Chinese Journal of Oceanology and Limnology
Vol. 23 No. 1, P. 29-38, 2005

Carbon dioxide partial pressure and carbon fluxes of air-water interface in Taihu Lake, China*

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Received Jan. 22, 2004; revision accepted Nov. 16, 2004

Abstract To obtain carbon dioxide (CO_2) flux between water-air interface of Taihu lake, monthly water samplers at 14 sites and the local meteorological data of the lake were collected and analyzed in 1998. Carbon dioxide partial pressures (pCO_2) at air-water interface in the lake were calculated using alkalinity, pH, ionic strength, active coefficient, and water temperature. The carbon fluxes at different sublakes and areas were estimated by concentration gradient between water and air in consideration of Schmidt numbers of 600 and daily mean windspeed at 10 m above water surface. The results indicated that the mean values of pCO_2 in Wuli Lake, Meiliang Bay, hydrophyte area, west littoral zone, riverine mouths, and the open lake areas were 1 807.8±1 071.4 (mean±standard deviation) μatm (1atm=1.013 25×10^5Pa), 416.3±217.0 μatm, 576.5±758.8 μatm, 304.2±243.5 μatm, 1 933.6±1 144.7 μatm, and 448.5±202.6 μatm, respectively. Maximum and minimum pCO_2 values were found in the hypertrophic (4 053.7 μatm) and the eutrophic (3.2 μatm) areas. The riverine mouth areas have the maximum fluxes (82.0±62.8 mmol/m^{2}a). But there was no significant difference between eutrophic and mesotrophic areas in pCO_2 and the flux of CO_2. The hydrophyte area, however, has the minimum (−0.58±12.9 mmol/m^{2}a). In respect to CO_2 equilibrium, input of the rivers will obviously influence inorganic carbon distribution in the riverine estuary. For example, the annual mean CO_2 flux in Zhihugang River estuary was 19 times of that in Meiliang Bay, although the former is only a part of the latter. The sites in the body of the lake show a clear seasonal cycle with pCO_2 higher than atmospheric equilibrium in winter, and much lower than atmospheric in summer due to CO_2 consumption by photosynthesis. The CO_2 amount of the net annual evasion that enters the atmosphere is 28.42×10^4 t/a, of which those from the west littoral zone and the open lake account for 53.8% and 36.7%, respectively.

Key words: carbon dioxide partial pressure, air-water interface, carbon flux, trophic area, river input, Taihu Lake

1 INTRODUCTION

Carbon dioxide (CO_2) is principal greenhouse gas. Its air-water exchange is important in terrestrial ecosystems for climate change (Frankignoulle et al., 1998; Schimel et al., 2001). The direction of CO_2 gas movement depends on the CO_2 concentration gradient between air and surface water. The amount of CO_2 exchange is related to the gas exchange coefficient, *k*. All lakes, with their small area but large atmospheric CO_2 flux are important to understand the CO_2 fluxes in continent (Meybeck, 1993). Fluxes, sources, and mechanisms of CO_2 in lakes have been previously studied and compared (Richey et al., 1978; Kling et al., 1992; Cole et al., 1994; Hope et al., 1996; Kelly et al., 2001). In those studies, each lake was usually considered as a homogenous water body regardless of its size. However in fact, some large lakes, especially those affected largely by human being activities have different trophic levels and pollution loading (Wang et al., 1998). Their trophic status may fluctuate in

* This research was supported by the Knowledge Innovation Project of Chinese Academy of Sciences (KZCX1-SW-01-15) and (KZCX1-SW-12)
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response to the input of carbon and nutrients from watershed due to fertilization, industrialization or urbanization, which result in different phytoplankton productivity and carbonate equilibrium systems (Richey et al., 1978; Wetzel, 2001). Generally, low CO_2 concentrations are observed during spring to autumn for high algal activity occurrence (Neal et al., 1998).

Taihu Lake (30°56'–31°34'N, 119°54'-120°36'E) is the third largest freshwater lake in China, situated on the ancient Changjiang (Yangtze) River delta, with the catchment of 36 500 km^2. This area is one of the most developed regions in China. It is situated in subtropic zone and a part of the Southeast Asian monsoon region with annual precipitation of 1 200–1 400 mm (Sun and Huang, 1993). Taihu Lake is a large, shallow and unstratified lake with area of 2 338 km^2, mean depth of about 2 m, and water residence time of approximately 300 days. The north part of the lake is the main water supply for Wuxi City with population about 1.1 million.

The whole Taihu Lake can be divided into five areas or types according to their geographic nature and environmental features (Fig.1): Wuli Lake, 5.3 km^2, is a closed and hypereutrophic lake in the north of Taihu Lake. It locates near the cityside of Wuxi and is surrounded by many scenic spots. Meiliang Bay is main drinking water source for Wuxi City but it is eutrophic and often suffers from algal bloom in summers. It has an area of 120 km^2, half of its bottom is covered by soft ooze. The mainly waters including most of the open areas and the areas around the West Dongting Island is the open lake with an area of 1 216.8 km^2. Its bottom is hard earth. West littoral zone, about 5 km wide and 336.7 km^2 in area, is from the northwest of the lake to the south, whose area is mainly discharged waters. Hydrophyte zone, about 659.3 km^2, including the bays (Gonghu

Fig.1 Distributions of different lake areas and sampling sites in Taihu Lake
I : Wuli Lake; II : Meiliang Bay; III: Open Lake; IV : West littoral zone; V : Hydrophyte zone

Lake and East Taihu Bay) in east of the lake and the part to the south of the West Dongting Island. 10% to 95% of its area is covered with aquatic vegetation, the dominant species are *Potamogeton malaianus* Miq, *Potamogeton maackianus* Benn. Vegitation flourishes from early April to late October every year. Except for Wuli Lake and Meiliang Bay, the other waters are mesotrophic (Huang et al., 2001). All areas have an average depth of 2 m except for the open lake (2.5m) and the East Taihu Lake (1.3m).

This paper is based on the data collected in waters in different trophic levels in Taihu Lake. This work aimed to quantify the annual spatial and temporal changes of pCO_2 and CO_2 fluxes in the areas of the lake in different tropic levels to determine the response of dissolved inorganic carbon to the CO_2 flux changes and annual CO_2 exchange amount at air-water interface in the whole lake.

2 MATERIALS AND METHODS

2.1 Sampling

Fourteen sampling sites were established in Taihu Lake (Fig.1). Sites 0 and 9 are in Wuli Lake. Sites 1–6 are in Meiliang Bay. Site 0 and Site 6 are in the estuaries of Liangxi River and Zhihugang River, respectively. Site 10 and 11 are distributed in the west littoral zone; Site 12 and 13 are located in East Taihu Bay and Gonghu Bay, respectively. The other two sites, 7 and 8, are in the open lake.

Water samples were collected monthly from mid January to mid December 1998 between 9:00–16:00 at 0.5m depth below the surface with a bilge sampler. Water temperature, pH, and electrical conductivity were measured *in situ*. Each water sample is filtrated through a glass filter (Whatman GF/C) when arriving at the laboratory and kept refrigerated in the dark. Chlorophyll a concentrations are determined using ethanol and corrected for phaeopigments using the technique developed by Marker (1994).

2.2 Meteorological measurement

Wind speed (10 meter above the surface), atmosphere pressure, temperature, and photosynthetic active radiation (PAR) are automatically recorded every 30 minutes with a meteorological observatory and precipitation is measured with a rain gauge in the Taihu Laboratory for Lake Ecosystem Research (TLLER) (Fig.1).

2.3 Water chemistry analysis

CO_2 partial pressure (pCO_2) is calculated for each sampling date from pH and alkalinity (Alk), with appropriate corrections for temperature, ionic strength, following Wetzel and Likens (1991) and Cole *et al.* (1994). Alk is measured by Gran titration in an open vessel (precision ±0.5%). pH is measured with an acid meter using a gel-filled combination, temperature-compensating electrode. Calibration is done using two buffers (pH 6.86 and 9.18). Ionic strength is calculated from K^+, Na^+, Ca^{2+}, $Mg2^+$, NH_4^+, Cl^-, F^-, SO_4^{2-}, NO_3^-, HCO_3^-, CO_3^{2-} and SiO_3^{2-} according to Butler (1991). NH_4^+ and NO_3^- are measured with a double tunnel Autolyzer (Skalar-SA1000); dissolved inorganic carbon (DIC) is calculated with $[CO_2]$, $[HCO_3^-]$, and $[CO_3^{2-}]$. In order to consider potential influence of biodynamics, we did hourly diel variation on pCO_2 in the Meiliang Bay (48 hours), the open lake (24 hours) and the hydrophyte zone (24 hours) in 21–27 April, 2003. The relative standard of pCO_2 deviations are 14.49, 19.24, and 10.84 μatm (1 atm=1.01325×10^5 Pa), respectively. So we can take the day samplers as diel ones. $[HCO_3^-]$ and $[CO_3^{2-}]$ are calculated from pH, Alk and the corrected carbonate first and second association constants (k_1 and k_2). All other ions are analyzed with ionic color spectrum meter (DX-100). Activity coefficients for carbonate were calculated used Butler method (1991).

3 CALCULATIONS

Gas exchange rate between surface waters and atmosphere depends on two main factors: concentration gradient between water and air, and gas exchange coefficient k at a given condition (Wanninkhof, 1992). k and the concentration gradient govern the flux (in mmol/m^{2}d) of CO_2 across the air-water interface can be parameterized:

$$\mathrm{Flux}=\mu k([CO_2]_w-[CO_2]_a) \qquad (1)$$

where k (m/s) is the gas exchange coefficient for CO_2 at a given temperature and ionic strength. The term $[CO_2]_w-[CO_2]_a$ is the concentration difference (mmol/m^3) between the water and the air. The $[CO_2]_w$ equals to pCO_2K_H. K_H is Henry's constant. There have been many experimental determinations of k in a variety of freshwater and marine systems. For the calculation of gas fluxes, we assumed a constant k_{600}, computed the Schmidt number for CO_2 at each temperature, and calculated k for CO_2 at each temperature (k is piston velocity for a given gas at a given temperature; k_{600} is the k for a gas with a Schmidt number of 600, e. g., CO_2 at 20 ℃). From k_{600} and temperature one can calculate k for any other Schmidt number and we made this temperature correction (MacIntyre et al., 1995).

The exchange coefficient, k, can be defined as a function of wind speed and temperature with a number of different parameterizations (Liss and Merlivat, 1986; Tans et al., 1990; Wanninkhof, 1992; Cole and Caraco, 1998). Two calculation methods were used, one is recommended by Cole and Caraco (1998) at daily-mean windspeed less than 3m/s, and another is recommended by Wanninkhof (1992) at daily-mean windspeed equal to or more than 3m/s:

$$k_{CO_2}=2.07+0.215\,U_{10}^{1.7} \qquad U_{10}<3\text{m/s};$$

rough wind regime (2)

$$k_{CO_2}=0.31\,U_{10}(k_{600}/600)^{-0.5} \qquad U_{10}\geq 3\text{m/s};$$

rough wind regime (3)

Where U_{10} is the wind speed at 10 m above water surface (m/s). Wind speed data collected every 10 min were averaged to give a mean daily value for k which, combined with individual monthly pCO_2 data in the water and air, and gave a monthly estimate of the flux. Atmospheric partial pressure of CO_2 was 365 μatm in 1998 according to Houghton and Ding (2001). The daily average wind speed at TLLER was used to estimate the velocity dependent gas exchange coefficient.

4 RESULTS

4.1 Principle for grouping sites

To summarize the results, data from the 14 sites in Taihu Lake has been put into 5 groups reflecting existing knowledge of ecological characteristics of the lake. Each group shares three "water quality regions" adopted by Vant et al. (1998). However, overall pattern of our data differs from that in earlier work. Our groupings are: Liangxi River (Site 0) reflecting the dominance of riverine and drainage inputs the Liangxi River to and from the Grand Canal. Meiliang Bay (Sites 1 to 5) is predominantly open water sites. River mouths and Wuli Lake (Sites 6, 9, 10 and 11) are those where riverine flows are the dominant influences. Open water (Sites 7 and 8) is well separated from the influences of inputs and probably reflects the lake as a whole. The submerged macrophyte sites (Sites 12 and 13) at Gonghu Bay and East Taihu Bay form the fifth group.

4.2 Meteorology

Except for rainfall, in 1998, light radiation, water temperature, and winds were relatively normal (Fig.2). The change of surface water temperature was similar to that of photosynthesis active radiation (PAR). The highest temperature and maximum radiation occurred during about mid July and mid August, respectively. Lake surface temperature was 3.6℃ (19 January) to 35.1℃ (12 August). The total mean temperature and standard deviation was 19.2±8.8℃, suitable for phytoplankton growth.

The wind patten was very different in different periods. The daily mean wind speed (DMWS) was 3.7 m/s. While the maximum DMWS was 8.7 m/s. During the summer and autumn, the dominant wind was southeast to southwest and daily mean wind speed was often less than 4 m/s. During the winter and spring, the dominant wind was north-westerly reaching instantaneous velocities higher than 13 m/s (while the mean wind speed was about 4 to 4.5 m/s). When the wind speed exceeded 3.6 m/s or so, the surface layer water would be broken (Liss and Merlivat, 1986). As Taihu Lake is a very shallow and large area spindrift could happen if wind speed is greater than 4.5 m/s.

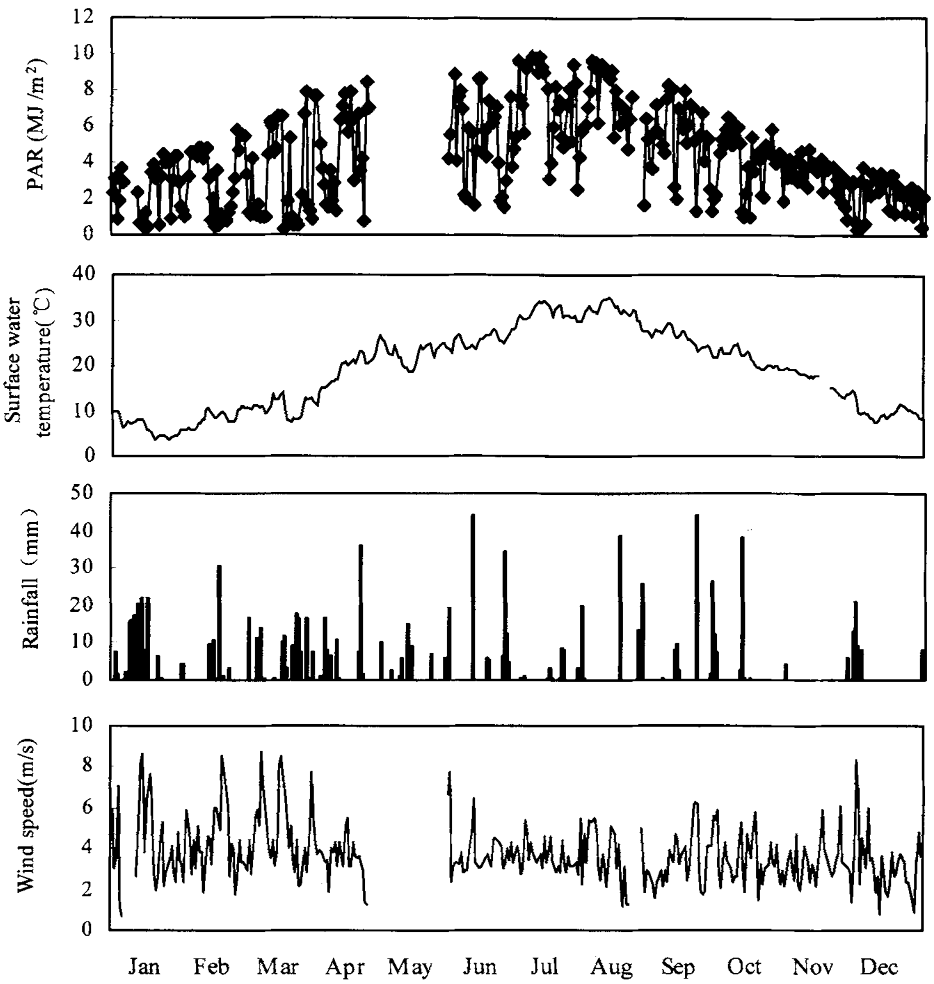

Fig.2 Daily light radiation, water temperature, rainfall and winds (measured at TLLER in 1998)

4.3 Spatial and temporal variations of pCO_2

All sites (Fig.3) show a clear seasonal variation in pCO_2. There are differences in size and time of maximum pCO_2 between the groups. For instance Sites 1–10 (excluding Site 6) show a very similar seasonal pattern. pCO_2 at these sites is the highest in late winter and early spring. In early summer, pCO_2 rapidly declines well below the atmospheric equilibrium value (as 365 μatm in 1998 according to Houghton and Ding, 2001). The similar seasonal pattern is confirmed also by covariance analysis between time series of pCO_2 at various sites (Table 1). Strong temporal correlation is observed between Sites 1 to 8 except for Site 6. Strong similarities in the pCO_2 are also shown at Sites 11, 12 and 13. This reflects the proximity of these sites to each other within main lake body and easy water exchange between them. On the other hand, they are far from river inflows and are less impacted by runoffs and groundwater discharges that sometimes occur.

Reflecting the influence of terrestrial processes, the pCO_2 at Sites 6, 9, 10 and to some extent 11 also (Fig.3) reaches the annual maximum in early spring. This coincides with the start of spring rainfall (Fig. 2) and the water from the nearby rural and urban areas. However, in Site 0, its maximum pCO_2 occurred in late fall, being 4053 μatm and poorly comparable with all other sites. This high value is 11.3 times of normal atmospheric CO_2 pressure. This abnormal high pCO_2 corresponded to the bulk labile organic intake from surrounding areas by summer rains in 1998. The organic matter bearing water passed Site 0 and caused high pCO_2 in the way of CO_2 metabolization. Higher alkalinity of 2.29±0.58 mmol/L and lower pH (7.90±0.23) are favorable for CO_2 dissolution in water through carbonate/bicarbonate equilibrium system.

Table 1 Covariance analysis between the time series of pCO_2 at the various sites

	0	1	2	3	4	5	6	7	8	9	10	11	12	13
0	1	0.103	-0.061	0.212	-0.023	-0.033	-0.100	-0.140	-0.176	0.113	0.212	0.071	-0.224	0.169
1		1	0.697	0.671	0.768	0.773	-0.219	0.650	0.599	0.562	0.639	0.656	0.449	0.523
2			1	0.551	0.694	0.705	-0.356	0.491	0.520	0.332	0.413	0.556	0.674	0.706
3				1	0.764	0.752	-0.074	0.476	0.402	0.590	0.656	0.692	0.509	0.524
4					1	0.905	-0.024	0.722	0.677	0.641	0.692	0.723	0.505	0.466
5						1	-0.097	0.135	0.674	0.577	0.644	0.693	0.537	0.544
6							1	0.166	-0.120	0.570	0.491	0.355	-0.141	-0.408
7								1	0.817	0.643	0.696	0.717	0.481	0.456
8									1	0.340	0.434	0.546	0.608	0.706
9										1	0.738	0.695	0.294	0.100
10											1	0.711	0.325	0.203
11												1	0.522	0.357
12													1	0.597
13														1

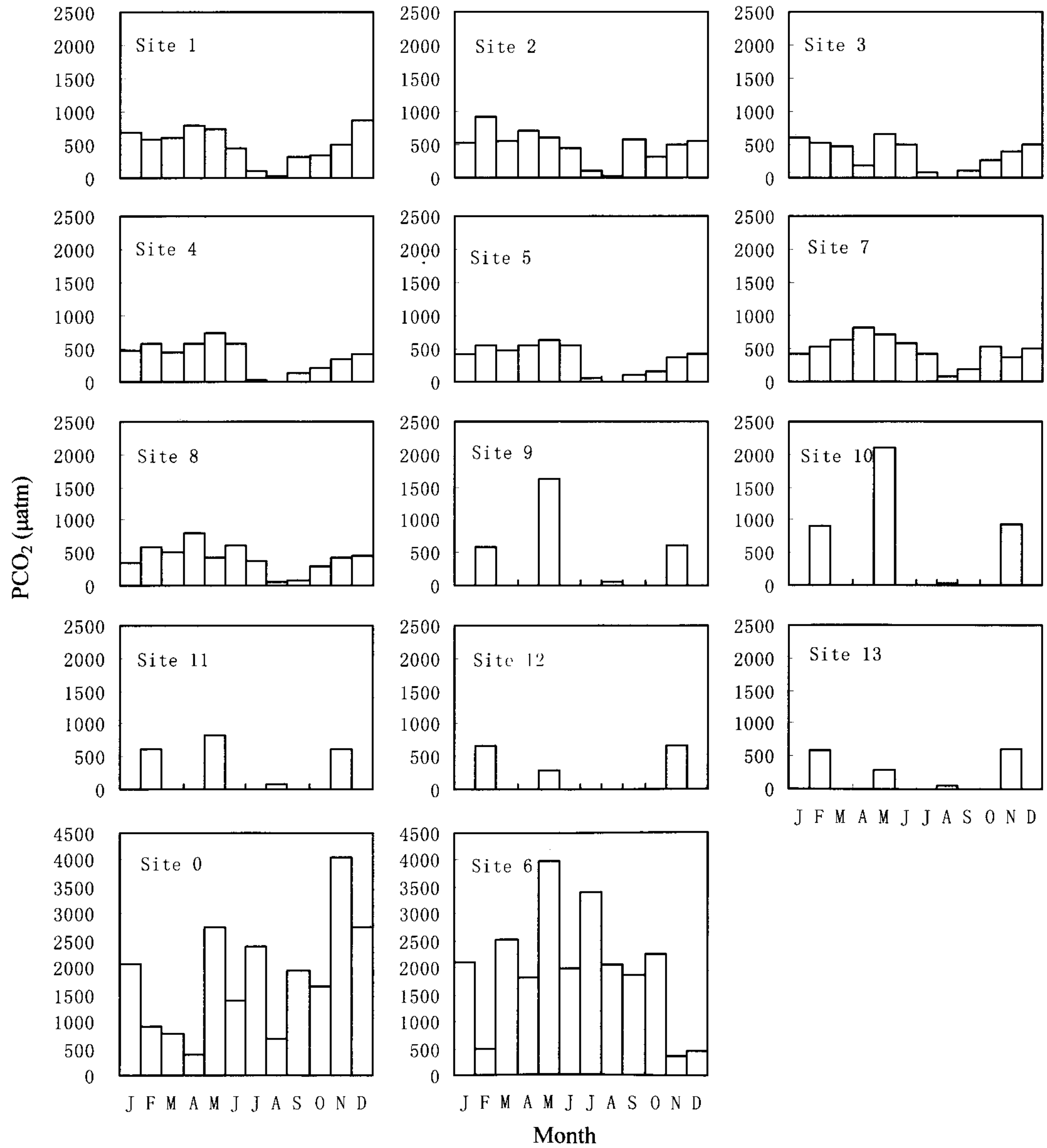

Fig.3 Monthly variation of CO_2 partial pressures at different sites (only four samples in the Sites 9–13)

Site 6 showed similar pattern with those having similar water environment such as Sites 9–10 which are directly affected by a river upstream. In early winter, Site 6 is shielded from WNW and ENE winds which affect the other sites. Hence the primary production is less limited by light and pCO_2 is reduced. Note especially (Table 1), the negative covariance between the most proximal sites (Sites 0 and 8) indicating poor correlation.

4.4 CO_2 flux

Bates and Merlivat (2001) revealed that use of high frequency wind speed data to estimate air-water CO_2 flux seems to yield more accurate results than those obtained by using high frequency $pCO_{2(water)}-pCO_{2(air)}$ data. Low frequency wind speed data are usually underestimated by about 20% to >600%. We used daily mean wind speed data to reduce this error. Atmospheric partial pressure of CO_2 was 365 μatm in 1998 (Houghton and Ding, 2001). The air-water fluxes of CO_2 in the five water bodies were calculated based on Eq.(1) and corrected based on Eq.(2) using daily wind speed (Fig.2). More than one order of magnitude difference was found in the CO_2 flux between the high area (Site 0 and 6, called as riverine estuary) and the low area (west littoral zone, hydrophyte zone, and the open lake). The maximum and minimum CO_2 flux was 82.0±62.8 mmol/(m^2a) (riverine estuary) and −0.58±12.9 mmol/(m^2a) (hydrophyte area). The annual mean CO_2 flux in the estuary was 17.8 times of that in Meiliang Bay (4.6±10.8 mmol/m^2a) although Site 6 and Site 1–5 are in the same bay. During the summer, a big CO_2 sink existed in Meiliang Bay and the hydrophyte area. The former was −4.71 to −10.62 mmol/(m^2d) in July through October and the latter was −4.02 and −13.49 mmol/(m^2d) in May through November, respectively.

5 DISCUSSIONS

5.1 CO_2 partial pressure in eutrophic area

The CO_2 partial pressure and chlorophyll a in the areas of Taihu Lake varies greatly. In eutrophic Meiliang Bay, during July to September, algal bloom developed with strong decrease of surface water pCO_2, most of which were under 360 μatm (Fig.3), indicating undersaturation of carbon dioxide. Algal bloom with the maximum chlorophyll a of 166.3 μg/L corresponded to pCO_2 low as 3.2μatm. Algae require an abundant and readily available source of carbon for its sustainable growth. Carbon supply is regulated by its availability. Abundant physiological evidence indicated that free CO_2 is most readily utilized by nearly all the algae. When free CO_2 supply is very low and HCO_3^- is abundant, a few species of algae have to take HCO_3^- (Robert et al., 1990). In Meiliang Bay, blooming algal growth required a rapid supply as the uptake rate is too fast for the carbonate system equilibrium to provide enough available carbon. In this situation, some algae would have to utilize carbon from the abundant HCO_3^- available. An obvious decrease of HCO_3^- was observed in Meiliang Bay around August, as a result of the increase of the chlorophyll a concentrations.

5.2 Difference of CO_2 flux among different lake areas

Monthly average fluxes of CO_2 were calculated based on monthly average pCO_2 and daily average wind speeds for the different site groups. Results are shown in Table 2. The riverine estuary areas have

Table 2 Annual average pCO_2 and net annual CO_2 fluxes in different areas of Taihu Lake

Sub-region	Average annual pCO_2 (μatm)*	CO_2 flux mmol/(m^2a) **	Area (km^2)	CO_2 exchange amount (t/a)
Wuli Lake	1 807.8±1 071.4	71.9±49.2	5.3	0.55×10^4
Meiliang Bay	416.3±2 17.0	4.6±10.8	117.0	0.86×10^4
West littoral zone	576.5±758.8	29.3±22.7	324.7	15.28×10^4
Hydrophytes	304.2±243.5	-0.58±12.9	659.3	-0.61×10^4
Open lake	448.5±202.6	5.3±10.2	1 216.8	10.36×10^4
Riverine mouths	1 933.6±1 144.7	82.0±62.8	15.0	1.98×10^4
Whole lake	–	–	2 338.1	28.42×10^4

* 1 atm=1.01325×10^5 Pa; ** positive indicates flux into the atmosphere.

the maximum fluxes (82.0±62.8 mmol/m^{2}a) which greatly exceed those of non-estuary areas. The hydrophyte area, however, has the minimum (−0.58±12.9 mmol/m^{2}a). But there was no significant difference between eutrophic (or hypertrophic) and mesotrophic areas in the flux of CO_2. For instance, annual CO_2 flux of the open lake is almost equal to that of Meiliang Bay.

The CO_2 amount of net annual evasion that enters the atmosphere from Taihu Lake is 28.42×10^4, of which the west littoral zone and the open lake accounts for about 53.8 percent and 36.7 percent. There is a remarkable seasonal variation in the magnitude of the monthly CO_2 fluxes in the Meiliang Bay and open water sites, evading in the winter, and invading in the summer. In contrast, the monthly fluxes in the more human being impacted sites such as Wuli Lake, and the river mouths (especially Sites 0 and 6) are almost invariable and generally higher (up to one order of magnitude) than the open water sites, but show little seasonality. In hydrophyte area, the annual mean CO_2 flux was negative, indicating invasion. Since the pH was usually high in eutrophic and hydrophyte areas in summer. CO_2 invasion might be controlled by so-called chemically enhanced diffusion (Wanninkhof and Knox, 1996). Determining the overall annual CO_2 emission from Taihu Lake is difficult because the fluxes at each site need to be evaluated first, which is sometimes arbitrary.

5.3 Input of inorganic carbon source *vs.* CO_2 flux

There are seven inflowing rivers locating at the west littoral zone and the Meiliang Bay. River waters usually which high inorganic or organic carbon content were discharged from the lands. Inorganic carbon, largely as dissolved carbon dioxide and bicarbonate, is the primary source of carbon for photosynthesis and of organic substances. These organic compounds are generated by cynobacteria, algae, and macrophytes in the lakes or rivers. During 1987–1988 and 1994–1995, 62 254 t/a and 81 700 t/a of total organic carbon entered the lake from these rivers, respectively (Sun and Huang, 1993; Huang et al., 2001). Zhihugang River and Liangxi River are the major rivers into the north of Taihu Lake. Zhihugang River alone delivered about 52% of the total polluted water into Meiliang Bay (Yuan et al., 1993). The organic carbon loadings of Liangxi River and Zhihugang River are accounted for 24%–29% of that of the whole lake. Although organic carbon is unusually resistant to biological mineralization (Tranvik, 1988), it can be largely decreased through various processes including settling of suspended organic matter, photodegradation, diffusion and so on. Although, a sharp downtrend of organic carbon content can be always observed from the river estuary to the main lake body in Taihu Lake, Liangxi River and Zhihugang River, as main contributors of carbon, add water with high inorganic and organic contents to the littoral zone. Part of the imported organic carbon is mineralized. Thus DIC and pCO_2 were increased. On the other hand, high concentrations of nutrients in river water stimulate algal blooms and photosynthetic uptake of CO_2 (Richey et al., 1978).

The air-water flux of CO_2 is insufficient to achieve equilibrium of CO_2 in the littoral zones. In shallow Taihu Lake, the effects of mineralization in biological production, water mixing in river estuaries, calcification and dissolution of calcareous material, and wind-induced perturbation on pCO_2 in lake surface water are obviously significant.

6 CONCLUSIONS

Great changes in CO_2 partial pressure are between different trophic or polluted areas in Taihu Lake. From May to December, the pCO_2 values in hypertrophic area could be twice as much as those of eutrophic and mesotrophic waters. The sites with indicate high pCO_2 principally situated in the river estuaries. The carbonate system of water chemistry and air-water exchange of CO_2 in the river estuary, located in the eutrophic area, show a similar behavior to those of the hypertrophic area. Undersaturated CO_2 values were due to CO_2 consumption by algal blooms in the eutrophic areas. In addition, there is about one order of magnitude difference in CO_2 fluxes from lows in Meiliang Bay (4.6±10.8 mmol/m^{2}a) and the open lake (5.3±10.2 mmol/m^{2}a) to Wuli Lake (71.9±49.2 mmol/m^{2}a). These results support the concept that a large lake may be considered as special uneven body, which can contain different carbonate equilibrium system. In Taihu Lake, CO_2 is from the surface to the atmosphere during most of the time. It participates in external drainage via rivers, and is a local major stable carbon sink, whose contribution to pCO_2 enhancement is very important in the carbonate compositions distribution in lake body. Future study of the seasonal inorganic and organic carbon outflow throughout the seasons from rivers, sediments, and aquatic organisms is recommended to quantify the carbon fluxes from different lake areas.

7 ACKNOWLEDGEMENTS

The authors thank all participants in the Taihu Laboratory for Lake Ecosystem Research (TLLER) for their support. We also thank Dr. Lu Zhang of Land and Water, CSIRO, Australia for valuable technical assistance.

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