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Eutrophic Lake Taihu as a significant CO_2 source during 2000–2015

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A B S T R A C T

Inland lakes receive growing attentions on eutrophication and their roles in global carbon cycle. However, understanding how inland lakes contribute to global carbon cycle is seriously hampered due to a shortage of long-term records. This study investigated the carbon dioxide (CO_2) flux from the Lake Taihu, a large (2400 km^2) and shallow (mean depth 1.9 m) eutrophic lake in subtropical region, based on a long-term (2000–2015) measurement of the partial pressure of carbon dioxide (pCO_2) at high spatiotemporal resolution. We found that the Lake Taihu was a significant source of atmospheric CO_2 with an average CO_2 emission flux at 18.2 ± 8.4 mmol m^{-2} d^{-1} (mean±1standard deviation) and a mean annual pCO_2 value of 778 ± 169 μatm. The highest pCO_2 and CO_2 flux were observed in eutrophic zone with a high external input of carbon and nutrient, and the lowest in non-eutrophic zones with no direct external input of nutrient and carbon. A substantial seasonal pattern in pCO_2 was observed, particularly in eutrophic pelagic area, and was significantly negatively correlated with chlorophyll *a*. Long-term measurement showed the interannual variation in annual lake CO_2 dynamics, which was highly sensitive to human-induced nutrient input. Watershed input of carbon and nutrient leads to the high CO_2 level, counterbalancing the in-lake primary production. All lines of evidence suggest that human activities may have predominate contribution to CO_2 source in the Lake Taihu, and this mechanism might be widespread in global freshwater lakes.

1. Introduction

Lakes are generally supersaturated with carbon dioxide (CO_2) due to internal metabolic activities and external loading, and thus play a disproportionately considerable role in the atmospheric CO_2 budget (Cole et al., 1994; Raymond et al., 2013; Tranvik et al., 2009). It is estimated that lakes occupy approximately 3.7% of global land area (Verpoorter et al., 2014) but emit 0.11–0.57 Pg C yr^{-1} as CO_2, which serve as a significant source of atmospheric CO_2 (Borges et al., 2015; Cole et al., 2007; Holgerson and Raymond, 2016; Tranvik et al., 2009). Meanwhile, some studies suggested that lake CO_2 production and emission will increase substantially due to warming (Hasler et al., 2016; Kosten et al., 2010; Marotta et al., 2014). Despite of high uncertainties, it is well established that CO_2 emission from lakes is an important component in the global carbon cycle (Cole et al., 2007; Tranvik et al., 2009).

Lake CO_2 emission varies strongly across regions and along latitudinal gradients (Marotta et al., 2009; Raymond et al., 2013; Sobek et al., 2005). However, the previous field measurements for lake CO_2 emission estimates are not geographically evenly distributed (Cole et al., 2007; Raymond et al., 2013). Lakes in the north temperate, boreal, and tropical zones are generally well-investigated (Buffam et al., 2011; Hastie et al., 2018; Marotta et al., 2009), however, few field measurements of lake CO_2 have been reported for subtropical zone with only 3% of sampled lakes from this zone (Sobek et al., 2005). Eventually there are numerous

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lakes in subtropical regions, it is estimated that approximately one third of lakes are located in subtropical zone that accounts for approximately 30% of total surface area (Lehner and Doll, 2004; Verpoorter et al., 2014). Therefore, long-term monitoring of C cycling in subtropical lakes is critically important for accurately quantifying C budget in global lakes.

Eutrophication is a serious environmental problem for inland lakes (Anderson et al., 2014; Sinha et al., 2017), yet its impacts on lake C budget remains under-investigated. Studies found that high nutrient loadings in eutrophic lakes stimulated mineralization and increased CO_2 emission (Kortelainen et al., 2006; Perga et al., 2016), and the external carbon input further enhanced the CO_2 emission (McDonald et al., 2013; Wilkinson et al., 2016). On the contrary, a few studies found that high nutrient increased primary production in eutrophic lakes, causing a decrease in CO_2 emission (Balmer and Downing, 2011; Gu et al., 2011; Pacheco et al., 2014; Schindler et al., 1997). A recent study reported a neutral impact of eutrophication on lake CO_2 emission in boreal lakes (Klaus et al., 2018). Considering accelerating eutrophication under changing climate (Sinha et al., 2017), more field measurements are needed to better understand eutrophication impacts on lake C budget. Meanwhile, understanding and predicting lake C cycling under a changing environment had been seriously hampered due to the shortage of long-term records (Perga et al., 2016; Seekell and Gudasz, 2016).

Lake Taihu, a typical large (area 2400 km^2), shallow (mean depth 1.9 m) and eutrophic lake with frequent cyanobacterial blooms, is located in the Yangtze River Delta, China featuring Marine monsoon subtropical climate. The specific objectives of this study are: (1) to evaluate the variations in lake pCO_2 based on long-term, monthly, and multi-sites measurements, (2) to determine the eutrophication impacts on pCO_2 variability, and (3) to quantify the Lake Taihu's contribution to the watershed CO_2 budget. This study presents one of the longest field records of large lake pCO_2 at high spatiotemporal resolution. Considering the dramatic changes in physical, chemical, and biological environments of Lake Taihu (Zhang et al., 2018), this study provides a unique opportunity to examine CO_2 flux in an inland freshwater lake.

2. Materials and methods

2.1. Study site

Lake Taihu (30°56′-31°33′ N, 119°52′-120°36′ E) has a surface area of 2400 km^2, a mean depth of 1.9 m, and a catchment area of 36500 km^2. The lake is surrounded by several large cities, and its catchment is one of the most industrialized and densely populated regions in China. The lake has a complex river network with 172 rivers or channels connecting to the lake, showing distinct inflow and outflow regions. Inflow rivers are located in the northern and western sides of the lake, and outflow in the eastern side of the lake. The lake is located in a subtropical climate, which is characterized by high water temperatures in summer and low temperatures in winter. The annual precipitation is approximately 1100 mm (Lee et al., 2014). Frequent eutrophication events have made Lake Taihu a priority environmental topic and a popular research site in the world (Duan et al., 2009; Zhang et al., 2018).

We divided the lake into three zones based on the eutrophic status and vegetation distribution (Fig. 1). Zone 1, including the northwest zone and Meiliang Bay, is the most eutrophic part of the lake due to pollutant discharge of inflowing rivers. Zone 3 is connected to the outflowing rivers of the lake and is inhabited by dense aquatic vegetation. Zone 2 is connected to a few inflowing rivers and represents a transitional region. This is evidenced from the mean total nitrogen concentrations of 3.25 mg L^{-1}, 2.14 mg L^{-1}, and 1.29 mg L^{-1} for zone 1, zone 2, and zone 3, respectively (Xiao et al., 2019), and in the average vegetation coverage of 3.8 kg m^{-2} in zone 3 and close to zero in zone 1 and 2 (Luo et al., 2016; Qin et al., 2007).

There are 29 sampling sites that are evenly distributed across the lake (Fig. 1). There are 9 sampling sites in zone 1, 12 in zone 2, and 8 in zone 3. It is worth noting that there are some sampling sites in zone 1 and zone 2 nearby inflow rivers, we further divided the two zones into two section, the river mouth and the pelagic area, in below analysis. Briefly, there are three sites in river mouth for zone 1 (the site of 6#, 10#, and 16#) and zone 2 (the site of 11#, 13#, and 14#, Fig. 1), respectively.

2.2. Data acquisitions

Long-term limnological observations of Lake Taihu, including the measurements of physical, chemical, and biological parameters, have been conducted by the Taihu Laboratory for Lake Ecosystem Research (TLLER; Zhang et al., 2018; Qin et al., 2019). The field measurements of the lake present some of the earliest limnological multi-sites observations in China. TLLER led the field sampling of the total 29 sampling sites across the lake, and field measurements covered the entire lake (zone 1, zone 2, and zone 3) since 2000. Thus, this study focused on the data collected during 2000–2015. Meanwhile, monthly measurements were conducted in zones 1 and 2, while seasonal measurements were conducted in zone 3 in February, May, August, and November. Each field survey across whole lake was generally completed between 9:00 and 17:00 in two consecutive days.

Variables considered in this study include water temperature (T_w), pH, alkalinity (Alk), dissolved oxygen (DO), total nitrogen (TN), ammonium nitrogen (NH_4^+-N), nitrate nitrogen (NO_3^--N), total phosphorus (TP), dissolved organic carbon (DOC), dissolved inorganic carbon (DIC), chlorophyll *a* (Chl-a), and water clarity. pH was measured with a composite electrode in the field, which was calibrated by standard buffer solution prior to measurement, T_w and DO were measured with a multi-parameter probe, Alk was measured via titration with hydrochloric acid standard solution, nutrient (TN, NH_4^+−N, NO_3^-−N, and TP) were measured with a spectrophotometer, DOC/DIC were measured with a TOC analyzer, Chl-a was determined spectrophotometrically, and water clarity was given by Secchi disk depth.

The measurements of Alk were conducted immediately on the sampling date. The surface-water samples for the analysis of nutrient, DO/DIC, Chl-a, and Alk were preserved in ice-chilled coolers while in the field. Then these samples were transported to laboratory for immediate filtration and measurements. The long-term dynamic and trend of the physical, chemical, and biological parameters from TLLER have been reported in previous studies (Duan et al., 2009; Xu et al., 2017a; Zhang et al., 2018).

2.3. Calculations of CO_2 partial pressure and flux

We calculated the partial pressure of CO_2 (pCO_2) based on pH, alkalinity, and *in situ* water temperature from the TLLER dataset. The details of the calculation equations are given in the supporting information (Text S1). Meanwhile, direct measurements of pCO_2 via headspace equilibration method were conducted in the lake since 2011, and the indirectly calculated pCO_2 was highly correlated with directly measured pCO_2 (Fig. S1: $R^2 = 0.76$, $p < 0.01$). Diel sampling for direct pCO_2 measurements was also conducted at eutrophic zone of Lake Taihu (the MLW site, Fig. S2) to investigate diel cycle in the CO_2 dynamics. The water samples were collected every 3 h for three consecutive days in each month from August 2012 to July 2013 (Fig. S2).

The CO_2 flux (F_c, mmol m^{-2} d^{-2}; a positive value indicates CO_2 emission from the water to atmosphere) across the water-air

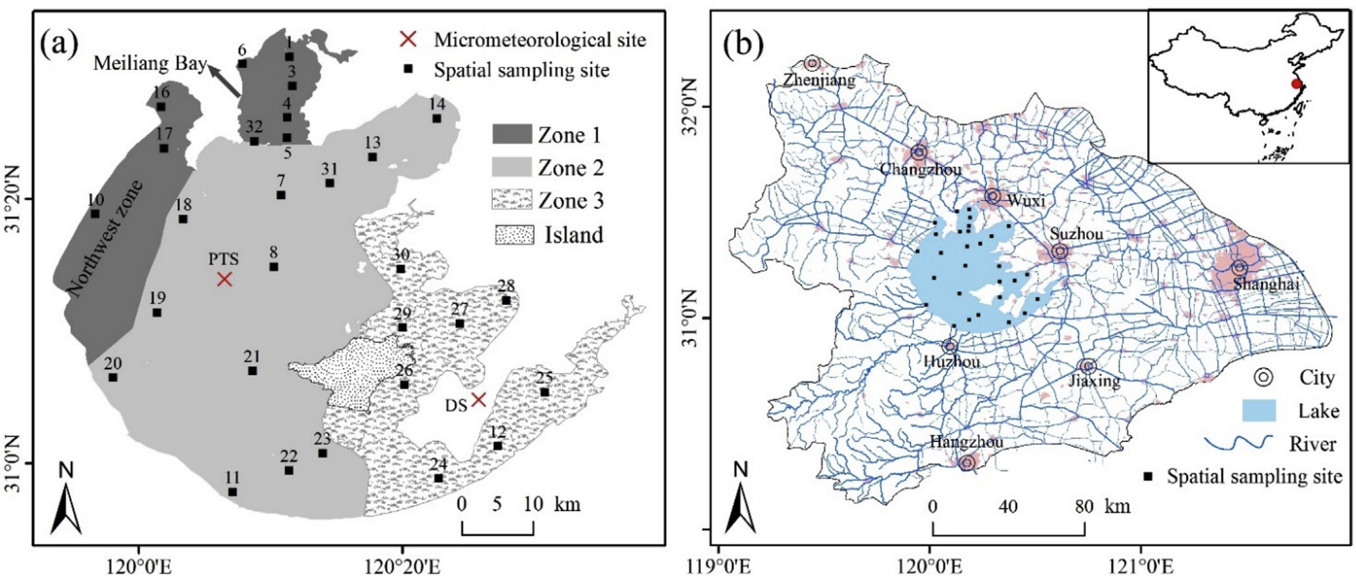

Fig. 1. Map showing the geographic location of study sites; (a) Map showing the three biological zones and the location of lake-survey sites at Lake Taihu. Zone 1: eutrophic zone, including the Northwest Zone and Meiliang Bay; Zone 2: transition zone; Zone 3: aquatic vegetation zone. Note that the lake was divided into three zones according to the geographic difference of nutrient concentrations, water quality indices, and vegetation biomass (Luo et al., 2016; Xiao et al., 2019). Black squares, spatial sampling sites, including the river mouth sites (6#, 10#, and 16# in zone 1; 11#, 14#, and 20# in zone 2) and open water sites; red crosses, micrometeorological site, including lake PTS (Pingtaishan) site and land DS (Dongshan) site. (b) Lake Taihu watershed and surrounding rivers and large cities. Water flows from the north and west to the east. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

interface was estimated based on the bulk diffusion model (Cole and Caraco, 1998):

$$F_c = k \times K_H \times (pCO_2 - p_a) \quad (1)$$

where K_H is the Henry's constant (mol L^{-1} atm^{-1}) adjusted for water temperature (Text S1), p_a is the partial pressure of CO_2 in atmosphere (μatm), local p_a was measured by CO_2 gas analyzer (Model G1301, Picarro Inc., CA, USA) in the study of Xiao et al., (2014). The gas transfer coefficient k (m d^{-1}) is dependent on wind speed based on the study of Cole and Caraco (1998), which was presented in details in the supporting information (Text S2).

For k calculation, we utilized the long-term wind speed data (2000–2015) from Dongshan (DS) station of the China Meteorological Observation Network, the data was downloaded from the China Meteorological Data Sharing Service System (http://cdc.nmic.cn). The DS station is located at the peak of Dongshan Mountain and is surrounded by Lake Taihu on three sides (Fig. 1). The wind speed at DS was lower than that at open area of Lake Taihu due to sheltering effect of the local terrain (Wang et al., 2014). Therefore, we corrected the long-term wind speed data based on the synchronous wind speed measurement at PTS, a micrometeorological site located in the center of Lake Taihu (Fig. 1; Lee et al., 2014), given the measured wind speed at the two sites were highly correlated (Fig. S3).

There are several formulations considering both wind speed and water vertical mixing velocity for the k calculation (e.g. MacIntyre et al., 2010; Read et al., 2012). We calculated the CO_2 gas transfer coefficient k using the models given by Read et al., (2012) and Podgrajsek et al., (2015) in the supporting information (Text S2). There were insignificant ($p > 0.05$) differences between k values calculated by the equation described by Cole and Caraco (1998) and those calculated by equations considering both wind shear and water mixing regime in Lake Taihu (Fig. S4). This is consistent with the study of Read et al., (2012) showing that the k value in the large lake was driven primarily by wind speed.

2.4. Statistic analysis

Simple linear regression was carried out to derive correlations between the environmental variables and pCO_2. Prior to linear regression analyses, these variables were log-transformed to ensure normality. The CO_2 flux over the lake was calculated based on the long-term pCO_2. Statistical analysis showed log-transformation CO_2 flux over the lake were normally distributed. A monthly zonal mean was calculated using all measurements within the corresponding zone from 2000 to 2015. The whole-lake pCO_2 and corresponding CO_2 flux were computed as an area-weighted zonal average. Monthly zonal mean variables, including pCO_2, CO_2 flux, and environmental variables, were summarized as means for spring (from March to May), summer (from June to August), autumn (from September to November), winter (December to February in next year), and annual mean from 2000 to 2015. The zone 1 and zone 2 were further separated into river mouth and pelagic area to distinguish the watershed impacts, respectively. A least significant difference post-hoc test was used to determine the differences among measured variables using SPSS (version 18.0), and differences at the $p < 0.05$ level were deemed statistically significant.

3. Results

3.1. Environmental variabilities

Lake Taihu showed dramatic spatial variability but reasonable temporal variability in chemical and biological properties based on multi-year's measurements (Figs. 2–3b). The highest concentrations of nutrient, DOC, DIC, and Chl-a with annual mean values of 4.04 mg L^{-1} (TN), 6.27 mg L^{-1}, 19.50 mg L^{-1}, and 30.42 μg L^{-1} were observed in zone 1, significantly ($p < 0.01$) higher than those in zones 2 and 3. On the contrary, the lowest DO with an annual mean value of 8.60 mg L^{-1} was observed in zone 1. For zone 1, the concentrations of nutrient, DOC, and DIC in river mouth with annual

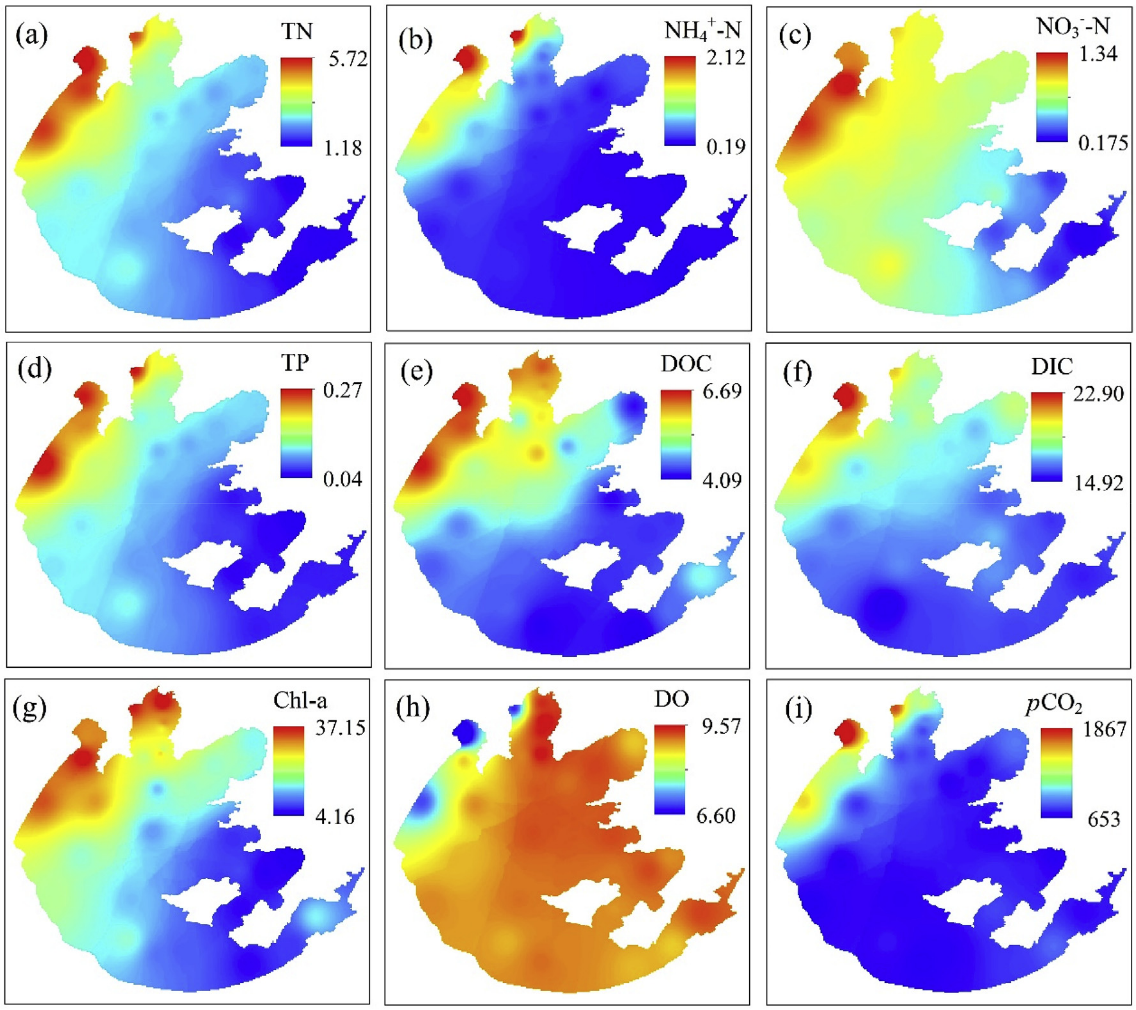

Fig. 2. Mean spatial variation of nutrient loading, water quality indices, dissolved carbon fraction across Lake Taihu during sampling period: (a) total nitrogen (TN, mg L^{-1}), (b) NH_4^+−N (mg L^{-1}), (c) NO_3^-−N (mg L^{-1}), (d) total phosphorus (TP, mg L^{-1}), (e) dissolved organic carbon (DOC, mg L^{-1}), (f) dissolved inorganic carbon (DIC, mg L^{-1}), (g) Chlorophyll *a* (Chl-a, μg L^{-1}), (h) dissolved oxygen (DO, mg L^{-1}), and (i) carbon dioxide partial pressure (pCO$_2$, μatm).

mean values of 5.35 mg L^{-1} (TN), 6.81 mg L^{-1}, and 21.09 mg L^{-1} were significantly ($p < 0.01$) higher than those in pelagic area due to the directly discharge via inflowing rivers (Table 1; Fig. S5). For zone 2, these variables showed nonsignificant ($p > 0.05$) difference between river mouth and pelagic area due to the less-polluted inflow rivers (Fig. S5).

In contrast to chemical and biological properties, water temperature and wind speed were remarkably uniform across the lake (Fig. S6). The spatial variability of water temperature is less than 1 °C during measurement period, and the annual mean water temperature was 17.9 ± 9.5 °C. The wind speed variation was <0.1 m s^{-1} across the entire lake with an annual mean value of 4.5 m s^{-1}. The temperature has increased, and wind speed has decreased significantly in recent decades (Fig. 3a).

3.2. Spatiotemporal variabilities of pCO$_2$

3.2.1. Inter-annual variability

The lake exhibited remarkable spatial and inter-annual variations in pCO$_2$ (Figs. 3c and 4). Spatially, the pCO$_2$ in zone 1 with an annual mean value of 1000 ± 260 μatm were significantly ($p < 0.01$) higher than those in the zone 2 (663 ± 148 μatm) and zone 3 (707 ± 176 μatm), and the highest pCO$_2$ occurred in river mouth of zone 1. Temporally, the inter-annual variability in pCO$_2$ was substantial and showed the similar trend for all three zones (Fig. 3c). However, the amplitude (maximum minus minimum) of pCO$_2$ in zone 1 (824 μatm) was larger than those in zone 2 (587 μatm) and zone 3 (646 μatm). For the entire lake, the highest pCO$_2$ appeared in 2010 with an annual mean value of 1053 ± 467 μatm and lowest in 2001 with an annual mean value of 455 ± 216 μatm.

The temporal variations in pCO$_2$ fall into three stages for the three zones: (1) from 2000 to 2004, the pCO$_2$ increased significantly; (2) from 2004 to 2010, the pCO$_2$ were relatively stable and then decreased since 2007, especially in zone 1; and (3) after 2010, the pCO$_2$ exhibited a decreasing trend. Multiple-year measurements showed the most dramatic inter-annual variabilities in pCO$_2$ were observed in river mouths (Fig. 4).

3.2.2. Seasonal and monthly variability

The pCO$_2$ showed substantial seasonal variations (Fig. 5 and Table S1). The pCO$_2$ was significantly different between summer and winter ($p = 0.01$), but not significantly different among spring, autumn, and winter ($p > 0.05$). The lake-wide mean pCO$_2$ was the lowest in summer with a value of 634 ± 224 μatm, and the highest in winter with a mean value of 874 ± 230 μatm. The spatial variations in pCO$_2$ were relatively weak in summer except some isolated

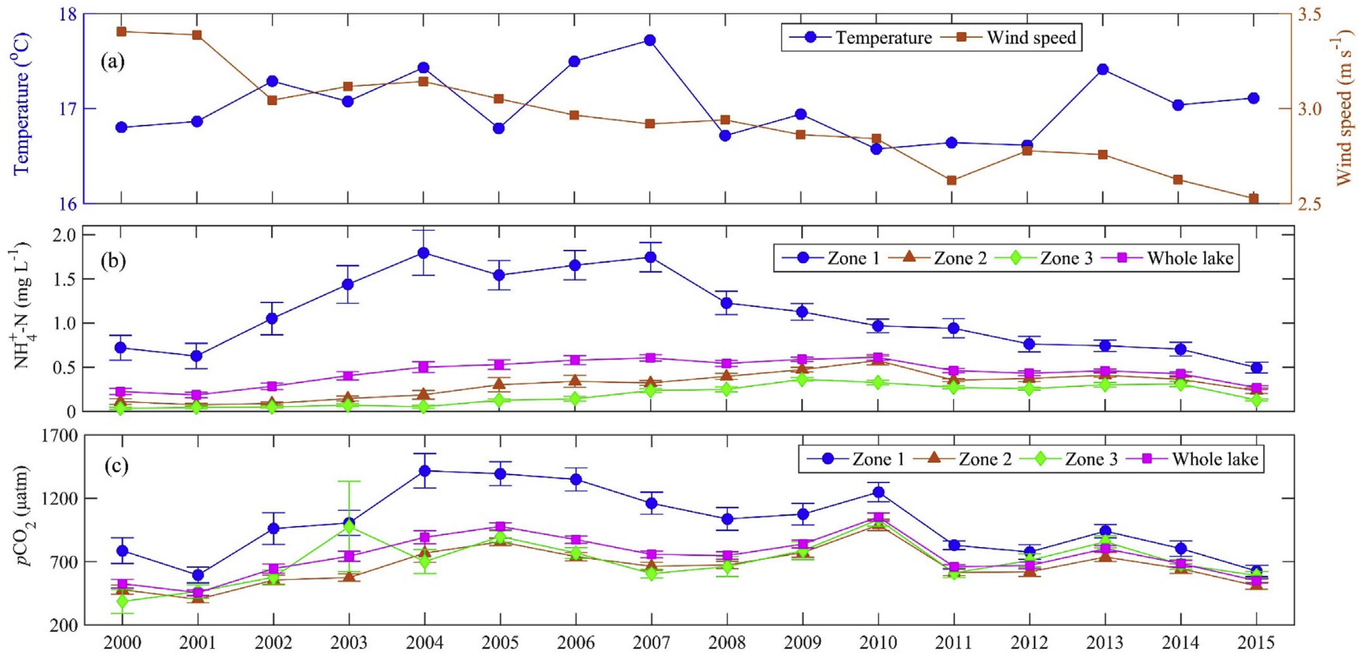

Fig. 3. Annul trend in meteorological factor (air temperature and wind speed, a), NH_4^+−N (b), and pCO_2 (c) from 2000 to 2015 for zone 1, zone 2, zone 3, and whole lake. Error bars indicated standard error. The meteorological factors were measured at DS station (Fig. 1).

Table 1
Key aquatic environment variables at different zones of the Lake Taihu during lake-survey period[a].

Zone		Statistics	pH	Alk mmol L^{-1}	TN mg L^{-1}	TP mg L^{-1}	DOC mg L^{-1}	DIC mg L^{-1}	Chl-a μg L^{-1}
Zone 1	River mouth	Range	6.90–9.49	0.91–3.38	0.12–21.93	0.04–3.60	0.01–18.5	3.87–34.64	1.41–497.18
		Mean ± SD	8.01 ± 0.37	2.10 ± 0.42	5.41 ± 2.60	0.26 ± 0.24	6.63 ± 2.93	21.09 ± 5.65	35.48 ± 59.40
	Open water	Range	6.90–10.10	0.69–3.06	0.05–18.77	0.02–2.70	0.81–23.58	5.62–29.5	0.56–491.15
		Mean ± SD	8.30 ± 0.37	1.86 ± 0.35	3.42 ± 1.90	0.14 ± 0.14	5.92 ± 2.58	18.70 ± 4.53	30.03 ± 40.75
Zone 2	River mouth	Range	7.63–9.07	0.78–2.24	0.03–6.07	0.02–0.63	0.62–13.32	5.21–28.50	0.98–189.72
		Mean ± SD	8.20 ± 0.26	1.69 ± 0.31	2.43 ± 1.09	0.10 ± 0.05	4.32 ± 1.98	17.33 ± 4.40	15.40 ± 17.19
	Open water	Range	7.40–9.06	0.09–2.78	0.03–10.19	0.01–1.09	0.35–33.59	4.57–30.03	0.55–462.30
		Mean ± SD	8.23 ± 0.24	1.65 ± 0.28	2.42 ± 1.25	0.10 ± 0.08	4.95 ± 2.44	17.10 ± 1.95	14.86 ± 26.20
Zone 3		Range	7.49–9.52	0.72–2.15	0.02–4.21	0.01–0.53	0.90–10.54	3.58–30.18	0.76–58.03
		Mean ± SD	8.16 ± 0.24	159 ± 0.27	1.45 ± 0.73	0.05 ± 0.04	4.41 ± 1.62	16.14 ± 4.52	7.50 ± 6.45

[a] Alk, Alkalinity; TN, total nitrogen concentration; TP, total phosphorus concentration; DOC, dissolved organic carbon; DIC, dissolved inorganic carbon; Chl-a, chlorophyll *a* concentration.

spots in river mouth showing pulse pCO_2 (Fig. 5).

The seasonal variation of pCO_2 was more remarkable in pelagic area than in river mouth in both zones 1 and 2 (Fig. 6). In zone 1, the mean pCO_2 in winter (964 ± 84 μatm) was 59% higher than the summer value (607 ± 71 μatm) for pelagic area, comparing with the 30% difference in river mouth area. In zone 2, the mean pCO_2 in winter (762 ± 71 μatm) was significantly ($p < 0.01$) higher than the summer value (575 ± 37 μatm) for pelagic area, while the differences between the two seasons (winter: 772 ± 60 μatm; summer: 665 ± 66 μatm) were nonsignificant ($p = 0.61$) for river mouth.

3.3. CO_2 flux

The lake CO_2 emission varied seasonally, annually, and spatially (Table S1 and Fig. 7). The annual mean CO_2 flux was 18.2 ± 8.4 mmol m^{-2} d^{-1}, adding up to an annual CO_2 emission of 0.7 ± 0.3 Tg yr^{-1}. Seasonally, peak CO_2 emission appeared in winter with a mean value of 24.4 ± 11.5 mmol m^{-2} d^{-1}, and summertime flux was the lowest with a mean value of 10.0 ± 11.8 mmol m^{-2} d^{-1}. Annually, the peak CO_2 flux (38.0 ± 16.7 mmol m^{-2} d^{-1}) appeared in 2005, which was approximately twenty times of the lowest flux in 2001. Spatially, zone 1 had significantly ($p < 0.01$) higher CO_2 flux (30.0 ± 14.5 mmol m^{-2} d^{-1}) than zone 2 (12.4 ± 6.7 mmol m^{-2} d^{-1}) and zone 3 (14.2 ± 8.8 mmol m^{-2} d^{-1}). The flux in river mouth (56.9 ± 29.1 mmol m^{-2} d^{-1}) was significantly ($p < 0.01$) higher than in pelagic area (19.0 ± 12.1 mmol m^{-2} d^{-1}) within the zone 1, however, there was no significant ($p = 0.73$) difference between river mouth and pelagic area in the zone 2.

3.4. Factors influencing pCO_2

Various factors affect the pCO_2 in the Lake Taihu. The Chl-a influenced the seasonal variation of the lake pCO_2 (Fig. 6). However, the zonal pCO_2 exhibited different responses to Chl-a. In zones 1 and 2, the lake monthly mean pCO_2 and were negatively correlated with Chl-a concentration for pelagic area, but the patterns were not profound in river mouth within these same zones (Fig. 8). In zone 3, pCO_2 showed low value in growing seasons (summer) and high value in winter overall, and the variation was not associated with Chl-a, but negatively associated with water clarity (Fig. S7), an index of aquatic vegetation biomass in Lake Taihu (Xiao et al., 2017).

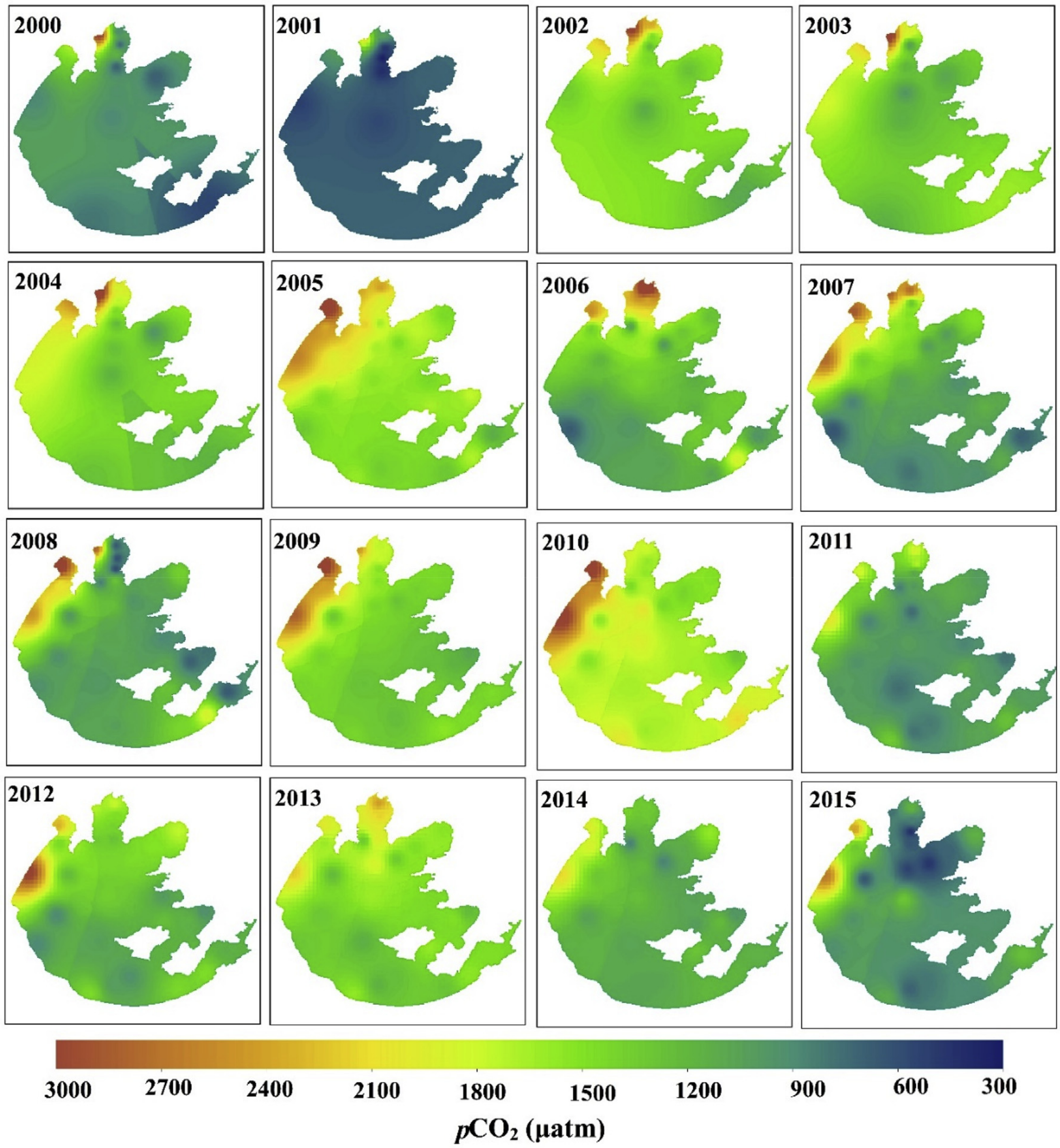

Fig. 4. Annual mean pCO_2 spatial distributions derived from whole lake survey data (2000–2015) for Lake Taihu.

The variability in pCO_2 was positively correlated with NH_4^+–N, NO_3^-–N,TN, TP, DIC, and DOC but negatively correlated with DO (Table 2 and Fig. S8). The annual mean pCO_2 was highly correlated with nutrient loadings, such as the TN in zone 1 ($r = 0.84$, $p < 0.01$, $n = 16$), zone 2 ($r = 0.80$, $p < 0.01$, $n = 16$), and zone 3 ($r = 0.58$, $p < 0.01$, $n = 16$) from 2000 to 2015. However, the inter-annual variability in pCO_2 was slightly affected by temperature and wind speed (Fig. 3).

4. Discussion

4.1. Whole lake CO_2 budget

The estimated CO_2 flux in the Lake Taihu ranged from −23.6 to 808.1 mmol m^{-2} d^{-1} with a mean value of 18.2 ± 8.4 mmol m^{-2} d^{-1} based on the gas transfer model. The mean CO_2 flux in the lake was slightly lower than those from lakes in the contiguous United States (McDonald et al., 2013), significantly lower than from boreal lakes with a mean value of 33.3 mmol m^{-2} d^{-1} (Hastie et al., 2018; Weyhenmeyer et al., 2015), and was higher than from temperature lake with mean value of 5.7–8.9 mmol m^{-2} d^{-1} (Buffam et al., 2011). For comparison, the mean CO_2 flux from global lakes was 20.9–28.6 mmol m^{-2} d^{-1} (Holgerson and Raymond, 2016; Tranvik et al., 2009). It should be noted that the mean pCO_2 and CO_2 emission flux in the Lake Taihu were generally higher than that in other eutrophic lakes (Table S2).

A large amount of CO_2 evasion into the atmosphere was confirmed in this study, indicating the Lake Taihu as a carbon source at an annual emission of 80 g C m^{-2} yr^{-1}. It is difference from a study on carbon burial that reported Lake Taihu as a carbon sink with carbon burial rate of 5 g C m^{-2} yr^{-1} (Dong et al., 2012). This discrepancy might due to the different approach in two studies. Extrapolating the estimated CO_2 flux of Lake Taihu in this study to others lakes in Yangtze River Delta given a large uncertainty

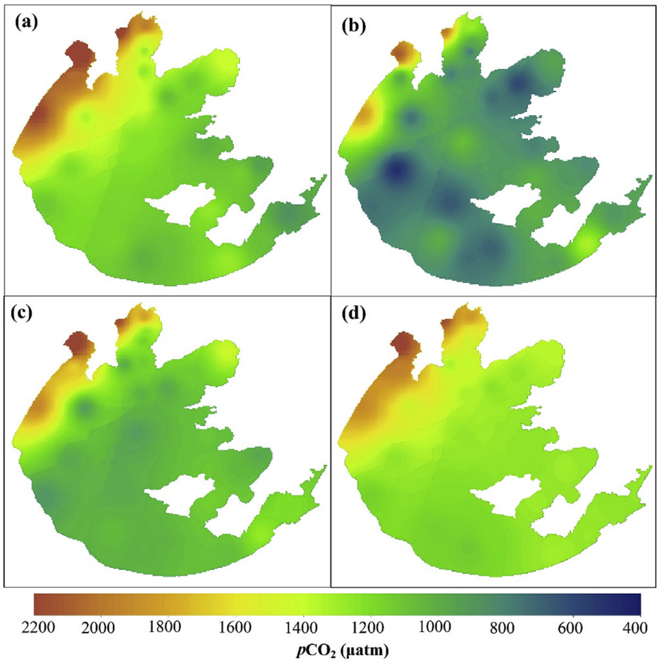

Fig. 5. Spatial variation of pCO_2 in (a) spring, (b) summer, (c) autumn, and (d) winter. Data shown as mean 2000–2015.

(48–309 g C m^{-2} yr^{-1}; Fig. 7b), these lakes with carbon burial rate of 5–373 g C m^{-2} yr^{-1} (Dong et al., 2012) were also likely to act as carbon sources. Since previous studies primarily focused on the eutrophication effects in the Yangtze River Delta, China (Duan et al., 2017; Paerl et al., 2011; Qin et al., 2010; Zhang et al., 2018), this study, to our best knowledge, is among the first attempts to report the eutrophication impacts on lake surface CO_2 dynamics at a high spatiotemporal resolution in this region. The results not only fill the gap of CO_2 flux in Lake Taihu, but also provide a valuable data sources for predicting how eutrophic lake CO_2 level evolve in a changing environment.

Previous study reported that lake CO_2 emission was highly associated with catchment productivity (Butman et al., 2016; Maberly et al., 2013). The net primary production (NPP) in Taihu basin with an area of 36500 km^2 is about 14.5 Tg C yr^{-1} (Xu et al., 2017b), the CO_2 emission from Lake Taihu alone accounted for 1.3% of the NPP based on our measurements. Extrapolating Lake Taihu measurements to the whole lake basin (~3160 km^2), we estimated that approximately 1.7% of NPP was lost to the atmosphere as CO_2, consisting with the study in English Lake District (Maberly et al., 2013). Taking the national total lake CO_2 emissions as 12.1 Tg C yr^{-1} (Li et al., 2018), the CO_2 emissions from Lake Taihu accounted for 1.6% of the national lake CO_2 budget across China.

Our data showed the lake pCO_2 varied seasonally and spatially, potentially resulting in the similar trend for CO_2 flux (Natchimuthu et al., 2017). It should be noted that a majority of lake CO_2 budget focus on warm season or use snapshot sampling regardless of

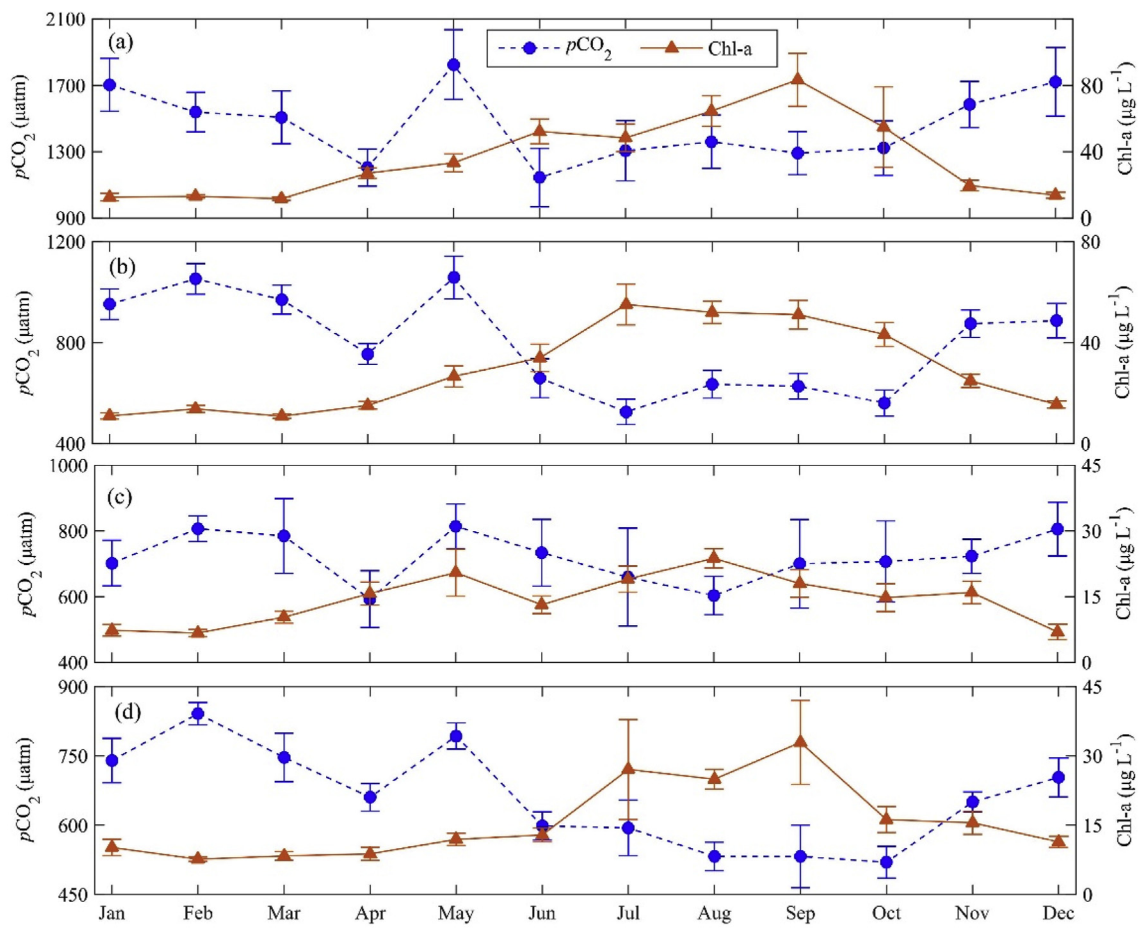

Fig. 6. Monthly mean pCO_2 and Chl-a concentrations in (a) river mouth of zone 1, (b) pelagic area of zone 1, (c) river mouth of zone 2, and (d) pelagic area of zone 2 from 2000 to 2015. Error bars indicate one standard error. The correlations between monthly mean pCO_2 and Chl-a for each zone were shown in Fig. 8.

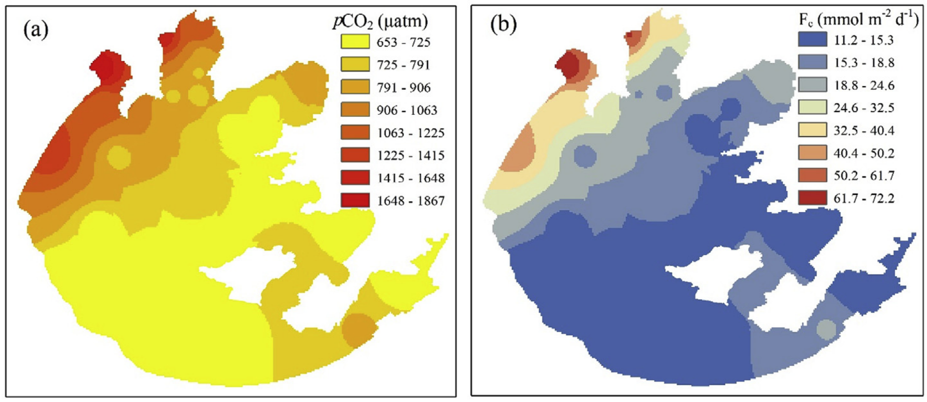

Fig. 7. Spatial variation of the mean pCO_2 and lake-air interface CO_2 exchange flux (F_c) during lake-survey period from 2000 to 2015.

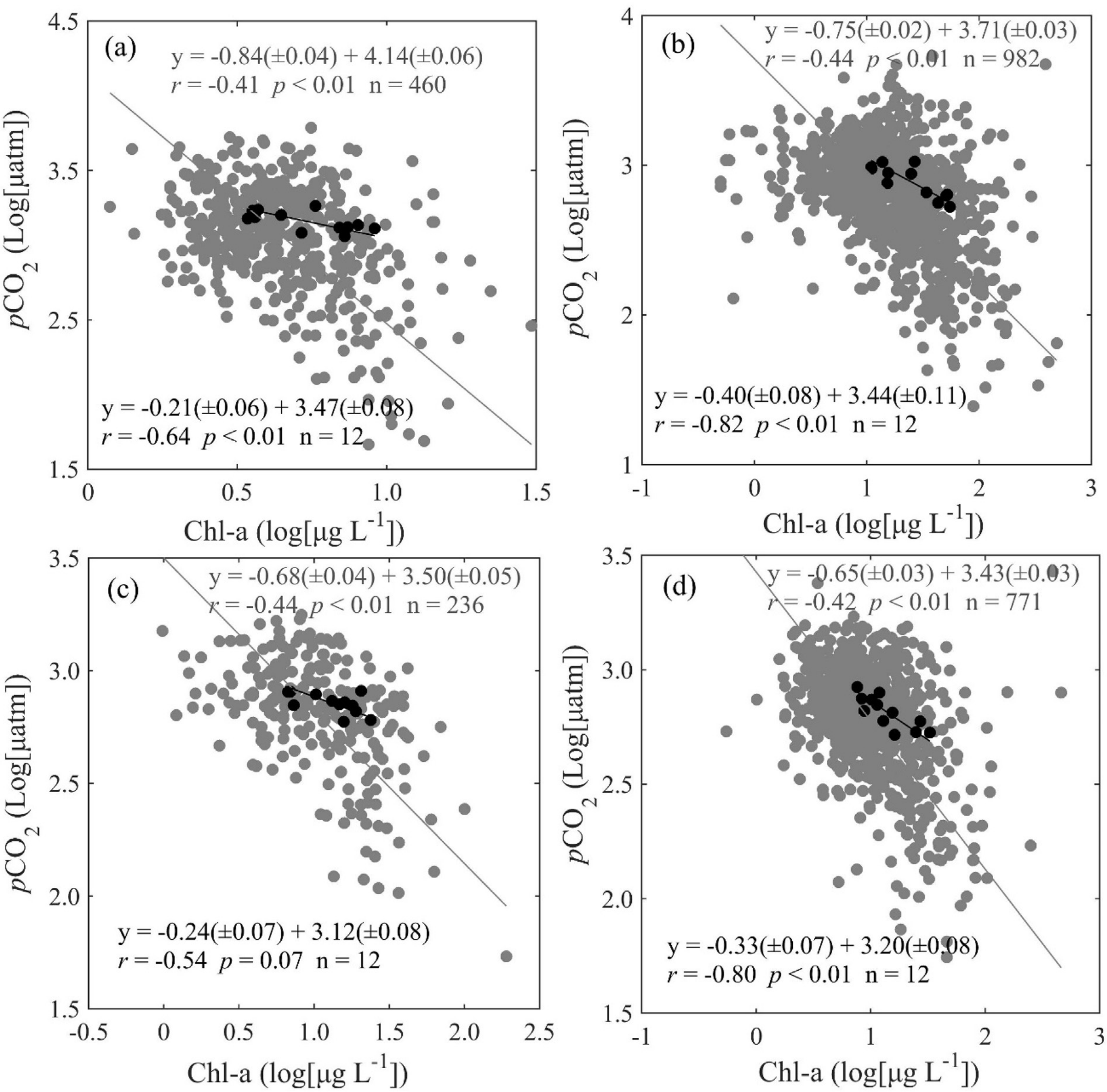

Fig. 8. Correlation of the normalized pCO_2 and normalized Chl-a concentration in river mouth in zone 1 (a), in open water in zone 1(b), in river mouth in zone 2 (c), and in open water in zone 2 (d). Black points represent the mean values for each month, and gray points represent all sampling data from 2000 to 2015. Parameter bounds on the regression coefficients are one standard deviation.

Table 2
Spatial correlation of the normalized pCO_2 against the major environmental variables[a].

	NH_4^+-N	NO_3^--N	TN	TP	DIC	DOC	DO
All data	0.44[b]	0.27[b]	0.39[b]	0.10[b]	0.21[b]	0.19[b]	−0.16[b]
Mean value	0.95[b]	0.44[c]	0.78[b]	0.82[b]	0.79[b]	0.78[b]	−0.92[b]

[a] All data indicated the all lake-survey data across space and time from 2000 to 2015. The total number are 3047 (NH_4^+-N), 3232 (NO_3^--N), 3055 (TN), 3056 (TP), 2280 (DIC), 2619 (DOC), and 3052 (DO). Mean value indicated the annual mean values for each spatial sampling site during lake-survey period, and the number is 29 for each variable.
[b] Correlation is significant at the 0.01 level.
[c] Correlation is significant at the 0.05 level.

temporal variability (Klaus et al., 2019; McDonald et al., 2013; Wen et al., 2017), which is likely to lead to biased estimates. Previous studies showed large variability in lake CO_2 flux across region (Li et al., 2018; Raymond et al., 2013; Sobek et al., 2005), our results suggest CO_2 flux varied dramatically within the single lake along the eutrophication gradients (Fig. 4). Annual mean CO_2 flux based on single sampling sites ranged from 62% to 397% of the whole-lake mean flux, suggesting the high underestimation or overestimation of lake CO_2 flux using single-site measurement (Natchimuthu et al., 2017; Seekell et al., 2014).

4.2. Causes of the lake CO_2 supersaturation

Eutrophic lakes were generally undersaturated for CO_2 and tend to be sink of atmospheric CO_2 due to the high primary production (Balmer and Downing, 2011; Gu et al., 2011; Schindler et al., 1997). However, the eutrophic Lake Taihu are presently supersaturated with CO_2 (778 ± 169 μatm) and acts as a source of atmospheric CO_2 (Fig. 7). This might be caused by the high external carbon and nutrient inputs.

Watershed-level carbon inputs may lead to the CO_2 supersaturation considering its complicated river network and high productivity (Perga et al., 2016; Rudorff et al., 2011). High DOC and DIC concentrations in inflow rivers were associated with the spatial distribution of the lake carbon fractionation (Fig. S5). The external DIC input may directly increase the CO_2 level, leading to the lake as a significant atmospheric CO_2 source (Kiuru et al., 2018; McDonald et al., 2013; Weyhenmeyer et al., 2015; Wilkinson et al., 2016). Additionally, the high river discharge of DOC can also raise the lake CO_2 level via supplying C substrate (Algesten et al., 2003; Larsen et al., 2011; Sobek et al., 2005). The pCO_2 in river mouth was about 2 times higher than that in pelagic areas in zone 1 with high external DIC and DOC input (Table S2), likely caused by the heavy external carbon input.

Human-driven nutrient inputs seem to elevate the lake pCO_2 level in this study. The lake pCO_2 value increased with nutrient (NH_4^+-N, NO_3^-−N, TN, and TP; Table 2 and Fig. S8), which was consistent with previous studies (Kortelainen et al., 2006; Li et al., 2012; Natchimuthu et al., 2017; Wang et al., 2017; Wen et al., 2017). Although nutrient loadings in lake can either increase CO_2 via enhancing respiration or decrease CO_2 via promoting primary production, the ultimate impact depending the balance of CO_2 production and consumption (Perga et al., 2016; Wang et al., 2017). The high human-driven loadings input, together with the high organic carbon, would stimulate microbial activities and enhance respiration, resulting in more CO_2 production in inland lakes. The inverse relationship between DO concentration and nutrient loadings (Fig. 2) and the significantly negative correlation between DO and pCO_2 (Fig. S8f and Table 2) also support this. The substantial nutrient loadings in the lake is mainly from the watershed input (Paerl et al., 2011; Qin et al., 2007), thus the high human-driven nutrient inputs may potentially stimulate respiration and then increase lake CO_2 level.

The substantial difference in pCO_2 value between zone 1 and the other two zones demonstrated the importance of external input in determining lake CO_2 level. The mean pCO_2 in zone 1 was 1000 ± 260 μatm, compared with 663 ± 148 μatm in zone 2 and 707 ± 176 μatm in zone 3 (Table S2). The physical factors, such as water temperature and wind speed, were remarkably uniform across different zones of the lake (Wang et al., 2014; Xiao et al., 2017; Fig. S6), however, zone 1 received a large amount of anthropogenic-derived nutrient and carbon via inflow rivers discharges, zone 2 and zone 3 were relatively clean due to no direct river pollutant inputs (Fig. 2; Fig. S5). These also suggested anthropogenic nutrient and carbon inputs increased CO_2 level and dominated spatial variation of CO_2 in the lake. The substantial portion of CO_2 derived from external input had been demonstrated by field measurements and modeling in others worldwide lakes (e.g. Kiuru et al., 2018; McDonald et al., 2013; Weyhenmeyer et al., 2015; Wilkinson et al., 2016), suggesting this mechanism might be widespread in global freshwater lakes.

4.3. Roles of human activities in pCO_2 variation

The weak influence of Chl-a on pCO_2 seasonal variation in river mouth may arise from the human-driven pollutant input. In regard to the seasonal variation in the lake pCO_2, our results showed significantly higher values occurred in the winter and lower values in the summer. Consistent with the field measurement in other lakes with algal blooms (Gu et al., 2011; Shao et al., 2015; Xing et al., 2005), the seasonal patterns were associated with the Chl-a concentration. However, the roles of Chl-a in determining pCO_2 seasonal variation varied among regions. The most apparent feature was the relatively weak correlation for the river mouths in zone 1 and zone 2 (Fig. 8). These river mouth regions hold the anthropogenic loadings, and the human-driven nutrient and carbon may have confounded the effect of Chl-a on the CO_2 dynamics.

Human activities may dominate the substantial inter-annual variability in the pCO_2. The lake pCO_2 inter-annual variability (Figs. 3 and 4) was partly explained by the reduction of external pollutant input from the watershed imposed by the local government (Paerl et al., 2011; Qin et al., 2007; Xu et al., 2017a). The "zero point action" aiming at industrial pollution control was implemented around 2000 and ultimately reduced the lake pollutant level (Paerl et al., 2011; Xu et al., 2017a), probably leading to the lowest pCO_2 in 2000−2001 (607 μatm in 2001; Table S1). The pCO_2 increased after 2001, this is partly attributed to pollutant discharge from watershed increased again after that (Xu et al., 2017a), evidencing from the increasing nutrient loading in the lake (Fig. 3b). A drinking water crisis in Lake Taihu took place in May 2007 due to a massive bloom of cyanobacteria (Qin et al., 2010), prompting a wide range of activities (e.g. closing a number of polluting factories) in an effort to reduce external pollutant loading. These measures may have led to a considerable decline in pollutant concentrations in recent years (Jeppesen et al., 2005; Xu et al., 2017a), and the associated decline in pCO_2.

Efforts aiming at eutrophication control may pose dramatic influences on lake CO_2 variability. Frequent eutrophication events in Lake Taihu had led to a series of environmental protection actions (Qin et al. 2010, 2019, Xu et al., 2017a; Paerl et al., 2011). Since 2007, these managements, including shutting off heavily polluted rivers, establishing wastewater treatment plants, closing small polluting factories, and resorting wetlands, have been widely implemented (Qin et al., 2019). These environmental investments may potentially decrease the external carbon input and the associated CO_2 level (Brigham et al., 2019; Fan et al., 2003). On the others hand, the nutrient loadings in the lake would be reduced due to these management practices (Jeppesen et al., 2005; Zhou et al., 2017), as shown in Fig. 4b, probably leading to the improving water quality and low CO_2 production rate (Brigham et al., 2019; Kortelainen et al., 2006; Wang et al., 2017).

The impacts of human activities on lake carbon biogeochemistry provide valuable information for management practices for the lake and upstream regions. The annual mean lake CO_2 dynamics were highly sensitive to human-driven pollutant (Fig. 3), suggesting the important role of watershed and lake management on lake CO_2 budget. Our findings suggested that measures should be taken to reduce watershed nutrient and carbon discharge into the lake to further decrease CO_2 emission (Jiang et al., 2018; Schrier-Uijl et al., 2011), and more studies should be carried out to investigate the underlying mechanisms to reach a harmonized relationship between lakes and human.

4.4. Uncertainties in pCO_2 and CO_2 estimations

The calculated pCO_2 may be overestimated due to the contribution of organic acids to alkalinity and the lower carbonate system buffering capacity at low pH (Abril et al., 2015). In this study, less that 1% of all the samples with pH below 7.4 and only two samples with pH below 7, together with the low DOC concentration (Table 1), suggested the overestimation could be neglected (Abril et al., 2015). Meanwhile, the calculated pCO_2 was highly correlated with directly measured pCO_2 (Fig. S1), implying a minor uncertainty in the calculated pCO_2.

The CO_2 flux in this study was based on the wind-dependent gas transfer coefficient k (Cole and Caraco, 1998). We showed that the k value does vary among methods, but the differences between them were nonsignificant (Fig. S4). The mean k value used for CO_2 flux estimation was 1.26 ± 0.42 m d^{-1}. We also compared part of the estimated CO_2 flux with those measured directly by eddy covariance method. There is a sampling site (5#) close to the micrometeorological site of Lake Taihu (Fig. S9a), in which the surface CO_2 flux was measured directly by eddy covariance method (Xiao et al., 2014). Part of the direct CO_2 flux measurement occurred under open-fetch and coincided approximatively with the field sampling at 5# site. The CO_2 flux estimated with the gas transfer model at 5# site was uncorrelated ($p = 0.21$) with the flux measured directly with the eddy covariance method at the micrometeorological site, but the CO_2 flux difference (Fig. S9b) between the two measurements was insignificant ($p = 0.93$).

The pCO_2 is a dominant variable influencing CO_2 flux (Natchimuthu et al., 2017). In this study, diurnal samplings showed the daytime pCO_2 was 2% lower than the whole-day value (Fig. S2), so the whole-lake pCO_2 and associated CO_2 flux may have been underestimated by a similar amount. The declining trend for pCO_2 from late morning (853 μatm at 9:00) to evening (709 μatm at 18:00) was found on annual basis (Fig. S2), which was consistent with the field measurement in other subtropical mesotrophic lake (Yang et al., 2019). Although each whole-lake survey was completed within the period, the annual mean pCO_2 across each spatial sampling site ranged from 653 μatm to 1867 μatm (Fig. 7a), suggesting the sampling time may have mirror influence on the spatial pattern (Fan et al., 2003).

5. Conclusions

Long-term measurement (2000–2015) confirmed the Lake Taihu as a significant source for atmospheric CO_2 with an annual mean CO_2 flux of 18.2 ± 8.4 mmol m^{-2} d^{-1} or 0.7 ± 0.3 Tg yr^{-1}, and the annual mean pCO_2 was 778 ± 169 μatm. The highest flux occurred in eutrophic zone with substantial external input of carbon and nutrient, and the lowest flux was in non-eutrophic zones without directly external input. The lake CO_2 dynamics also varied seasonally and annually, emphasizing the importance of yearlong and spatially distributed sampling to achieve unbiased lake CO_2 budget. High watershed input of carbon and nutrient is likely to lead to CO_2 emission rather than higher primary production in the lake, but the underlying mechanisms deserve further investigation.

Long-term field measurements of CO_2 dynamics in the highly heterogeneous Lake Taihu will shed light on the effect of eutrophication and watershed management on CO_2 emissions from freshwater lakes. This study presented one of the longest field records of large lake CO_2 at a high spatiotemporal resolution, which not only provides useful information for the regional budget of CO_2 from lakes, but also provides a valuable example to predict lake CO_2 fluxes under a changing environment.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.watres.2019.115331.

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