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Dynamic and high methane emission flux in pond and lake aquaculture

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ABSTRACT

Freshwater aquaculture systems are recognized as significant contributors to atmospheric methane (CH₄) emissions, yet accurate quantification remains challenging due to high variability across different aquaculture types and the scarcity of high-frequency observations. To address these gaps, we conducted synchronous in-situ measurements of CH₄ emissions from two typical aquaculture types - pond aquaculture and lake aquaculture - in the Yangtze River Delta, China, using the eddy covariance technique, capturing CH₄ flux from hourly to annual scales. Our high-resolution measurements revealed a weak diurnal pattern in CH4 flux, although daily mean flux varied considerately at the two systems (lake aquaculture: 0.04 to 6.60 μ g CH₄ m⁻² s⁻¹; pond aquaculture: 0.08 to 15.4 μ g CH₄ m⁻² s⁻¹). CH₄ flux was significantly higher in the pond aquaculture compared to the lake aquaculture, with the annual mean value of 5.08 μ g CH₄ m⁻² s⁻¹ (120 g CH₄-C m⁻² yr⁻¹) and 1.52 μ g CH₄ m⁻² s^{-1} (36 g CH₄-C m⁻² yr⁻¹), respectively. Further analysis suggested that smaller size of the ponds, combined with higher nitrogen and carbon loadings and elevated chlorophyll-a concentrations, likely contributed to substantial emissions from the pond aquaculture. Ebullition was identified as the primary emission pathway, accounting for 70 % and 60 % of the total CH₄ emissions from the pond and the lake system, respectively. While CH₄ flux increased significantly with increasing temperature, the flux during the warming phase of the year was lower than during the cooling phase at equivalent temperatures. Additionally, CH₄ flux in the pond system was less sensitive to temperature than in the lake system (Q_{10} for pond: 2.29; Q_{10} for lake: 5.75), likely due to the compound influence of biotic factors. Our findings underscore the importance of incorporating high-resolution emissions data from diverse aquaculture systems to accurately estimate the CH₄ budget of freshwater aquaculture.

1. Introduction

The renewed rapid growth of atmospheric methane (CH₄) (10.2 ppb/ year) has motivated research on freshwater ecosystems, which are regarded as the largest natural source of CH₄ (Rosentreter et al., 2021; Rocher-Ros et al., 2023; Thanwerdas et al., 2024). Among freshwater natural ecosystems like boreal wetlands and tropical rivers are significant CH₄ sources (Borges et al., 2015, 2022; Saunois et al., 2024), these human-impacted ecosystems have received considerable attention because their CH₄ emissions are sensitive to human activities (Xiao et al., 2017; Beaulieu et al., 2019; Rocher-Ros et al., 2023). Significant CH₄ emissions rates have been reported for freshwater aquaculture due to intense management practices (Yuan et al., 2019; Kosten et al., 2020). The total CH₄ emission from the global freshwater aquaculture systems is estimated to be 6 to 14 Tg CH₄ yr⁻¹ (Yuan et al., 2019; Rosentreter et al., 2021). Importantly, it is projected that aquaculture production worldwide will expand rapidly to keep pace with the escalating demand for protein sources (FAO, 2022; Zhang et al., 2022b), which will further amplify the role in aquacultural CH₄ emission in the global CH₄ cycle (Yuan et al., 2019; Gephart et al., 2021; Naylor et al., 2021; Dong et al., 2022). Thus, accurate quantification of CH₄ emissions from aquaculture ecosystems is necessary for mitigating CH₄ emissions from human-

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Received 30 October 2024; Received in revised form 4 January 2025; Accepted 17 January 2025 Available online 1 February 2025 0022-1694/© 2025 Elsevier B.V. All rights are reserved, including those for text and data mining, AI training, and similar technologies. dominated freshwater systems (Tong et al., 2021; Chen et al., 2023; Saunois et al., 2024; Yang et al., 2024a, b).

One major reason for the large uncertainty is that aquaculture CH₄ emission varies significantly among different aquaculture types. Pond aquaculture and lake aquaculture, the two predominant types of freshwater aquaculture, exhibit distinct emission dynamics and intensities. However, the IPCC only uses a single emission factor (EF: 183 kg CH₄ $ha^{-1} yr^{-1}$) to obtain the global freshwater aquaculture emission estimate (IPCC, 2021); This estimate is likely biased due to the lack of integration across diverse aquaculture types (Yuan et al., 2019; Dong et al., 2022). In theory, lake aquaculture, characterized by its larger size and natural water exchange, should emit less CH₄ per unit surface area due to the dilution of organic matter (Holgerson and Raymond, 2016). Other factors, such as low primary production rate and stable thermal stratification in large lake systems may also contribute to low CH4 emissions (Borges et al., 2022). However, aeration and dredging, two common management practices in pond aquaculture, can substantially decrease CH₄ emission (Yang et al., 2018, 2019; Fang et al., 2022; Nijman et al., 2022). Since these practices are not used in lake aquaculture, possibility exists that some pond systems may emit less CH₄ than lake aquaculture in the same climate. Currently, there is a focus on pondbased CH₄ emissions (Tong et al., 2021; Yang et al., 2019, 2024b; Yuan et al., 2021; Zhao et al., 2021), yet there is a scarcity of CH₄ emission observations in lake aquaculture (Pu et al., 2022). This imbalance in research likely skews our understanding of CH₄ emission processes in freshwater aquaculture.

Large temporal variabilities in CH₄ flux are another factor that contributes to the uncertainty in the CH₄ emission estimates. Methane emission from aquaculture systems is influenced by complex biogeochemical processes and is shaped by biotic factors (e.g., nutrients loading) and abiotic factors (e.g., temperature). These factors vary across a spectrum of time scales, leading to short-term (hourly) fluctuations in CH₄ flux and long-term trends that span seasons and years (Podgrajsek et al., 2014; Natchimuthu et al., 2016; Sieczko et al., 2020; Zhang et al., 2022c; Yang et al., 2024). Despite considerable temporal fluctuations in the flux, most studies have relied on snapshot samplings and daytime-only measurements at a low sampling frequency (monthly or seasonal) to obtain annual total emission rates (Kosten et al., 2020; Zhang et al., 2023b). The lack of temporal resolution may represent a large source of uncertainties (Wik et al., 2016; Duan et al., 2023). CH₄ flux from natural freshwater ecosystems often exhibit substantial diurnal fluctuations, with differences between different times of day exceeding 200 % (Xiao et al., 2014a; Zhang et al., 2019a; Hu et al., 2020; Tan et al., 2021). But the diel variation of aquacultural CH₄ flux is poorly understand due to the lack of continuous and high-frequency measurements (Kosten et al., 2020).

China is a hotspot region of CH₄ emissions from aquaculture. It is the world's largest freshwater aquaculture producer, contributing over 60 % of global aquaculture production and 58 % of CH₄ emissions from freshwater aquaculture systems among the top 21 global producers (Yuan et al., 2019; FAO, 2022). With the increasing need for protein diet, China's freshwater aquaculture is projected to further increase in both production and area in the coming decades (Cao et al., 2015; FAO, 2022). The Yangtze River Delta (YRD) in eastern China is an important region for freshwater aquaculture, accounting for 26 % of the country's total aquaculture area (China Fishery Statistical Yearbook, 2023). Because both pond and lake aquaculture are practiced, the YRD region offers an opportunity to compare and contrast emission rates and patterns between these two systems under the same climate conditions. Although previous studies have compared aquacultural CH₄ emission across different types in China (Yuan et al., 2019; Zhang et al., 2022d; Dong et al., 2022), they have been based on meta-analysis of data collected over different climate zones and without consideration of temporal variations. Furthermore, continuous measurements have been rare. In recent years, aquaculture in China has gradually shifted from lake to pond systems, driven by ecological restoration policies. This shift

caused an 18 % reduction in lake and reservoir aquaculture area and a 10 % increase in pond aquaculture from 2016 to 2022 (China Fishery Statistical Yearbook, 2023). Synchronized observations in both systems are needed to determine whether this policy-driven shift has resulted in an overall reduction in CH_4 emission.

In this study, we employed the eddy covariance (EC) technique to simultaneously measure CH₄ flux at an aquaculture pond complex and at an aquacultural lake in the YRD. The EC technique, which provides insitu measurement of the flux at half-hourly to hourly timesteps, have been employed in studies of natural lake and reservoir systems (e.g., Eugster et al., 2011; Podgrajsek et al., 2014; Zhang et al., 2019; Waldo et al., 2021; Hounshell et al., 2023). However, the application of this technique in aquaculture systems is still limited. There are two motivations for this study. Firstly, we are interested in temperaturedependent behaviors of the flux. Previous studies have suggested that the flux sensitivity to temperature may vary across different aquaculture types (Davidson et al., 2015; Zhu et al., 2020). Measurements at high temporal resolutions should help resolve this issue, therefore allowing more robust projection of how aquaculture CH₄ emission may respond to future climate warming. Secondly, ebullition, an important CH₄ emission pathway, is size dependent (Bastviken et al., 2004; Yang et al., 2020). Even though the eddy covariance technique measures the total CH₄ flux, it is now possible to isolate the ebullition component using high frequency (e.g. 10 Hz) data collected by eddy covariance (Iwata et al. 2018). Because aquaculture ponds are much smaller than aquaculture lakes, eddy covariance collected in these two systems provides a good opportunity to further investigate the size-dependence of the ebullition contribution to the total CH4 flux. Specifically, the objectives of this study are: (1) to compare temporal patterns of CH₄ flux between the two aquacultural systems; (2) to quantify their annual CH₄ emission and the ebullition contribution; and (3) to investigate biotic and abiotic factors that regulate the CH₄ flux across multiple time scales.

2. Materials and methods

2.1. Sites

The aquaculture pond and the aquaculture lake were both situated in the YRD, Eastern China, a region characterized by a subtropical humid monsoon climate (Fig. 1a). The mean air temperature and annual rainfall were 18.0 °C and 1290 mm at the lake site, and 17.5 °C and 1330 mm at the pond site in 2018. The pond site was in the middle of four fishponds (31° 58' N, 118° 15' E), each having a mean water depth of 1.8 m and an area of about 0.7 ha (Fig. 1b). The lake site was situated in the southeast corner of Lake Taihu (31° 03'N, 120° 28' E, Fig. 1c), with a mean water depth of 1.3 m, in an extensive aquaculture area of about 10600 ha in size (Qin et al., 2007; Pu et al., 2022). Both aquaculture systems were established in the 1980 s and have never been dredged.

Both aquaculture systems utilized a polyculture practice prevalent in the YRD region. At the pond, the polyculture system was dominated by six fish species mixing with clams. They received a commercial diet three times daily. To promote phytoplankton growth, chicken manure was applied twice per year, once in early February and again in late November. The lake aquaculture primarily stocked Chinese mitten crab, and was supplemented with black shrimp and giant river prawn. The annual feed input were 8570 kg C/ha and 6400 kg C/ha at the pond and the lake system, respectively.

2.2. CH_4 flux and supporting measurements

The EC observation took place at the two sites using two identical EC systems. Each EC system consisted of an open-path CO_2/H_2O infrared gas analyzer (Model EC150, Campbell Scientific Inc., Logan, Utah, USA), a three-dimensional sonic anemometer/thermometer (Model CSAT3A, Campbell Scientific Inc.), and an open-path CH₄ analyzer (Model LI-7700, LI-COR Inc., Lincoln, Nebraska, USA). They measured the fluxes



Fig. 1. (a) Map showing site locations in the Yangtze River Delta; (b) and (c) satellite images of the fish ponds and Lake Taihu; (d) and (e) photographs of the instrument setup and the measurement platform. Stars in b and c mark measurement locations.

of momentum, sensible heat, latent heat or water vapor, CO_2 and CH_4 . The observation height was approximately 1.0 m and 5.0 m above the water surface at the pond and the lake, respectively. Raw data were collected using a data logger (Model CR3000, Campbell Scientific Inc.) at a sampling frequency of 10 Hz. The results presented in this study span the period from January to December 2018 at the pond and from March to December 2018 at the lake.

Ancillary meteorological data were collected at the sites. Air temperature and air humidity were measured using a temperature/humidity probe (Model HMP155, Vaisala Inc., Helsinki, Finland). Wind speed and direction were observed using the three-dimensional sonic anemometer and a wind vane (Model 05103, R M Young Company, Traverse City, Michigan, USA). The four components of the surface net radiation were monitored with a four-way net radiometer (Model CNR4, Kipp&Zone B. V., Delft, the Netherlands). Water temperature at 20 cm below the water surface and sediment temperature at the depth of 5 cm were observed using temperature probes (Model 109-L, Campbell Scientific Inc.). All parameters were recorded at 1 Hz and archived as half-hourly averages by a datalogger.

In addition to micrometeorological variables, water chemical properties were measured at several locations. At the pond site, pH and dissolved oxygen concentration (DO) were observed at the 20 cm depth using a multi-parameter probe (Model: YSI 650MDS, YSI Inc., Yellow Springs, Ohio, USA), typically once per season. Water samples were collected at the same locations and the same time for analysis of dissolved organic carbon (DOC), total phosphorus (TP) and total nitrogen (TN), NO⁻ $_3$.N, NH⁺ $_4$.N, Chl-a concentration. At the lake, surface water samples were collected at a depth of 50 cm during each season: winter in February, spring in May, summer in August, and autumn in November. These samples were stored on ice while in the field and then transported to the Taihu Laboratory for Lake Ecosystem Research for analysis of water quality parameters (Zhang et al., 2016; Xiao et al., 2024).

2.3. Data processing and gap filling

The standard procedure embedded in the EddyPro software (version 6.2.1; LI-COR Inc.) was used to process the 10 Hz EC data to derive half-hourly fluxes. A number of processing steps were implemented in EddyPro. A series of quality control criteria were applied to screen raw

data for spikes (Vickers and Mahrt, 1997). Time lags resulting from spatial separations between the analyzers and the sonic anemometer were compensated using a cross-covariance maximization method (Horst and Lenschow, 2009). A double coordinate rotation was executed to force the mean vertical wind velocity to zero for each 30-min period (Lee et al., 2004). The WPL correction was applied to eliminate the effect of density fluctuations arising from variations in water vapor and temperature (Webb et al., 1980). The spectroscopic effect caused by temperature, pressure, and water vapor fluctuations on the shape of the CH₄ absorption line was rectified using the method outlined by McDermitt et al. (2011). High-frequency damping losses were assessed and corrected using the method proposed by Moncrieff et al. (1997). The 0-1-2 quality flag system developed by Mauder and Foken (2004) was utilized to classify flux data. Each 30-min datapoint was given a quality flag, categorized as 0 (best quality), 1 (good quality), or 2 (bad quality). Data with flags 0 and 1 were kept for further analysis. Flag 2 marks periods with the CH₄ relative signal strength indicator (RSSI) value below 10 %, instrument maintenance, or rain interference; these periods were excluded. Furthermore, a flux footprint analysis conducted at the pond sites revealed that about 80 % of the footprint fell within the targeted ponds, and the footprint peak was always located inside the pond boundary (Zhao et al., 2019; Zhao et al., 2021). Outside these ponds were also wet surfaces (rice paddies) whose CH4 flux was similar in magnitude to the pond flux (Section 4.2). Of the three gases we measured (CH₄, CO₂ and H₂O), CH₄ flux was the least affected by the surface source heterogeneity (Zhao et al., 2019). Overall, 44 % and 43 % of the CH₄ flux observations passed these data quality control checks at the lake and the pond sites, respectively.

The daily flux was determined by averaging all valid half-hourly observations on days with the 30-min flux data above 40 %. The valid daily CH₄ flux represented 56 % and 31 % of the total observational periods at the lake and pond, respectively. We employed the random forest (RF) method to fill the gaps in the daily CH₄ flux. Monthly and annual fluxes were determined with the gap-filled daily flux. As a supervised machine learning algorithm, RF is the optimal choice for completing the CH₄ flux dataset, especially for long gaps. This is because of its robustness against over-fitting, which is a key concern when using artificial neural networks, especially when dealing with limited training periods or noisy data (Kim et al., 2020; Mahabbati et al., 2021). All valid

data were first divided into two subsets: a training set (70 %) and a validation set (30 %). The RF model was initially built based on the training dataset and then was used to predict the CH₄ flux of the validation set. The input data used as predictors include global radiation, sediment temperature, wind speed, atmospheric pressure, relative humidity, precipitation, and day of year. These variables were obtained from ancillary micrometeorology measurements. Overall, the agreement between the prediction and the observation is satisfactory, with a coefficient of determination (R^2) of 0.92 and 0.85 and a root mean square error (RMSE) of 0.44 µg CH₄ m⁻² s⁻¹ and 0.48 µg CH₄ m⁻² s⁻¹ for the lake and the pond, respectively.

2.4. Estimating CH₄ ebullition

A partitioning method developed by Iwata et al. (2018) was utilized to isolate the ebullitive flux from the total CH₄ flux measured with the EC instruments. This method has been successfully applied in various ecosystems, including lakes, rice paddies, and bogs (Hwang et al., 2020; Pu et al., 2022; Taoka et al., 2020; Ueyama et al., 2022). It relies on the local-scale similarity between the CH₄ concentration and other scalar components, like H₂O concentration and air temperature, in the wavelet domain. It assumes that the ebullitive flux can be characterized by dissimilar fluctuations between the CH₄ concentration and a reference scalar. In this study, air temperature was chosen as the reference scalar. Wavelet coefficients between the CH₄ concentration and air temperature were calculated using a wavelet transform. An iteratively re-weighted linear regression was applied between the converted wavelet coefficients of CH₄ concentration and air temperature at the same scale. If wavelet coefficients significantly deviated from the fitted regression line and were greater than three times the minimum of root mean squared deviation for each month, this signal was considered to be ebullitive.

2.5. Determining temperature sensitivity

The temperature sensitivity of CH₄ flux is commonly characterized by the Q_{10} factor, which quantifies the proportional change over a 10 °C increase in temperature. We employed the Q_{10} method described by DelSontro et al. (2016), focusing on the "ecosystem-level" Q_{10} . This approach accounts for the cumulative effects of multiple biotic and abiotic factors that influence CH₄ flux. The Q_{10} for CH₄ flux was estimated using the formula $Q_{10} = e^{10b}$, where *b* represents the slope of the linear relationship between temperature and the natural logarithm of the observed CH₄ flux. This calculation was applied to daily flux data.

2.6. Data analysis

Single-variable regression was conducted to investigate the relationships between CH₄ fluxes with environmental factors across different time scales. The best-fit regression equation for CH₄ flux and temperature, along with *p* value and adjusted linear correlation *R*, were presented. Student's *t*-test was used to determine the significant differences between the two aquaculture types, with a significance level set at p < 0.05. All data processing and statistical analysis were performed with the MATLAB software (MathWorks, Inc., Natick, MA, USA).

3. Results

3.1. Temporal variations in environmental conditions

Significant differences in water quality parameters were observed between the pond and the lake, with the pond exhibiting markedly higher levels across all metrics (p < 0.05, Fig. 2, Table S1). The annual mean concentrations of nutrient indicators, including TN, TP, NH⁺ 4.N, and NO⁻ 3.N at the pond exceeded those at the lake by two to four times, with the NH⁺ 4.N concentration showing the largest difference of 13 times (Fig. 2a–d, Table S1). This nutrient enrichment at the pond was caused by abundant feed, small waterbody size and stagnant conditions. The mean DOC and Chl-a concentration at the pond were significantly higher, by an average of five times more, than those at the lake (Fig. 2e–f).

Seasonally, the concentrations of TN, $\rm NH^+$ 4.N, and $\rm NO^-$ 3.N were generally lower in the summer and the autumn than the winter and the spring at the two sites, primarily attributed to the higher denitrification rate, rainfall dilution, and phytoplankton uptake (Xu et al., 2010). At the pond, the reductions were significant, with TN decreasing by 39 %, $\rm NH^+$ 4.N by 15 %, and $\rm NO^-$ 3.N by 71 %. At the lake, the reductions were less pronounced but still substantial: TN by 18 %, $\rm NH^+$ 4.N by 49 %, and $\rm NO^-$ 3.N by 65 %. Additionally, the DOC concentration at the pond showed a substantial reduction of 43 % in the summer and the autumn compared to the winter and the spring, whereas the seasonality was not as evident at the lake.

In contrast to water quality, there were no large differences in physical factors such as water temperature (T_w) , sediment temperature (T_s) , and net radiation (R_n) between the pond and the lake, except for



Fig. 2. Seasonal mean value of (a) NO⁻₃.N concentration, (b) NH⁺₄.N concentration, (c) total phosphorus (TP) concentration, (d) total nitrogen (TN) concentration, (e) dissolved organic concentration (DOC), and (f) Chlorophyll *a* (Chl-a) concentration in the water column at the lake and the pond site.

wind speed (Table S1). Both the pond and the lake exhibited similar and significant diurnal and seasonal patterns in these factors (Fig. 3). Specifically, both systems displayed diurnal and seasonal patterns in T_w and $T_{\rm s}$, with generally higher values during the daytime than at night and during the summer than during the winter (Fig. 3, Table S1). The lake and pond sites showed similar annual mean values of $T_{\rm w}$ of 18.8 °C and 18.5 °C and T_s of 19.9 °C and 19.7 °C, respectively. The net radiation also followed a strong temporal pattern, reaching its peak during the month of July. The annual mean R_n values were comparable, at 105.7 W/m^2 and 103.2 W/m^2 at the lake and the pond site, respectively. The wind speed showed different seasonal and diurnal patterns between the sites. Higher values were observed during the daytime than night and in the winter and the spring than in the summer and the autumn at the pond; these patterns were reversed at the lake (Table S1). The annual mean wind speed at the lake site (3.17 m/s) were significantly higher than that at the pond site (2.04 m/s), primarily due to the large open water fetch at the lake site.

3.2. Diurnal and seasonal CH₄ flux

The two systems show different diurnal pattern of CH₄ flux (Fig. 4, Table S2). In the lake system, the flux was higher at night (20:00 to 04:00 local time), by an average 9 %, than during the day (08:00 to 16:00 local time). The most pronounced diurnal difference occurred in the winter, with the nighttime flux exceeding the daytime flux by 29 %, followed by 17 % in the autumn and 7 % in the summer (Table S2). In contrast, the pond exhibited a slightly higher flux in the daytime, by an average of 4 %, than at night. The diurnal difference at the pond peaked at 13 % in the autumn, followed by 7 % in the winter, 2 % in the summer, and 1 % in the spring.

In terms of the CH₄ flux seasonality, both systems exhibited peak values in the summer (June to August; Fig. 5a & b and Table S2), with a seasonal mean of 3.35 μ g CH₄ m⁻² s⁻¹ at the lake and 9.61 μ g CH₄ m⁻² s⁻¹ at the pond. The summer season accounted for 56 % and 50 % of the annual total emission at the lake and the pond, respectively. The lowest flux was observed during winter months (January, February, and December), the seasonal mean value being 0.26 μ g CH₄ m⁻² s⁻¹ at lake and 0.62 μ g CH₄ m⁻² s⁻¹ at the pond. The daily CH₄ flux at the lake ranged from 0.04 μ g CH₄ m⁻² s⁻¹ to 6.60 μ g CH₄ m⁻² s⁻¹, with an annual mean value of 1.52 μ g CH₄ m⁻² s⁻¹. At the pond, the variation

range and the mean value were both greater, with the daily flux ranging from 0.08 μ g CH₄ m⁻² s⁻¹ to 15.4 μ g CH₄ m⁻² s⁻¹ and the annual mean of 5.08 μ g CH₄ m⁻² s⁻¹. Seasonally, the CH₄ flux at the pond was 6.0, 2.9, 3.4, and 2.4 times higher than at the lake in the spring, summer, autumn, and winter, respectively (Table S2). It is noted that the negative half-hourly flux in Fig. 5 was caused by random errors, which is also present in other eddy covariance studies (e.g., Eugster et al., 2011; Podgrajsek et al., 2014; Waldo et al., 2021).

The annual gap-filled flux at the pond (1200 kg CH₄-C/ha) was approximately three times higher than at the lake (360 kg CH₄-C/ha). This difference was largely due to the frequent high half-hourly flux episodes (> 5 μ g CH₄ m⁻² s⁻¹) observed at the pond. Approximately 38 % of half-hourly measurements surpassed 5 μ g CH₄ m⁻² s⁻¹ at the pond, whereas only 8 % of the observations were above this threshold at the lake (Fig. 5c).

3.3. CH₄ ebullition

Over the annual period, ebullition was the major pathway for CH₄ emissions at both sites, contributing 60 % and 70 % (median value) or 60 % and 67 % (mean value) to the total CH₄ emission at the lake and pond, respectively (Fig. 6a). There was no significant difference of the ebullition ratio between the two systems across different seasons (Fig. 6a), but the ebullitive rate was different (Fig. 6b). The ebullitive rate demonstrated clear seasonal variation at both sites, with the highest ebullition flux observed between July and September. The monthly mean ebullition rate at the lake and the pond was in the range of 0.07 to 2.43 µg CH₄ m⁻² s⁻¹ and 0.24 to 9.04 µg CH₄ m⁻² s⁻¹), respectively. The annual mean ebullition rate at the pond (4.33 µg CH₄ m⁻² s⁻¹) was much higher than at the lake (1.17 µg CH₄ m⁻² s⁻¹).

3.4. Factors influencing CH₄ flux

Among various meteorological factors, CH_4 flux at both sites showed a significant and positive correlation with temperatures at half-hourly, daily, and monthly scale (Table S3). At the daily scale, the relationship was exponential (Fig. 7a), with sediment temperature having the highest *R* value (lake: 0.82; pond: 0.80), followed by water temperature (lake: 0.77; pond:0.77) and air temperature (lake: 0.74; pond: 0.69). In other words, variations in sediment temperature explained over 60 % of



Fig. 3. (a) Diurnal and (b) daily variation of environmental variables: T_w: water temperature, T_w: sediment temperature, WS: wind speed, R_n: net radiation.



Fig. 4. Diurnal variation in half-hourly CH_4 flux (Mean \pm Standard error) in four different seasons.

the flux variations. The CH₄ flux was more sensitive to temperature at the lake (Q_{10} : 5.75) than at the pond (Q_{10} : 2.29). Notably, there was a clear temperature hysteresis at both sites (Fig. 7a), with a higher daily flux value occurring during the cooling phase than during the warming phase of the year at the same temperature. The cooling phase occurred roughly between peak daily sediment temperature (roughly August 10th) and December 31st and warming phase between the January 1st to August 9th. The hysteresis was most evident when the temperature was between 15 and 25 °C (Table S4). The Q_{10} during the warming phase was 3.00 and 8.17 for the pond and the lake, respectively. The cooling phase exhibited lower Q_{10} values, specifically 1.95 for the pond and 3.25 for the lake. Similar trends were also captured at the monthly scale (Fig. 7b).

At our sites, the correlations between the seasonal mean CH₄ flux and NH-N, TP, DOC and DO concentrations were not significant, in part due to large scatter among a small sample size. In Fig. 8, we combine our seasonal data with data found in the published literature (Table S5) to explored the correlation between CH₄ flux and these water quality factors. A significant and positive relationship between CH₄ flux and NH⁺ 4.N and TN was found across the aquaculture systems (Fig. 8a–b). The correlation coefficient was 0.41 and 0.52 for NH⁺ 4.N and TN (p < 0.05), respectively. Although there was a trend of increasing CH₄ flux with higher DOC content, the correlation was not significant (r = 0.25, p = 0.20, Fig. 8c). Chl-a showed a robust positive correlation with CH₄ flux

(r = 0.61, p < 0.05, Fig. 8d).

4. Discussion

4.1. Importance of high-resolution CH₄ measurements

Understanding the temporal variability in CH_4 flux is crucial for developing effective sampling strategies and for gaining mechanistic understanding of CH_4 emission processes. This study covered a wide range of timescale, from the 10 Hz eddy timescale to diurnal and seasonal timescales. At the eddy timescale, the data carried the imprints of ebullition flux, allowing the total flux to be partitioned into ebullition and non-ebullition contributions.

At the diurnal scale, previous studies have reported large diurnal variations in CH_4 flux in unmanaged natural water bodies, but they disagree with regard to timing of the peak flux occurrence (Podgrajsek et al., 2014; Jammet et al., 2017; Zhang et al., 2019; Sieczko et al., 2020; Waldo et al., 2021; Hounshell et al., 2023). Using an automated chamber system, Sieczko et al. (2020) found that CH_4 flux at four dimictic lakes in the boreal and north temperate regions, with maximum water depths ranging from 4.5 m to 11 m, was, on average, 2.4 times higher during the day than at night. Other studies showed that the nighttime CH_4 flux exceeded the daytime flux by a factor of 1 to 3 in shallow ponds and shallow lakes (Podgrajsek et al., 2014; Jammet et al., 2017; Zhang et al.,



Fig. 5. (a-b) Temporal variations of CH₄ flux at three timescales: gray dots: half-hourly; black dots: daily; blue lines: monthly. (c) Probability density distribution of half-hourly CH₄ flux. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



Fig. 6. (a) Boxplot of half-hourly CH₄ ebullition ratio. The notch, the lower boundary, and the upper boundary correspond to the median, the first quartile (25th percentile), and the third quartile (75th percentile), respectively, and the whiskers extend to \pm 1.5 times the interquartile range. (b) Monthly mean CH₄ ebullitive rate, with error bars representing \pm one standard deviation.



Fig. 7. (a) Relationship between CH_4 flux (F_m) and sediment temperature (T_s) at the daily and (b) monthly scale. Lines represent regression equations given in Table S4.

2019). These variations underscore the risk of over- or underestimating CH₄ flux when relying solely on daytime measurements to temporal extrapolation. In comparison to these unmanaged systems, our continuous, high-resolution observations showed less pronounced diurnal patterns at the two aquaculture systems. Averaged over the year, the difference between the daytime flux and the nighttime flux was 4 % for the pond and -9 % for the lake. Given that most studies in aquaculture systems have predominantly concentrated their sampling between 09:00 to 12:00 (Kosten et al., 2020; Yang et al., 2024), our findings imply a potential 6 % to 8 % bias in the annual CH₄ emission estimate if only daytime data are used.

At multi-day to seasonal timescales, the total CH_4 flux and the ebullitive flux were characterized by peaks in the warm season and low values in the cold season at both sites. The seasonal pattern was consistent with those observed in other aquaculture systems (Tong et al., 2021; Yang et al., 2020; Yang et al., 2023; Yuan et al., 2021; Fang et al., 2022; Zhang et al., 2022a). In line with Wik et al. (2016), the diurnal variability in CH₄ flux at our sites was weaker than the day-to-day variability, with the daily flux varying by over 10 times in the same season. This significant day-to-day fluctuation, rather than diurnal changes, is likely the main source of large uncertainty in past CH_4 flux estimates, which often relied on sparse observations spread out over the year or short-term (a few weeks) campaigns. Our results show a notable

reduction in the standard deviation of the annual flux as sampling duration increases (Fig. S1). Here we conducted a bootstrap analysis to evaluate the effect of various sampling scenarios on the annual CH₄ emission estimates (Fig. 9). In our analysis, we define the "chance of high-accuracy flux estimates" as the fraction of estimates that fall within \pm 20 % of the observed annual mean. For example, for the annual mean value is 1.52 μ g CH₄ m⁻² s⁻¹, a high-accuracy flux estimate would be any value between 1.22 μ g CH₄ m⁻² s⁻¹ and 1.82 μ g CH₄ m⁻² s⁻¹. A 100 % probability indicates that all estimates based on a certain sampling scenarios fall within this range. Conversely, a 0 % probability would indicate that none of the estimates fall within this range. Drawing upon the insights from Wik et al. (2016), we propose a minimum sampling duration of 80 days for pond aquaculture and 120 days for lake aquaculture, evenly distributed throughout the year, to ensure accurate CH₄ emission estimates (Fig. 9).

4.2. Annual CH₄ emission

We show that the pond and the lake were large sources of atmospheric CH₄, with the annual flux of 120 g CH₄-C m⁻² yr⁻¹ and 36 g CH₄-C m⁻² yr⁻¹, respectively. These flux levels are three to eight times greater than the median annual fluxes from inland (unmanaged) waterbodies reported in a recent synthesis study utilizing EC



Fig. 8. Relationships between CH₄ flux and (a) NH⁺ 4.N concentration, (b) TN concentration, (c) DOC concentration, and (d) Chl-a concentration. Open symbols denote seasonal observed data at our sites, while closed symbols denote data in other published studies on aquacultural systems, representing the annual mean values for each system (Supplementary Table S5 and S6).



Fig. 9. Probability of achieving high-accuracy CH_4 flux estimates as a function of the number of sampling days for pond (a) and lake (b) site. Arrows indicate the sampling days required for a 95 % confidence level in estimating fluxes within \pm 20 % of the observed annual mean.

measurements (Knox et al., 2019). They exceed the annual emissions in other ecological zones of Lake Taihu (eutrophic zone: $1.35 \text{ g CH}_4\text{-C m}^{-2} \text{ yr}^{-1}$; submerged vegetation zone: $6.12 \text{ g CH}_4\text{-C m}^{-2} \text{ yr}^{-1}$; Xiao et al., 2014b; Zhang et al., 2019) by one to two orders of magnitude. According to the *meta*-analysis provided by Zhang et al. (2022d), Zhang et al.

(2024), the annual CH₄ flux from aquaculture pond is also substantially higher than those of other aquaculture systems regardless of size or waterbody/wetland types. Specifically, the CH₄ emissions from other aquaculture systems are 88 g CH₄-C m⁻² yr⁻¹ with size smaller than 1 ha and 27 g CH₄-C m⁻² yr⁻¹ with size larger than 1 ha; For waterbody/

wetland types used for aquaculture, the CH₄ emissions reported by these authors are 80 g CH₄-C m⁻² yr⁻¹ (ditches), 60 g CH₄-C m⁻² yr⁻¹ (paddy fields), 5 g CH₄-C m⁻² yr⁻¹ (reservoirs), and 3 g CH₄-C m⁻² yr⁻¹ (lakes). We also note that our pond and the lake values bracket those reported for ecosystems heavily impacted by human activities and using multi-years EC observation, such as small eutrophic reservoirs (42 g CH₄-C m⁻² yr⁻¹, Liu et al., 2023; 60 g CH₄-C m⁻² yr⁻¹, Zhang et al., 2024). The IPCC emission factor for fish aquaculture is 183 kg CH₄ ha⁻¹

The IPCC emission factor for fish aquaculture is 183 kg CH₄ ha⁻¹ yr⁻¹ or 13.7 g CH₄-C m⁻² yr⁻¹. However, our study revealed the CH₄ EF was not uniform across different aquaculture types within the same climate zone. Specifically, both sites showed EFs significantly higher than the IPCC default value, with the pond being 8.8 times higher and the lake site 2.6 times higher. This finding is consistent with the *meta*-analysis results, which show that different aquaculture types produce varying amounts of CH₄ (Yuan et al., 2019; Dong et al., 2022; Zhang et al., 2022d; Zhang et al., 2024). Additionally, we also observed large temporal variations in EF values at both sites, with differences ranging from 4.5 to 7 times between the aquaculture period (April to November) and non-aquaculture period (January to March and December). These results suggest that using a single EF for all aquaculture types and period in CH₄ budget accounting may be inappropriate.

4.3. Role of ebullition

The high CH₄ emissions in our aquaculture systems were largely linked to the high ebullition ratio. Previous studies have established the dominant role of CH₄ ebullition in the total CH₄ emission from inland aquatic ecosystems. Generally, the ebullitive flux accounts for over 50 % of the total emission (e.g., Bastviken et al., 2008; Sawakuchi et al., 2014; Wu et al., 2019). In organic-rich shallow zones, the ebullition ratio can be as high as 98 % (DelSontro et al., 2016; Yang et al., 2020). Our results, from the EC partitioning method, corroborates this, revealing that ebullition contributes 60 % and 70 % of the total CH₄ flux at the lake and the pond, respectively. These high ebullition ratios can be attributed to factors that favor bubble formation and the subsequent CH₄ emission, such as low hydrostatic pressure, shallow water depths, and abundant organic inputs (Zhu et al., 2016; Fang et al., 2022).

In natural ecosystems, the ebullition ratio is size-dependent, typically with smaller water bodies showing higher ratios than larger ones (Bastviken et al., 2004; DelSontro et al., 2016). This size-dependence behavior was absent in this study. The two systems differ in size by several order of magnitudes (pond area 0.6 ha versus lake area 10600 ha), but their ebullition ratios were comparable. The size independence is also suggested by previous studies of aquaculture systems, where aquaculture sizes varied widely, from 0.09 ha to 2.7 ha, and ebullition ratios were consistently high, ranging between 80 % and 98 % (Yang et al., 2020; Yuan et al., 2021; Tong et al., 2021; Fang et al., 2022). In addition to size-dependence, water depth is a key factor influencing ebullition, with shallower waters showing higher flux rates than deeper ones (Bastviken et al., 2004; DelSontro et al., 2016). The two systems had similar water depth (lake: 1.3 m; pond: 1.8 m), which may be an explanation for this comparable ebullition ratio between the two systems.

4.4. Potential causes of high pond CH₄ emission

Significant differences in CH_4 emissions were observed between the pond and the lake aquaculture systems, with the pond system showing a threefold higher annual flux rate than the lake system. This disparity is consistent with findings from a *meta*-analysis by Zhang et al. (2022d), which reported substantial higher CH_4 emissions from pond aquaculture compared to lake and reservoir aquaculture. One key factor contributing to these higher CH_4 emissions is elevated nutrient loading in ponds (Dong et al., 2022; Yang et al., 2019, 2022, 2024b; Zhang et al., 2022d). Previous studies have highlighted the roles of allochthonous nutrient inputs and in-situ high productivity in enhancing CH_4 emissions in aquatic ecosystems (Rasilo et al., 2015; Sepulveda et al., 2018; Beaulieu et al., 2019; Wang et al., 2021; Zhang et al., 2023a) and some pristine lakes (Borges et al., 2022). In our study, the primary difference between the lake and the pond systems was the severity of human activities, given their similar climatic conditions. We hypothesize that the greater feed supply to the pond system (annual input: 8570 kg C/ha) compared to the lake system (annual input: 6400 kg C/ha) provided more substrates for methanogenesis, therefore driving higher CH_4 emissions.

Additionally, concentrations of TN and TP, which are indicators of anthropogenic nutrient input, are known to correlate with increased CH₄ emission by accelerating in-situ CH₄ production (Tang et al., 2021; Wu et al., 2023). Our findings support this relationship, with the pond system showing a threefold increase in TN and TP concentrations across all seasons compared to the lake system. Moreover, the concentration of NH⁺ 4.N, another marker of nutrient enrichment, likely contributed to CH₄ production and slowed CH₄ oxidation due to preferential oxidation by methane monooxygenase (Holmes et al., 1995; Xiao et al., 2020; Zhou et al., 2020). The 13-fold higher NH⁺ 4.N concentration in the pond system compared to the lake system further supports the assertion that nutrient enrichment, primarily from feed and animal waste application, substantially enhanced CH₄ emission in the pond system. This is further corroborated by the significant and positive relationships between CH₄ flux and TN as well as NH⁺ 4.N concentrations revealed by a metaanalysis (Fig. 8).

High phytoplankton biomass and high primary productivity are additional factors likely driving the high emission from the pond system. Previous research has identified Chl-a, an indicator of phytoplankton biomass and productivity, as a strong predictor of CH₄ emissions (Bastviken et al., 2004; Rasilo et al., 2015; Beaulieu et al., 2019). Elevated Chl-a concentrations can supply newly produced organic matter, providing the essential precursors for CH₄ production (Huttunen et al., 2003; Schroll et al., 2023). In this study, the Chl-a concentration was much higher in the pond system (85.0 µg L⁻¹) than in the lake system (17.8 µg L⁻¹), and we observed a strong positive correlation between CH₄ flux and the Chl-a concentration in both systems (r = 0.61, p < 0.05), supporting the notion that high Chl-a conditions are conducive to increased CH₄ emissions.

Another potential explanation for the high CH₄ emission from the pond system was its small size. Our findings, in agreement with previous studies (Holgerson and Raymond, 2016; Grinham et al., 2018; Zhang et al., 2022d), demonstrate that the size of water bodies plays a critical role in determining CH₄ flux. Smaller bodies are typically associated with higher CH₄ emissions due to their higher perimeter-to-area ratios and shallower depths, which enhance the retention of allochthonous organic carbon and decrease the likelihood of methane oxidation (Holgerson and Raymond, 2016). While lake aquaculture is characterized by larger open waters and greater water circulation, the more enclosed and stagnant conditions of ponds foster the accumulation of DOC and nutrients, creating favorable environment for methanogenesis. Indeed, our measurements revealed much higher DOC concentrations in the pond system (15.46 mg/L) than in the lake system (3.37 mg/L; Fig. 2e). Although the relationship between the DOC concentration and CH₄ flux was not statistically significant, higher CH₄ fluxes were observed during periods of elevated DOC concentrations in both aquaculture systems, suggesting that the size of a waterbody may regulate CH₄ emissions. Additionally, the CH₄ emissions recorded in our study exceeded the predictions of the area-dependent scaling relationship proposed by Holgerson and Raymond (2016) by two to three orders of magnitude. This discrepancy may be attributed to the substantial organic inputs typical of aquaculture systems, which act as a potent stimulant for CH₄ production. These findings highlight the importance of considering not only the size of the water body but also the specific conditions of the system when estimating annual CH₄ emissions.

4.5. Sensitivity of CH_4 flux to temperature

The significant exponential relationships between CH₄ flux and temperature, observed across varying time scales at both the pond and the lake site, underscores the important role of temperature in regulating CH₄ emissions in these aquaculture systems. This pattern aligns with findings across a wide range of ecosystems (Yvon-Durocher et al., 2014; Aben et al., 2017; DelSontro et al., 2018; Jansen et al., 2020). However, unlike previous studies, our high-resolution in-situ measurements at both the pond and the lake aquaculture systems revealed the presence of a "hysteretic effect" - a phenomenon where the sensitivity of CH4 flux to temperature varies throughout the year (Davidson et al., 2018; Kariyapperuma et al., 2018; Jansen et al., 2020; Bao et al., 2021; Chang et al., 2020; Chang et al., 2021). Specifically, CH₄ flux was higher during the cooling phase of the year compared to the warming phase at the same temperature. This was likely due to increased substrate availability from unused feed material and phytoplankton accumulation during the cooling phase, which simulates methanogenesis (Chang et al., 2020; Chang et al., 2021).

Considering the strong temperature dependence of CH₄ flux, temperature is commonly used as a predictor for estimating annual CH₄ emissions in various ecosystems (Aben et al., 2017; Jansen et al., 2020). However, assuming a static relationship between temperature and CH₄ flux could lead to large uncertainties. For instance, during the warming phase, CH₄ flux could be overestimated by 14 % at the lake site and 20 % at the pond site, while during the cooling phase, underestimations could range between 17 % and 30 %. Such large seasonal biases highlight the importance of incorporating the seasonal hysteretic dynamics into predictive models to improve the accuracy of CH₄ emission estimates in diverse aquaculture systems.

This study also identified a notable difference in the temperature sensitivity (Q_{10}) of CH₄ emissions between the lake and the pond aquaculture systems. The lake system exhibited a high Q_{10} of 5.75, which exceeds the global wetland average Q_{10} of 2.23 based on EC measurements in 47 wetlands (Delwiche et al., 2021). In contrast, the pond system's Q_{10} of 2.29 closely aligned with the global average, indicating a more moderate response to temperature fluctuations. The difference in Q_{10} values between these two aquaculture systems is likely due to the more pronounced impact of nutrient enrichment on CH₄ emissions in lake systems. This nutrient enrichment can amplify the temperature's role in CH₄ production, as suggested by long-term mesocosm studies (Davidson et al., 2015). High temperature sensitives have also been observed in other aquatic ecosystems, including boreal lakes, northern ponds, urban ponds, rivers, and streams (Rasilo et al., 2015; DelSontro et al., 2016; Rocher-Ros et al., 2023; Bauduin et al., 2024).

These findings underscore the need for a comprehensive approach that accounts both biotic factors (e.g., nutrient loading) and abiotic factors (e.g, temperature) to accurately predict CH_4 emission in aquaculture systems. Future research should focus on the complex interactions between temperature, nutrient loading, and eutrophication, and how they collectively regulate CH_4 emissions across various aquaculture settings.

4.6. Implications of the present study

Several studies have explored variations in CH₄ emissions across different fish species, management practices, and measurement locations (Ma et al., 2018; Yang et al., 2019; Fang et al., 2022; Nijman et al., 2022; Zhang et al., 2022d; Znachor et al., 2023). However, less attention has been given to comparing different types of aquaculture system. Our study addresses this gap by comparing CH₄ flux between two major systems – lake-based and pond-based – in the YRD, China.

The substantial differences in CH_4 flux between the two systems underscore the need for type-specific investigations. Currently, a major transition from lake to pond aquaculture is under way, driven by local policy initiatives. Using the total lake aquaculture area (688,500 ha) in China in 2020 and the difference in the annual flux rate of 840 kg CH_4 -C/ha between the pond and the lake, our findings suggest that replacing all lake aquaculture systems with pond-based ones would increase CH_4 emission by 0.6 Tg CH_4 -C per year. This represents a 30 % rise from the current national freshwater aquaculture systems total of 1.9 Tg CH_4 -C per year (Zhang et al., 2024). Although this transition accounts for a relatively small percentage (1 %) of the country's total CH_4 emissions (49 Tg CH_4 -C yr^{-1} , Chen et al., 2022), it underscores the environmental trade-offs involved in different aquaculture system types and stresses the need for a balanced approach that integrates economic, ecological, and climate considerations.

As with many other studies, this work has some limitations. First, while our results corroborate previous research showing that aquaculture systems are large sources of atmospheric CH₄ (Yuan et al., 2019; Tong et al., 2021; Zhao et al., 2021; Yang et al., 2024), we also found that the transition from lake aquaculture to pond aquaculture could result in a threefold increase in CH₄ emission. This finding bridges a knowledge gap and provides empirical data that can help refine emission inventories. However, the influence of management practices (aeration, drainage, and dredging) requires further exploration.

Second, our study focused a one-year observation period. Previous studies have demonstrated considerable inter-annual variability in CH_4 emissions (Fang et al., 2022; Yang et al., 2024). Longer-term, multi-year observations are necessary for a more accurate comparison between the lake and the pond aquaculture systems.

Last, the field measurements in this study were limited to a specific region and climate, and specific fish species. Despite the high-frequency observations with identical instrumentation, the conclusions drawn from the comparison of one lake and one pond may lack sufficient generalizability to represent broader aquaculture systems. To improve the accuracy of CH_4 estimation at these larger scales, further comparative pond-lake studies should focus on diverse locations, climate, and aquaculture practices.

5. Conclusions

This study presents the first comparative analysis of CH₄ flux from lake and pond aquaculture systems, based on simultaneous one-year eddy covariance measurements. Results show minimal diurnal variability in CH₄ flux at both sites, but large day-to-day variability, underscoring the need for high-frequency observations to capture these fluctuations. Both pond and lake aquaculture farms were large CH₄ sources, with annual rates of 120 g CH₄-C m⁻² yr⁻¹ for the pond system and 36 g CH₄-C m⁻² yr⁻¹ for the lake system. The three-fold increase in CH₄ emission from the lake to the pond aquaculture highlights the importance of considering type-specific differences when assessing aquaculture-related CH₄ emissions. The higher CH₄ flux in the pond system was likely driven by a combination of biotic factors, such as high nutrient and carbon loadings and Chl-a concentration, and abiotic factors like the smaller surface area and stagnant conditions compared to the lake.

Our findings showed that sediment temperature was a key driver of CH_4 flux variability across multiple time scales (half-hourly, daily, and monthly) in both systems. Intriguingly, we observed an asymmetric response of CH_4 flux to sediment temperature at both sites: the flux was higher during the cooling phase compared to the warming phase at the same temperature. Additionally, the pond site exhibited a lower Q_{10} value than the lake site, possibly due to the dampening effect of high nutrient loading on temperature sensitivity.

CRediT authorship contribution statement

Jiayu Zhao: Writing – original draft, Funding acquisition, Formal analysis, Data curation, Conceptualization. Mi Zhang: Data curation. Yini Pu: Methodology, Data curation. Lei Jia: Data curation. Wei Xiao: Funding acquisition, Data curation. Zhen Zhang: Data curation. Pei Ge: Data curation. **Jie Shi:** Data curation. **Qitao Xiao:** Writing – review & editing, Funding acquisition, Data curation. **Xuhui Lee:** Writing – review & editing, Data curation, Conceptualization.

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.jhydrol.2025.132765.

Data availability

Data will be made available on request.

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