A significant net sink for CO$_2$ in Tokyo Bay

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Most estuaries and inland waters are significant source for atmospheric CO$_2$ because of input of terrestrial inorganic carbon and mineralization of terrestrially supplied organic carbon. In contrast to most coastal waters, some estuaries with small freshwater discharge are weak source or sometimes sink for CO$_2$. Extensive surveys of pCO$_2$ in Tokyo Bay showed that the overall bay acts as a strong net sink for atmospheric CO$_2$. Although small area was a consistent source for CO$_2$, active photosynthesis driven by nutrient loading from the land overwhelmed the CO$_2$ budget in the bay. Here we show a comprehensive scheme with a border where air-sea CO$_2$ flux was $\pm 0$ between nearshore waters emitting CO$_2$ and offshore waters absorbing CO$_2$. The border in Tokyo Bay was extremely shifted toward the land-side. The shift is characteristic of highly urbanized coastal waters with an extensive sewage treatment system in the catchment area. Because highly urbanized coastal areas worldwide are expected to quadruple by 2050, coastal waters such as Tokyo Bay are expected to increase as well. Through extrapolation of Tokyo Bay data, CO$_2$ emission from global estuaries would be expected to decrease roughly from the current 0.074 PgC year$^{-1}$ to 0.014 PgC year$^{-1}$ in 2050.

Despite their relatively small areal coverage, coastal waters significantly contribute to the global account of CO$_2$ exchange due to their dense biological activities$^1$–$^3$. Coastal waters have been regarded as CO$_2$ sources to the atmosphere because of their input of terrestrial organic carbon and subsequent mineralization$^2$–$^3$. However, current estimates of the coastal CO$_2$ budget are based on observations in only limited types of marine systems, irrespective of the large heterogeneity in such biogeochemical settings. In fact, Kuwae et al.$^4$ have recently discussed the possibility of net carbon fixation in certain types of coastal systems including those affected by intense anthropogenic activities. Furthermore, the available observational data from a particular system are generally insufficient to cover the large spatial and temporal variability of the coastal carbon cycle, thus resulting in an uncertain estimate of the carbon budget. More data from various systems with sufficient areal and temporal coverage are thus needed.

We conducted extensive observations of the partial pressure of carbon dioxide (pCO$_2$) in Tokyo Bay from March 2007 to December 2010 during 49 observational cruises. Tokyo Bay is a semi-enclosed embayment with an area of approximately 1320 km$^2$ and a mean water depth of 19 m. The bay is bounded by highly urbanized areas. The bay has suffered from severe cultural eutrophication since the late 1950s, along with rapid development in catchment areas including the Tokyo metropolis. Sewer systems with secondary or tertiary treatment cover most of the urban regions, and yet phytoplankton blooms persist throughout the year and anoxic bottom waters consistently appear during the summer. In some estuary, carbonate dissolution contribute to coastal CO$_2$ concentration about 20%$^5$. As regard to Tokyo Bay, shellfish at the bottom water are the major calcifier and calcifying plankton rarely dominates$^6$. In addition, surface waters at the bay are oversaturated with respect to calcium carbonate saturation throughout the year$^7$. Therefore, we suppose that carbonate dissolution do not contribute to coastal CO$_2$ in the bay.

Results and Discussion

All of the pCO$_2$ data points ($n = 21,076$) from our observations are plotted against salinity in Fig. 1, and each point in the figure represents the value of pCO$_2$ at an interval of one minute during the observation. R/V Seiyo-maru observed most of the area during 40 cruises from March 2007 to December 2010 on a monthly basis. R/V Hiyodori observed the innermost head of the bay during nine cruises during the period from May 2009 to October 2010. The pCO$_2$ values of surface bay ranged from 10 to 7218 μatm. The pCO$_2$ values for 21,076 data

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indicated that a total of 16,345 points were under-saturated with respect to the atmospheric equilibrium; oversaturation was found only in 4,731 data points.

The air-sea CO$_2$ fluxes were calculated from all of the pCO$_2$ data (see Methods). The results obtained from different times were binned into a 500 m $\times$ 500 m horizontal resolution grid and then averaged together (Fig. 2a). The northwestern head of the bay was the only area with consistent positive CO$_2$ flux to the atmosphere, whereas the central bay and bay mouth were a sink for atmospheric CO$_2$. The location where CO$_2$ flux was $\pm$ 0 roughly coincided with the location where the average annual surface salinity was 25 (Fig. 2b). The areas where salinity was above and below 25 were estimated according to methodology described in Ninomiya et al.$^8$. The average CO$_2$ flux (positive values indicate efflux into the air) for areas where salinity was less than 25 (81 km$^2$) was 15.2 mmolC m$^{-2}$ day$^{-1}$. In contrast, the average CO$_2$ flux for areas where salinity was greater than 25 (1239 km$^2$) was $-10.6$ mmolC m$^{-2}$ day$^{-1}$. The area weighted annual CO$_2$ flux in Tokyo Bay had a rate of $-8.8$ mmolC m$^{-2}$ day$^{-1}$; the bay as a whole was a strong net sink for atmospheric CO$_2$. The annual CO$_2$ flux in Tokyo Bay was calculated to be $-5.2 \times 10^9$ gC year$^{-1}$.

The seasonal variations of pCO$_2$ and related parameters at three representative stations in the bay are presented in Fig. 3. The parameters include the observed pCO$_2$, pCO$_2$ normalized to average temperature (19°C), temperature, salinity and chlorophyll $a$ (Chl $a$) concentration. In the northwestern head of the bay (St. TPE), where salinity was lowest among the three stations, pCO$_2$ values generally exceeded the atmospheric equilibrium. There was no distinct pattern observed for the seasonal variation of pCO$_2$ (93–1920 μatm), and the variation of pCO$_2$ was not correlated with Chl $a$ ($R^2 = 0.03$, $P > 0.1$, $n = 39$). At the central bay (St.F6) and the bay mouth (St.06), low values of pCO$_2$ were observed during spring and summer (70–336 μatm), whereas pCO$_2$ values were close to atmospheric equilibrium values during autumn and winter (264–449 μatm). In terms of the seasonal

![Figure 1. All pCO$_2$ (μatm) data points (n = 21,706) plotted against salinity.](image1)

![Figure 2. Map of (a) sea-air flux (mmol C m$^{-2}$ day$^{-1}$) and (b) salinity, binned into a 500 m $\times$ 500 m horizontal resolution grid. Positive flux values (red) represent outgassing of CO$_2$ to the atmosphere, and negative values (blue) represent uptake. Maps were created using Generic Mapping Tools software (GMT v4.5.12; http://gmt.soest.hawaii.edu/)](image2)
Figure 3. (a) Map of the study site. The ship routes of all cruises are shown as blue lines. Data were measured with a sampling frequency of one minute (n = 21076). Circles indicate the three representative stations (06, F6, and TPE). TPE stands for Tokyo Port Entrance. Isobaths of 20-, 50- and 200-m are also shown. (b) Seasonal variations of pCO₂ (μatm) and atmospheric pCO₂ (the dotted line). (c) pCO₂ normalized to average temperature (19°C). (d) Temperature (°C) and salinity and (e) Chl a (μg L⁻¹) at the three representative stations during the observation period (March 2007 – December 2010). Map (a) was created using Generic Mapping Tools software (GMT v4.5.12; http://gmt.soest.hawaii.edu)37.
variation of pCO₂ in the central bay and bay mouth, a negative correlation was found between pCO₂ and Chl a \( (R^2 = 0.64, P < 0.001, n = 77) \). The correlation suggests that active photosynthesis reduced pCO₂ during spring and summer, hence suggesting that pCO₂ in surface waters is mainly controlled by biological activity. The stratified water column prevents the CO₂ supply from the deeper waters with high CO₂ levels. In autumn and winter, the low photosynthetic rate along with the well-mixed water column resulted in pCO₂ values close to the atmospherically equilibrium. Temperature did not significantly contribute to the seasonal variation of pCO₂. The pCO₂ values normalized to annual average temperature \( (19{°}C) \), which accounted for the temperature dependence of equilibrium constants and the solubility coefficient\(^9\), did not affect the seasonal variation of pCO₂.

In June 2010, pCO₂ decreased to a low of 10 μatm, and Chl a concentration was >300 μg L\(^{-1}\), which we found to be the lowest reported value in a marine environment. Based on the low observed CO₂ concentrations, the exhaustion of gaseous CO₂ in seawater resulted in the limitation of the CO₂ supply to algal cells, which may limit the cells growth. In such CO₂ limiting conditions, phytoplankton would need to take up bicarbonate using the proton pump mechanism and the carbonic anhydrase\(^10\).

The CO₂ absorption of 5.2 × 10\(^{10}\) gC year\(^{-1}\) in Tokyo Bay was in accordance with the overall carbon budget of the bay. A mass balance model estimated the total organic carbon (TOC) influx from the rivers to Tokyo Bay to be 8.1 × 10\(^{10}\) gC year\(^{-1}\) and the TOC efflux from the bay to the open ocean to be 9.4 × 10\(^{10}\) gC year\(^{-1}\). On the basis of actual observations, the amount of organic carbon burial was estimated to be 4.2 × 10\(^{10}\) gC year\(^{-1}\). A different box model estimated the dissolved inorganic carbon (DIC) influx to Tokyo Bay to be 11.2 × 10\(^{10}\) gC year\(^{-1}\) and the DIC efflux from the bay to be 13.4 × 10\(^{10}\) gC year\(^{-1}\). By combining these two estimates, an additional carbon input of 7.7 × 10\(^{10}\) gC year\(^{-1}\) will be required, a value roughly equal to our estimate of net CO₂ uptake. In addition, the above analysis also suggests that the eventual sink of the fixed carbon from CO₂ absorption in Tokyo Bay would be both from its burial in bay sediments and export to outer oceanic areas.

Because our observations were conducted on monthly basis, our estimates of CO₂ flux may have failed to report on events on a daily and/or weekly time-scale. We acknowledge that our results regarding the net CO₂ uptake in the bay might be compromised if these short-term temporal events contributed to significant CO₂ emission or outgassing from surface waters. We postulated two types of outgassing events in Tokyo Bay and found that such outgassing events should be insignificant to our carbon budget. First, we postulated a sudden vertical mixing event in early autumn when stratification was weakened. The observed pCO₂ values of the bottom waters during the stratification season from June to September were 400–940 μatm, with a typical water-column average of approximately 600 μatm. Even if the outgassing from seawater at this level of CO₂ occurred in the entire area of the bay for 30 days, the amount of CO₂ efflux under typical wind speed would be 0.6 × 10\(^{10}\) gC, which would account for 12% of the air-sea CO₂ exchange in the bay (5.2 × 10\(^{10}\) gC). Second, we postulated coastal upwelling events, which are generally observed in the northeastern part of the bay in late summer. These upwelling events cause conspicuous milky turquoise waters\(^11\) due to the oxidation of hydrogen sulfide in anoxic bottom waters. We observed these bottom waters on 16 and 23 September, 2010, in which pCO₂ values ranged from 765 to 1,161 μatm with an average of 967 μatm and the average CO₂ flux was estimated to be 54.0 mmolC m\(^{-2}\) day\(^{-1}\). Because of the conspicuous water color, the area and duration of the upwelling events are well described in the bay; the events were observed for an average of 11.4 days a year from 2007 to 2010, with a maximum area of 80 km\(^2\). On the basis of this average duration and area, the observed CO₂ flux would yield an annual flux of 5.9 × 10\(^{10}\) gC year\(^{-1}\). This value accounts for only 1.2% of the air-sea CO₂ exchange in the entire bay.

This study clearly demonstrated that Tokyo Bay as a whole is a net sink for atmospheric CO₂; however, this finding may contradict those of many studies on coastal waters. Most inland waters and estuaries have been reported to be significant sources of CO₂ to the atmosphere due to respiration of terrestrial organic carbon\(^2,3,16\) and terrestrial input of freshwater CO₂\(^1\). In contrast, continental shelf areas, which are laid offshore of coastal areas, have generally been reported to be sinks for atmospheric CO₂ due to nutrient input through coastal waters and from pelagic deep waters\(^2,3,18,19\). Oceanic basins that are further offshore are either weak sinks or weak sources of atmospheric CO₂, depending on the biogeochemical settings of the basin\(^20\). Although it is confined to a very small inner part of the bay, Tokyo Bay certainly has an area of CO₂ emission, and the CO₂ emission mechanism in this area is common to that of other coastal areas. The net CO₂ absorption in the main body of the bay is driven either by a mechanism similar to that of continental shelf regions or by biological CO₂ fixation with a terrestrial contribution. On the basis of these considerations, we propose a generalized scheme for the CO₂ budget in a continuing water system composed of nearshore water emitting CO₂, an outer water absorbing CO₂, and pelagic water with a neutral CO₂ budget (Fig. 4).

In this scheme, there would be a border where the air-sea CO₂ flux is ±0 between the nearshore waters emitting CO₂ and an outer waters absorbing CO₂. On the land-side of this border, CO₂ emission due to biological degradation of terrestrial organic matter would exceed CO₂ uptake due to photosynthesis. The CO₂ emission would decrease toward the offshore side, and photosynthetic CO₂ uptake would exceed emissions on the offshore side of this border. The location of this border may shift either offshore or inshore. In fact, several studies have observed that some waters in estuaries with small freshwater discharge are weak sources or sometimes weak sinks of CO₂\(^21-23\). In addition, some continental shelves with large freshwater discharge have been reported to be sources of CO₂ to the atmosphere\(^24\). We interpret that these rather atypical observations are associated with the shift of the aforementioned border. The shift is likely caused by the different terrestrial organic carbon load accompanied by freshwater discharge\(^24\). Other factors that may affect the border shift and intensity of CO₂ flux include hydrographic and geomorphological characteristics such as a stratified estuarine system\(^22\), a microtidal estuarine system\(^25\), an open/enclosed nature of the coast\(^22,28\), and a submerged aquatic vegetation in shallow coastal waters\(^27\).

In the case of Tokyo Bay, the border is extremely shifted inshore. The shift reflects the relatively low organic carbon supply from land and the active organic matter production driven by the massive nutrient supply from land. The seasonal stratification and semi-enclosed nature of the embayment should further facilitate the net uptake of CO₂ in Tokyo Bay. The low organic matter supply and large nutrient supply from land may seem
contradictory. We believe that this imbalance between organic matter and nutrient supply is largely derived from
the secondary sewage treatment in the catchment area of Tokyo Bay. Currently, secondary-treated effluent flow-
ing into Tokyo Bay accounts for 50% of the total freshwater discharge. Sewage treatment plants along Tokyo
Bay remove organic carbon from freshwater at a rate of $7.7 \times 10^{10}$ gC year$^{-1}$. This value is comparable to the
uptake of CO$_2$ observed in this study ($5.2 \times 10^{10}$ gC year$^{-1}$). As the quantity of dissolved organic carbon (DOC)
flowing into the bay has decreased by 60% in the last 40 years, the quality of DOC has become more recalcitrant
due to improved sewage treatment. The decrease in turbidity due to the decrease in organic carbon inflow has
also enhanced phytoplankton activity from the improvement of light availability. The effect of sewage treatment
has also been reported in other estuary systems; Amann et al. have recently reported that pCO$_2$ decreased from
7000 to 2500 μatm at the oxygen minimum zone in 1986 and 2007 due to the installation of sewage treatment
plants in the Elbe estuary.

Urbanized coastal waters that have a relatively large coverage of secondary sewage treatment tend to act as a
strong net sink for atmospheric CO$_2$. The progress made in the urbanization of coastal areas with improved sew-
ge treatment is common in many parts of the world. This development will have an impact on the future budget
of marine CO$_2$. Moreover, approximately 40% of the world's population is currently settled in coastal zones,
and developed urban areas with completed sewer systems are expected to expand rapidly. According to a UNEP
report, highly developed coastal areas occupied only 15% of the world coastal zones in 2002, but this figure is
expected to rise to 60% in 2050. Therefore, the CO$_2$ budget characteristic of Tokyo Bay is expected to be observed
in more marine coastal areas in the future. Although it may be difficult to predict future trends, we assume that
highly urbanized coastal waters currently occupy 15% of the world coastal zones and are expected to occupy 60%
by 2050. When the CO$_2$ fluxes in highly urbanized coastal waters and ordinal coastal waters are the same values
as that of Tokyo Bay ($-3.2$ moles C m$^{-2}$ year$^{-1}$) and previously published data (7.7 moles C m$^{-2}$ year$^{-1}$), respectively,
the CO$_2$ emissions from global coastal waters will be estimated to be approximately 0.074 PgC year$^{-1}$ at present
and 0.014 PgC year$^{-1}$ in 2050. The emission of CO$_2$ to the atmosphere from estuaries is calculated to be 21% and
85% lower than previous estimates. As a result, the CO$_2$ budget of the global coastal waters would be expected to
be a sink rather than a source.
Methods
The ship routes of all cruises were presented in Fig. 3 (a). Each point in this figure represents a pCO₂ value measured at a one-minute interval. Observations of pCO₂, salinity, and temperature were conducted during 40 cruises of the R/V Seiyo-maru and 9 cruises of the R/V Hiyodori. Measurements of pCO₂, salinity, and temperature were taken with a sampling frequency of one minute. Surface seawater was pumped up from the ship's bottom at ca. 2.0 m depth. Our pCO₂ measuring system consisted of a NDIR analyzer (LI-820, Li-Cor) and a membrane equilibrator. The membrane equilibrator was composed of multi-layered composite hollow-fiber membrane modules (MHF module, Mitsubisi Rayon Co., Ltd.). The equilibrator was made of 6 MHF modules to create more equilibrator. The membrane equilibrator was composed of multi-layered composite hollow-fiber membrane modules (Seiki Co., Ltd. at Seiyo-maru and YSI 6920 at Hiyodori). The net flux of CO₂ across the air-sea interface was determined by the method given by Wanninkhof. Wind speed data were obtained from the Japan Coast Guard (http://www6.kaiho.mlit.go.jp/tokyowan/) from stations (Tsurugisaki, Kannonzaki, Honmoku, and Tokyo 13 gouchi) close to the observation areas. The samples for chlorophyll a (Chl a) measurement were collected at each stations using bucket and filtered through precombusted (450 °C, 3 h) GF/F filters. After filtration, chlorophyll pigments were extracted using N, N-dimethylformamide, and the concentrations of Chl a were determined by the fluorometric method (fluorometer used TD-700, Turner Desings).

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Author Contributions
A.K. designed the experimental setup, carried out part of the experiments, and wrote the manuscript; Y.M.
carried out a portion of the experiments; and J.K. contributed to the design of the experiments and discussion of
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