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Accepted Version

Methane dynamics of aquaculture shrimp ponds in two subtropical estuaries, Southeast China: Dissolved concentration, net sediment release, and water oxidation

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Abstract Aquaculture ponds are important atmospheric CH_4 sources with strong temporal variability, yet the variation in CH_4 concentrations and other biogeochemical processes (e.g., CH_4 production and oxidation) among estuaries and aquaculture stages are poorly understood. In this study, we assessed the CH_4 sediment release, oxidation, and dissolved concentrations of aquaculture ponds in two subtropical estuaries in the different growth stages of shrimps. Overall, porewater CH_4 concentrations and sediment CH_4 release rates from the shrimp ponds varied greatly between different stages and followed the order: middle stage > final stage > initial stage. Water column CH_4 concentrations, and overlying water CH_4 oxidation rates indicated a growth pattern during the research period. There were also large variations in pond sediment CH_4 release rates, and dissolved concentration among estuaries. These variations can be mainly attributed to the interactions of salinity and other abiotic factors (e.g., pH and substrate availability). Furthermore, lower sediment CH_4 release rates, and high shrimp survival rate and yield were observed in Jiulong River Estuary with high water salinity compared to that in Min River Estuary with low water salinity. The results implies that increasing the salinity level of shrimp ponds and improving feed utilization efficiency might be an important strategy to mitigate CH_4 emissions and increase shrimp production. The result will also provide scientific data and support for preparation of wetland resources protection, the sustainable development of fishery economy, and CH_4 reduction in coastal zone. Our results emphasize the potential importance of the CH_4 biogeochemical cycle in aquaculture ponds and its contribution to the global CH_4 cycle. In the future work, the need to obtain high-frequency and continuous field measurements over the long term at multiple spatial scales, and to further identify the effect of microbial mechanisms on CH_4 biogeochemical cycle in coastal aquaculture ponds to reveal the causalities of spatial and temporal variations.

1. Introduction

Worldwide concern regarding global climate change and its effects on various environments requires a better understanding of greenhouse gas (GHG) emissions [*Tong et al.,* 2010]. As one of major final products in the process of organic matter degradation [*Bridgham et al.,* 2013], methane (CH_4) is an important GHGs in the air [*IPCC,* 2013]. At present, atmospheric CH_4 levels have increased threefold since pre-industrial times, reaching 1845±2 ppb in 2015 [*WMO,* 2016]. CH_4 has accounted for approximately 20% of global radiative forcing, while the atmospheric concentrations are still growing [*WMO,* 2016]. Quantifying the potential source of CH_4 from various system is an important basis for predicting future CH_4 emissions.

In terms of atmospheric equilibrium, CH_4 is usually over-saturated in inland and coastal aquatic system [*Bastviken et al.,* 2011*; Blees et al.,* 2015*; Dutta et al.,* 2015*; Xing et al.,* 2004*; Yang et al.,* 2015a]; thus, they are considered potentially significant sources of atmospheric CH_4 [*Bastviken et al.,* 2011*; Musenze et al.,* 2016*; Yang et al.,* 2011]. Recent estimates suggest that global freshwater CH_4 emissions to be 103 Tg CH_4 yr^{-1} [*Bastviken et al.,* 2011], accounting for nearly 25% of total CH_4 emission to the atmosphere [*Musenze et al.,* 2016]. Despite the significance, it is far from clear about various CH_4 sources [*Martinez-Cruz et al.,* 2017; *Natchimuthu et al.,* 2016] and CH_4 cycling in these aquatic system [*Martinez-Cruz et al.,* 2017]. This is particularly true in the case of CH_4 dynamics in aquaculture system.

Similar to other aquatic systems (e.g., lakes, reservoirs and rivers) and wetlands [*Bastviken et al.,* 2008*; Lai,* 2009*; Musenze et al.,* 2016], CH_4 fluxes from aquaculture system is controlled by the production of methanogens, consumption by methanotrophs, and transport via different pathways (e.g., diffusive and bubble), which are in turn, affected by a series of biotic and abiotic variables. In addition to this, a large proportion of produced CH_4 is dissolved in porewater *in situ* high pressure, with the consequence of marked CH_4 super-saturation. Dissolved CH_4 is released through diffusion from the sediment to the atmosphere, while the residual CH_4 undergoes ebullition as gas bubbles [*Dutta et al.,* 2013; *Neue et al.,* 1997]. Furthermore, the surface sediment generally retains a large amount of organic matter from aquatic animal feces and residual feed [*Chen et al.,* 2016*; Yang et al.,* 2017a], which will favor the production of CH_4. Therefore, the boundary between water and sediment, i.e., the sediment-water interface (SWI), plays a critical role in producing, transferring, circulating, and storing CH_4 in aquaculture ponds [*Xiong et al.,* 2017]. The rapid development of the aquaculture industry has prompted some scholars began to perceive the importance of studying CH_4 biogeochemical cycling in aquaculture systems. However, most attempts have been made in CH_4 exchange across the water-air interface and the influencing factors [e.g., *Chen et al.,* 2016*; Hu et al.,* 2014*; Hu et al.,* 2016*; Yang et al.,* 2015c*;Yang et al.,* 2018b]. However, detailed studies quantifying CH_4 fluxes and their correlative biogeochemical processes (e.g., sediment CH_4 release and CH_4 oxidation) in aquaculture system have received little attention until now. Carrying out such studies is quite significant for sustainable development and achieving a balance between carbon reduction and aquaculture development.

With the leveling off of capture fisheries production from the 1970s, the aquaculture industry has played an important role to satisfy the growing requirement of aquatic foods, for example fish and shellfish [*Hu et al.,* 2012]. Recent estimates suggest that global aquaculture

contributes to an increasing proportion of the global fish production from around 10% in 1985 to 40% in 2014 [*FAO,* 2016]. Approximately 90% of the world's aquaculture production occurs in the Asia-Pacific region [*FAO,* 2016], and land-based aquaculture ponds are considered the most important part of fish and shrimp production in marine and freshwater environment [*Yang et al.,* 2017a]. As a world leading aquaculture producer, China's total cultivation area and production were up to 58,579 km^2 [*Verdegem and Bosma,* 2009] and 29.4 million metric tons (Mt) in 2015 [*Fisheries Department of Agriculture Ministry of China,* 2015]. Shrimp pond is one of the major parts of China's aquaculture ponds, with wide distribution around the coastal regions [*Yang et al.,* 2017a]. In these ponds, water is generally influenced by the interactions of seawater and precipitation; hence, there are large variations in salinity between estuaries. Previous studies have indicated that salinity could affect the spatial-temporal variation of CH_4 production and emission through its effects on extracellular enzyme activities, carbon mineralization rates and methanogens activities in coastal wetland [e.g., *Hu et al.,* 2017*; Poffenbarger et al.,* 2011*; Sun et al.,* 2013*; Vizza et al.,* 2017*; Welti et al.,* 2017]. Therefore, salinity was considered as an factor influencing CH_4 dynamics in the current study. Moreover, previous studies have identified a number of factors controlling the spatial-temporal variation of CH_4 biogeochemical processes (e.g., CH_4 production, oxidation and emission) from natuarl wetland, rice fields and aquatic eosystems, including temperature [*Knox et al.,* 2016; *Olsson et al.,* 2015; *Palma-Silva et al.,* 2013; *Whalen,* 2005; *Xing et al.,* 2005], redox level or dissolved oxygen level [*Huttunen et al.,* 2003; *Liu et al.,* 2015], pH [*Wang et al.,* 1993; *Hu et al.,* 2017], salinity [*Poffenbarger et al.,* 2011; *Vizza et al.,* 2017; *Welti et al.,* 2017], water table [*Dinsmore et al.,* 2009; *Olsson et al.,* 2015; *Yang et al.,* 2013b], primary production [*Whiting and Chanton,* 1993; *Xiao et al.,* 2017], substrate availability [*Allen et al.,* 2007; *Venkiteswaran et al.,* 2013], and competitive electron acceptors [*Purdy et al.,* 2003; *Yang et al.,* 2019]. Furthermore, in an earlier study, *Yang et al.* [2018] found that CH_4 emissions from the aquaculture shrimp ponds into the atmosphere in the subtropical estuaries varied significantly among the different aquaculture stages, with the highest fluxes occurring during the middle stage. As a result of this, there could also be large variations in other CH_4 biogeochemical processes from aquaculture ponds between estuaries. Unfortunately, there is little available information that compares CH_4 dynamics and its influence factors in aquaculture ponds between estuaries [*Yang et al.,* 2018b].

To increase our understanding of the dynamics of CH_4 biogeochemical cycles in shrimp ponds and also improve the accuracy of flux estimates upscaled from local ponds to regional scales, this study made the attempt to research CH_4 dynamics (e.g., CH_4 dissolved concentrations, sediment CH_4 release, and overlying water CH_4 oxidation) in aquaculture shrimp ponds during the culture period in two subtropical estuaries in Fujian Province, Southeast China. There are two hypotheses in the current study: (1) the CH_4 biogeochemical cycles in the shrimp ponds varied greatly due to the remarkable change in environmental variables (e.g., DO, temperature, pH and substrate availability) between different culture stages; (2) CH_4 dissolved concentrations, sediment CH_4 release fluxes, and overlying water CH4 oxidation rates changed markedly due to the difference in salinity and substrate supply between two estuaries. This study could provide scientific foundation for improving GHG inventories and contribute to the ongoing evaluation by the IPCC Task Force on National Greenhouse Gas Inventories (TFI), particularly the urgent need of estimation of GHG

emission from various types of flooded land including aquaculture ponds [*Yang et al.*, 2017]. Moreover, this study can provide one major reference for a more accurate prediction, and effective regulation of GHG emission from the aquaculture ponds in the future.

2. Materials and Methods

2.1. Study Site Descriptions

Our study area was located in the Shanyutan and Humao in the Min River Estuary (MRE) and the Jiulong River Estuary (JRE), southeast China (Figure 1). Min River Estuary has a typically subtropical monsoon climate, warm and wet in summer. The mean annual temperature is around 19.6 °C and the average annual precipitation is about 1,350 mm [*Tong et al.*, 2010]. Jiulong River Estuary has a subtropical oceanic climate. The mean annual air temperature is approximately 21.0 °C and the average annual rainfall is around 1371 mm [*Wang et al.*, 2013]. The tides are typical semi-diurnal. The mean tide ranges are approximately 4.5 m and 4.0 m in these two estuaries. The catchment areas of the MRE and JRE are 60,092 and 14,741 km^2, respectively, with mean annual discharges of approximately 58.6×10^9 m^3 and 12.4×10^9 m^3 y^{-1} [*Zhou et al.*, 2016].

Three shrimp (*Litopenaeus vannamei*) ponds were randomly selected in each estuary. These ponds had been converted from the tidal marshes in recent years. The shrimp ponds in MRE and JRE were constructed in 2012 and 2006, respectively. Removing marsh vegetation, the shallow pond basins with steep sides and homogenous depth were created. The water in the shrimp ponds is a mix of fresh water drawn locally and sea water pumped from the coast. Aquaculture in most of the shrimp ponds starts in June and ends in November. *L. vannamei* were fed in both morning (07:00 a.m.) and afternoon (16:00 p.m.) with an artificial diet (Yuehai™, Guangzhou, China) which contained 42% of crude protein. The feed amount was estimated according to experience and shrimp response to previous feeding [*Casillas-Hernández et al.*, 2006; *Liu et al.*, 2015] and the feeding rates were controlled at around 10-16, 50-55, and 40-45 kg ha^{-1} d^{-1} from the initial stage, middle stage, and to the final stage. More details about the shrimp ponds and their management practices can be found in *Yang et al.* [2017a].

2.2. Collection and Analysis of Water Samples

To research shrimp production in different stages (initial, middle, and final stages), field sampling campaigns were performed in June, August, and October 2015. Sampling for the respective estuaries was done separately in each of the three sampling campaigns. Sample collection was performed in estuaries within one day between different estuaries. Three sampling sites were in each pond and water was sampled at three different depths: the surface (approximately 10 cm below the surface), the middle (between surface and bottom layer), and the bottom (approximately 5 cm above the surface sediment). On each sampling day, water was collected in morning (08:00 and 11:00), afternoon (14:00), and evening (17:00).

2.2.1. Dissolved carbon Concentrations in the Water Column

To measure the dissolved carbon concentrations, water samples were collected using a 5 L Niskin water sampler, and transferred to 250 mL polyethylene bottles. To inhibit microbial activity, about 2 mL of saturated $HgCl_2$ solution was injected into each bottle of water sample [*Taipale and Sonninen*, 2009; *Zhang et al.*, 2013]. All water samples were subsequently stored in an ice box, transported to the laboratory within 4 h, and analyzed within one week.

After filtering through a 0.45 μm cellulose acetate filter (Biotrans™ nylon membranes), the samples were analyzed for the levels of dissolved inorganic carbon (DIC), dissolved organic carbon (DOC), and total organic carbon (TOC) and using a SHIMADZU TOC-$V_{CPH/CPN}$ analyzer (Shimadzu, Kyoto, Japan).

2.2.2 Physicochemical and Biological Variables of Water Column

Water temperature and pH were measured *in situ* using a portable pH/mV/Temp system (IQ150, IQ Scientific Instruments, USA). Salinity and dissolved oxygen (DO) concentrations at different water depths were determined using a salinity meter (Eutech Instruments-Salt6, USA) and a multi-parameter controller (HORIBA, Japan), respectively. To estimate chlorophyll-*a* (Chl-*a*) concentrations, samples were also collected in 250 mL polyethylene brown bottles and stored in dark and cold environment during transporting back to laboratory. The Chl-*a* samples were extracted using 90% (V/V) acetone for 24 h and determined using a UV-visible spectrophotometer (Shimadzu UV-2450, Japan). The Chl-*a* concentration was calculated based on measurements of absorbance at 665 and 750 nm [*Qu et al.,* 2007]. In addition, meteorological data including wind speed, air temperature, and atmospheric pressure were measured *in situ* using a meteorological instrument (NK3500 Kestrel, USA).

2.2.3. Dissolved CH_4 Concentration in Water Column

A headspace equilibration technique was used to determine dissolved CH_4 concentration. Water samples were collected using a 60 mL pre-weighed serum glass bottle, completely filling them with an air-tight water sampler that limits gas exchange and prevents formation of gas bubbles [*Abril et al.,* 2007*; Cotovicz et al.,* 2016]. To inhibit microbial activity, the bottles were sealed and 0.2 mL of saturated $HgCl_2$ solution was injected into each water sample [*Cotovicz et al.,* 2016; *Dutta et al.,* 2015]. All water samples were stored in ice cooler, transported back to the laboratory, and analyzed within three days of collection. In the laboratory, a headspace was generated. A total of 30 mL ultra-pure nitrogen (N_2) was injected through the top septum of the sampling bottles, a needle was penetrated into the bottom septum, allowing the discharge of an equal volume of water [Wang et al., 2015]. The sampling bottles were then shaken vigorously for 30 min on a mechanical shaker to equilibrate CH_4 between the air and water phase. CH_4 concentrations in the headspace were measured using a gas chromatograph (GC-2010, Shimadzu, Kyoto, Japan) equipped with a flame ionization detector (FID). The levels of CH_4 in the shrimp pond water samples were estimated based on the solubility coefficients [*Wanninkhof,* 1992; *Yamamoto et al.,* 1976]. The detailed calculation process of CH_4 concentration and saturation followed the description of *Wang et al.* [2015].

2.3. Collection and Analysis of Sediment Samples

On each sampling campaign, twelve intact short sediment cores were sampled in each pond using a surface-operated coring device (Core-60, Austria). The device includes a Plexiglas tube, a core cylinder and a one-way check valve to preserve the integrity of overlying water and sediment [*Han et al.,* 2014]. The intact cores included 15 cm sediments and 15 cm overlying water. Sediment samples were immediately sealed and stored vertically in a 4 °C cooler and transported back to the laboratory within 4 h. In the laboratory, the core of sediment samples was divided into four parts.

2.3.1. Sediment Physicochemical Parameters

Sediment pH was measured using a pH meter (Orion 868, USA) with a soil-to-water ratio of

1:2.5, and sediment salinity was determined using a salinity meter (Eutech Instruments-Salt6, USA) with a soil-to-water ratio of 1:5 [*Yang et al.*, 2017a]. Sediment porosity (Φ) was determined according to the water content of sediment samples which was calculated by the sediment weight difference before and after baking in an oven at 105 °C for 24 h [*Yang et al.*, 2017a; *Zhang et al.*, 2013]. Sediment total carbon (TC) was analyzed via a CHN Elemental Analyser (Elementar Vario MAX CN, Germany) on freeze-dried and ground subsamples passing through a 2 mm sieve [*Sun et al.*, 2013].

2.3.2. CH_4 Concentration and Physicochemical Parameters of Sediment Porewater

Triplicate sediment samples from each pond were measured for porewater CH_4 concentration and physicochemical properties. The collection method of dissolved CH_4 concentration in porewater followed the description of *Dutta et al.* [2015]. Briefly, duplicate 6 cm^3 subsamples were collected using 10 plastic syringes with the needle attachment end removed. The subsample plugs from the syringes were immediately extruded into 60 mL glass serum vials and sealed with blue butyl stoppers and aluminum crimp caps. Approximately 24 mL of degassed deionized water and 0.5 mL of saturated $HgCl_2$ solution was then added to serum vials. To inhibit oxidation, porewater samples were processed in nitrogen-filled glove bags [*Matos et al.*, 2016]. To obtain an equilibrium between the slurry and the headspace, the mixtures were shaken on a mechanical shaker [*Dutta et al.*, 2015]. CH_4 concentration in the headspace was measured using a gas chromatography (GC-2010, Shimadzu, Kyoto, Japan). The calculation formula of porewater CH_4 concentration followed the description of *Ding et al.* [2005].

Porewater was extracted from the rest of the sediment by centrifugation (4,000 rpm, 10 min Hereaus Omnifuge 2000 RS). These porewater samples were divided into two groups. A group of porewater samples were filtered through cellulose acetate filters with pore size of 0.45 μm (Biotrans™ nylon membranes) [*De Vittor et al.*, 2012], and the filtrates were analyzed for the concentrations of TOC, DOC, and DIC via a SHIMADZU TOC-VCPH/CPN analyzer. Another group of porewater samples (unfiltered) was used to determine the salinity via a salinity meter (Eutech Instruments-Salt6, USA).

2.3.3. Incubation and Measurement of Sediment CH_4 Release Fluxes

The final set of three sediment cores were used for incubation and measurement of CH_4 relese fluxes across the sediment-water interface (SWI). The incubation device (Figure S1) was structured following *Chen et al.* [2014a] and *Cowan et al.* [1996]. With intact structures, approximately 15-cm-long sediment cores were placed in Plexiglas® incubation chambers (diameter 6 cm, height 30 cm) (Figure S1) which were sealed with a Teflon plunger to prevent exposure to the air. The chambers were then completely filled with overlying water up to 15 cm above the sediment surface. Before the incubation, DO levels of the overlying water in the chambers were adjusted to achieve the level of DO *in situ*. Finally, the incubation chambers were incubated in a constant temperature oscillation incubator (QHZ-98A, China) device for 9 h. The incubation temperature was consistent with *in situ* temperature. During the initial time (0 h) of incubation, 60 mL of overlying water from the chambers were withdrawn using a 100 mL airtight polypropylene syringe connected to rubber tubing, and the water was immediately transferred to pre-capped 60 mL infusion vials. Subsequent samples were collected from the chambers after 3, 6, and 9 h of the incubation. To inhibit microbial activity, approximately 0.5 mL of saturated $HgCl_2$ solution was injected to each sample vial.

Headspace equilibrium technique was used to measure dissolved CH_4 concentration in incubated water samples. Water samples were processed and measured following the same procedure as Section 2.2.2. CH_4 release fluxes across the SWI were determined according to the concentration variations in overlying water versus time [*Wang et al.*, 2015] (Eq. 1):

$$F_{\text{S-W}} = \frac{dc}{dt} \times V \times S \qquad (1)$$

where $F_{\text{S-W}}$ is the flux of CH_4 (μmol m^{-2} h^{-1}) at the sediment-water interface; $\frac{dc}{dt}$ is variation slope of CH_4 concentration over sampling time (μmol L^{-1} h^{-1}); V is the volume of overlying water in the incubation chamber (L); and S is the cross-sectional area of the sediment core (m^2).

2.4. Incubation and Measurement of Dissolved CH_4 Oxidation in the Overlying Water

On each aquaculture stage, CH_4 oxidation (MOX) was also determined as the decrease of CH_4 concentration over time in Plexiglas® incubation chambers [*Almeida et al.*, 2016; *Bastviken et al.*, 2008; *Dutta et al.*, 2015; Figure S1]. The 588.75 mL incubation chambers (n = 3 for each pond) were completely filled with overlying water samples from the 5 L Niskin bottles using transparent silicon tubes, overflowing each chamber to prevent any air spaces and/or bubbles formation. Finally, the incubation time, conditions, water sampling, and measurement of CH_4 oxidation in incubation chambers were performed as described in Section 2.3.3. CH_4 oxidation rates (μmol CH_4 m^{-2} h^{-1}) in the incubation chambers were calculated *via* regressions of concentration and time (Eq. 1; *Wang et al.*, 2015).

2.5. Statistical Analyses

Data were transformed to normal distributions when the selected attributes were skewed. Differences in the researched variables among three growth stages of shrimps in each estuary were calculated using one-way Analysis of Variance (ANOVA). For a given estuary and season, the differences in environmental variables, CH_4 concentration, and CH_4 saturation among the three water depths were also tested using ANOVA. This research tested the statistical differences in studied variables between the two estuaries' shrimp ponds during the study period by repeated measures analysis of variance (RMANOVA). Correlations between biogeochemical dynamics of CH_4 and individual environmental variables were tested by Pearson correlation analysis. All statistical analyses were conducted using SPSS version 17.0 (SPSS, Inc., USA). Significance was considered at the level of 0.05. Results are reported as mean ± 1 SE. Graphs were generated using OriginPro 7.5 (OriginLab Corporation, USA). Conceptual diagrams were plotted using EDraw Max version 7.3 (EdrawSoft, Hong Kong, China).

3. Results

3.1. Dissolved CH_4 Concentration in Sediment Porewater and Water Column

There were large variations in pond sediment porewater CH_4 concentration between estuaries and among observation stages (Figure 2). Porewater CH_4 concentration from the shrimp pond sediment in the MRE and JRE changed between 13.47 and 95.27 μmol L^{-1}, and 2.29 and 29.52 μmol L^{-1}, respectively, with the minimum and maximum in the initial and middle stages (Figure 2), respectively. Overall, average porewater CH_4 concentration from sediment in the MRE (51.51±4.41 μmol L^{-1}) was significantly larger than that in the JRE (11.50±1.56

μmol L^{-1}) ($F_{df=1}$ = 15.717, p<0.01; Figure 2).

The CH_4 concentrations in the pond water column from the MRE and JRE changed between 0.18±0.01 μmol L^{-1} and 1.20±0.11 μmol L^{-1}, and between 0.15±0.01 μmol L^{-1} and 0.62±0.03 μmol L^{-1}, respectively (Figure 3), corresponding to the saturation varied between (226.9±10.0) % and (1525.3±132.8) %, and between (207.7±9.9) % and (974.42±46.37) %. CH_4 concentrations and saturations from all estuaries showed significant (p<0.01) temporal variation in the order of: final stage > middle stage > initial stage (Figure 3). Average CH_4 concentrations and saturations in the pond water column in the MRE were significantly higher (p<0.05) than those in the JRE during the initial and final stages, but only slightly higher (p>0.05) in the middle stage (Figure 3). The mean values of CH_4 concentration and saturation grew gradually from the surface to bottom waters, but in most cases the differences were insignificant (p>0.05) (Figure 3).

3.2. CH_4 Release Fluxes Across the Sediemnt-Water Interface

The dynamics of sediment CH_4 release fluxes from the shrimp ponds in the MRE and JRE are shown in Figure 4a. The two estuaries showed similar temporal patterns, with minimum and maximum release fluxes occurring at the initial and middle stages, respectively (Figure 4a). The CH_4 release fluxes over the study period ranged from between 5.26 to 168.81 μmol m^{-2} h^{-1}, and from 2.93 to 56.74 μmol m^{-2} h^{-1}, respectively. Overall, the mean CH_4 release fluxes from the shrimp ponds in the MRE amounted to 84.52±11.58 μmol m^{-2} h^{-1}, which was higher than that in the JRE, at 11.27±2.43 μmol m^{-2} h^{-1} ($F_{df=1}$ = 59.576, p<0.01).

3.3. Oxidation Rates of CH_4 in the Overlying Water

CH_4 oxidation rate dynamics in the overlying water of the shrimp ponds are shown in Figure 4b. In the MRE, CH_4 oxidation rate varied significantly with aquaculture stages (p<0.05) and followed the order: final stage (3.07±0.41 μmol m^{-2} h^{-1}) > middle stage (1.99±0.11 μmol m^{-2} h^{-1}) > initial stage (1.27±0.17 μmol m^{-2} h^{-1}) (Figure 4b). The oxidation rates of CH_4 in the JRE also followed the same trend; however, the oxidation rates among different aquaculture stages did not differ significantly (p<0.05; Figure 4b). Overall, the mean CH_4 oxidation rate from the shrimp ponds were higher in the MRE than those in the JRE (2.23±0.22 *vs* 1.97±0.23 μmol m^{-2} h^{-1}), but the difference was not significant ($F_{df=1}$ = 0.214, p=0.413).

3.4. Physicochemical Properties of Sediment and Water

Environmental parameters of the shrimp ponds in the studied estuaries are shown in Table S1 and Figure S2. Sediment temperature was highly variable among aquaculture stages, with significantly higher temperature occurring at the middle stages (p<0.05) (Table S1 and Figure S2). Sediment TC, porewater concentration of TOC, DOC and DIC, and the concentration of DO and Chl-*a* in the water column also followed a similar trend (Table S1 and Figure S2). Salinity in the porewater and water column changed significantly (p<0.05) in different aquaculture stages and followed the order: initial stage > middle stage > final stage (Table S1 and Figure S2).

Overall, the means of sediment TC, pH, DO concentration, Chl-*a* concentration, atmospheric pressure, and porewater TOC and DOC concentration in the MRE were significantly larger than those in the JRE (p<0.05) (Table S1 and Figure S2); however, the means of salinity, temperature and porewater DIC concentration in the MRE were significantly lower than those in the JRE (p<0.05) (Table S1 and Figure S2). Compared with the MRE, the JRE had slightly lower wind speed and higher air temperature (p>0.05) (Table S1). In addition, the differences

in physicochemical properties of water at different water depths were statistically indistinguishable in each estuary during the three stages (p>0.05) (Figure S2).

4. Discussion

4.1. Dissolved CH_4 in Sediment Porewater and Water Column

In this study, porewater CH_4 from the shrimp pond sediment in two estuaries varied greatly in different stages and the highest concentrations appeared in the middle stage (Figure 2). CH_4 concentrations were significantly positively correlated with sediment temperature, porewater TOC, DOC and DIC levels (p<0.05 or p<0.01; Table 1). The results indicate that higher sediment temperature and the availability of organic matter are important factors contributing to the elevated porewater CH_4 concentrations during the middle stage. Similar results have been found in coastal marshs [*Dutta et al.*, 2015; *Xiang et al.*, 2015]. This may be due to that higher temperatures stimulate organic matter decomposition, thus providing a larger source of available substrate for microbial production of porewater CH_4 [*Ding et al.*, 2004], and consequently producing more CH_4. This inference is further supported by a similar trend between the changes in CH_4 release rates and porewater concentrations in the shrimp ponds (Figure 2 and Figure 4a).

There was a clear temporal pattern of CH_4 concentrations in shrimp pond water, much higher concentrations appearing in the final stage (Figure 3). The significantly negative relationship between CH_4 concentrations and salinity over the study period indicated the influence of salinity on CH_4 levels in this ecosystem (Figure 5a). This finding is consistent with results from other estuaries [*Cotovicz Jr. et al.*, 2016; *Dutta et al.*, 2015]. Moreover, atmospheric pressure over the shrimp ponds also greatly varied in temporal dynamics (Table S1) [*Yang et al.*, 2018b], similar to the trend of the CH_4 concentration in water (Figure 3). The result reveals that atmospheric pressure could be another important factor responsible for temporal variation of water column CH_4 concentration (Figure 5b) by affecting CH_4 solubility in water [*Chen et al.*, 2014b; *Fu et al.*, 1996]. Furthermore, the shrimp ponds are a semi-intensive and closed ecosystem characterized by high stocking densities, low feed conversion rates and low water exchange frequency [*Yang et al.*, 2017a]. Therefore, the increase in the ponds' water CH_4 concentrations over time was probably related to the CH_4 accumulation in water column due to sustained CH_4 production and release from the sediment to the water column.

This study also found significant differences in sediment porewater (or water column) CH_4 concentrations between the two estuaries during all sampling stages (p<0.05; Figure 2 and Figure 3). The higher CH_4 concentration in the MRE is likely due to the low salinity and high organic matter in the sediment (Table S1), leading to high CH_4 production rates and high release across the SWI (Figure 4a). This inference is confirmed by the significantly positive relationship between CH_4 release fluxes and salinity, TC content, or CH_4 concentration (Table 2). This correlation was also observed by *Borges* and *Abril* [2011], *Rao* and *Sarma* [2016], and *Dutta et al.* [2013]. Methanogenic archaea is pH sensitive and develops quickly at a neutral or weakly alkaline environment in the coastal wetland [*Chang* and *Yang*, 2003]. Compared to the MRE, sediment pH in the JRE was relatively lower (average pH = 6.32) (Table S1), which may not favor the growth of methanogens, consequently inducing lower CH_4 concentrations (Figure 2 and Figure 3) by reducing CH_4 release fluxes (Figure 4a). Thus, higher CH_4 concentrations that occurred in MRE ponds, to some extent, were also dependent

on the differences in sediment pH between the two estuarial ponds.

4.2. Dynamic Variation of CH_4 Release Fluxes from Shrimp Ponds Sediment

Temporal variations in CH_4 release fluxes from the sediment were found in the current research (Figure 4a) and other wetlands and lakes [e.g., *Avery et al.*, 2003*;Bergman et al.*, 2000*; Hu et al.*, 2017*; Lofton et al.*, 2015*; Vizza et al.*, 2017*; Xing et al.*, 2005*; Yang et al.*, 2008*; Yang et al.*, 2017b]. It is generally acknowledged that temporal patterns of CH_4 release are governed by temporal changes in temperature affecting the production of substrate precursors and microbial activity [e.g., *Bergman et al.*, 2000*; Inglett et al.*, 2012*; Segers*, 1998*; Yang et al.*, 2015a]. According to correlation analysis (Table 2), sediment temperature is also a critical abiotic factor affecting the seasonal variation of sediment CH_4 release fluxes in the present study. In aquatic ecosystems, the majority of CH_4 is produced by the decomposition of organic matter under anaerobic conditions in the sediment [*Xiao et al.*, 2017]. Thus, the organic matter supply in the sediment is an important factor influencing CH_4 production and its subsequent release [e.g., *Bergman et al.*, 2000*; Whiting and Chanton*, 1993*; Xiao et al.*, 2017]. Although this study did not directly measure organic matter, we observed similar trends (Figure 4a and Table S1) and a significantly positive relationship between the sediment TC contents and pond CH_4 release fluxes in the MRE and JRE (Table 2). The results reveal that the temporal differences in pond sediment CH_4 release fluxes among the three stages in subtropical estuaries were also dependent on organic matter supply.

Average sediment CH_4 release fluxes from the coastal shrimp pond in the JRE were significantly smaller than those in the MRE (11.27±2.43 *vs* 84.52±11.58 μmol m^{-2} h^{-1}; $p<0.05$), indicating a strong spatial variability in CH_4 production from the coastal shrimp pond sediment. Similarly, in coastal marsh studies, the spatial variability of CH_4 production and release was attributed to the direct or indirect effects of salinity [e.g., *Poffenbarger et al.*, 2011*; Sun et al.*, 2013*; Vizza et al.*, 2017*; Welti et al.*, 2017] and substrate supply [e.g., *Duc et al.*, 2010*; Kim et al.*, 2015*; Wertebach et al.*, 2016*; Xing et al.*, 2006*; Yang et al.*, 2011*; Yang et al.*, 2017b]. In the present study, porewater salinity in the MRE was lower than that in the JRE (1.89±0.21% *vs* 7.72±0.75‰), a result of the effect of freshwater dilution caused by the interaction of precipitation, evaporation, and surface runoff. The mean levels of sediment TC in the JRE were also significantly smaller than those in the MRE ($p<0.05$, Table S1). Among the variables measured, the CH_4 release fluxes values were best correlated with porewater salinity and sediment TC in the two estuaries (Table 2). These results indicated that the difference in pond sediment CH_4 release fluxes between the MRE and JRE could also be attributed to salinity and substrate supply.

4.3. CH_4 Oxidation in the Overlying Water of Shrimp Ponds

CH_4 oxidation in water has been reported in a few freshwater ecosystem (e.g., lake, river, and reservoir) studies [e.g., *Almeida et al.*, 2016*; Bastviken et al.*, 2008*; Matoušů et al.*, 2017*; Striegl and Michmerhuizen*, 1998]. The oxidation rates observed in these water columns showed very large variability among aquatic ecosystems. In this study, CH_4 oxidation rates in the overlying water in the shrimp ponds were calculated based on the time-dependent reduction in dissolved CH_4 concentrations in the incubated water samples. During the experimental period, none of the samples indicated net CH_4 production in the shrimp pond water column, suggesting the oxidation of CH_4 in the shrimp ponds (Figure 4b). However, an unexpected but interesting result of the current study was that CH_4 oxidation rates were

always maintained at a relatively lower level, even though the water column contained a high level of DO (Figure S2). The CH_4 oxidation rates in the overlying water of the shrimp ponds at two subtropical estuaries ranged between 0.36 and 5.66 μmol m^{-2} h^{-1}, much lower than those reported in freshwater ecosystems (0.04 - 0.16 mol m^{-2} h^{-1}) [e.g., *Bastviken et al.,* 2008*; Matoušů et al.,* 2017; *Sawakuchi et al.,* 2016]. This may be attributed to the shrimp ponds' polyhaline characteristics that maintain a relatively higher salinity range (Figure S2) resulting in a weak CH_4 oxidation in the water column [*Dutta et al.,* 2015].

Despite relatively lower water column CH_4 oxidation rates in the shrimp ponds, there were large variations in CH_4 oxidation among aquaculture stages (Figure 4b). A similar phenomenon was also reported in other aquatic environments. Multiple studies found that the temporal changes of CH_4 oxidation rates were governed by temperature variation that affected methanotroph activity [e.g., *Dutta et al.,* 2015*; Lofton et al.,* 2014*; Matoušů et al.,* 2017; *Osudar et al.,* 2015]. However, CH_4 oxidation rates in the subtropical shrimp ponds did not seem to be affected by the seasonal variability in temperature (Figure 4b and Figure S2). Similar results were also observed by *Abril.* [2007] and *Roland et al.* [2017], who found that the water column substrate (i.e., CH_4 concentrations) played a crucial role affecting the temporal variability in the CH_4 oxidation rates. In this study, CH_4 concentrations in water of two estuaries increased in the order: final stage > middle stage > initial stage (Figure 3), which was similar to the trend of CH_4 oxidation rates (Figure 4b). This study also found the strongest relationships between CH_4 oxidation rate measurements from each estuary and the corresponding water column CH_4 concentrations (Figure 6a). Therefore, variation of CH_4 oxidation rates from the shrimp ponds were possibly controlled by water column substrate (CH_4 concentrations) shifts. Because CH_4 oxidation also needs oxygen, the process of CH_4 oxidation is a section of the biological oxygen demand [*Matoušů et al.,* 2017]. Thus, we hypothesize that the temporal patterns in CH_4 oxidation rates from the shrimp ponds, to some extent, are dependent on the supply of dissolved oxygen, which is probably more favorable for the methanotrophs growth. Although methanotroph data are unavailable in the present study, the CH_4 oxidation rates from the ponds' overlying water showed a significantly positive relationship with dissolved oxygen (Figure 6b), which lends indirect support to the above hypothesis.

4.4. Implications of Aquaculture Pond CH_4 Biogeochemical Cycling

In an earlier study, *Yang et al.* [2018] estimated that CH_4 emissions from the aquaculture shrimp ponds into the atmosphere in the two subtropical estuaries ranged from 2.5 to 189.4 mg m^{-2} h^{-1}, with mean values of 47.8 mg m^{-2} h^{-1}. It is worth noting that the CH_4 emissions fluxes in subtropical estuarine aquaculture ponds were substantially higher than those from the freshwater aquaculture systems [e.g., *Da Silva et al.,* 2018; *Hu et al.,* 2014, 2016*; Liu et al.,* 2015*; Wu et al.,* 2018*;*] and were also one to three orders of magnitude higher than those observed in most reservoirs and lakes in temperate, boreal, and Arctic regions [e.g., *Gerardo-Nieto et al.,* 2017*; Huttunen et al.,* 2002*; Natchimuthu et al.,* 2016; *Zhu et al.,* 2010]. The range of CH_4 emissions from aquaculture shrimp ponds in the two subtropical estuaries were also substantially higher than those from the coastal marsh in Liaohe Delta (0.32 to 6.40 mg m^{-2} h^{-1}) [*Olsson et al.,* 2015], Yellow River estuary (−0.80 to 0.48 mg m^{-2} h^{-1}) [*Sun et al.,* 2013], Min River estuary (0.80 to 13.28 mg m^{-2} h^{-1}) [*Tong et al.,* 2010], and Mobile Bay

estuary (0.00 to 4.00 mg m^{-2} h^{-1}) [*Wilson et al.*, 2015]. In addation, *Yang et al.* [2017b] found that the conversion of brackish marsh in the Min River Estuary to shrimp ponds could considerably increase CH_4 emissions during the culture period. These results indicate that subtropical estuarine aquaculture ponds could be important sources of atmospheric CH_4 and thus should not be overlooked in greenhouse gas accounting for their contributions to global climate change. China's economy developed rapidly in the last four decades with the cost of environmental degradation [*Yang et al., 2013a; Yang et al., 2018a*], while environmental protection has received increasing attention in the last years [*Yang 2014; Yang et al., 2015b*]. Considering the world momentum of mitigating global warming, effective approaches are needed to reduce CH_4 emissions from aquaculture ecosystem in both China and other countries.

Our previous study have reported that also there were large variations in CH_4 emissions fluxes from aquaculture ponds among estuaries, with high fluxes occurred Min River Estuary (86.01 mg m^{-2} h^{-1}) and low fluxes occurred Jiulong River Estuary (9.64 mg m^{-2} h^{-1}) [*Yang et al.*, 2018b]. The variations pattern of CH_4 emission fluxes was highly similar to that of sediment CH_4 release rates (Figure 4a) and porewater CH_4 concentrations (Figure 2). The result indicate that high CH_4 emissions were accompanied by high sediment CH_4 production and porewater CH_4 concentrations. The high variability of CH_4 emission and other biogeochemical processes from mariculture ponds is commonly related to multiple environmental factors, but low salinity is necessary to produce high CH_4 emission fluxes. In addition, the mean survival rate and yield of shrimp in the MRE ponds amounted to 65% and 0.35 kg m^{-2}, respectively, which were lower than those in the JRE ponds, at 70% and 0.41 kg m^{-2}, respectively [*Yang et al.*, 2018b]. This implies that increasing the salinity level of aquaculture ponds might be an important strategy to mitigate CH_4 emissions and increase aquaculture animal (e.g., shrimp) production. Further studies are merited to investigate the effect of salinity on methanotroph in aquaculture ponds.

4.5. Limitation and Future Research

Same as many studies, there are limitations in the current study. Firstly, CH_4 dynamics investigation was conducted in two estuaries in June, August, and October during the aquaculture period. The limited number of studying times and estuaries might limit the wide application of our results. Obviously, future studies including longer research period and more sites, especially, covering different aquaculture pond type, specific aquaculture management practice and reclamation history, will expand our understanding of CH_4 dynamics in mariculture systems. Second, this study examined the effect of key environmental factors on CH_4 dynamics in aquaculture ponds, while the contribution of microbial abundance and activity (methanogens, methanotrophs, and sulfate reducting bacteria), and methanogenic substrates may also be very important to control CH_4 biogeochemical cycling in estuarine aquaculture ponds. Thus, more studies are still needed to further understand the driving mechanism for CH_4 biogeochemical cycles in estuarine aquaculture ponds. Recently, *Oliveira Junior et al.* [2019] reported that fish bioturbation can significantly decrease CH_4 emission by increasing sediment oxygenation. This study analyzed CH_4 dynamics based on laboratory incubation experiments. Future *in situ* study on bioturbation effect will further improve our understanding of CH_4 dynamics in aquaculture ponds.

5. Conclusions

This study researched the variations of dissolved concentrations, net sediment release, and overlying water oxidation of methane in three aquaculture stages in shrimp ponds in two estuaries, MRE and JRE, in Southeast China. Methane dynamics in subtropical aquaculture ponds were controlled by multiple environmental factors including physicochemical factors, such as salinity, temperature, DO, and meteorological parameter, and biological factors, such as biological metabolism. Despite higher CH_4 oxidation rates in the MRE than those in the JRE, CH_4 release rates in the MRE pond sediments were high and also large levels of CH_4 accumulated in the porewater because of low porewater salinity, high sediment organic matter content, and the use of noncompetitive substrates by methanogenic microorganisms [*Chuang et al.*, 2016]. This resulted in large CH_4 release from surface sediment to water column and then to the atmosphere. Combined with the previous study [*Yang et al.*, 2018b], our results indicate that aquaculture ponds in the subtropical estuaries are some potential "hotspots" of CH_4 biogeochemical cycling and may contribute a substantial fraction to China's total wetland CH_4 emission. Achieving a tradeoff between carbon mitigation and aquaculture development will be a big challenge. The high spatial CH_4 biogeochemical cycle variation between estuaries implies that increasing the salinity level of ponds water and higher feed utilization efficiency would help to mitigate CH_4 emissions from aquaculture ponds. However, this study results from a limited area across a limited time period. In the future work, the need to strengthen the frequency of sampling *in situ* at longterm and different spatial scales, and to further identify the effect of microbial abundance and activity on CH_4 biogeochemical cycle in coastal aquaculture ponds to reveal the causalities of spatial and temporal variations.

Acknowledgements

This research was financially supported by National Science Foundation of China (No. 41801070, 41671088), Research Grants Council of the Hong Kong Special Administrative Region, China (CUHK458913), CUHK Direct Grant (SS15481), Open Research Fund Program of Jiangsu Key Laboratory of Atmospheric Environment Monitoring & Pollution Control, and Minjiang Scholar Programme. We would like to thank Wei-Ning Du and Jing-Yu Zhang of the School of Geographical Sciences, Fujian Normal University, for their assistance in the field. The data used in this study are available in the Article File, Data Set File and Supporting Material File.

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Table 1. Pearson correlation analysis between porewater CH_4 concentration and environmental parameters from the shrimp ponds in the Min River Estuary (MRE) and Jiulong River Estuary (JRE)[a].

Data set	Porewater parameters				Sediment parameters		
	Salinity	**TOC**	**DOC**	**DIC**	**Temperature**	**pH value**	**TC**
MRE	NS	0.775**	0.596**	0.558**	0.714**	NS	NS
JRE	NS	0.744**	0.433*	0.696**	0.402*	NS	NS
All data	-0.731****	0.643**	0.631**	NS	NS	0.527****	0.654****

[a] MRE, Min River Estuary; JRE, Jiulong River Estuary. All data in the table represent all estuaries. NS means "not significant". The symbols * and ** indicate significant correlations at the 0.05 and 0.01 levels, respectively. *n*=27 for environmental parameters, and porewater CH_4 concentration from the shrimp ponds in each estuary.

Table 2. Pearson correlation analysis between sediment properties, porewater CH_4 concentration and CH_4 release fluxes across the Sediment-water interface from the shrimp ponds in the Min River Estuary (MRE) and Jiulong River Estuary (JRE) [a].

	Sediment properties					**Porewater**	
	Temperature	**pH**	**Porosity**	**TC**	**C/N ratio**	**Salinity**	**CH_4 concentration**
MRE	0.473*	NS	0.469*	0.811**	0.752**	-0.649**	0.372*
JRE	0.443*	NS	NS	0.520**	0.595**	NS	0.283*
All data	NS	0.366**	NS	0.859**	0.854**	-0.604**	0.676**

[a] MRE, Min River Estuary; JRE, Jiulong River Estuary; All data in the table represent all estuaries. NS means "not significant". The symbols * and ** indicate significant correlations at the 0.05 and 0.01 levels, respectively. n = 27 for sediment parameters, CH_4 production rate, porewater CH_4 concentration and CH_4 fluxes from the shrimp ponds in each estuary.

Figure captions

Figure 1. Location of the study area and sampling sites in the Min River Estuary and Jiulong River Estuary, Southeast China.

Figure 2. Dynamics variation of sediment porewater CH_4 concentration in the shrimp ponds in the Min River Estuary (MRE) and Jiulong River Estuary (JRE). The boxes, center line, and whiskers represent the $25^{th} - 75^{th}$ percentiles, median value, and 5^{th} and 95^{th} percentiles, respectively. The square represents the area-weighted average (n = 9). Lowercase letters indicate significant differences (p<0.05) in mean value among growth stages on the same sampling estuary, while uppercase letters indicate significant differences (p<0.05) in mean value among estuaries on the same sampling stage.

Figure 3. Vertical variation of water CH_4 concentration and saturation in the shrimp ponds in the Min River Estuary (MRE) and Jiulong River Estuary (JRE). Bars represent mean±SE (n = 36). The various lowercase letters on the bars indicate significant differences (p<0.05) among sampling depths within each aquaculture stage.

Figure 4. Dynamics variation of (a) CH_4 release fluxes across the sediment-water interface (SWI), and (b) overlying water CH_4 oxidation rate from the shrimp ponds in the Min River Estuary (MRE) and Jiulong River Estuary (JRE). The boxes, center line, and whiskers represent the $25^{th} - 75^{th}$ percentiles, median value, and 5^{th} and 95^{th} percentiles, respectively. The square represents the area-weighted average (n = 9). Lowercase letters indicate significant differences (p<0.05) in mean value among growth stages on the same sampling estuary, whereas uppercase letters indicate significant differences (p<0.05) in mean value among estuaries on the same sampling stage.

Figure 5. Relationship between (a) water salinity, (b) atmospheric pressure and water column CH_4 concentration from the shrimp ponds in the Min River Estuary (MRE) and Jiulong River Estuary (JRE). All data in the figure represent all estuaries. Values of CH_4 concentration were calculated as the means of three depths and four sampling times at the same aquaculture stage. *Circles* + *Bars* represent mean±SE (n = 12).

Figure 6. Relationship between (a) water CH_4 concentration, (b) DO concentration and overlying water CH_4 oxidation rate from shrimp ponds in the Min River Estuary (MRE) and Jiulong River Estuary (JRE). All data in the figure represent all estuaries.

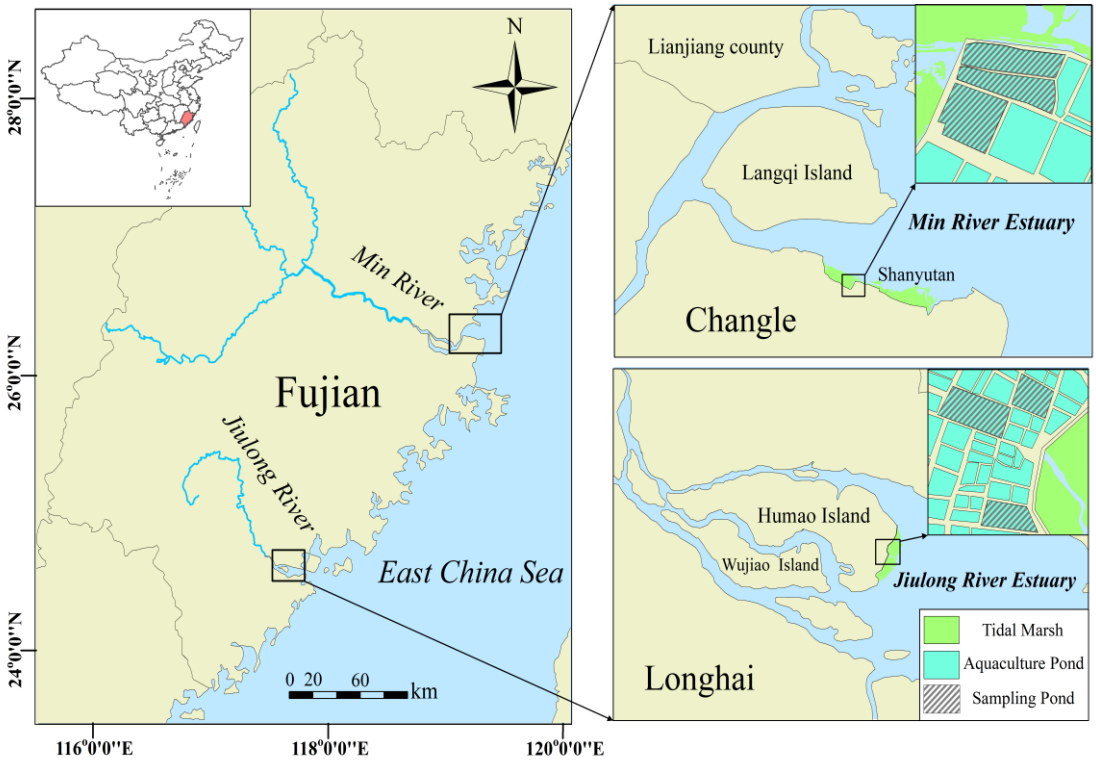
N
28°0'0"N
26°0'0"N
24°0'0"N
116°0'0"E
118°0'0"E
120°0'0"E
Min River
Jiulong River
Fujian
East China Sea
0 20 60
km
Lianjiang county
Langqi Island
Min River Estuary
Shanyutan
Changle
Humao Island
Wujiao Island
Jiulong River Estuary
Longhai
Tidal Marsh
Aquaculture Pond
Sampling Pond

Fig. 1

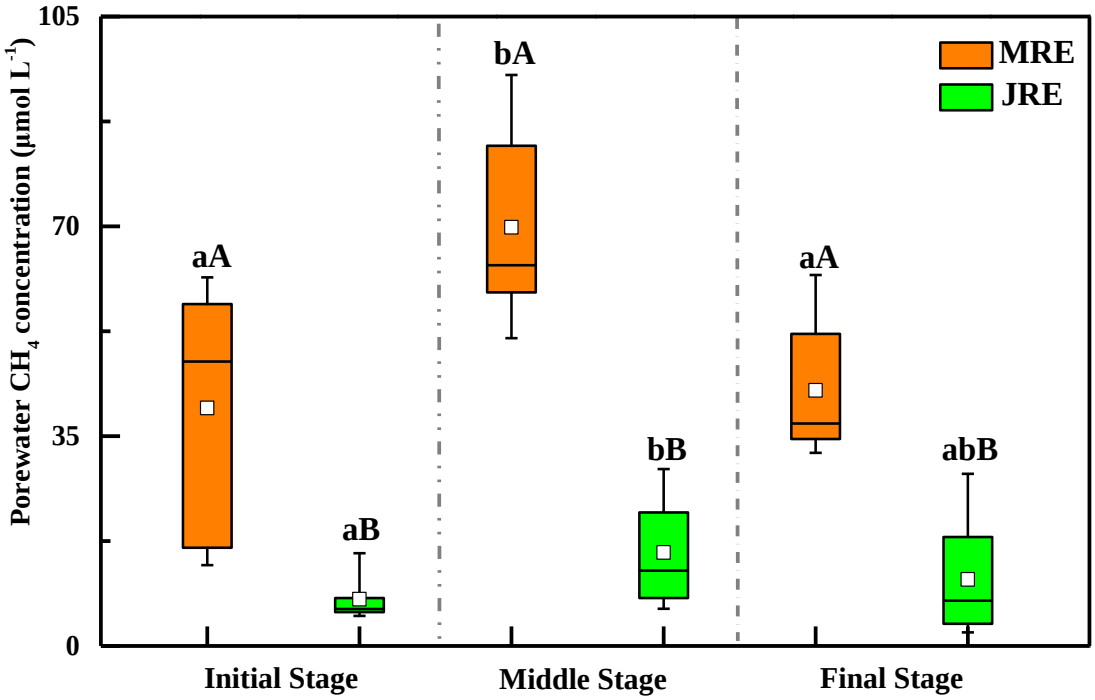
105
70
35
0
Porewater CH4 concentration (μmol L-1)
aA
aB
bA
bB
aA
abB
MRE
JRE
Initial Stage
Middle Stage
Final Stage

Fig. 2

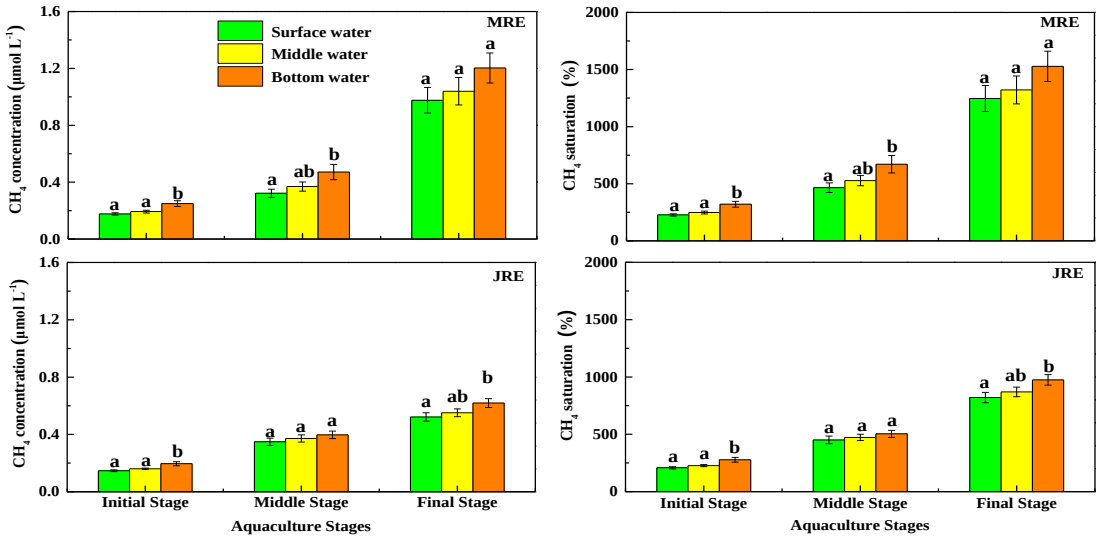
MRE
JRE
Surface water
Middle water
Bottom water
CH4 concentration (μmol L-1)
CH4 saturation (%)
Initial Stage
Middle Stage
Final Stage
Aquaculture Stages

Fig. 3

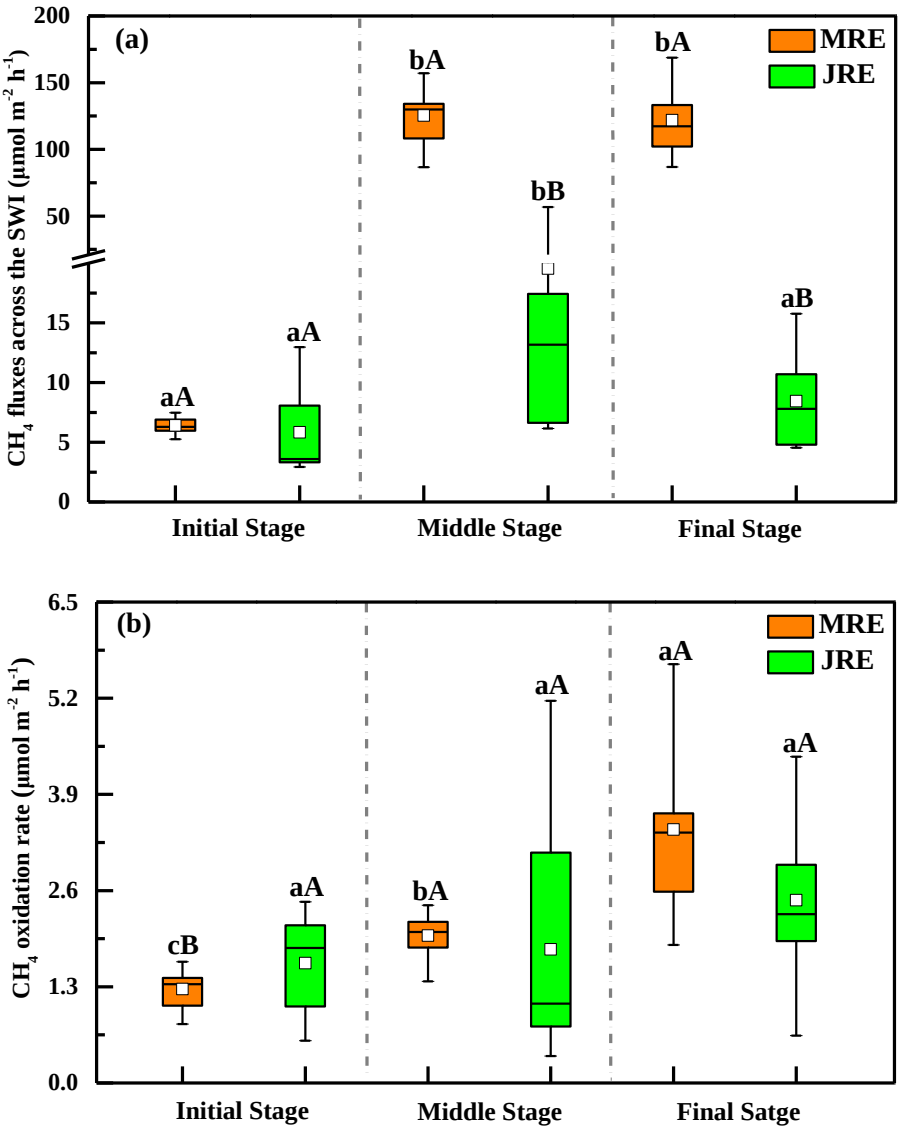
(a)
MRE
JRE
CH_4 fluxes across the SWI (μmol m^{-2} h^{-1})
200
150
100
50
15
10
5
0
aA
aA
bA
bB
bA
aB
Initial Stage
Middle Stage
Final Stage
(b)
MRE
JRE
CH_4 oxidation rate (μmol m^{-2} h^{-1})
6.5
5.2
3.9
2.6
1.3
0.0
cB
aA
bA
aA
aA
aA
Initial Stage
Middle Stage
Final Satge

Fig. 4

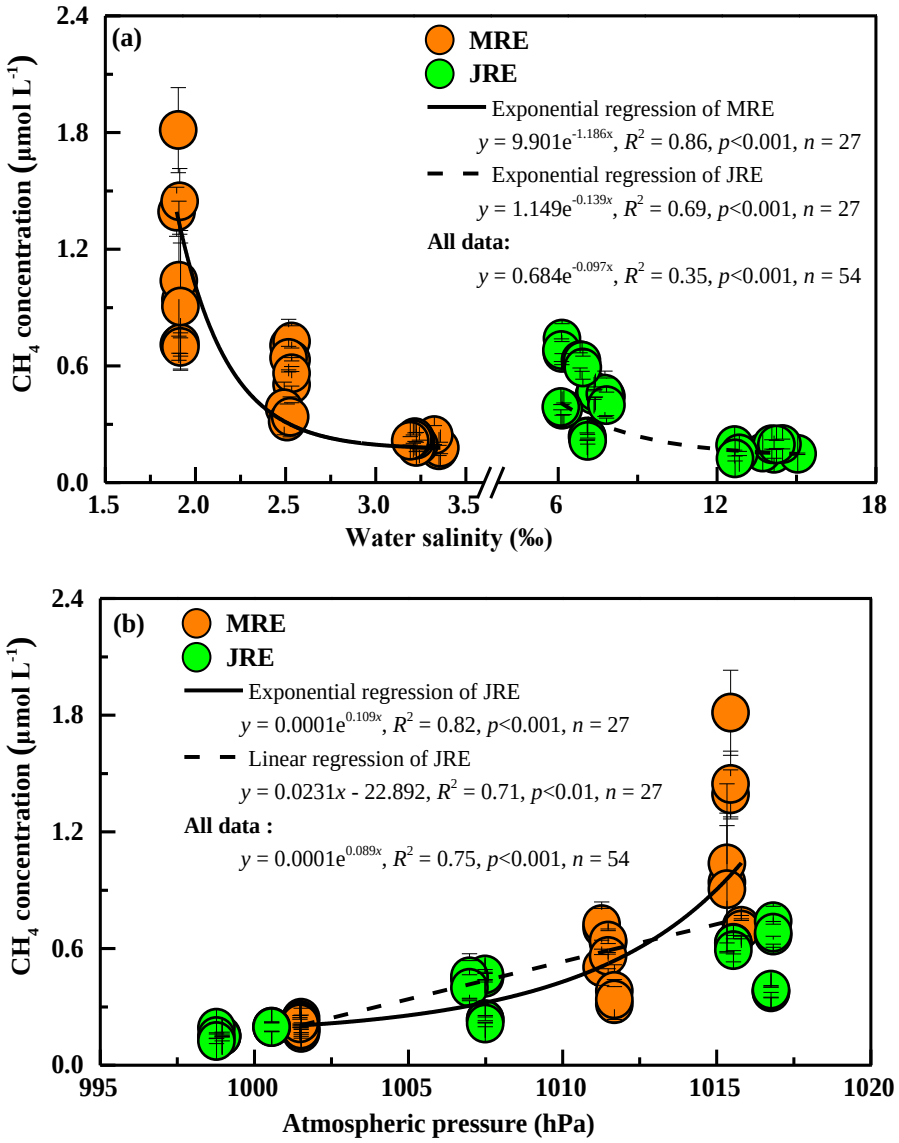
(a)
MRE
JRE
Exponential regression of MRE
y = 9.901e-1.186x, R2 = 0.86, p<0.001, n = 27
Exponential regression of JRE
y = 1.149e-0.139x, R2 = 0.69, p<0.001, n = 27
All data:
y = 0.684e-0.097x, R2 = 0.35, p<0.001, n = 54
CH4 concentration (μmol L-1)
Water salinity (‰)
(b)
MRE
JRE
Exponential regression of JRE
y = 0.0001e0.109x, R2 = 0.82, p<0.001, n = 27
Linear regression of JRE
y = 0.0231x - 22.892, R2 = 0.71, p<0.01, n = 27
All data :
y = 0.0001e0.089x, R2 = 0.75, p<0.001, n = 54
CH4 concentration (μmol L-1)
Atmospheric pressure (hPa)

Fig.5.

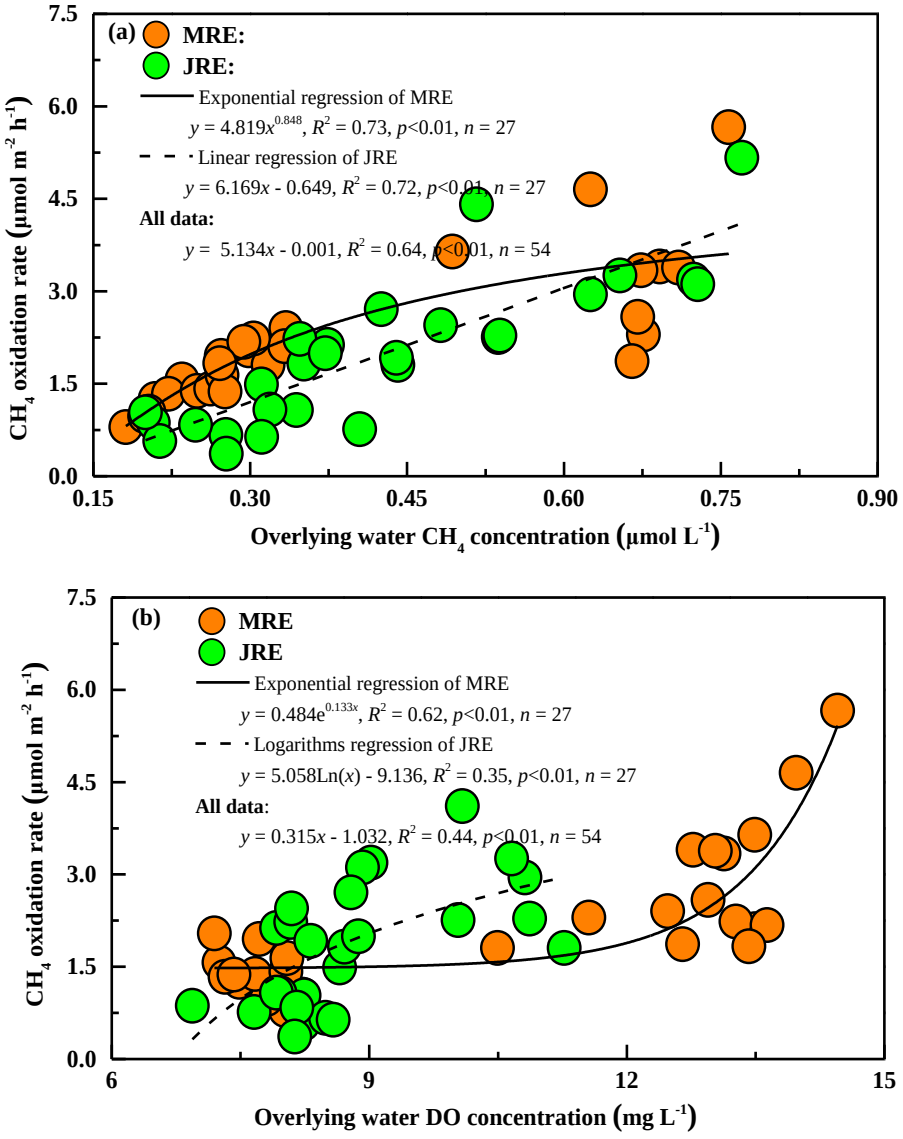
(a)
MRE:
JRE:
Exponential regression of MRE
y = 4.819x^0.848, R2 = 0.73, p<0.01, n = 27
Linear regression of JRE
y = 6.169x - 0.649, R2 = 0.72, p<0.01, n = 27
All data:
y = 5.134x - 0.001, R2 = 0.64, p<0.01, n = 54
CH4 oxidation rate (μmol m-2 h-1)
Overlying water CH4 concentration (μmol L-1)
7.5
6.0
4.5
3.0
1.5
0.0
0.15
0.30
0.45
0.60
0.75
0.90
(b)
MRE
JRE
Exponential regression of MRE
y = 0.484e^0.133x, R2 = 0.62, p<0.01, n = 27
Logarithms regression of JRE
y = 5.058Ln(x) - 9.136, R2 = 0.35, p<0.01, n = 27
All data:
y = 0.315x - 1.032, R2 = 0.44, p<0.01, n = 54
CH4 oxidation rate (μmol m-2 h-1)
Overlying water DO concentration (mg L-1)
7.5
6.0
4.5
3.0
1.5
0.0
6
9
12
15

Fig. 6