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Original Research

Re-estimating China's lake CO_2 flux considering spatiotemporal variability

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ABSTRACT

The spatiotemporal variability of lake partial carbon dioxide pressure (pCO_2) introduces uncertainty into CO_2 flux estimates at the lake water-air interface. Knowing the variation pattern of pCO_2 is important for obtaining accurate global estimation. Here we examine seasonal and trophic variations in lake pCO_2 based on 13 field campaigns conducted in Chinese lakes from 2017 to 2021. We found significant seasonal fluctuations in pCO_2, with decreasing values as trophic states intensify within the same region. Saline lakes exhibit lower pCO_2 levels than freshwater lakes. These pCO_2 dynamics result in variable areal CO_2 emissions, with lakes exhibiting different trophic states (oligotrophication > mesotrophication > eutrophication) and saline lakes differing from freshwater lakes (-23.1 ± 17.4 vs. 19.3 ± 18.3 mmol m^{-2} d^{-1}). These spatiotemporal pCO_2 variations complicate total CO_2 emission estimations. Using area proportions of lakes with varying trophic states and salinity in China, we estimate China's lake CO_2 flux at 8.07 Tg C yr^{-1}. In future studies, the importance of accounting for lake salinity, seasonal dynamics, and trophic states must be noticed to enhance the accuracy of large-scale carbon emission estimates from lake ecosystems in the context of climate change.

1. Introduction

Inland water carbon fluxes (1.40–3.88 Pg C yr^{-1}) are nearly equivalent to global ocean and land sinks, accounting for 13–37% of the global carbon dioxide (CO_2) emissions from the fossil fuel combustion and industrial processes, thereby wielding considerable influence in the global carbon cycle [1–5]. Although the global CO_2 emissions from lakes are lower than those from rivers, lakes, as the main constituent of inland waters, play an important role as the atmospheric CO_2 sources and sinks [3,6–8]. However, these high carbon fluxes from lakes seem unreasonably high, given they need to be sustained by organic carbon inputs from land. Independent estimates of carbon transfer from land to inland waters are insufficient to sustain such high fluxes. Currently, global estimations of CO_2 emissions from lakes are obtained by multiplying local or regional CO_2 emissions by the global lake area; thus, biased regional averages can lead to inaccurate global results. Large uncertainties exist in the global lake CO_2 flux estimates, ranging from 0.29 to 0.81 Pg C yr^{-1} [5,9,10]. A study has pointed out that CO_2 emissions may likely have been substantially overestimated by a factor of 9–18 in Africa [6]. These discrepancies arise from many factors, including alterations in the lake area [11–13] and pronounced disparities in the adopted regional CO_2 emission values. For example, the lack of data for tropical lakes was a major impediment to obtaining accurate global estimations [14].

Local or regional CO_2 emissions are often calculated using the inter-lake mean partial carbon dioxide pressure (pCO_2) and gas transfer velocity. Unlike the fairly uniform atmospheric pCO_2, lake pCO_2 varies significantly both spatially and temporally and depends on the lake size, salinity, and trophic state [15–19]. The results of CO_2 fluxes obtained using the inter-lake mean pCO_2 values may overestimate or underestimate CO_2 emissions because they ignore seasonal variations, trophic states, and lake saline gradients. In northern lakes, the major sustained CO_2 emissions exhibit significant seasonal differences and patterns, which need to be considered in reassessing the lake's CO_2 emissions under future climate warming scenarios [20]. An accurate estimate of the annual global greenhouse gas emissions from lakes, considering the differences

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in lake size and productivity, indicated that the traditional upscaling approach overestimated CO_2 emission [10]. A study on greenhouse gas emissions from African lakes also pointed out that CO_2 fluxes were related to the lake size and productivity [14]. Studies on selected lakes, such as the Sete Cidades Lake and Lizard Lake, elucidated that the lake's trophic states significantly influenced the lake's pCO_2 and CO_2 emissions [15,21]. Nowadays, lake eutrophication is considered a serious global environmental problem, especially in eastern Asia [22–25]. Gradually intensifying eutrophication has dramatically changed the ecological function and carbon cycle of lakes [23,26–28]. Future lake carbon emission studies should thus pay more attention to the lake nutrient levels and seasonal pCO_2 patterns. Lakes with high nutrient content and blooming algae growth could function as net sinks for atmospheric CO_2, especially when they are small and enriched with oxygen from primary production [29,30].

Saline lakes have also been recognized as important regional and global climate contributors. The global CO_2 emissions to the atmosphere from saline lakes are estimated to be 0.11–0.15 Gt C yr^{-1} [1,31]. However, a recent study conducted in the Qingzang Plateau presented a contrary view that saline lakes, which absorb large amounts of CO_2 from the atmosphere during the ice-covered periods, may turn saline and thus become carbon sources with future climate warming [32]. This proposition was further confirmed by a long-term CO_2 exchange flux analysis associated with the changes in the lake numbers and area [33]. Global saline lakes emitted 0.11–0.15 Pg C yr^{-1} to the atmosphere in 2008 [1]. However, with the escalating global climate change, the biological-physical-chemical processes affecting the carbon exchange processes at the water–air interface may change gradually. Thus, determining whether saline lakes function as carbon sinks or sources requires further investigation.

China has a total of 2693 lakes with an area of over 1 km^2. These lakes account for 6.2% of the world's total lake surface area, specifically within the latitudes ranging from 25° to 54° N [34]. While estimates of the CO_2 emissions from the lakes in China are available from several sources [35–37], they were largely generated using the inter-lake mean pCO_2 values, which failed to consider salinity and seasonal variations and the effect of trophic differences on lake pCO_2. This study measured *in situ* lake pCO_2 over different seasons and trophic states during 2019–2022. Our research objectives encompassed four key aspects: (1) explore the distribution patterns of pCO_2 values for different seasons, trophic states, and fresh or saline lakes; (2) examine the estimation accuracy of the areal CO_2 emissions in the studied lakes, ignoring the pCO_2 differences related to the trophic and seasonal changes; (3) refine the CO_2 flux estimates from Chinese lakes, relating it to the area of lakes with different trophic states and saline lakes; and (4) discuss the implications of this study results for the carbon budget of inland waters in China. Knowing the dependence of CO_2 released from lakes on the season, salinity, and trophic states is important for understanding the lake carbon cycle under future climate changes.

2. Materials and methods

2.1. Field sampling and measurements

China, situated in eastern Asia, predominantly occupies the temperate climatic zone, with only marginal portions extending into the subtropical and tropical zones. The country boasts an impressive tally of approximately 2700 lakes, each spanning an area exceeding 1 km^2 in China, contributing to a cumulative lake expanse covering 81,415 km^2 [12,34]. Thirteen field research campaigns were conducted on Chinese lakes between 2017 and 2021 to measure the pCO_2 values. Detailed information on the sampling campaigns has been provided in Table S1. To compare the seasonal variations of pCO_2 in the lakes, seven lakes with different trophic states, which are ice-free all year round, were sampled in January, April, July, and November of 2019. These included the Baipenzhu Reservoir, Zhelin Lake, Xinfengjiang Reservoir, Fengshuba Reservoir, Poyang Lake, Gaoyou Lake, and Hongze Lake. To compare the effect of trophic variations on lake pCO_2, a total of 43 lakes were sampled in summers between 2017 and 2021 (Table 1). These lakes were distributed over five different regions of China: eight in Northeast China, four in the Yangtze River region, five in the Huang-Huai-Hai region, four in the Greater Pearl region, and twenty-three in the Qingzang Plateau (Table 1). The sampled lakes in the Qingzang Plateau included eight freshwater lakes and fifteen saline lakes.

Gas samples were collected at 6–10 sampling stations from every lake on average using the headspace equilibrium method as described in Supplementary Materials [7,38,39]. The measurements of CO_2 concentrations were made with a gas chromatography analyzer (Agilent, 7890B). Approximately 2 L of water was collected at every station at the water depth of 0.5–1 m for measuring water quality parameters in the laboratory by a standard procedure [40,41], including total nitrogen, total phosphorus, chemical oxygen demand, and chlorophyll *a*. The water samples were placed in a portable refrigerator and transported to the laboratory within 24–48 h. During gas and water sampling, water pH, salinity, water temperature, and electrical conductivity were all determined *in situ* using a portable multi-parameter water quality analyzer (YSI 6600, U.S.). Wind speed at 1.0 m above the water surface was measured by a portable anemometer (Pro'sKit MT-4615, China), while lake clarity was measured by a Secchi disc. The water and gas sampling were conducted between 9:00 a.m. and 2:00 p.m.

2.2. Calculation of CO_2 fluxes at the lake water-atmosphere interface

The measurements of CO_2 concentrations in surface water were converted into pCO_2 according to Henry's law [42]. Areal (i.e., per unit surface area) CO_2 flux at the lake water and atmosphere interface (FCO_2, mmol m^{-2} d^{-1}) was calculated from the water–air gas concentration gradient (pCO_2–$pCO_{2,air}$) using the following equation:

$$FCO_2 = k \times k_H \times \left(pCO_2 - pCO_{2,air}\right) \qquad (1)$$

where k is the gas transfer velocity (m d^{-1}), k_H is Henry's constant for CO_2 corrected for temperature and pressure (0.029433 atm mol^{-1} m^{-3} at 25 °C), pCO_2 is the gas partial pressure of CO_2 in surface water (μatm), and $pCO_{2,air}$ is the concentration of CO_2 in air (μatm). A positive FCO_2 corresponds to CO_2 emission from water to the atmosphere, whereas a negative value indicates that carbon is absorbed in water. The detailed calculation process is explained in Supplementary Materials. The relative underestimation or overestimation of FCO_2 (%) was calculated as follows:

$$\textit{Relative underestimation or overestimation}\ (\%) = \frac{FCO_{2,\mathrm{mean}} - FCO_2'}{FCO_{2,\mathrm{mean}}} \times 100 \qquad (2)$$

where $FCO_{2,\mathrm{mean}}$ (mmol m^{-2} d^{-1}) is the areal CO_2 flux calculated based on the mean pCO_2, and FCO_2' (mmol m^{-2} d^{-1}) is the areal CO_2 flux calculated based on the mean pCO_2 of lakes in the same season/trophic state/salinity level. A positive value indicates overestimation, whereas a negative value indicates underestimation.

The total annual CO_2 flux (F, Tg C yr^{-1}) was calculated using the lake area and areal CO_2 flux using the following formula:

$$F = F_{CO_2} \times area \times Days \qquad (3)$$

where FCO_2 (mmol m^{-2} d^{-1}) is the areal CO_2 flux, *Days* is the total number of days in a year, and *area* (m^2) is the total lake area.

2.3. Data analysis

The transportation of terrestrial organic carbon (OC) mediated by inland waters in China was quantified as the sum of the annual total carbon flux from inland waters (lakes, reservoirs, and rivers), burial in sediment, and total OC amount transported to the ocean. The data on the carbon flux from inland waters in China was taken from Li et al. (2018) and Ran et al. (2021) [35,36]. The total OC amount from Chinese rivers transported to the ocean was obtained from Liu et al. (2020) [43]. The OC burial in Chinese lakes and reservoirs was calculated from the total OC burial amount of the global lakes and reservoirs and the areal proportion of Chinese lakes and reservoirs to the global area [36,44]. The OC burial in Chinese rivers was found using two main approaches. The first approach involved multiplying the total OC burial amount from global rivers by the areal proportion of Chinese rivers to global rivers [31,44]. The second approach mirrored this method, focusing on the total OC burial amount of the Yangtze River while considering its net discharge proportion with all Chinese rivers [41,45].

The trophic states of lakes were assessed using the comprehensive trophic status index (TSI). The detailed formulas were provided in the Supplementary Materials. In this study, several mean pCO_2 values were adopted: (1) the annual mean pCO_2 value, the mean value from all sampled sites over four seasons of the year; (2) the seasonal mean pCO_2 value, the mean value of all sampled sites in the same season; and (3) the trophic mean pCO_2 value, the mean value of all sampled sites in the same trophic state. The changes in pCO_2 values over different seasons or lakes with

Table 1
Detailed information on the sampling lakes.

Regions	Number	Lake name	Latitude (°)	Longitude (°)	Lake area (km^2)	k (m d^{-1})	Sites
Northeast	8	Jingyuetan	43.788769	125.44754	4.3	2.8	8
		Xingxingshao	43.637612	126.06041	32	4.4	8
		Songhua Lake	43.615488	126.72799	550	3.1	10
		Chagan Lake	45.245856	124.27755	307	5.8	10
		Taipingchi	44.019061	124.94094	118	6.9	8
		Baishan	42.526073	127.07135	300	3.0	10
		Xianghai	45.055601	122.29974	71	3.1	5
		Xingkai Lake	45.232661	132.42817	4380	3.5	10
Yangtze River	4	Zhelin Lake	29.260031	115.42981	308	4.6	10
		Poyang Lake	29.553269	116.12792	3960	4.0	10
		Caizi	30.836308	117.04511	226	4.0	8
		Chaohu	31.675051	117.33582	780	4.4	10
Huang-Huai-Hai	5	Gaoyou	32.818524	119.30265	760	3.6	8
		Hongze Lake	33.156228	118.72333	2070	3.7	8
		Wuhai Lake	39.566222	106.78333	118	3.7	5
		Ulansuhai Nur	40.915390	108.86715	300	2.9	10
		Xiaolangdi	34.940083	112.32916	159.6	3.5	10
Greater Pearl	3	Baipenzhu	23.099826	115.11084	6.5	3.7	10
		Xinfengjiang	23.772185	114.51584	304.5	3.3	10
		Fengshuba	24.449706	115.40236	30	5.0	10
Qingzang Plateau	23	Qinghai Lake	36.790240	100.27367	4625	3.6	10
		Yamdrok Lake	29.156343	90.63348	638	1.8	5
		Kara nor	38.226776	97.61404	625	1.9	5
		Namtso Lake	30.926865	90.97337	1920	3.0	8
		Tuosu Lake	37.124402	96.90319	57.4	2.4	8
		Puma Yumco	28.578587	90.50908	295	1.6	5
		Siling Co	31.711048	88.68664	2391	1.8	6
		DagzeCo	31.879300	87.51037	244.7	1.9	6
		Zhari Namco	30.971675	85.43461	1023	3.5	6
		Tangra Yumco	31.050834	86.60711	835	1.7	8
		Chaidan Lake	37.452675	95.51069	30	2.2	4
		Baimalamu Tso	30.778772	90.96806	1.45	1.7	4
		Pa Mucco	31.164661	90.60645	180	1.9	6
		Phong Tso	31.524897	90.98188	187	2.1	4
		Jiang Tso	31.529219	90.79425	150	2.1	4
		Dongji Cuona	35.303879	98.65557	232	1.5	6
		Longyangxia	36.135446	100.88058	158.4	2.2	10
		Lijiaxia	36.148261	101.77941	10.45	3.5	10
		Grenco	31.053208	88.54472	475.9	2.9	4
		Eling Lake	35.062261	97.70428	610	2.6	6
		Tso Ngön	31.685849	88.70192	244	3.3	6
		Wuru Tso	31.740235	87.87465	342.7	2.7	3
		Keluke Lake	37.276993	96.91403	57.4	2.1	10

different trophic states were analyzed with ANOVA or a non-parametric test using SPSS 23.0 for Windows.

3. Results

3.1. Seasonal variations in CO$_2$ in lakes with different trophic states

The $p\mathrm{CO_2}$ values in the studied lakes varied between 217 and 801 μatm across all seasons (Fig. 1), with the lake annual mean $p\mathrm{CO_2}$ values ranging from 412 ± 212 to 556 ± 207 μatm (Table S2). The annual mean $p\mathrm{CO_2}$ values at all sampled lakes were greater than that of the atmosphere (411 μatm in 2019). The $p\mathrm{CO_2}$ values showed significant seasonal variations regardless of the trophic state (Fig. 1). The highest seasonal mean $p\mathrm{CO_2}$ values occurred in winter and were 1.3–1.6 times higher than the annual mean value, while the lowest values in summer were only 0.5–0.7 times that of the annual mean value (Fig. S1). Under-saturated seasonal mean values were observed in summer and autumn (Fig. 1). On the whole, there was no obvious variation of $p\mathrm{CO_2}$ values observed in the lakes with different trophic states ($p > 0.05$).

Based on the annual mean $p\mathrm{CO_2}$ values, k and k$_\mathrm{H}$, the areal CO$_2$ emissions ($F\mathrm{CO}_{2,\mathrm{mean}}$) were calculated for the lakes with the values ranging between 0.1 and 24.3 mmol m^{-2} d^{-1} (Table S2) while ignoring the seasonal differences in $p\mathrm{CO_2}$ values. When the seasonal variations were considered, the CO$_2$ emissions were calculated using the mean $p\mathrm{CO_2}$ value for the corresponding season. The high mean $p\mathrm{CO_2}$ in winter resulted in higher areal CO$_2$ emissions ($F\mathrm{CO}_{2,\mathrm{season}}$ = 22.2–48.5 mmol m^{-2} d^{-1}) than the annual mean $p\mathrm{CO_2}$ values. There was thus a discrepancy between the annual $F\mathrm{CO}_{2,\mathrm{season}}$ and $F\mathrm{CO}_{2,\mathrm{mean}}$ in the lakes. When substituting the annual mean $p\mathrm{CO_2}$ values with the *in situ* $p\mathrm{CO_2}$ values from one season to calculate $F\mathrm{CO}_{2,\mathrm{mean}}$, the results may be underestimated compared to the flux values calculated using the spring or winter $p\mathrm{CO_2}$ values (spring: 4.0–208.5%, winter: 185.0–1152.6%), or overestimated compared to the flux values calculated using the summer and autumn $p\mathrm{CO_2}$ values (summer: 114.1–933.7%, autumn: 105.2–368.6%) (Fig. 2).

Fig. 1. Seasonal variation of $p\mathrm{CO_2}$ values (μatm) in lakes with different trophic states. Oligo-1: Baipenzhu Reservoir; Oligo-2: Zhelin Lake; Meso-1: Xinfengjiang Reservoir; Meso-2: Fengshuba Reservoir; Meso-3: Poyang Lake; Eutro-1: Gaoyou Lake; Eutro-2: Hongze Lake.

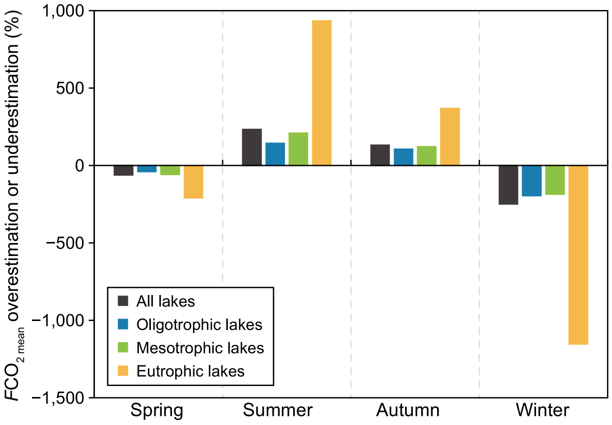

Fig. 2. Differences between $F\mathrm{CO_2}$ values calculated based on annual mean $p\mathrm{CO_2}$ values and seasonal mean $p\mathrm{CO_2}$ values.

3.2. Analysis of CO$_2$ variations in lakes with different trophic states

Ran et al. (2021) [36] assigned all lakes in China to different regions based on the hydrologic units, climate, and geomorphological conditions. Although the above seasonal analysis of $p\mathrm{CO_2}$ values revealed no obvious variations with the trophic states, the lakes in the same region varied in $p\mathrm{CO_2}$ with the trophic states (Fig. 3, Fig. S2). Regardless of the regional classification of freshwater lakes, eutrophic lakes had the lowest $p\mathrm{CO_2}$ values (173.3–593.3 μatm), while the oligotrophic lakes displayed the highest (354.9–725.3 μatm).

Based on the mean $p\mathrm{CO_2}$ values in all sampled lakes, k and k$_\mathrm{H}$, the areal CO$_2$ emissions ($F\mathrm{CO}_{2,\mathrm{mean\text{-}T}}$) were obtained for the lakes as −24.3 to +37.2 mmol m^{-2} d^{-1} (Table S3), when the trophic differences in $p\mathrm{CO_2}$ values were ignored. Using the $p\mathrm{CO_2}$ values corresponding to variable trophic state, the highest areal CO$_2$ emissions were obtained for the oligotrophic lakes (−18.4 to +47.1 mmol m^{-2} d^{-1}, mean value = 11.1 mmol m^{-2} d^{-1}) and the lowest for the eutrophic lakes (−54.8 to +24.3 mmol m^{-2} d^{-1}, mean value = −14.7 mmol m^{-2} d^{-1}) (Table S3). If the calculation of CO$_2$ emissions ignored the trophic differences in $p\mathrm{CO_2}$, compared to using the eutrophic $p\mathrm{CO_2}$ values, the results ($F\mathrm{CO}_{2,\mathrm{mean\text{-}T}}$) would be overestimated by 35.1–161.9%; compared to using oligotrophic and mesotrophic $p\mathrm{CO_2}$ values, $F\mathrm{CO}_{2,\mathrm{mean\text{-}T}}$ would be underestimated by 7.4–189.7% (Fig. 4).

3.3. Analysis of CO$_2$ in saline and freshwater lakes

When the $p\mathrm{CO_2}$ values in lakes from the Qingzang Plateau were analyzed, the saline lakes showed a lower mean value than the

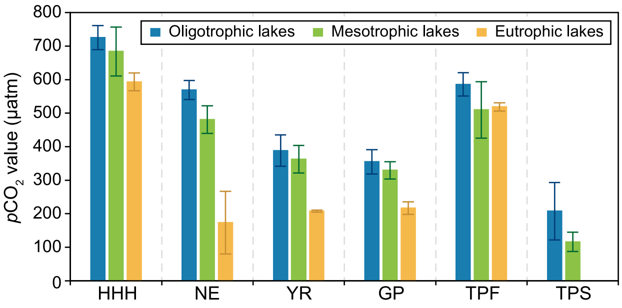

Fig. 3. Comparison of $p\mathrm{CO_2}$ values between different trophic states. HHH: Huang-Huai-Hai; NE: Northeast China; YR: Yangtze River; GP: Greater Pearl; TPF: Qingzang Plateau (Fresh); TPS: Qingzang Plateau (Saline).

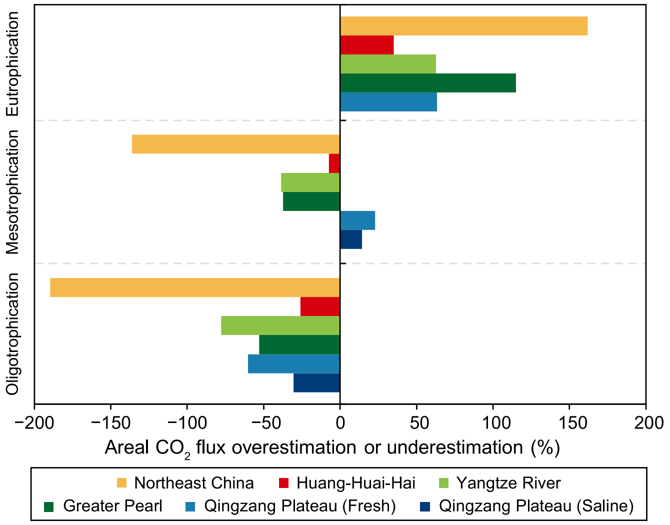

Fig. 4. Differences between areal CO_2 fluxes calculated using the same trophic pCO_2 and mean pCO_2 of all lakes with different trophic states. The vertical axis was the lakes with different trophic states.

freshwater lakes (mean ± SD = 170.9 ± 62.9 vs. 539.4 ± 11.05 μatm, SD: standard deviation) (Figs. S3 and S4). In this region, the areal CO2 fluxes ranged from −61.5 to 5.6 mmol m^{-2} d^{-1} (mean ± SD = −23.1 ± 17.4 mmol m^{-2} d^{-1}) in saline lakes and between 5.9 and 61.9 mmol m^{-2} d^{-1} (mean ± SD = 19.3 ± 18.3 mmol m^{-2} d^{-1}) in freshwater lakes (Fig. 5). Almost all saline lakes located in the Qingzang Plateau investigated in this study showed high CO_2 uptakes, with the areal CO_2 fluxes lower than that calculated with the mean pCO_2 of both freshwater and saline lakes (−12.2 mmol m^{-2} d^{-1}). Compared to using the mean pCO_2 values for saline or freshwater lakes, the areal CO_2 emissions calculated using the mean pCO_2 values (combining freshwater and saline lakes) would cause a flux overestimation of about 89.1% or a 258.2% underestimation.

3.4. Carbon budget calculation for inland waters in China

In this study, the transportation of terrestrial OC mediated by inland waters in China was quantified based on refining annual total CO_2 flux from lakes using statistical models and datasets available in published literature [35,36,41,43,45,46] (Fig. 6). The results showed that the processing of carbon in inland waters in China resulted in an annual emission flux of 78.1–123.3 Tg C and burial of 34.3–55.9 Tg C [31,36,47]. Given the annual OC transport of 14.8 ± 5.1 Tg by Chinese rivers into marginal seas [43], the total amount of OC imported to Chinese inland waters from the terrestrial environment should be approximately 119.0–195.1 Tg. Among this, 22–36% is stored in sediment, 50–79% is emitted to the atmosphere as CO_2 and CH_4, and only 6–13% is discharged into oceans via rivers.

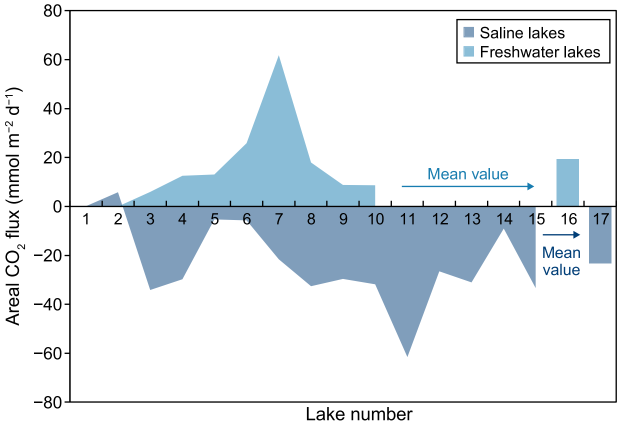

Fig. 5. Areal CO_2 fluxes in freshwater and saline lakes in the Qingzang Plateau. The x-axis shows sampled lake numbers, including eight freshwater and fifteen saline lakes.

4. Discussions

4.1. Influence of seasonal change on CO_2 flux estimation

CO_2 emissions from lakes constitute a crucial component of the global carbon cycle, and the temporal dynamics of sustained CO_2 emissions have attracted the attention of the academic community [20,48]. The pCO_2 values in lakes often show significant seasonal variability with lower values in summer in high-latitude regions for several reasons, including strong photosynthesis of phytoplankton, water temperature dynamics, and thermal stratification of the water column [46,49]. This seasonal variability of pCO_2 should be fully considered when calculating the CO_2 fluxes [20]. Most current studies estimated the areal CO_2 emissions relying on the surface water pCO_2 and extrapolating the areal CO_2 emissions to the annual total CO_2 flux through the water surface area. However, the frequent practice of using the mean pCO_2 values may result in underestimation or overestimation of the final areal CO_2 fluxes [1,50,51]. The results presented in this study have pointed out the differences between the calculated areal CO_2 emissions when using the mean pCO_2 values and seasonal pCO_2 values (Fig. 2). Thus, it can be assumed that most gas samples from lakes were acquired in summer and autumn, but the areal CO_2 flux calculations were conducted based on the annual mean pCO_2 values, overestimating the final results by 105.2–933.7% (mean 280.1%) compared to the fluxes derived from the seasonal pCO_2. It is worth noting that the CO_2 flux bias induced by the seasonal variability of pCO_2 may not necessarily be to tropical lakes due to the "endless summer" characteristic of the tropics [14,52].

Only limited large-scale studies have addressed the contribution of seasonal variations in pCO_2 to the CO_2 flux. In China, Ran et al. (2021) considered both dry and wet seasonal differences in pCO_2 when analyzing the CO_2 emissions from Chinese reservoirs and lakes and arrived at a flux value of 8.4 Tg C yr^{-1} [36]. However, this value was only half that Li et al. (2018) obtained, who estimated the CO_2 flux from Chinese lakes to be 16.0 Tg C yr^{-1} based on the mean value of the collected pCO_2 [35]. The areal CO_2 flux estimation by Ran et al. (2021) was also inconsistent with the value calculated using the mean pCO_2 by Wen et al. (2017) [37]. However, according to the present study, Wen et al. (2017) may have underestimated the areal CO_2 flux due to their reliance on predominantly summer and autumn pCO_2 values [37]. The primary reasons for the estimation uncertainty in those previous studies were the seasonal pCO_2 data paucity and inaccurate water surface area measurements. Nowadays, remote sensing technology enables effective and accurate observation of the spatial-temporal changes in water surface area [11–13]. Therefore, knowing the seasonal variations in pCO_2 should facilitate more accurate flux estimations.

Additionally, a significant daily variation in pCO_2 has been observed in some lakes [19,53,54]. The CO_2 evasion estimates from lakes should also consider the daily variability of pCO_2. It is a critical step to reduce uncertainties in CO_2 flux estimations of lakes. The diurnal pCO_2 is related to the daily cycles of metabolic activity, with a consistently declining trend of pCO_2 from early mornings to late afternoons [8,19,49,53]. Previous studies have also confirmed a

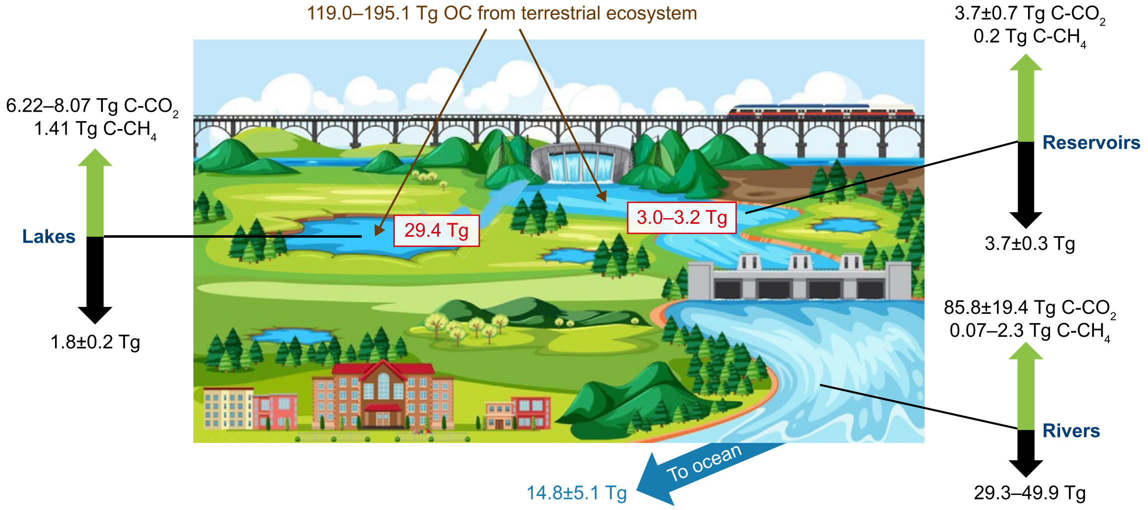

Fig. 6. Schematic diagram of terrestrial carbon transport by inland waters (rivers, lakes, and reservoirs) in China. The green arrows represent the carbon emission from the inland waters, the black arrows represent the carbon burial from the inland waters, and the blue numbers represent the organic carbon storage in waters.

close correlation between daily changes in pCO_2 and solar radiation, water temperature, and the lake trophic status [19,49,55,56]. In this study, the authors actively scheduled all sampling campaigns between 9:00 a.m. and 12:00 p.m., aiming to minimize the impact of pCO_2 daily variation to the greatest extent possible.

4.2. CO_2 flux in different trophic state lakes

In the region with distinct seasons, due to the high primary production in summer, pCO_2 values show a declining trend with increasing trophic states, with the eutrophic lakes always showing lower pCO_2 values than mesotrophic and oligotrophic lakes in the same region [57]. This phenomenon was also observed in the current study, but it only applied to lakes located in the same region (Fig. 3). The trophic differences in lake pCO_2 led to the estimation uncertainty of CO_2 fluxes calculated using primarily the mean pCO_2 for sampled lakes, especially that the majority of them were eutrophic. Previous study has found that eutrophication may increase the total amount of greenhouse gases emitted from lakes [10], but CO_2 emissions to the atmosphere from natural lakes will decline substantially with the intensified eutrophication [21,29,47]. Some African pristine lakes are naturally eutrophic without man-made nutrient inputs and are also the sinks of CO_2 [14,52,58]. Accordingly, there are grounds to hypothesize that areal CO_2 fluxes from lakes reported in previous studies without considering trophic levels might have been overestimated due to the increasingly severe lake eutrophication. However, it should be noted that eutrophic lakes account for 63.1% of the total number globally, but they cover a smaller fraction of the Earth's surface area, only 30.5%. In contrast, mesotrophic and oligotrophic lakes account for 69.5% of the total surface area. Especially in Asia, mesotrophic lakes occupy the highest proportion (71.2%) [24]. The annual total areal CO_2 flux estimated from the areal CO_2 flux (ignoring trophic levels) and the area of lakes may be underestimated due to the larger area contribution of oligotrophic and mesotrophic lakes.

In China, mesotrophic lakes account for over 60% of the total lake number [59]. Thus, we deduced that our current estimation of the annual total CO_2 flux from China lakes might be underestimated due to ignoring the trophic differences in $pCO_{2.}$ We assumed that k and k_H were consistent with those used in the previous study by Ran et al. (2021). To rectify this, we adjusted CO_2 fluxes for each region for the corresponding contribution of eutrophic, mesotrophic, and oligotrophic lake areas to the total lake area [25]. Finally, we obtained CO_2 fluxes in the Greater Pearl River, Yangtze River, Huang-Huai-Hai, Northeast China, and Qingzang Plateau as 0.12, 1.20, 0.82, 2.10, and 3.90 Tg C yr^{-1}, respectively. The revised CO_2 flux from the Chinese lakes was about 9.35 Tg C yr^{-1} (without adjusting the CO_2 flux in northwest China). This last value was higher than the CO_2 flux of 8.4 Tg C yr^{-1} reported recently by Ran et al. (2021) [36]. Future research on lake carbon emissions should pay more attention to the nutrient levels in lakes because eutrophic lakes may act as a net sink for the atmospheric CO_2, and oligotrophic and mesotrophic lakes occupy a higher area proportion and contribute more to the CO_2 emissions [29].

4.3. Role of saline lakes in lake carbon emissions

Saline lakes comprise approximately a quarter of the Earth's lake surface area and approximately 44% of their total volume [1,60]. These lakes contain vast amounts of dissolved carbon, primarily due to their locations at hydrological terminals, and always play a significant role in lake carbon cycling thanks to the active organic-inorganic carbon conversion process [61–63]. A published study has illuminated that CO_2 emissions from saline lakes amounted to about 18% of the global lake CO_2 flux occurring over about 23% of the lake surface [3], which implied that the role of saline lakes in lake carbon emissions cannot be evaluated as equivalent status with the area. In a regional estimate of CO_2 emissions from Chinese lakes, the CO_2 emissions to the atmosphere from saline lakes (per unit lake surface area) were predicted to be smaller than that of freshwater lakes [37]. However, the existing estimations of CO_2 flux from global lakes often do not distinguish between saline and freshwater lakes, resulting in inaccurate annual total lake carbon budgets. According to the results presented in this study, excluding saline lakes from sampling campaigns in the Qingzang Plateau would result in an overestimation of the lake CO_2 flux due to the lower pCO_2 of saline lakes in the region. A study considering both the ice-covered and ice-free periods showed that saline lakes in the Qingzang Plateau represented a sizable CO2 sink [48,64].

Due to climate change, the contribution of northern saline lakes to global carbon emissions may be reduced in the future due to the declining ice cover and increasing pH levels [50,65]. A previous study has also shown that increasing atmospheric temperature could decrease CO_2 emissions from hardwater lakes [65]. Thus, the role of saline lakes in the greenhouse gas emissions from lake ecosystems is drawing increasing attention. In China, saline lakes contribute to about 55% of the total lake area and 68% of the total water storage. About 86% of Chinese saline lakes were in the Qingzang Plateau [66]. The study concluded that the areal CO_2 emissions derived from the mean pCO_2 values (combining freshwater and saline lakes) would cause an approximate 89.1% overestimation compared to the saline lake pCO_2 values. We used this result and adjusted the total CO_2 flux of 8.4 Tg C yr^{-1} from China lakes, according to Ran et al. (2021), to a rough value of 6.22 Tg C $yr^{-1,}$ ignoring the trophic status of the lakes. Saline lakes with a total area of 27,600 km^2 approximately account for 70% of the total lake area on the Qingzang Plateau. According to the area and the overestimation rate, we further amended the total CO_2 flux for the Qingzang Plateau (3.90 Tg C yr^{-1}, which has been corrected considering the trophic states) to 2.61 Tg C yr^{-1}. As a result, the annual total CO_2 emissions from Chinese lakes further changed from 9.35 to 8.07 Tg. However, this final value did not consider the proportions of saline lakes in other regions and thus will need to be further refined.

4.4. Implications for carbon budget for inland waters in China

This study found that CO_2 emissions from lakes vary significantly with changing seasons and lake eutrophic states. Saline lakes may contribute less CO_2 emissions to the atmosphere than the mean CO_2 emissions from all lakes. Chinese lakes alone may annually emit 6.22–8.07 Tg C of CO_2, lower than previously reported 8.4 Tg C and 15.98 Tg C by Ran et al. (2021) and Li et al. (2018), respectively [35,36]. Under climate change and intense human activities, eutrophication is considered a global environmental problem, which has also recently become serious in China [67–69]. On the other hand, eutrophication may convert a lake from a CO_2 source to a sink [21,47]. Meanwhile, in response to climate change, as important carbon gas emitters, lakes are also undergoing rapid changes, increasing in the area globally, thus producing more carbon emissions [12]. There are inherent feedback loops between the carbon cycle and climate change, and the contributions of CO_2 emissions from Chinese lakes to global climate change must be further explored.

Although inland waters occupy an exceedingly small portion of Earth's surface, they are a “hot spot” of carbon exchange and transformation [70]. Research has pointed out that CO_2 emissions from inland waters are on a similar scale to the land-ocean net carbon exchanges [6,71]. The concept of “active pipe” of inland waters was proposed by Cole et al. (2007) [31], which conceptualized inland waters as a unidirectional OC conduit from soils to sea. During this transmission, part of OC is transported to the atmosphere and sediments via biogeochemical processes. According to the quantitative results of the present study, the annual emission flux and burial of OC in inland waters in China were 78.1–123.3 and 34.3–55.9 Tg C, respectively. The estimated ratio of burial to emission was similar to that of the global inland waters reported by Tranvik et al. (2009) (0.6 Pg buried to 1.4 Pg emission). The inland waters in China correspond to about 3.5% of the global inland water area (1.67×10^5 vs. 47.86×10^5 km^2) (Maavara et al., 2017; Zhang et al., 2017), and the terrestrial OC input amounts to that of Chinese inland waters (119.0–195.1 Tg) was about 2.3–3.8% of the global terrestrial OC transportation (5.1 Pg) [71]. The estimate was broadly consistent with the area proportion. The estimated value was reached without subtracting the amount of carbon fixated from the atmosphere via photosynthesis by aquatic vegetation and algae. Moreover, the mineralization of OC and emission to the atmosphere from inland waters are generally accepted by some researchers as explanations for the OC loss during passage through the continuum of inland waters from soils to the sea [72]. However, CO_2 emissions from inland waters were not only driven by carbon input from the drained land. Wetland CO_2 pump may also contribute disproportionately to CO_2 emissions from inland waters [73]. Also, the sedimentation and burial in freshwater systems are additional reasons for the OC losses [74]. The rough estimates of the OC storage in Chinese lakes (29.4 Tg) and reservoirs (3.0–3.2 Tg) derived in this study have indicated that the amount of carbon stored in water was astonishingly high. Acknowledging this will help to fully elaborate the carbon budgets for inland waters in China [75,76].

In recent decades, most efforts have been devoted to precisely quantifying the terrestrial OC and its fate at the regional and global scales [5,31,64,77]. However, our understanding of carbon budgets for Chinese inland waters has remained inadequate, primarily due to limited studies and a lack of nationwide comprehensive investigations. This research represents a pioneering endeavor to quantify the “active pipe” function of Chinese inland waters schematically shown in Fig. 6. Further, the refined approach to lake CO_2 emission estimation proposed in this study and the results for the terrestrial OC budgets in Chinese inland waters have the potential to serve as a valuable template for enhancing the precision of carbon emission assessments in inland waters worldwide.

5. Conclusions

In this study, we investigated CO_2 emissions from Chinese lakes, each characterized by different trophic states and saline gradients, throughout four seasons of the year. Our investigation encompassed a total of 13 sampling campaigns. The pCO_2 values from lakes exhibited seasonal variations, with winter and summer registering the highest and lowest average values, respectively. There was an obvious trophic variation trend in the pCO_2 values but only for lakes from the same region. Saline lakes showed lower pCO_2 values and areal CO_2 emissions than freshwater lakes in the Qingzang Plateau (-23.1 ± 17.4 vs. 19.3 ± 18.3 mmol m^{-2} d^{-1}). Such variations in pCO_2 with seasons, trophic states, and salinity complicate the CO_2 emission calculations. However, this study quantified the influence of these factors on CO_2 emissions. From the quantitative analysis, the study indicated that the current annual total CO_2 fluxes from lakes in China might be overestimated when lake tropic states and salinity are ignored. Nevertheless, our conclusions are drawn from a dataset of limited scope. To improve the accuracy of regional and large-scale estimations of carbon emissions and carbon budgets in lake ecosystems affected by seasons, salinity, and trophic states, further investigations into CO_2 emissions from lakes in other parts of the world are warranted.

CRediT authorship contribution statement

Zhidan Wen: Conceptualization, Methodology, Writing - Original Draft, Field Investigation, Funding Acquisition. **Yingxin Shang**: Methodology, Field Investigation, Visualization. **Lili Lyu**: Laboratory Measurement, Data Curation. **Hui Tao**: Visualization, Data Curation. **Ge Liu**: Field Investigation, Visualization. **Chong Fang**: Data Curation. **Sijia Li**: Visualization. **Kaishan Song**: Conceptualization, Writing - Reviewing & Editing, Funding Acquisition, Supervision.

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

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