical-cyclone-mediated  $\text{CO}_2$  efflux ( $67 - 179 \text{ mmol C m}^{-2}$ , calculated based on estimates by Bates (2002)) to the  $\text{CO}_2$  flux ( $329 \text{ mmol C m}^{-2}$  calculated from climatology by Takahashi et al. (2002)) estimated for all ocean basins of  $40$  to  $10^\circ\text{N}$  and  $10$  to  $40^\circ\text{S}$ . This may be because the effect of vertical mixing of DIC into the mixed layer during tropical cyclone events was not considered in the estimation by Bates (2002) and/or most of the  $\text{CO}_2$  efflux due to tropical

cyclones occurred in the Pacific Ocean basin (Bates, 2002). In any case, these results indicate that the upper ocean physics associated with typhoon passage has a significant influence on the carbon cycle in the upper ocean. Future studies, coupled with precise measurements of DIC changes, will be required to understand its detailed mechanism.

#### 4. Conclusions

It is difficult to directly monitor the parameters essential for estimating CO<sub>2</sub> flux through a severe storm by observations made on any platform other than the buoy. In the present study, the data on  $p\text{CO}_2^{\text{sea}}$  and  $p\text{CO}_2^{\text{air}}$  along with maritime meteorological parameters for 191 days were acquired by an automatic measuring system mounted on the buoy moored in the East China Sea, without any maintenance. During the observation period, three typhoons passed by the moored buoy. The data for all parameters including  $p\text{CO}_2$  were collected without any break under severe conditions during the passage of the typhoons. The variations in surface properties were different through the different processes, depending on where the typhoon passed through. The magnitudes of the fluxes during the passage of typhoons were determined by the wind speed, the amount and persistency of the DIC supply, and the duration of positive  $\Delta p\text{CO}_2$ . It is noted that during the passage of the intense typhoon, the main driver for the variations of the CO<sub>2</sub> flux changed from wind speed to  $p\text{CO}_2^{\text{sea}}$  before and after the minimum SLP. The DIC entrainment due to vertical turbulent mixing contributed to the enhancement of the efflux by holding  $\Delta p\text{CO}_2$  positive. In intense typhoons, a mechanism was found whereby a discontinuation of efflux CO<sub>2</sub> was triggered by the decrease in  $p\text{CO}_2^{\text{sea}}$  due to sea-surface cooling just after the closest approach of the typhoon to the buoy site, irrespective of high DIC levels. It is necessary to clarify these working processes in detail in future studies for a better understanding of the air-sea CO<sub>2</sub> gas exchange enhanced during an intense

storm.

In the present study, the efflux enhanced by the three typhoons increased the efflux of CO<sub>2</sub> from the ocean during summer to autumn by 150% and reduced the absorption of atmospheric CO<sub>2</sub> during the study period by 30%. Typhoons that passed at some distance from the mooring site also had a significant impact on the enhancement of air-sea CO<sub>2</sub> flux through long-lasting vertical turbulent mixing in the upper ocean, indicating that their influence could extend over a wide range. It is suggested that the contribution of typhoons to the carbon cycle in the western subtropical North Pacific is significant. Recent studies suggest that tropical cyclones will be stronger in future warm oceans (e.g., Oouchi et al., 2006). The intensification of tropical cyclones could contribute to an acceleration in the reduction of ocean uptake of CO<sub>2</sub>, consequently exerting positive feedback and increasing global warming.

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## Figure captions

Fig. 1. Location of the JMA moored buoy in the East China Sea ( $28^{\circ}10'N$ ,  $126^{\circ}20'E$  ; depth, 136 m) and configuration of  $CO_2$  measurement system mounted on the buoy.

Fig. 2. Structure of seawater intake and air-sea equilibrator of the cascade type, installed inside the sensor hole on the JMA buoy. The overall height of the cylinder is 1.7 m. The lower half (1.0 m) of the cylinder was set below the sea surface.

Fig. 3. Schematic diagram of the gas flow line.

Fig. 4. Time series of surface properties observed at the JMA buoy in the East China Sea from June 29, 1997 to January 6, 1998.

(a) Sea-level pressure, (b) wind speed (at 7.5 m), (c) water temperature at depths of 1, 50 and 100 m, (d)  $pCO_2^{air}$  and  $pCO_2^{sea}$ , (e)  $n-pCO_{2,T=25}$ , (f)  $\Delta pCO_2$ , and (g)  $CO_2$  flux.

The  $pCO_2$  values were expressed as normalized to an SLP of 1 atm. The normalization of  $pCO_2^{sea}$  to a temperature of  $29^{\circ}C$  was performed using the equation by Weiss et al. (1982). The  $CO_2$  flux values covered three hours and were calculated by using the gas exchange-wind relationships for a short term from Wanninkhof (1992) and wind-speed data observed every three hours on the same buoy. Positive values for  $CO_2$  flux indicate efflux of  $CO_2$  from the ocean and negative values indicate influx into the ocean.

Fig. 5. Storm tracks of typhoons T9711 and T9713 in August and T9719 in September, 1997.

Fig. 6. Time series of surface properties during the passage of typhoon T9711.

(a) Sea-level pressure, (b) wind speed (at 7.5 m), (c) water temperature at depths of 1, 50 and 100 m, (d) SST (1 m), (e)  $p\text{CO}_2^{\text{air}}$ ,  $p\text{CO}_2^{\text{sea}}$  and  $n\text{-}p\text{CO}_{2, T=29}$ , (f)  $\Delta p\text{CO}_2$ , and (g)  $\text{CO}_2$  flux.

Fig. 7. As Fig. 6, but for typhoon T9713.

Fig. 8. As Fig. 6, but for typhoon T9719.

Fig. 9. Contributions of wind speed (blue circle),  $p\text{CO}_2^{\text{sea}}$  (red square) and  $p\text{CO}_2^{\text{air}}$  (green triangle) to the  $\text{CO}_2$  flux (gray bold line) during the typhoon events. (a) T9711, (b) T9713, and (c) T9719. The contribution of each parameter was calculated based on the corresponding term in the right side of Eq. 2;  $\frac{\partial E(W)}{\partial t} \cdot \Delta p\text{CO}_2$  for wind speed,  $\frac{\partial p\text{CO}_2^{\text{sea}}}{\partial t} \cdot E(W)$  for  $p\text{CO}_2^{\text{sea}}$  and  $-\frac{\partial p\text{CO}_2^{\text{air}}}{\partial t} \cdot E(W)$  for  $p\text{CO}_2^{\text{air}}$ . The sum of contributions of three parameters is equal to the flux,  $\frac{\partial F}{\partial t}$ . Arrows show the time of the minimum SLP. The correlation of each parameter with the  $\text{CO}_2$  flux is as follows:  $r^2 = 0.90$  ( $p < 10^{-7}$ ) in T9711,  $r^2 = 0.84$  ( $p < 10^{-4}$ ) in T9713, and  $r^2 = 0.18$  ( $p = 0.08$ ) in T9719 for wind speed before the minimum SLP;  $r^2 = 0.89$  ( $p < 10^{-4}$ ) in T9711,  $r^2 = 0.74$  ( $p = 0.001$ ) in T9713, and  $r^2 = 0.49$  ( $p = 0.004$ ) in T9719 for  $p\text{CO}_2^{\text{sea}}$  after the minimum SLP;  $r^2 = 0.45$  ( $p = 0.003$ ) for  $p\text{CO}_2^{\text{sea}}$  and  $r^2 = 0.14$  ( $p > 0.1$ ) for wind speed in T9719 over the entire period (2 days) of typhoon passage.

Table 1

Statistics of typhoons compiled around the buoy site and in the western subtropical North Pacific

| Location                                         | Buoy site            |                                       | Western subtropical North Pacific<br>(10-30°N, 120-160°E) <sup>b</sup> |
|--------------------------------------------------|----------------------|---------------------------------------|------------------------------------------------------------------------|
|                                                  | 1997<br>(This study) | Average <sup>a</sup> for<br>1961-2004 | Average for 1961-2004                                                  |
| Period                                           |                      |                                       |                                                                        |
| Number (per year)                                | 3                    | 3.6                                   | 0.78 <sup>c</sup>                                                      |
| Duration (days per year)                         | 2.8                  | 3.6                                   | 3.8                                                                    |
| Maximum wind speed<br>(m s <sup>-1</sup> )       | 31.1                 | 37.9                                  | 30.9                                                                   |
| Summer efflux<br>(mmol C m <sup>-2</sup> )       | 150                  | 249 <sup>d</sup>                      | 132 – 176 <sup>d</sup>                                                 |
| Efflux by typhoons<br>(mmol C m <sup>-2</sup> )  | 90                   | 173 <sup>e</sup>                      | 121 <sup>e</sup>                                                       |
| Contribution of typhoons to<br>summer efflux (%) | 60                   | 69                                    | 69 – 92                                                                |

<sup>a</sup> Values compiled in a box of 5° x 5° around the buoy site (28°10'N, 126°20'E) were annually averaged for 1961-2004.

<sup>b</sup> Values compiled in the region of 10 to 30°N, 120 to 160°E were annually averaged for 1961-2004. Values are expressed as those per year for a box of 5° x 5° and can directly be compared with those for the buoy site.

<sup>c</sup> 24.8 typhoons per year passed through the region of 10 to 30°N, 120 to 160°E were divided among 32 boxes of 5° x 5°.

<sup>d</sup> Positive values of CO<sub>2</sub> flux were taken from the climatology by Takahashi et al. (2002) and were summed up for the source season in each 4° x 5°. For the region of 10 to 30°N, 120 to 160°E, efflux values summed up in the respective 4° x 5° boxes were latitudinally averaged in the range of 120 to 160°E. The standard deviation of efflux value in 10 to 30°N ranged in 63 to 112 mmol C m<sup>-2</sup>.

<sup>e</sup> Calculated from the efflux value by typhoons estimated in this study, based on the differences in maximum wind speed and duration between observation and statistical average. With regard to wind speed, the value was converted using the *E(W)* equation from Wanninkhof (1992).

















