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Ebullitive CH_4 flux and its mitigation potential by aeration in freshwater aquaculture: Measurements and global data synthesis

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ABSTRACT

Freshwater aquaculture ponds constitute one of the important anthropogenic sources of atmospheric methane (CH_4). Nevertheless, estimates of global CH_4 emissions from freshwater aquaculture have large uncertainties due to a lack of data from different aquaculture types. Furthermore, despite that ebullition is a major pathway of CH_4 in aquatic systems, the quantification of ebullitive CH_4 fluxes from typical freshwater aquaculture ponds has been poorly represented. Here, field measurements of CH_4 fluxes over two years were taken to quantify ebullitive CH_4 fluxes from inland freshwater fish and crab aquaculture ponds in subtropical China. Ebullitive CH_4 fluxes averaged 15.97 ± 1.57 and 11.22 ± 1.26 mg m^{-2} d^{-1} in the fish and crab ponds in the first experimental year, respectively, and were 22.86 ± 2.30 and 21.95 ± 2.19 mg m^{-2} d^{-1} in the second year. During aquaculture period, ebullition dominated the emission pathways of CH_4, accounting for 83% and 98% of the total CH_4 emissions in the fish and crab ponds, respectively. Ebullitive CH_4 fluxes exhibited considerable spatial variations, with the lowest flux rates captured at the aeration area due to aerator-use in both the fish and crab ponds. Dissolved oxygen and dissolved organic carbon were the two primary factors that drove ebullitive CH_4 fluxes in both aquaculture ponds. By incorporating global measurement data, we further assessed the CH_4 mitigation potential of aerator use in freshwater aquaculture and revealed the dominant role of ebullition in this mitigation contribution. Together with the rice-based aquaculture, aerator use could reduce CH_4 emissions from freshwater aquaculture ponds globally by 71% and in China by 63%.

1. Introduction

Methane (CH_4) is a potent long-lived atmospheric greenhouse gas (GHG), accounting for 16–25% of global radiative forcing (IPCC, 2014; Etminan et al., 2016). Global averaged atmospheric CH_4 concentration is increasing steadily and has exceeded pre-industrial levels by about 150% and reached up to 1875 ppb in 2019 (NOA, 2020). Aquatic systems constitute one of the major sources of global CH_4 (Bastviken et al., 2011; Holgerson and Raymond, 2016; Natchimuthu et al., 2014; Raymond et al., 2013). A recent estimate projected that global CH_4 emissions from aquatic systems could contribute 53% of the global anthropogenic and natural emission sources (Rosentreter et al., 2021). Among aquatic systems, freshwater wetlands have been identified as the largest aquatic source of CH_4 (138–165 Tg yr^{-1}), representing 35–55% of the aquatic total (Rosentreter et al., 2021).

To meet the growing animal protein demand of the increasing world population, global aquaculture production has increased rapidly since the late 1950 s and has come up to 82.1 million tons in 2018 (FAO, 2020). Of which, over 60% (51.3 million tons) is produced from freshwater aquaculture. Furthermore, fed aquaculture has outpaced non-fed

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aquaculture, accounting for 69.5% of total aquaculture production in 2018 (FAO, 2020). With this rapid development of global industrial-scale aquaculture, freshwater aquaculture has become an increasing concern as an important source of CH_4 (Hu et al., 2012; Williams and Crutzen, 2010; Yuan et al., 2019). By compiling a worldwide database from freshwater aquaculture, for instance, Yuan et al. (2019) estimated global CH_4 emissions from freshwater aquaculture to be 6.04 Tg in 2014; however, uncertainties exist for this preliminary estimate due to an extremely limited data (Yuan et al., 2019).

In general, CH_4 fluxes from aquatic systems are determined by two processes, i.e., CH_4 production from sediment and the delivering of CH_4 from sediment to water surface through gas bubble ebullition, molecular diffusion, and/or plant stem-mediated transport (Bastviken et al., 2008; Laini et al., 2011; Tranvik et al., 2009). Diffusive CH_4 fluxes are determined by the water-air CH_4 concentration gradient and the piston velocity (Borges et al., 2018; Cole et al., 1998; Wu et al., 2019). Among these, ebullition has been considered as a dominant pathway of CH_4 emissions from aquatic systems, which has gained much attention in rivers (Wu et al., 2019; Sawakuchi et al., 2014), lakes (Attermeyer et al., 2016; Huttunen et al., 2001; Natchimuthu et al., 2016; Walter et al., 2018; Wik et al., 2013; Zhu et al., 2016), reservoirs (Beaulieu et al., 2018; Delsontro et al., 2010; Sturm et al., 2014), and marine aquaculture ponds (Yang et al., 2020). However, relatively few ebullitive CH_4 flux measurements have been taken from inland freshwater aquaculture ponds, while a recent study showed that CH_4 ebullition was up to 80% of total CH_4 emissions from inland freshwater crab aquaculture ponds (Yuan et al., 2020).

Freshwater aquaculture ponds receive abundant organic materials from residual aquaculture feeds, excrements, and remains of cultured animals, which greatly stimulate CH_4 production from aquaculture sediment by providing organic substrates to microbial communities (Yuan et al., 2019; Walter et al., 2008; Davidson et al., 2018). Different from other aquatic systems, freshwater fed aquaculture undergoes typical agricultural practices, such as the periodical aeration and materials feeding. These agricultural practices would incur high spatio-temporal variations in CH_4 emissions from different freshwater aquaculture types and from different functional stocking areas within an aquaculture pond, leading to the current estimate of CH_4 ebullition from inland freshwater aquaculture having a lot of uncertainty (Yang et al., 2020; Ma et al., 2018). As noted by Yuan et al. (2019), the preliminary estimate of global CH_4 emissions from freshwater aquaculture has uncertainties derived primarily from insufficient field measurements (only 34 measurements compiled from 17 publications) (Yuan et al., 2019). Therefore, field measurements of CH_4 fluxes from freshwater aquaculture by taking into account various management practices and covering complete flux components are highly needed. In addition, various controlling factors have been documented to influence diffusive CH_4 fluxes from inland freshwater aquaculture ponds, such as the temperature, water dissolved oxygen (DO) and dissolved organic carbon (DOC) (e.g., Liu et al., 2016). However, the factors key to driving ebullitive fluxes of CH_4 have been rarely explored in aquatic systems, especially in aquaculture systems, which thus needs to be incorporated in the upcoming studies with diverse aquaculture management patterns.

Here, in-situ measurements of CH_4 fluxes were taken in the two typical inland freshwater aquaculture ponds of fish and crab in southeast China over the period of 2017–2019. The main objectives of this study were to 1) quantify ebullitive CH_4 fluxes from inland freshwater aquaculture ponds; 2) assess the effect of aeration management on CH_4 emissions (especially for ebullition) by examining spatiotemporal variations in CH_4 fluxes from aerated, undisturbed and feeding areas within aquaculture ponds; and 3) generalize the mitigating potential of aerator use for CH_4 emissions from inland freshwater aquaculture ponds by synthesizing the global literature data on CH_4 fluxes from various freshwater aquaculture types with contrasting management practices.

2. Materials and methods

2.1. Study site

This study was conducted in the semi-intensive fish and crab aquaculture ponds, which are located at the experimental farm of Nanjing Agricultural University, Xinghua, Jiangsu province, China (32°52′N, 119°50′E) over the period of 2017–2019 (Fig. S1). The fish and crab ponds were converted from paddy fields in 2007 and they have experienced aquaculture production for ten years until the establishment of this field experiment. Each aquaculture pond was well-equipped with aeration and feeding devices. The experimental region is characterized by a subtropical monsoon climate, with an annual mean temperature of 17.8 °C and precipitation of 1090 mm during the two-year field experimental duration. Sediment of aquaculture ponds was classified as hydromorphic and the detailed water/sediment parameters were summarized in Tables S1.

2.2. Experimental design

Two-year parallel field experiments were conducted in neighboring crab (1.26 ha) and fish ponds (2.7 ha) from August 2017 to August 2019. To determine the spatiotemporal variations in CH_4 emissions from fish and crab ponds over the two-year experimental period, we collected gas and surface water/sediment soil samples simultaneously from three typical measurement sites consisting of the aerated stocking area (AA), undisturbed stocking area (UA) and feeding stocking area (FA). Gas flux measurements and water samples were collected once a week, and sediment soil samples were taken twice a month. To minimize the potential disturbance to aquaculture systems, boardwalks were fixed above the water surface at each measurement site of the fish and crab ponds. The management practices in the fish and crab ponds are detailed in Table S2. Briefly, the fish pond was dominated by cultivating the Carassius auratus gibelio mixed with Hypophthalmichthys molitrix, representing 80% and 20% of the annual total aquaculture stocking volume, respectively. The fish were fed four times per day at 08:00 a.m., 11:00 a.m., 14:00 p.m. and 17:00 p.m., and annual total input of C and N from fish compound diet amounted to 1015 kg C ha^{-1} and 325 kg N ha^{-1}, respectively. Water level was kept at 1–2 m all year round and no yearly drainage events have occurred in the fish pond. The crab pond was characterized by cultivating the Chinese mitten crab (Eriocheir sinensis) fed by crab compound diet, which was fed twice a day at 10:00 a.m. and 17:00 p.m. Annual total input of C and N from crab compound diet amounted to 640 kg C ha^{-1} and 205 kg N ha^{-1}, respectively. The water depth of the crab pond ranged from 0.7 m to 1.2 m and drainage events typically occurred from 29/Dec/2017–04/May/2018 and from 23/Nov/2018–6/May/2019 in the crab pond. Both fish and crab ponds were equipped with aeration and feeding devices. More information about the management options over the two-year aquaculture cycle was detailed in Table S2.

2.3. CH_4 flux measurements

The fluxes of CH_4 were measured using the floating chamber method during the flooding period and the static chamber method during the drainage period (Fang et al., 2021; Zou et al., 2005). Three sampling chambers and floating panels were placed as replication for CH_4 flux measurement at each sampling area of fish and crab ponds and gas samples were collected 0, 5, 10, 15 and 20 min after floating chamber closure. The deployment of chamber and floating panels, sampling procedures and further detailed information can be found in our previous studies (Liu et al., 2016; Wu et al., 2018; Fang et al., 2021).

Over the two-year experimental period, CH_4 fluxes were measured once a week except twice a week during the period of initial drainage or flooding and gas sampling was conducted at 08:00–10:00 local time to minimize diurnal variation in CH_4 fluxes for both fish and crab ponds;

this time was chosen because the soil and water temperature during this time is close to the daily average (Liu et al., 2016; Wu et al., 2018). The CH_4 concentration was analyzed using a modified GC (Agilent 7890 A) equipped with a flame ionization detector (FID) (Zou et al., 2005). The CH_4 flux was determined by a nonlinear fitting approach, and the averaged CH_4 fluxes and standard deviations were calculated from three replicated chambers at each sampling area (Liu et al., 2016). Annual total CH_4 emissions were sequentially accumulated from fluxes between every two adjacent intervals of measurements (Wu et al., 2019).

2.4. Diffusive and ebullitive flux components

Diffusive CH_4 fluxes across the water surface into the atmosphere were calculated by using the water-air gas transfer equation (Bastviken et al., 2011, 2004; Sawakuchi et al., 2014). During gas sampling, surface water samples (0–30 cm depth) were collected using PTE vials (100 ml), which were injected by Helium (He, 99.999%) gas to stop biological alteration of water samples to measure the dissolved CH_4 concentrations from three typical measurement sites both in the fish and crab ponds. The diffusive CH_4 flux (F) was calculated using the following equation (Eq):

$$F = k(C_m - C_e)1000 \quad (1)$$

Where C_m is the dissolved CH_4 concentration measured in the surface water (mol m^{-3}), C_e is the CH_4 concentration in surface water in equilibrium with the CH_4 partial pressure in the floating chamber, and k is the gas transfer velocity (m d^{-1}) (Cole et al., 1998).

However, in Eq. (1), the flux is partially driven by the change in concentration, which will decrease with time in the chambers as the internal concentration increases. Therefore, this simple calculation will underestimate the instantaneous flux rate. To reduce this error, we solved for k to estimate the instantaneous flux.

Then, according to the common gas law (PV = nRT), we used dP/dt instead of ($P_t - P_0$) to make the equation continuous. The total CH_4 fluxes in floating chambers were therefore determined by the Eq. 2:

$$F = \frac{dP}{dt} \times \frac{V}{ART} \times 1000 \quad (2)$$

Where dP/dt is the slope of CH_4 accumulation in the chamber (Pa d^{-1}) over the sampling period, V is the sampling chamber volume (m^3), R is the universal gas constant (8.314 m^3 Pa K^{-1} mol^{-1}), T is temperature (K).

The CH_4 concentration was calculated by Henrys Law ($C = K_h P$), where K_h is the Henry's Law constant for CH_4 (mol m^{-3} Pa^{-1}), and P is the partial pressure of CH_4 in the sampling chamber. By combining Eq. (1) and Eq. (2), k is calculated by the following equation:

$$k = \frac{dP}{dt} \times \frac{V}{K_h ART(P_m - P_0)} \quad (3)$$

Further, as Bastviken et al. (2004) described, we used the distribution and variance of the apparent piston velocities to determine which chambers captured ebullition. To compare the k values for any gas and temperature, the calculated apparent k values for each chamber were normalized to Schmidt numbers of 600 (k_{600} values) (Jahne et al., 1987; Wanninkhof, 1992) as follows:

$$k_{600} = k\left(\frac{600}{Sc}\right)^{-\frac{1}{2}} \quad (4)$$

Where Sc is the kinematic viscosity of water divided by diffusion coefficient of the gas.

Ebullition caused the calculated apparent k_{600} values to be significantly higher than chambers only receiving diffusive fluxes, which can help to identify chambers with diffusive CH_4 fluxes only. For each measurement, the minimum K_{600} could only be attributed to diffusive fluxes and the diffusive fluxes had a constant rate at the given time and area. Hence, the chamber only receiving diffusive fluxes had lower and similar flux rates at the same sampling site, which can be used to distinguish the ebullitive and diffusive CH_4 flux components. However, as noted by Sawakuchi et al. (2014), when all chambers display a large difference in flux rates at a given sampling site, it indicates that all chambers could have received ebullition. In this case, the minimum value similar to the diffusion at nearby sites was assumed as diffusion. We thus calculated the ratio of $k_{600\text{-}CH4}$ and minimum $k_{600\text{-}CH4}$ ($k_{600\text{-}CH4}$/minimum $k_{600\text{-}CH4}$) at each measurement site for each chamber to determine flux component sources.

In our study, the frequency distribution of this ratio ($k_{600\text{-}CH4}$/minimum $k_{600\text{-}CH4}$) for all chambers showed two distinct groups, i.e., one between 1.0 and 2.5 and another > 2.5 in both the fish and crab ponds (Fig. S2). Therefore, a ratio of 2.5 was defined as the threshold to indicate significant attribution of ebullition to total CH_4 fluxes in the fish and crab ponds. For the chambers receiving ebullition, by using the averaged k_{600} from the chambers only receiving diffusion and Eq. (1), we calculated the diffusive CH_4 fluxes and the remaining CH_4 fluxes from each chamber were attributed to ebullition.

2.5. Global data synthesis

To test the generality of our findings on the potential of aerator use to reduce CH_4 emissions from freshwater aquaculture ponds, we conducted a global synthesis of literature-sourced measurement data on CH_4 emissions from inland freshwater aquaculture. We searched the data (cut-off date on Feb 15, 2021) from the Web of Science using different combinations of keywords ('greenhouse gas' OR 'CH_4 'OR 'methane' AND 'aquaculture' OR 'fish' OR 'crab' OR 'shrimp' OR 'rice-fish' OR 'rice-crayfish' OR 'rice-crab' OR 'aquaponics'). Only field measurements of CH_4 fluxes from inland freshwater aquaculture were included, while indoor simulated or modeling data were not considered in this analysis. We extracted the original data on CH_4 fluxes and recorded the location, latitude and longitude, experimental period, and other key aquaculture parameters (Dataset S1). The final dataset was comprised of 66 field measurements from 27 publications related to CH_4 emissions (Dataset S1).

To estimate global CH_4 emissions from inland freshwater aquaculture wetlands and examine the potential of aerator use for mitigating CH_4 emissions, we classified aquaculture wetlands into rice-based aquaculture (including rice-fish, rice-crab and rice-crayfish) and mono-aquaculture systems (including fish, crab, shrimp and mixed). For mono-aquaculture, we divided these data into two subgroups, i.e., aquaculture ponds with and without aerator use. We estimated global CH_4 emissions from inland freshwater aquaculture by multiplying the mean CH_4 emission rates by aquaculture areas and differentiated the contributions from rice-based aquaculture and mono-aquaculture. To evaluate the mitigation potential of global CH_4 emissions due to aerator use from aquaculture, we assumed that aerator use is either not or fully adopted in mono-aquaculture ponds due to unavailable respective aquaculture area information. Similarly, we also estimated the provincial rates of CH_4 emissions and assessed the mitigation potential of aerator use to CH_4 emissions from inland freshwater aquaculture in China. Ultimately, we mapped the spatial distribution of CH_4 mitigation potential after aerator use in freshwater aquaculture ponds from global top 21 producers and China.

2.6. Other data measurements

Sediment soil samples (0–20 cm) were collected from two ponds before field experiment to measure initial pH, total organic carbon (TOC), total nitrogen (TN), dissolved organic carbon (DOC), extractable NH_4^+-N and NO_3^--N (Table S1). The sediment soil/water pH was measured at a volume ratio of 1:2.5 (soil: water) with a pH detector (PHS-3 C mv/pH detector, Shanghai, China). The sediment soil NH_4^+-N and NO_3^--N contents were measured by using a flow analyzer system (Auto Analyzer 3, German) after soil samples were extracted with 50 ml 2 M KCL Solution (1: 10, w/v). The sediment soil and water temperature was measured with mercury thermometers while gas sampling. The sediment dissolved organic carbon (DOC) was measured with ultraviolet-enhanced persulfate digestion and infrared detection (Phoenix8000, Teledyne Tekmar). The chemical oxygen demand (COD) concentration of surface water (0–30 cm) was determined by HACH reaction kits (Loveland, CO, Germany). Surface water dissolved oxygen (DO) concentration was monitored with a portable digital DO-meter (TY1055–12, America) during gas sampling. A LECO TruSpec CN analyzer (LECO Corp., St. Joseph, MI) was used to measure the total carbon (TC) and total nitrogen (TN) contents of aquaculture feed. The yields of aquaculture were weighed upon harvest.

2.7. Statistical analysis

Annual CH_4 emissions as affected by aquaculture type, experimental year, and their interaction were examined by a two-way analysis of variance (ANOVA). Spearman correlation analyses were used to examine the correlations between ebullitive CH_4 fluxes and water/sediment parameters. Linear stepwise regression models were used to fit ebullitive CH_4 fluxes with physicochemical parameters. The significance of the regression coefficients was tested by Student's t-test at the 0.05 probability level. All statistical analyses were performed using SPSS version 22.0 (SPSS Inc., USA) and all statistical plots were generated using Origin 2021 (OriginLab Corp. USA).

3. Results

3.1. Surface water and sediment parameters

Overall, water temperature and dissolved oxygen (DO) concentration shared similar seasonal patterns, which did not significantly differ among three functional areas (Fig. 1). Over the two-year observation period, water temperature averaged 19.4 °C and 20.4 °C in the fish and crab ponds, respectively. Seasonal patterns of water DO concentration showed a trade-off relationship with water temperature (Fig. 1a-b and d-e), with an annual average of 9.79 mg L^{-1} and 7.72 mg L^{-1} in the fish and crab ponds, respectively. Water DO concentration at the AA site was the highest among the three sampling sites, which averaged 9.64 mg L^{-1} and 8.01 mg L^{-1} in the fish and crab ponds, respectively.

However, seasonal patterns of sediment DOC contents showed large variations in both ponds over the two-year experiment period (Fig. 1c and f). In general, sediment DOC contents initially decreased steadily during the aquaculture period in the fish (Apr–Dec) and crab ponds (May–Nov). Thereafter, sediment DOC contents decreased dramatically and remained at relatively low levels following the aquaculture period. Moreover, over the two-year period, the annual mean of sediment DOC contents was the highest at the FA site, relative to at the other two sampling sites in both the fish and crab ponds.

3.2. Dissolved CH_4 concentrations and flux components

Seasonal variations of dissolved CH_4 concentrations and ebullitive CH_4 fluxes showed similar patterns (Fig. 2). Both dissolved concentrations and ebullitive fluxes of CH_4 were consistently greater in summer and autumn than in winter and spring seasons, with the highest rates in August and the lowest in February. Over the two-year period, the dissolved CH_4 concentrations averaged 0.11 μmol L^{-1}, ranging from 0.01 to 0.52 μmol L^{-1} in the fish pond and from 0.01 to 0.30 μmol L^{-1} in the crab pond. Among sampling sites, dissolved CH_4 concentrations were the greatest at the FA site, with an annual average of 0.19 μmol L^{-1} and 0.11 μmol L^{-1} in the fish and crab ponds, respectively.

Annual total CH_4 fluxes (diffusive plus ebullitive) differed with pond type, with an average of 24.26 mg m^{-2} d^{-1} in the fish pond, greater than that of 19.23 mg m^{-2} d^{-1} in the crab pond (Table 1). Over the two experimental years, annual CH_4 fluxes were significantly affected by aquaculture type and experimental year, but not their interaction (Fig. 3). Among three sampling sites in the two aquaculture ponds, annual ebullitive CH_4 emissions were the highest at the FA site, while they were lowest at the AA site (Fig. 3). Annual average diffusive and ebullitive CH_4 fluxes were significantly greater in the second than in the first year (Table 1). The seasonal average of ebullitive CH_4 fluxes ranged from 0 to 3.81 mg m^{-2} h^{-1} in the fish pond, compared to a range of 0–3.52 mg m^{-2} h^{-1} in the crab pond (Fig. 2). In the fish pond, diffusive

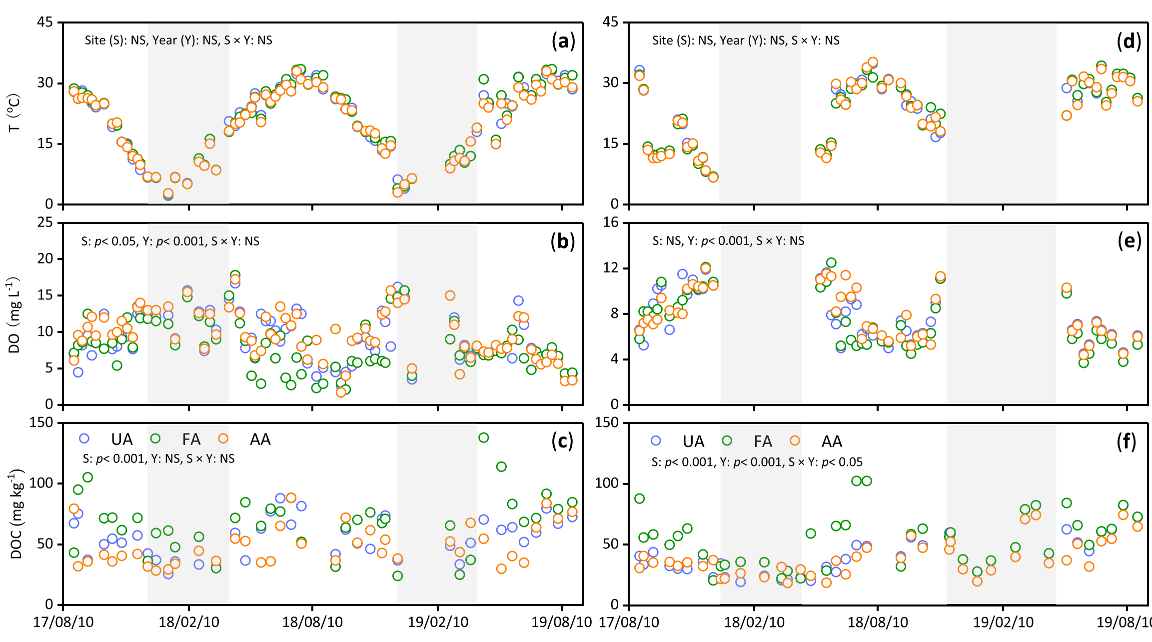

Fig. 1. Seasonal dynamics of water temperature (T), water dissolved oxygen (DO), sediment dissolved organic carbon (DOC) in fish pond (a–c) and crab pond (d–f). UA, undisturbed stocking area; FA, feeding stocking area; AA, aerated stocking area.

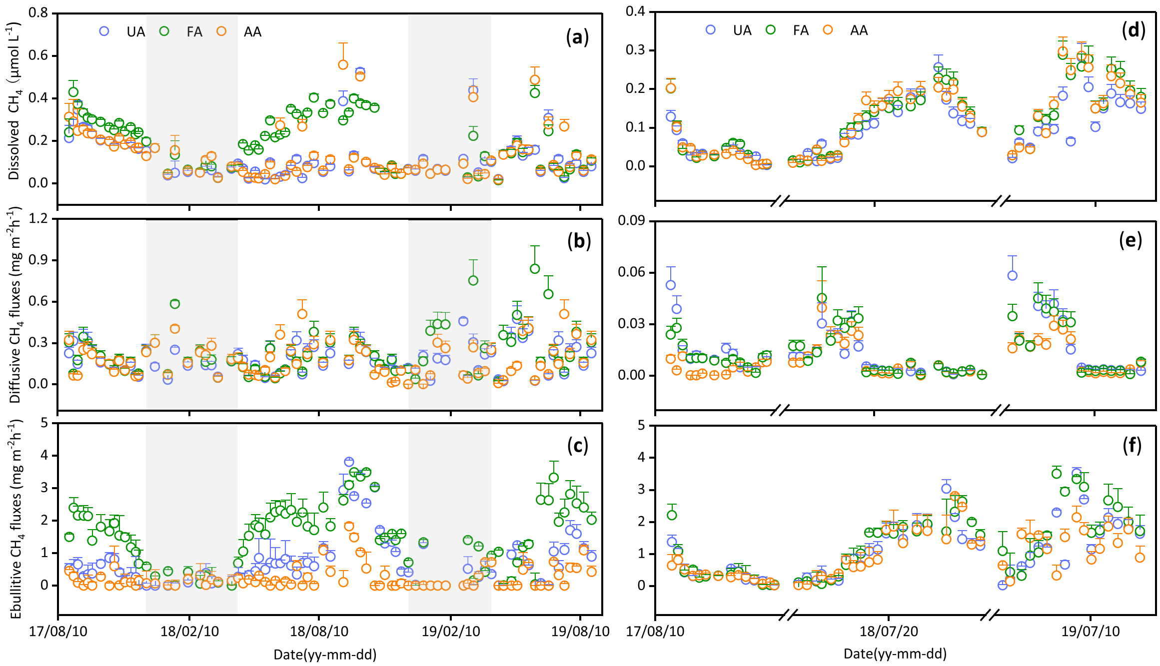

Fig. 2. Seasonal dynamics of dissolved CH_4 concentration, diffusive CH_4 fluxes and ebullitive CH_4 fluxes (mean ± SE) in fish pond (a–c) and crab pond (d–f). UA, undisturbed stocking area; FA, feeding stocking area; AA, aerated stocking area.

Table 1
CH_4 fluxes (Mean ± SE, mg m^{-2} d^{-1}), diffusive and ebullitive components during the aquaculture period and non-aquaculture period from 2017 to 2019 in the fish and crab ponds.

Aquaculture types		First year			Second year		
		aquaculture	non-aquaculture	average	aquaculture	non-aquaculture	average
Fish pond	Total fluxes	24.52 ± 1.94	8.26 ± 0.82	20.16 ± 1.57	35.40 ± 3.04	12.92 ± 1.86	28.66 ± 2.39
	Diffusive fluxes	4.12 ± 0.78	4.37 ± 0.82	4.19 ± 0.79	5.98 ± 0.86	5.36 ± 0.81	5.80 ± 0.83
	Ebullitive fluxes	20.40 ± 1.93	3.89 ± 0.74	15.97 ± 1.57	29.42 ± 2.92	7.56 ± 1.77	22.86 ± 2.30
	Ebullition (%)	83	47	79	83	59	80
Crab pond	Total fluxes	15.89 ± 1.52	4.42 ± 0.87	12.69 ± 1.21	39.83 ± 2.40	11.76 ± 1.48	26.7 ± 1.94
	Diffusive fluxes	0.33 ± 0.02	4.42 ± 0.87	1.47 ± 0.46	0.70 ± 0.05	11.76 ± 1.48	4.76 ± 0.92
	Ebullitive fluxes	15.56 ± 1.53	0	11.22 ± 1.26	39.13	0	21.95 ± 2.19
	Ebullition (%)	98	0	88	98	0	82

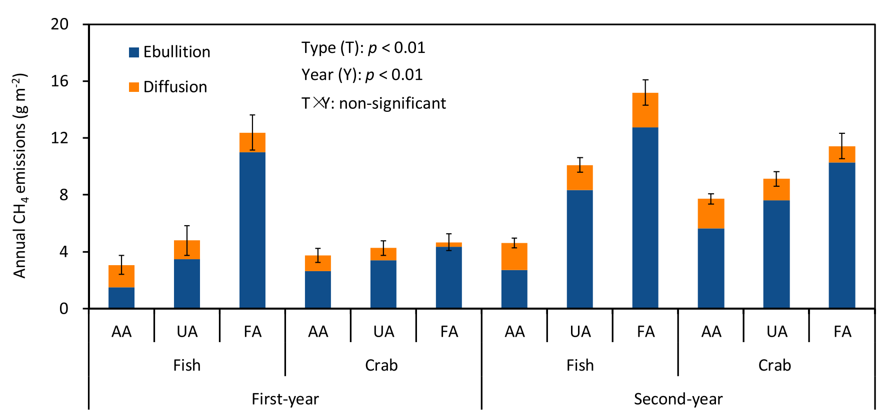

Fig. 3. Annual total of CH_4 emissions, diffusive and ebullitive components at three sampling sites in the fish and crab ponds over the two experimental years. Bars represent the mean ± SE (n = 3). The type and year represent the aquaculture type and experimental year, respectively. The statistical significance was examined at $p = 0.05$ possibility level.

CH_4 fluxes showed no significant difference between the aquaculture (April-November) and non-aquaculture periods (December-March) for each experiment year (Table 1). In contrast, annual ebullitive CH_4 fluxes averaged 20.40 mg m^{-2} d^{-1} and 29.42 mg m^{-2} d^{-1} during the aquaculture period, which were significantly higher than those of 3.89 mg m^{-2} d^{-1} and 7.56 mg m^{-2} d^{-1} during the non-aquaculture period in the first and second experimental years, respectively (Table 1). However, in the crab pond no ebullitive CH_4 fluxes occurred during the non-aquaculture period, while diffusive fluxes of CH_4 were greatly stimulated during this period due to the pulse release of sediment trapped CH_4 in both observation years (Table 1).

On average, ebullition contributed approximately 79% and 80% to the total CH_4 fluxes in the fish pond, comparable to the proportion of 88% and 82% in the crab pond over the first and second experimental years, respectively (Table 1). In the fish pond, almost 83% of CH_4 fluxes were attributed to ebullition during the aquaculture period when averaged across two years, while only 47% and 59% of CH_4 fluxes resulted from ebullition during the non-aquaculture period in the first and second experimental years, respectively (Table 1). Moreover, compared with the fish pond, ebullition contributed a larger proportion to the total flux rate in the crab pond, with an average of 98% during the aquaculture period across the two experiment years (Table 1).

In both aquaculture ponds, ebullitive CH_4 fluxes showed a large variation among three sampling sites, with the lowest rate at the AA site (Fig. 2). Over the two years, annual ebullitive CH_4 emissions at the AA site were 83% and 34% lower than those at the FA site in the fish and crab ponds, respectively (Fig. 3). Among the three sampling areas, ebullition contributed the lowest proportion at the AA site, with a fraction of 55% and 72% of total CH_4 emissions in the fish and crab ponds, respectively (Figs. 3 and 5). The largest contribution of ebullition was observed at the FA site, reaching up to 86% and 91% of total CH_4 emissions in the fish and crab ponds, respectively (Figs. 3 and 5). Relatively, ebullition contributed 79% and 82% to the total CH_4 emissions at the UA site in the fish and crab ponds, respectively (Figs. 3 and 5).

3.3. Relationships between ebullitive CH_4 fluxes and environmental factors

In general, ebullitive CH_4 fluxes were positively related to water temperature, water COD, and sediment DOC, while they were negatively dependent on water DO (Table S3). Nevertheless, these responses to physicochemical parameters varied across the different aquaculture stocking areas, such as the positive response to water COD was only observed at the UA and FA sites in both ponds, and the positive response to sediment DOC only occurring at the AA site in the fish pond.

According to regression analysis, the incorporation of water DO, water COD and sediment DOC together could explain 49% of the seasonal variation in ebullitive CH_4 fluxes from the fish pond (Table 2). Different from the fish pond, water DO and temperature were identified as two primary factors driving ebullitive CH_4 fluxes from the crab pond (Table 2) and can explain 63% of the variation in ebullitive CH_4 fluxes.

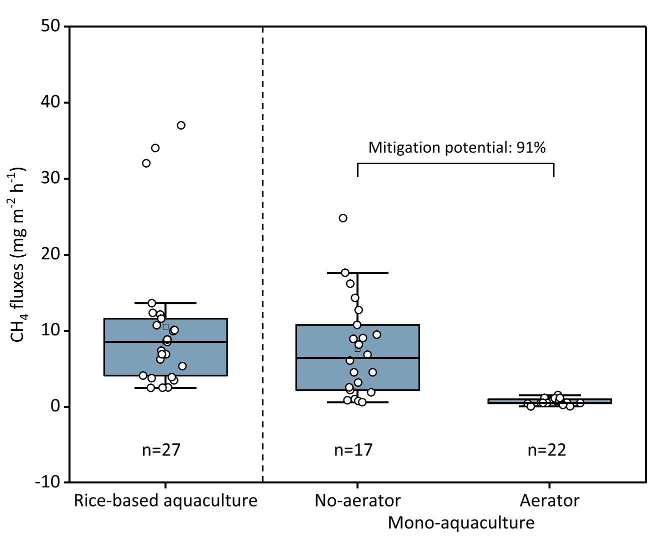

Fig. 4. Comparison of CH_4 fluxes between rice-based aquaculture and mono-aquaculture ponds with and without aerator use based on data synthesis. The square and black solid lines, lower and upper edges, and bars and hollow circle represent the mean and median values, 25th and 75th, 10th and 90th percentiles and distribution of data, respectively. The number of measurements (n) is shown next to the x-axis.

3.4. Global CH_4 budgets and mitigation potential

With the postulation of no aerator-use in all the mono-aquaculture ponds, the total CH_4 emissions from the global top 21 producers in 2014 were estimated to be 7.41 Tg CH_4, consisting of 1.67 Tg CH_4 from rice-based aquaculture and 5.74 Tg CH_4 from mono-aquaculture (Table S4). For mono-aquaculture, the globally synthesized data showed that the mean CH_4 fluxes from unaerated aquaculture were 7.57 mg m^{-2} h^{-1}, which was significantly greater than that from aerated aquaculture (0.65 mg m^{-2} h^{-1}) (Fig. 4). We further estimated the global CH_4 mitigation potential after the full use of aerator in all mono-aquaculture ponds, projecting that global CH_4 emissions could be reduced by 71% to 2.16 Tg CH_4 (Table S4). The spatial distribution of global CH_4 mitigation potential from inland freshwater aquaculture production due to aerator-use was mapped in Fig. 6a.

In China, the estimated CH_4 emissions from inland freshwater aquaculture were 5.01 Tg CH_4, with 0.98 Tg CH_4 from rice-based aquaculture and 4.03 Tg CH_4 from mono-aquaculture, which contributed 68% of global CH_4 emissions from freshwater aquaculture in 2014 (Table S4). Moreover, we further estimated the latest CH_4 emissions from inland freshwater aquaculture from China in 2019, generating an emission total of 4.92 Tg CH_4, with a contribution of 1.53 Tg CH_4 from rice-based aquaculture and 3.39 Tg CH_4 from mono-aquaculture (Table S5). Among provinces in China, Hubei (656 Gg CH_4) showed as the largest provincial emitter of CH_4, followed by Hunan (499 Gg CH_4), Anhui (488 Gg CH_4) and Jiangsu (407 Gg CH_4) (Table S5). The full use of aerator in mono-aquaculture has a potential to reduce CH_4 emissions by 63% to 1.82 Tg CH_4 in 2019 (Table S5).

4. Discussion

4.1. High contribution of CH_4 ebullition in freshwater aquaculture

Ebullition has been found to dominate the pathways of CH_4 emissions from aquatic systems (Wu et al., 2019; Sawakuchi et al., 2014). For instance, Sawakuchi et al. (2014) reported ebullition contributed over 50% of total CH_4 emissions from the Amazonian River and Wu et al. (2019) found that 44–56% of overall CH_4 emissions were derived from ebullition in rivers that were impacted by agriculture. Moreover, the contribution of ebullition to total CH_4 emissions from lakes has been found to be in the range from 36% to 80% (Natchimuthu et al., 2016). Compared with rivers and lakes, inland freshwater aquaculture ponds, characterized by relatively low hydrostatic pressure and shallow water depth, tend to benefit the formation of bubbles (Zhu et al., 2016; Casper et al., 2000; Delsontro et al., 2011). In addition, the bioturbation by aquaculture organisms can further facilitate the release of bubbles from sediment, which causes high contributions of ebullitive CH_4 fluxes to the total fluxes in aquaculture systems (Yang et al., 2020). Across the two experimental years, the rate of ebullition in our study from inland freshwater fish and crab ponds were generally greater than that in rivers or lakes. This is in line with results from marine aquaculture, as reported by Yang et al. (2020), where the contribution of ebullition was over 90% of total CH_4 emissions from shrimp ponds during the aquaculture period. Moreover, a recent study conducted at crab ponds in the Tai Lake basin, China reported a similar contribution of 81% for CH_4 ebullition to the emission total (Yuan et al., 2020). Thus, our findings confirmed the dominant role of ebullition in inland freshwater aquaculture characterized by different species and functional stocking areas.

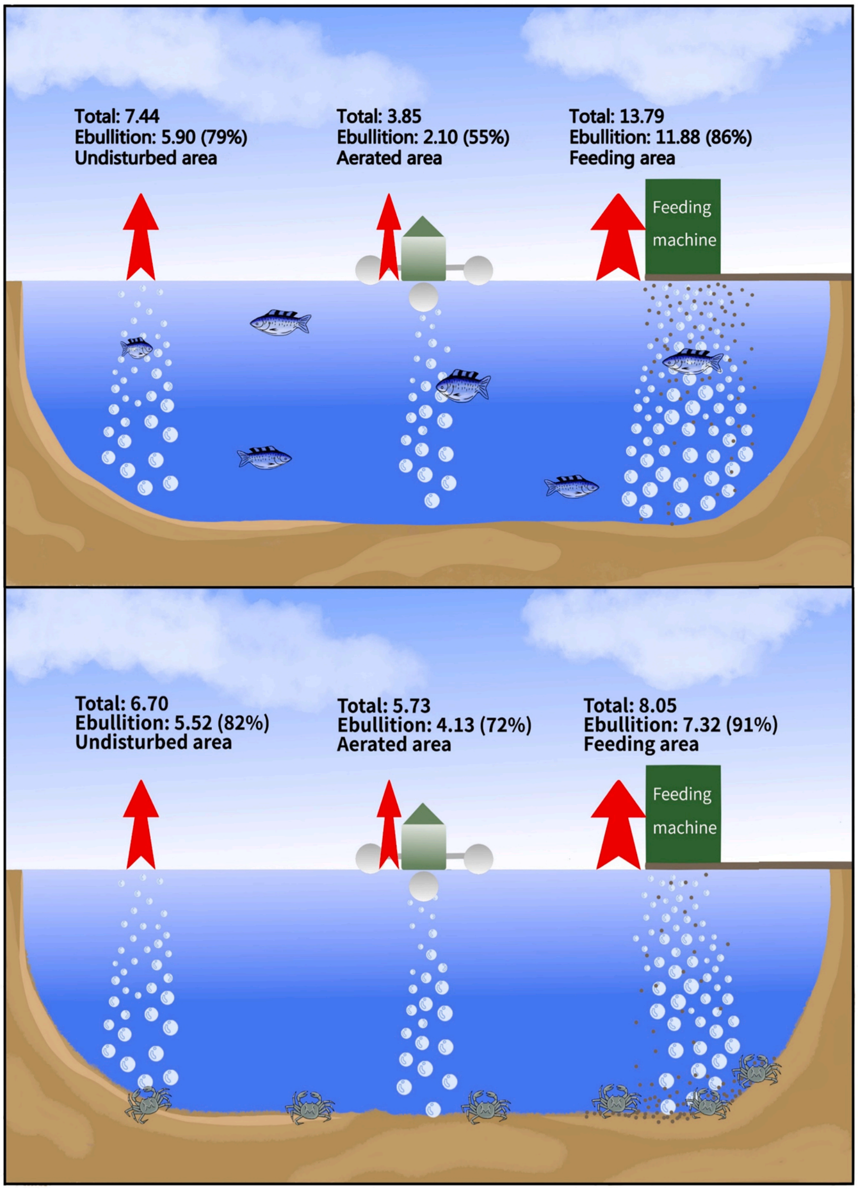

Fig. 5. Summarizing diagram of the spatial variations in annual total and ebullitive CH_4 emissions (g m^{-2}) and ebullition proportion within the aquaculture fish and crab ponds.

Table 2
Multiple linear regressions of ebullitive CH_4 fluxes against water temperature (T), dissolved oxygen (DO), chemical oxygen demand (COD) and sediment dissolved organic carbon (DOC) in fish and crab ponds.

Aquaculture types	Equation	n	r^2	*p*-value			
				T	DO	DOC	COD
Fish pond	Y = 0.012DOC + 0.008COD - 0.103DO + 0.705	102	0.49	$p = 0.443$	$p < 0.001$	$p < 0.001$	$p < 0.01$
Crab pond	Y = 0.044 T - 0.180DO + 1.397	60	0.63	$p < 0.05$	$p < 0.05$	$p = 0.082$	$p = 0.129$

4.2. Spatio-temporal variation in ebullitive CH_4 fluxes

Ebullitive CH_4 fluxes showed similar seasonal patterns in both the fish and crab ponds, independent of different sampling areas, with the highest rates observed in summer seasons and the lowest rates obtained in winter seasons, which is in line with previous studies in marine and inland freshwater aquaculture ponds (Yang et al., 2020; Yuan et al., 2020). Seasonal patterns of ebullitive CH_4 fluxes were generally consistent with the trends of water temperature and sediment DOC, while exhibiting a trade-off relationship with water DO in both aquaculture systems. Specifically, ebullitive CH_4 emissions primarily occurred during the aquaculture period, while only a small fraction of ebullitive CH_4 emissions was attributed to the non-aquaculture period (Table 1). Presumably, organic C input from aquaculture feeding materials during the aquaculture period increased sediment DOC availability as substrate for CH_4 production (Yuan et al., 2019). In addition, the disturbance of fish and crab routine activities during the aquaculture period might stimulate the bubble formation and the release of CH_4 from

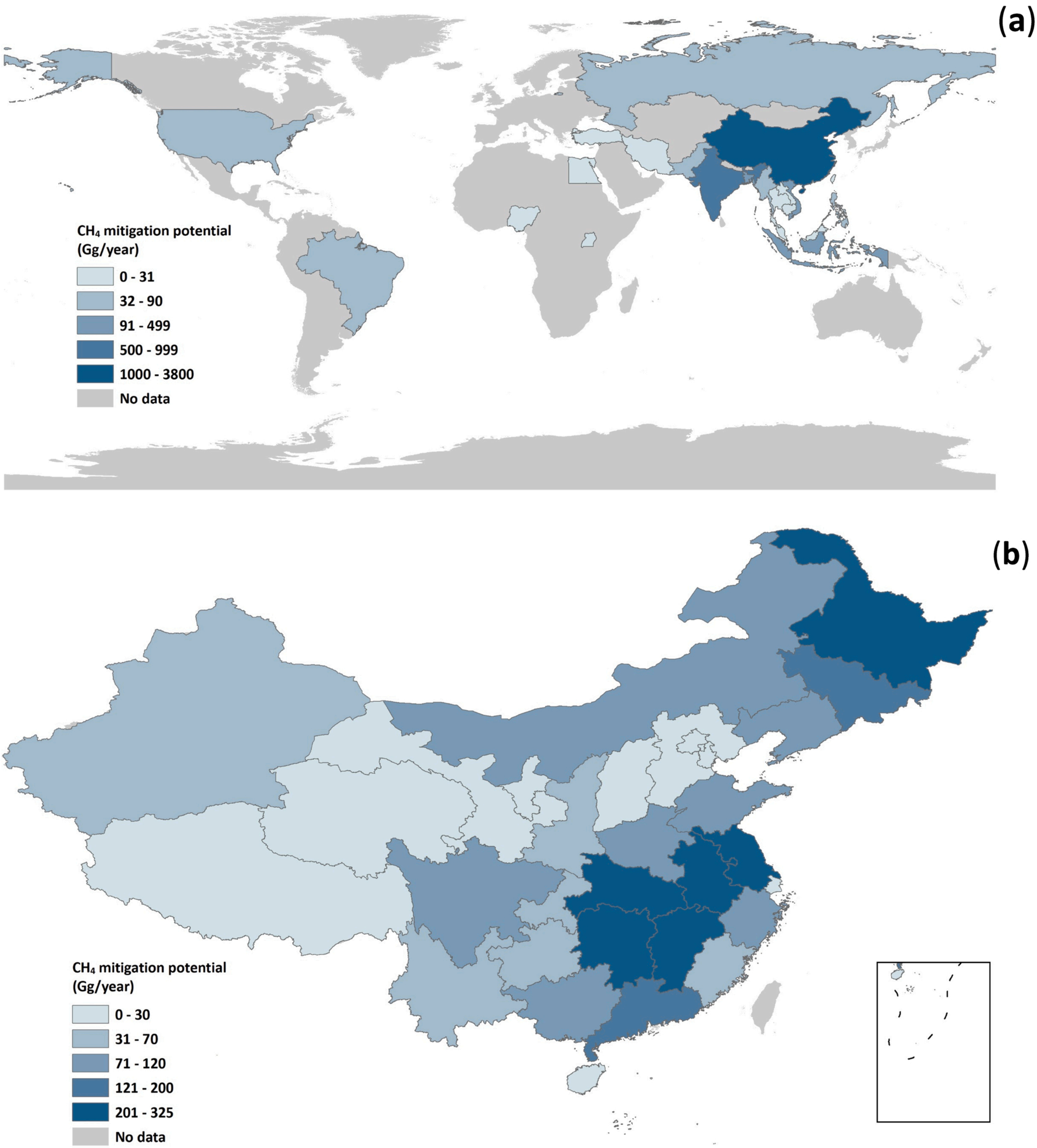

Fig. 6. Spatial distribution of CH_4 mitigation potential (Gg) from global top 21 producers (a) and China (b) after aerator use in inland freshwater aquaculture.

the sediment (Yang et al., 2020; Yuan et al., 2020). The relatively higher contributions of ebullition to total CH_4 emissions in the crab than fish ponds (Table 1) are most likely attributed to the differences in bioturbation by crabs versus fish (Yuan et al., 2020), i.e. benthic organisms, such as crabs highly disturb the sediment through routine activities, which can enhance the formation of bubbles and their release from sediment (Yang et al., 2020; Yuan et al., 2020).

Moreover, ebullitive CH_4 emissions showed obvious interannual variations, with significantly higher annual CH_4 emissions in the second than in the first experimental year for both the fish and crab ponds (Fig. 3). This difference was largely ascribed to the combined effect of higher water temperature, higher sediment DOC, and lower water DO in the second than in the first year in both aquaculture ponds (Fig. 1).

Among the three sampling areas, a higher ebullitive contribution of CH_4 at each functional stocking area was found in the crab pond than that at the corresponding area of the fish pond (Table 1). Consistently, the lowest contribution of ebullition was found at the AA site (55–72%) and the highest at the FA site (86–91%) in both aquaculture ponds. The contribution of ebullition at the UA site between two aquaculture ponds had no significant difference (fish 79% vs. crab 82%). As mentioned above, a stronger bioturbation to sediment from the crab than fish activities can lead to a larger contribution of CH_4 ebullition, and this effect seems to be amplified at the FA site in both ponds due to the gathering of aquaculture organisms. Our findings of the highest ebullitive CH_4 emissions at the FA site in freshwater aquaculture are in agreement with the results reported in a mariculture shrimp pond (Yang et al., 2020). However, the lower ebullitive contribution to CH_4 emissions at the AA site in both aquaculture ponds benefited primarily from the use of the aerator that inhibited CH_4 production and in turn its bubble formation for enhanced CH_4 oxidation in the water column due to relatively higher DO concentration (Yang et al., 2020). In addition, ebullitive CH_4 fluxes were found to negatively correlate with water DO in both aquaculture ponds (Table S3). Hence, given the higher rate of CH_4 ebullition in inland freshwater aquaculture than other aquatic ecosystems, a full understanding of its role in driving climate change should incorporate these spatio-temporal variations in ebullitive CH_4 fluxes across diverse aquaculture types and functional stocking areas.

4.3. Factors driving ebullitive CH_4 emissions

Generally, ebullitive CH_4 fluxes showed significant positive correlations with water temperature, water COD and sediment DOC, while having a negative correlation with water DO in both the fish and crab ponds. In addition, compared to the fish pond, a stronger dependence of ebullitive CH_4 fluxes on these parameters were found in the crab pond (Table S3). Presumably, the high water temperature could stimulate the formation of CH_4 bubbles in sediment and enhance their transport through the water column to the atmosphere (Frei et al., 2007; Stadmark and Leonardson, 2005). Similarly, ebullitive CH_4 fluxes were also positively related to water COD in the fish pond. As reported by others, high COD concentration in the aquaculture water column can lead to an accumulation of oxygen-consuming soluble microbial products, which would help to develop anaerobic aquatic environments that promote sediment CH_4 production (Hu et al., 2012; Liu et al., 2016; Jarusutthirak et al., 2006). Moreover, ebullitive CH_4 fluxes in the two aquaculture ponds were positively related to sediment DOC (Table S3), suggesting the dominant role of sediment DOC in regulating CH_4 ebullition in aquaculture ponds due to enriched C substrate from its decomposition (Liu et al., 2016; Singh et al., 2000). Furthermore, a strong negative dependence of ebullitive CH_4 fluxes on water DO was consistently observed in both aquaculture ponds (Table S3), similar to studies from lakes and reservoirs (Bastviken et al., 2002; Huttunen et al., 2006). High water DO concentration could enhance CH_4 oxidation in the water column of aquatic systems but simultaneously inhibit methanogenesis in the sediment (Schrier-Uijl et al., 2010). Linear stepwise regression models were used to further explore the key factors driving ebullitive CH_4 fluxes in both aquaculture ponds. We found differential combinations of physiochemical parameters for predicting ebullitive CH_4 fluxes from the two aquaculture ponds (Table 2).

4.4. Mitigation potential by aeration in freshwater aquaculture

Over the two-year period, we quantified the potential of aerator use for reducing CH_4 emissions in inland freshwater aquaculture ponds. We found that the decrease in CH_4 emissions due to aerator use was mainly realized via influencing the pathway of ebullition rather than diffusion. In addition, we highlighted that the aerator use may actually mitigate CH_4 emissions from the whole aquaculture pond, rather than only from the specific areas with aeration events. According to our literature survey, the full use of aerator in mono-aquaculture ponds could reduce global CH_4 emissions from mono-aquaculture pond down to 0.49 Tg yr^{-1}, corresponding to a mitigation potential up to 91% (Fig. 4 and Table S4). Presumably, the increase of water DO concentration in aquaculture ponds due to aerator use inhibited CH_4 production and then the bubble formation in sediment, whereas it simultaneously enhanced CH_4 oxidation in the water column (Yang et al., 2020; Fang et al., 2021). The estimated CH_4 emissions from the global top 21 producers in 2014 accounted for 2.2% of the total global anthropogenic CH_4 emissions and the full use of aerator in mono-aquaculture ponds could reduce CH_4 emissions by 71% globally. In China, the full use of aerator in mono-aquaculture ponds could reduce total CH_4 emissions by over 60% when averaging the mitigation potential estimated both in 2014 and 2019. According to the spatial distribution of global and China CH_4 mitigation potentials, high CH_4 mitigation potential was observed to mainly occur in China, especially in the East and Northeast of China (Fig. 6).

4.5. Limitations of this study and future implications

Limitations of course existed for this study. First, the field measurements of this study were limited to a typical location, climate and specific aquaculture species. Future studies should focus on CH_4 emissions from large-scale mixed aquaculture ponds. Second, our study highlighted the option of aerator use for mitigating CH_4 emissions in freshwater aquaculture ponds. However, more field experiments with detailed management information are needed from different countries and regions under various extents of oxygen supply status. Finally, due to the fact that the information of aerator use in most global aquaculture systems were missing or not available, the current mitigation potential in this study was assessed based on the postulation that no aerator was used in all the mono-aquaculture ponds, relative to the full use of aerator in these systems, which may overestimate the global CH_4 mitigation potential from aeration in freshwater aquaculture.

5. Conclusions

This study explored the contribution of ebullitive CH_4 fluxes to total CH_4 emissions from different functional areas in inland freshwater fish and crab ponds. Our results revealed that the contribution of ebullition to total CH_4 emissions in the crab pond (82–88%) was higher than that in the fish pond (79–80%) due to the relatively stronger bioturbation of crab to aquaculture sediment. The lowest contribution of ebullition was consistently found at the aerated aquaculture areas (55–72%). We found that this decrease in CH_4 emissions due to aerator use was mainly realized via influencing the pathway of ebullition rather than diffusion and the use of aerator could be an effective strategy for CH_4 mitigation from inland freshwater aquaculture systems worldwide.

CRediT authorship contribution statement

S.W.L., J.W.Z. and X.T.F. designed the investigation. X.T.F. conducted the experiment. X.T.F. and T.R.Z. extracted the data from literature and constructed the database. X.T.F., S.W. and S.W.L. performed the statistical analyses, created the figures and wrote the paper. Johan Six and Matti Barthel provided support and encouragement for this research. All authors contributed to improving and finalizing the manuscript.

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. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.agee.2022.108016.

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