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pCO_2 and carbon fluxes across sea-air interface in the Changjiang Estuary and Hangzhou Bay*

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Abstract Partial pressure of CO_2 (pCO_2) was investigated in the Changjiang (Yangtze River) Estuary, Hangzhou Bay and their adjacent areas during a cruise in August 2004, China. The data show that pCO_2 in surface waters of the studied area was higher than that in the atmosphere with only exception of a patch east of Zhoushan Archipelago. The pCO_2 varied from 168 to 2 264 μatm, which fell in the low range compared with those of other estuaries in the world. The calculated sea-air CO_2 fluxes decreased offshore and varied from −10.0 to 88.1 mmol m^{-2} d^{-1} in average of 24.4 ± 16.5 mmol m^{-2} d^{-1}. Although the area studied was estimated only 2×10^4 km^2, it emitted $(5.9 \pm 4.0) \times 10^3$ tons of carbon to the atmosphere every day. The estuaries and their plumes must be further studied for better understanding the role of coastal seas playing in the global oceanic carbon cycle.

Keyword: partial pressure of carbon dioxide; spatial distribution; sea-air exchange; Changjiang Estuary; Hangzhou Bay

1 INTRODUCTION

Increasing in carbon dioxide (CO_2) level in the atmosphere has become a concern in many scientists as perhaps a serious global environmental problem to the human beings (Haugan, 1997). The carbon transport by rivers has been well-documented as a part of the global carbon cycle (Meybeck, 1993; Abril et al., 2000). Estuaries are important transitional zones between land and sea. With rapid development of industry and agriculture in recent decades, huge amounts of nutrients with inorganic and organic carbon from both natural and anthropogenic sources have been introduced into ocean via shallow coastal waters every year, especially estuaries (Wu, 1999; Schlünz and Schneider, 2000). It was found over 30 years ago in pioneering studies by Park et al. (1969), Kelley and Hood (1971), and Kelley et al. (1971) that some coastal ecosystems, such as estuaries and upwelling systems, act (at least temporally) as strong sources of CO_2 to the atmosphere.

The Changjiang (Yangtze River) Estuary, about 120 km long and over 90 km wide at its outer limit, is a mesotidal estuary and characterized by the complexity of morphology associated multi-order bifurcations (Li and Zhang, 1998; Yang et al., 2003). Hangzhou Bay, the estuary of the Qiantang River, is dominated by macrotides. The sedimentary dynamic process in the bay is characterized with the alternation of rapid erosion and deposition in tidal cycles. Both the Changjiang Estuary and Hangzhou Bay are within the Changjiang Delta Region and near the metropolis Shanghai, the most important economic development zone in China.

The areas surrounding the Changjiang Estuary and Hangzhou Bay have been the focus of element biogeochemical research for many years. In recent years, environmental changes in the Changjiang Estuary and Hangzhou Bay resulted from rapid development in the surroundings have caught public

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attention. However, information for clear understanding of dissolved inorganic carbon behaviors in these areas remains insufficient. The main objective of this paper is to describe the pCO_2 spatial distribution in surface waters of the Changjiang Estuary, the Hangzhou Bay, and their adjacent areas, estimate the air-sea fluxes of CO_2, and discuss the main factors impacting them.

2 MATERIALS AND METHODS

The field investigation was carried out from August 5 to 10, 2004, on board of *R/V Zhe Hai Huan Jian*. 37 stations were set in the areas of the Changjiang Estuary, Hangzhou Bay and nearby regions and covered almost entire salinity gradient (Fig.1). Water samples were collected with Go-Flo sampler just below the sea surface.

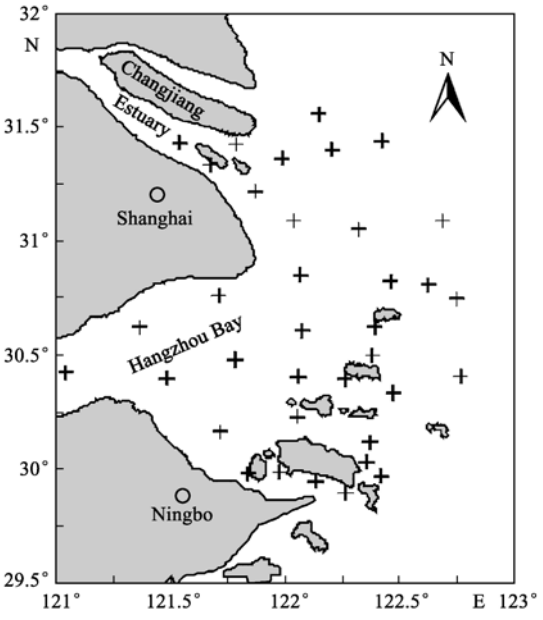

Fig.1 The study area and the sites deployment

Salinity (S), temperature (T), pH and total alkalinity (TA) of the samples were measured *in situ*. S and T were measured using a Sea-Bird SBE 19plus CTD probe. For pH measurements, an ORION Ross type combination electrode was used and calibrated on the NBS scale. Precision for pH measurements was 0.03 pH units. Samples for total alkalinity (TA) were collected in 500-ml glass bottles according to the DOE (Department of Energy, USA) procedures (Dickson and Goyet, 1994) and analyzed onboard as quickly as possible by Gran titration with the precision of approximately 0.1%–0.3%.

Values of carbon dioxide partial pressure (pCO_2) and total inorganic carbon (TCO_2) in the water column were calculated from TA and pH measurements, with the dissociation constants of carbonic acid from Mehrbach et al. (1973). The CO_2 solubility was calculated according to Weiss (1974).

The flux of CO_2 across the air-sea interface (F) was calculated from the difference of CO_2 partial pressure between the water surface and the air (ΔpCO_2), and the CO_2 transfer velocity (k) using the equation:

$$F = k \times \alpha \times \Delta pCO_2$$

where α is the CO_2 solubility at *in situ* temperature and salinity (Weiss, 1974). The magnitude of the flux is mainly imposed by the value of k, which is a function of various parameters and processes such as wind speed, turbulence at the interface, air bubbles, and surface organic films, etc (Borges et al., 2004). The direct measurements of the physical component of gas exchange in rivers and estuaries were fewer than those in streams, lakes and marine systems (Raymond and Cole, 2001). Raymond and Cole (2001) suggested that under a lack of direct estuarine k measurements condition, k_{600}, i.e. the k for CO_2 at 20°C in freshwater, that is, k at a Schmidt number of 600, should be in the range of 3−7 at average wind speeds (4.6±0.28 m s^{-1}), tidal velocity (0.34±0.28 m s^{-1}), and estuary depth (>10 m). Therefore, as an attempt, a k value of 7 cm h^{-1} should cover the average physical mixing condition in the studied area considering the Changjiang Estuary and Hangzhou Bay as a mesotidal and macrotidal region respectively, and should be adequate for the purpose of tracing biogeochemical processes in this work. ΔpCO_2 imposes the direction of the flux and mainly depends on the pCO_2 in surface water since atmospheric pCO_2 is relatively homogeneous and much less variable. Tan et al. (2004) measured the atmospheric pCO_2 in the west East China Sea (ECS) near the investigated spot of this study in the summer of 2001, and gained an average value of 375 μatm. Considering the rise in atmospheric CO_2 concentration with time, an atmospheric pCO_2 of 380 μatm was used to estimate the CO_2 flux across sea-air interface. A positive flux value indicates a transfer of CO_2 from surface water to the atmosphere, while a negative one represents the reverse.

3 RESULTS AND DISCUSSION

3.1 Spatial distribution of pCO_2

A typical estuary can be divided into two sections, namely inner and outer estuary (Abril and Borges, 2004). Inner estuary is a region between the limit of the tidal influence and the river mouth; outer estuary is the plume of freshened water which floats on denser coastal seawater and can be traced miles away from geographical mouth of the estuary and it is also called an "estuarine plume" (Kechum, 1983). The surface area of a plume is commonly defined on the basis of salinity in surface waters. Salinity value of 1 lower than the adjacent oceanic basin is arbitrarily used as the offshore boundary (Borges and Frankignoulle, 2002). The characteristic value of surface salinity of the East China Sea, with which the Changjiang Estuary and Hangzhou Bay are connected, was >33 (Wang and Xu, 2004). In this cruise, the distribution of isohaline indicated that the investigated area covered both Changjiang and Qiantang River estuaries and their plumes (Fig.2a).

Fig.3 shows the distribution of pCO_2 across the sampled region. A distinct spatial difference in

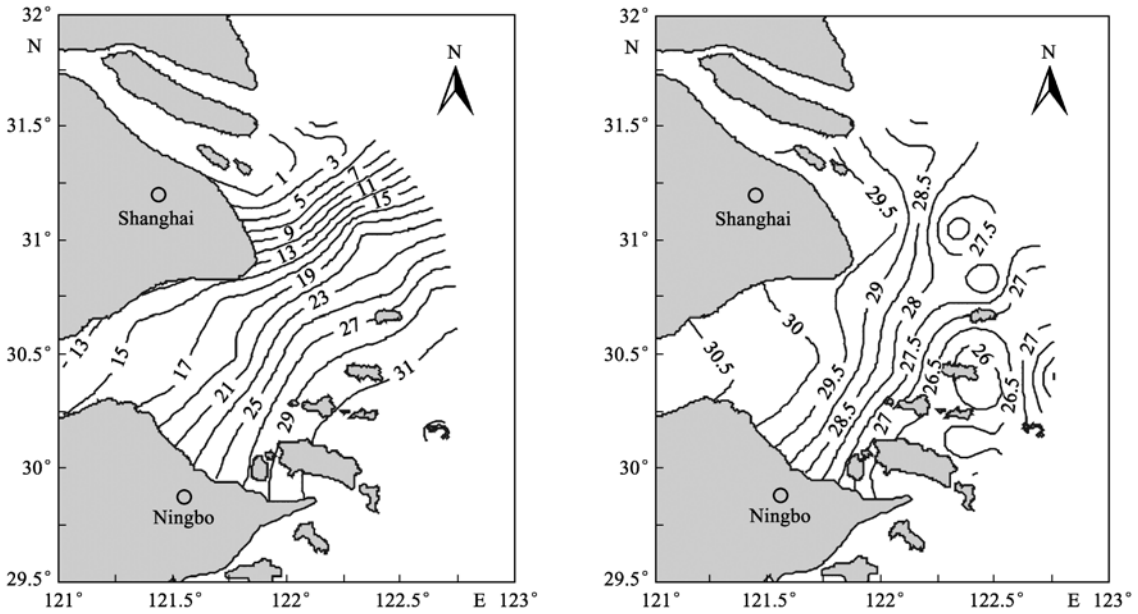

Fig.2 Isohaline (a) and isotherm (b, °C) distributions in surface waters

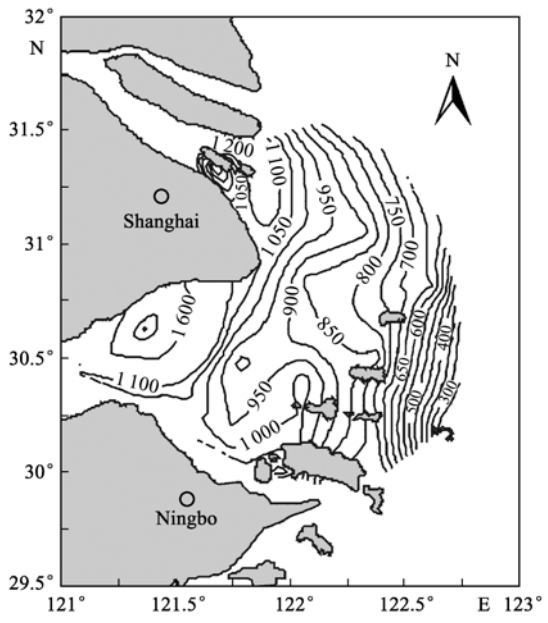

Fig.3 Distribution of pCO_2 isolines (μatm) in surface waters

overall distribution of pCO_2 values was shown with the minimum and maximum of 168 and 2 264 μatm respectively. The pCO_2 decreased offshore while salinity rose. However, the maximum of pCO_2 did not coincide with the minimum of salinity (at the mouth of the Changjiang Estuary) as expected if the input of over-saturated water from the Changjiang was the only process controlling the distribution of pCO_2. The inner Changjiang Estuary and Hangzhou Bay were entirely over-saturated with CO_2. The variation in pCO_2 values was >1 000 μatm. The estuarine plume region was also over-saturated in CO_2, except for a small area in the east.

Estuaries are known for significant supersaturation of CO_2 with respect to the atmosphere. pCO_2 as high as 15 500 μatm was once reported in the Scheldt Estuary (Belgium and Netherlands), which is >40 times pCO_2 of the present atmosphere equilibrium (Hellings et al., 2001). Such high pCO_2 values are believed resulted

from intricate biological and physicochemical processes that characterize estuarine dynamics (Hellings et al., 2001). Investigations in the 1990's indicated that, in Europe, the estuaries emitted between 30 and 60 million tons of carbon per year to the atmosphere, representing 5%−10% of anthropogenic CO_2 emissions from the Western Europe at that time (Frankignoulle et al., 1998). The surface pCO_2 in the inner Changjiang Estuary was similar to those of some European estuaries in spring such as Elbe (Germany) and Gironde (France) (Frankignoulle et al., 1998), and was much lower than those of some well-studied estuaries such as Pearl River Estuary (China) and Scheldt Estuary (Frankignoulle et al., 1998; Hellings et al., 2001; Zhai et al., 2005).

The mechanism by which estuarine systems can sustain such high levels of pCO_2 remains unclear. Abril et al. (2000) indicated that heterotrophic activity and acidification due to nitrification within estuarine zone were major factors for the total estuarine emission to the atmosphere, while the excess CO_2 transport by rivers followed by ventilation in the estuary was a minor one. Cai and Wang (1998) believed that the combined effects of pelagic and benthic respiration, photodegradation, and the mixing of seawater and acidic river water were insufficient to sustain the high pCO_2 values and then high water-to-air fluxes in the estuaries they studied, they suggested that the CO_2 input from organic carbon respiration in tidally flooded salt marshes controlled the CO_2 concentration. This explanation is consistent with a subsequent mass balance study of biogenic gases (Cai et al., 1999).

Therefore, pCO_2 distribution pattern in estuary is resulted from the combination of various processes: the production/degradation/export of organic carbon, the production/dissolution/export of carbonates, the input of dissolved inorganic carbon by vertical mixing processes and/or freshwater runoff and the thermodynamic effects related to both water temperature variations and water mass mixing.

Compared with the inner estuary, the outer estuary is substantially different properties in terms of CO_2, where intense phytoplankton bloom can consume significant amount of dissolved CO_2. pCO_2 in estuarine plume depends not only on its primary production/respiration balance but also on the quantity of excess dissolved CO_2 advected from the inner estuary. Thus, CO_2 atmospheric exchanges in plumes are affected by many parameters, among which river discharge, the degree of heterotrophy in the inner estuary, the availability of nutrients and light, and the stratification of the water column are important ones (Abril and Borges, 2004).

In the sampling area of this study, light penetrates deeper and deeper into water west to east with deposition of suspended particles, and the region near the eastern boundary of the studied area has similar characteristics to the sea to which it connected. The light and nutrients from the surroundings of the Changjiang Estuary and Hangzhou Bay are favorable for phytoplankton blooms (Cloern, 1996). Thereby, this was a possible factor causing the pCO_2 distribution pattern described in this study.

Temperature affects the equilibrium constants of dissolved inorganic carbon and, in particular, the solubility coefficient of CO_2, so that pCO_2 rises by about 4% with the increase of every 1°C in temperature (Borges and Frankignoulle, 2002). Fig.2b shows that, in the sampling region, surface water temperature decreased from the west to east, similar to that of pCO_2 (Fig.3). Thus, this was another possible factor causing the pCO_2 distribution pattern shown in this paper.

Table 1 compares pCO_2 ranges in various estuaries of the world. pCO_2 concentration instead of CO_2 flux was compared for removing the uncertainties in using different k values. The Changjiang Estuary and Hangzhou Bay fell in the low end of pCO_2 reported for estuaries. Part of the discrepancy may be due to the lack of seasonal pCO_2 data in many estuaries including the ones we researched. Besides, large differences in organic matter concentrations among estuaries are another important reason for the variation in estuarine CO_2 supersaturation. For example, the dissolved organic carbon (DOC) concentrations in the Satilla and Altamaha River Estuaries were 25–50 and 10 mg L^{-1} respectively (Cai and Wang, 1998), and 5 mg L^{-1} in average in the low salinity region of the York River Estuary (Raymond and Bauer, 2000), whereas the maximum was only 3.5 mg L^{-1} obtained in May 2003 in the entire salinity gradient of the Changjiang Estuary (Gao and Song, unpublished data).

3.2 Sea-air CO_2 flux

The exchange of CO_2 between an aquatic ecosystem and the overlying atmospheric is an area of intense interest for scientists. An aquatic system can be a significant CO_2 source or sink on a global or regional scale; and the magnitude and direction of the CO_2 flux in an ecosystem can be very

Table 1 pCO_2 ranges reported in estuaries of the world*

Estuary (location)	pCO_2 range (μatm)	References
Scheldt (Belgium/Netherlands)	~100−15 500 **	Borges and Frankignoulle (2002); Hellings et al. (2001)
Saja-Besaya (Spain)	264−9 728 #	Ortega et al. (2005)
Satilla (US-Georgia)	420−8 200 #	Cai and Wang (1998); Cai et al. (1999)
Altamaha (US-Georgia)	380−7 800 #	Cai and Wang (1998)
Sado (Portugal)	450−5 700 **	Frankignoulle et al. (1998)
Thames (UK)	465−5 200 #	Frankignoulle et al. (1998)
Pearl River (China-Guangdong)	~360−4 785 **	Zhai et al. (2005)
Ems (Germany/ Netherlands)	525−3 755 **	Frankignoulle et al. (1998)
Loire (France)	~600−~2 900 **	Abril et al. (2003)
Gironde (France)	440−2 860 **	Frankignoulle et al. (1998)
Mandovi-Zuavi (India) *	400−2 500 #	Sarma et al. (2001)
Hudson (US-New York)	503−2 270 #	Raymond et al. (1997)
Changjiang (China-Shanghai/Jiangsu)	168−2 264 **	This study
Douro (Portugal)	385−2 200 **	Frankignoulle et al. (1998)
Rhine (Netherlands)	375−1 990 **	Frankignoulle et al. (1998)
York (US-Virginia) *	352−1 896 #	Raymond et al. (2000)
Tamar (UK)	380−2 200 #	Frankignoulle et al. (1998)
Rappanhannock (US-Virginia) *	474−1 613 #	Raymond et al. (2000)
Urdaibai (Spain)	256−1 569 #	Ortega et al. (2005)
James (US-Virginia) *	284−1 361 #	Raymond et al. (2000)
Elbe (Germany)	340−1 100 **	Frankignoulle et al. (1998)
Columbia (US-Oregon)	560−950 #	Park et al. (1969)
Potomac (US-Maryland) *	646−878 #	Raymond et al. (2000)
Asón (Spain)	246−436 #	Ortega et al. (2005)

Note: The pCO_2 range was obtained by taking the lowest and highest values for each estuary reported in the reference(s) except for those marked with an asterisk, which was obtained by averaging the lowest and highest values for each transect. The estuaries are ranked by the high limit. ** denotes that the data were gained from both inner and outer estuary; # denotes that the data were gained only from the inner estuary.

informative to the metabolism and the links between the aquatic system and its watershed (Raymond and Cole, 2001).

The sea air flux calculations provided an overview of CO_2 sea-air exchange in the studied area (Fig.4). The results indicated that F values varied between −10.0 and 88.1 mmol m^{-2} d^{-1} with an average of 24.4±16.5 (mean±standard deviation) mmol m^{-2} d^{-1}.

The present study did not take the diel fluctuation into account in calculating CO_2 flux as all measurements were made daytime. Higher flux values could be gained if diel variation was considered, particularly in summer, because nighttime CO_2 levels are generally higher owing to the absence of primary production. However, limited studies in diel cases in the Hudson and York Rivers indicated that the range of pCO_2 over a summer diel cycle was considerably smaller than the observed range in seasonal and spatial measurements (Raymond et al., 1997; Raymond et al., 2000).

As the sea-air exchange of CO_2 is a dynamic process in estuarine regions, to estimate the ability of a region to absorb atmospheric CO_2 accurately, sufficient spatial and temporal data about the difference of CO_2 partial pressure between water

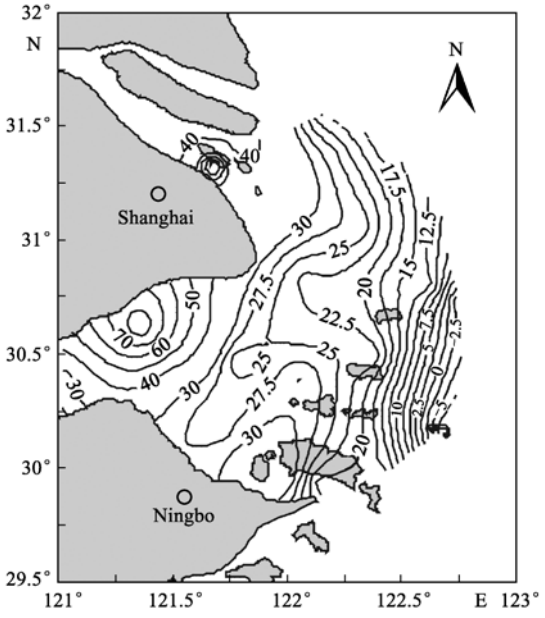

Fig.4 Distribution of F isolines (mmol m^{-2} d^{-1}) **in surface waters**

and air as well as synchronizingly-obtained hydrological, meteorologic, chemical and biological data are necessary (Hu and Yang, 2001).

In view of the great geographical heterogeneity of pCO_2 distribution in continental shelf areas, to estimate the CO_2 flux between water and air interface of a whole area based on limited observations, deviation will come into being inevitably. The deviation would be even more if the role of global marginal shelf seas on sources and sinks of atmospheric CO_2 were evaluated based on these data (Hu and Yang, 2001; Cai and Dai, 2004; Zhai et al., 2005).

pCO_2 in ECS surface seawaters has been extensively studied. Although significant CO_2 sinks were observed certainly in the ECS shelf using sea-air pCO_2 difference and it is generally considered that the ECS is an annual net carbon sink, controversies still exist about whether on ECS is a source or sink of carbon in a given season and the ability of the ECS to absorb atmospheric CO_2 (Tsunogai et al., 1997; Zhang et al., 1997; Peng et al., 1999; Tsunogai et al., 1999; Wang et al., 2000; Hu and Yang, 2001; Song, 2004). Except for the fact that they used different calculation methods, which can cause difference to some extents, the difference in the studied time and area is another reason leading to the controversies, owing to the fact that spatio-temporal diversity is an important characteristic for pCO_2 distribution in surface seawaters.

The data of this study show that the sea-air CO_2 flux was quite high in the Changjiang Estuary, Hangzhou Bay and their adjacent areas. Although the studied area was estimated to be only 2×10^4 km^2, it could emit $(5.9\pm4.0)\times10^3$ tons of carbon to the air every day. Therefore, these areas must be taken into account for understanding the ability of ECS in absorption of atmospheric CO_2. However, none of sea-air CO_2 flux researches performed in the ECS have covered the whole Changjiang Estuary and nearby oligohaline and mesohaline areas.

4 CONCLUSION

For the period and area sampled, the surface water pCO_2 was higher than atmospheric pCO_2 with an only exception of a patch area east of Zhoushan Archipelago, and varied within the range of 168 to 2264 μatm. The pCO_2 range fell in the low range of other estuaries in the world. The calculated sea-air CO_2 fluxes decreased offshore and varied from -10.0 to 88.1 mmol m^{-2} d^{-1} with an average of 24.4 ± 16.5 (mean ± standard deviation) mmol m^{-2} d^{-1}. During the sampling period, the studied area as a whole acted as a CO_2 source and emitted estimatedly $(5.9\pm4.0)\times10^3$ tons of carbon to the air every day.

If fully quantitative studies of the Changjiang Estuary are undertaken, this marked spatio-temporal variability must be considered in field programs. Marked changes in estuarine master variables can lead to significant changes in the speciation, reactivities and transport pathways of other chemical constituents. As the Changjiang River is the largest source of freshwater into the East China Sea, accurate river-sea flux estimates should be made based on detail understanding of these factors.

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