Large increase in CH_4 emission following conversion of coastal marsh to aquaculture ponds explained by gas transport pathways

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A B S T R A C T

Reclamation of coastal wetlands for aquaculture use has been shown to have opposite effects on sediment CH_4 production potential and CH_4 emission flux, but the underlying reason remained unclear. In this study, we compared sediment properties, CH_4 production potential, emission flux, and CH_4 transport pathways between a brackish marsh and the nearby reclaimed aquaculture ponds in the Min River Estuary in southeastern China. Despite that the sediment CH_4 production potential in the ponds was significantly lower than the marsh, CH_4 emission flux in the ponds (17.4 ± 2.7 mg m^{-2} h^{-1}) was 11.9 times higher than the marsh (1.3 ± 0.2 mg m^{-2} h^{-1}). Plant-mediated transport accounted for 75% of the total CH_4 emission in the marsh, whereas ebullition accounted for 95% of the total CH_4 emission in the ponds. CH_4 emission flux in both habitat types was highest in the summer. These results suggest that the increase in CH_4 emission following the conversion of brackish marsh to aquaculture ponds was not caused by increased sediment CH_4 production, but rather by eliminating rhizospheric oxidation and shifting the major transport pathway to ebullition, allowing sediment CH_4 to bypass oxidative loss. This study improves our understanding of the impacts of modification of coastal wetlands on greenhouse gas dynamics.

Keywords: Methane (CH_4) production; CH_4 emission; Gas transport pathway; Coastal wetland; Aquaculture ponds

1. Introduction

Human activities since the industrial revolution have significantly increased greenhouse gas emissions that have drastically changed the global climate (IPCC, 2021). Among the different greenhouse gases (GHGs), methane (CH_4) has a global warming potential (GWP) 45 times that of carbon dioxide (CO_2) over a 100-year time horizon (Neubauer and Megonigal, 2015), and CH_4 contributes ~20% of the global radiative forcing (IPCC, 2013). The atmospheric concentration of CH_4 has risen to 1909 ppbv in 2022 (NOAA, 2022), exceeding the preindustrial level by ~150%. To increase food production, natural landscape is increasingly converted to farmlands and aquaculture ponds, which changes the hydrology, nutrient cycles, soil properties and biodiversity on a large scale (Andreetta et al., 2016; Gao et al., 2019; Lin et al., 2022), often with an increase in CH_4 emission (IPCC, 2019; Tan et al., 2020). Comparing CH_4 production and emission and their environmental drivers between natural and modified lands is a key to understanding and predicting the effects of farming activities on climate change.

Despite covering only 5–8 % of the global land area, natural wetlands contribute 20–30 % of the global terrestrial carbon sequestration (Lal, 2008; Mitsch et al., 2013) and therefore play a crucial role in mitigating global climate change (Kayranli et al., 2010; Lu et al., 2017; Nahlik and Fennessy, 2016). Coastal wetlands alone are estimated to sequester approximately 44.6 Tg C yr^{-1} thanks to their high sedimentation rate (Chmura et al., 2003). However, large areas of coastal wetlands around the world have been lost or degraded in the past century due to land development and modification (Murray et al., 2019), which

may increase the release of terrestrial CO_2 and CH_4 to the atmosphere (Pendleton et al., 2012; Tan et al., 2020; Verhoeven and Setter, 2010), with potential dire consequences on the climate.

Coastal wetlands in mainland China cover about 5.79 M ha across its southern and eastern seaboards. In the recent decades, China has experienced among the worst coastal wetland degradation due to continuous population and economic growth, rapid urbanization and infrastructure development (Sun et al., 2015). A large area of its coastal wetlands has been converted for agroforestry uses since the 1950s (He et al., 2021; Wang et al., 2014) and the reclamation of wetlands into aquaculture ponds is also widespread along the coast (Ren et al., 2019; Duan et al., 2020). In vegetated wetlands, plant-mediated transport of sediment CH_4 can be a dominant pathway for CH_4 emission to air (Bhullar et al., 2013; Jeffrey et al., 2019). However, construction of earthen aquaculture ponds not only removes the wetland vegetation, but also changes the hydrology from free-flowing water to standing water, modifies the sediment physical properties and microbial communities, and alters nutrient and organic loading to the sediment, all potentially affect CH_4 production and emission (Tan et al., 2020).

The Shanyutan Wetland (26°00′36″–26°03′42″ N, 119°34′12″–119°40′40″ E) is the largest tidal wetland within the Min River estuary in southeastern China (Figure 1). About 30% of its area has been converted into shrimp aquaculture ponds since 2011. A study has shown that CH_4 emission from the aquaculture ponds was much higher than the nearby vegetated wetland (Yang et al., 2017); yet, a follow-on study showed that the sediment CH_4

81 production potential in the ponds was significantly lower than the vegetated wetland (Yang
82 et al., 2022). To resolve this paradox, we hypothesized that conversion of vegetated wetland
83 to aquaculture ponds changes the transport pathways of sediment CH_4, leading to an overall
84 higher emission flux in the aquaculture ponds.

85 To test our hypothesis, we compared CH_4 emission flux and the main transport
86 pathways between the aquaculture ponds and nearby wetland over a 33-month period. We
87 also collected monthly samples of sediment and porewater from both habitat types over a
88 10-month period to measure sediment properties and CH_4 production potential. The results
89 improve our understanding of how conversion of vegetated wetland to aquaculture ponds
90 impacts the climate by changing sediment CH_4 production and transport mechanisms and
91 subsequent emission to air.

92 ## 2. Materials and methods

93 *2.1. Study area and sampling frequency*

94 The Shanyutan Wetland region is characterized by a subtropical humid monsoonal
95 climate, with 1,390 mm annual mean precipitation, 19.6 °C air temperature and 77.0%
96 relative humidity (Yang et al., 2020). A large area of the wetland has been cleared of
97 vegetation (mainly *Cyperus malaccensis* and *Spartina alterniflora*) and reclaimed for
98 shrimp (*Litopenaeus vannamei*) aquaculture over the past decade (Yang et al., 2020). A
99 brackish *C. malaccensis* marsh stand and three nearby aquaculture ponds were selected for
100 the study (Figure 1c).

101 To measure plant-mediated CH_4 emission in the marsh, two sets of 1 m × 1 m plots (in

triplicate; < 5 m distance between plots) were established. One set of the plots was used as shoot clipping treatment (SCT) where the aboveground vegetation was removed and petroleum jelly was used to seal the clipped end of the stems to prevent gas release (Tong et al., 2012). The plants in the other set were left intact as the unclipped control treatment (UCT) (Hu et al., 2016; Tong et al., 2012).

The selected shrimp aquaculture ponds were 1.0–3.0 ha in area with a mean water depth of ~1.5 m during the farming period. The ponds were filled with seawater drawn from the estuary. Shrimp farming started in May and ended in November, producing a single crop. The shrimp were fed commercial food pellets daily. Further details of the aquaculture pond system and farming practice can be found elsewhere (Yang et al., 2020).

CH_4 flux measurements were taken 1–2 times each month between April 2019 and December 2021. Sediment and porewater samples were collected once every month between April 2019 and January 2020. Sediment samples (top 15 cm; triplicate) were collected using a steel corer (internal diameter 5 cm). Sediment porewater (top 15 cm) in the brackish marsh was sampled by *in situ* dialysis (Strack and Waddington, 2008; Tong et al., 2018). Porewater within the pond sediment was extracted by centrifugation in the laboratory (Bodmer et al., 2020; Matos et al., 2016). All field samples were stored in an ice-packed cooler and transported back to the laboratory within 4–6 hr.

2.2. CH_4 emission flux measurements

Within the marsh stand, gas emission samples from the SCT and UCT plots were collected using a static chamber (Tong et al., 2012; Xiang et al., 2015). The static chamber

was made of transparent polyvinyl chloride (PVC), with a top chamber (35 cm length × 35 cm width × 100 cm height) and a bottom collar (35 cm length × 35 cm width × 30 cm height). The bottom collar was inserted into the marsh sediment, leaving only 5 cm above the sediment surface. Gas samples from the aquaculture ponds were collected using a floating chamber (*FC*) as described in previous studies (Natchimuthu et al., 2016; Yang et al., 2020). The *FC* was made from an inverted plastic basin (polyethylene/plexiglas®), covering an area of 0.1 m^2 and a volume of 5.2 L, and was fitted with Styrofoam on the sides for floatation.

On each sampling date, gas samples were taken from each chamber headspace with a syringe into 50 mL aluminum-foil gas sample bags (Dalian Delin Gas Packing Co., Ltd., China) at 0, 15, 30 and 45 min intervals. Upon return to the laboratory, the CH_4 concentrations in the gas sample bags were measured on a Shimadzu GC-2010 gas chromatograph (Kyoto, Japan) with a flame ionization detector (FID). The CH_4 fluxes (mg m^{-2} h^{-1}) were calculated according to Hirota et al. (2004) and Yuan et al. (2021).

2.3. Determination of CH_4 transport pathways

Within the marsh stand, the CH_4 flux measured in the UCT plot was a combination of diffusive, ebullitive and plant-mediated CH_4 fluxes, whereas the CH_4 flux measured in the SCT plot represented the combination of diffusive and ebullitive CH_4 fluxes. Therefore, plant-mediated CH_4 flux was deduced from the difference in CH_4 flux between UCT and SCT plots (Ding et al., 2005; Hu et al., 2016).

The CH_4 flux measured by the *FC* method (F_T) in the aquaculture ponds represented

144 the combination of ebullitive flux (F_E) and diffusive flux (F_D) (Chuang et al., 2017; Wu et
145 al., 2019). To separate the two components, F_D was independently determined from surface-
146 water dissolved CH_4 concentrations and wind-dependent gas exchange velocity (k)
147 according to the thin boundary layer model (Cole and Caraco 1998; Musenze et al., 2014;
148 Wanninkhof et al., 1992). To determine dissolved CH_4 concentrations, triplicate bubble-free
149 water samples were collected from each pond (at 20 cm depth) using pre-weighed serum
150 glass bottles, and upon return to the laboratory, dissolved CH_4 concentrations were
151 measured using the headspace equilibration method on a gas chromatograph (GC-2010 with
152 FID, Shimadzu, Kyoto, Japan) (Borges et al., 2018; Musenze et al., 2014; Yang et al., 2019a,
153 2020). Diffusive flux (F_D; mg m^{-2} h^{-1}) across the water-air interface was calculated as:

154 $$F_D = k_x \times (C_W - C_{eq}) \times M \times 1000 \quad \text{(Eq. 1)}$$

155 Where k_x is the gas transfer coefficient (m h^{-1}) calculated from wind speed (Cole and Caraco,
156 1998), C_W is the dissolved CH_4 concentration (μmol L^{-1}), C_{eq} is the air-equilibrated
157 dissolved CH_4 concentration (μmol L^{-1}) in surface water for the *in situ* temperature and
158 salinity, M is the molar mass of CH_4 (16 g mol^{-1}). Afterward, ebullitive flux (F_E) was
159 calculated as the difference between F_T and F_D (Chuang et al., 2017; Xiao et al.,2017).

160 *2.4. Measurement of sediment CH_4 production potential*

161 Triplicate sediment samples (top 15 cm) collected from each site were used in
162 anaerobic slurry incubation experiment to measure CH_4 production potentials (Bodmer et
163 al., 2020; Minick et al., 2021; Vizza et al., 2017). In the laboratory, ~30 g of each sediment
164 sample and 30 mL of *in situ* water were added to a 200 mL glass incubation bottle. The

bottle was flushed with pure nitrogen gas (N_2) for 5–8 min to create an anoxic condition (Song et al., 2021; Vizza et al., 2017; Zhou et al., 2022) and then sealed with a sterile butyl rubber stopper. Subsequently, the bottles were incubated for 15 days at in *situ* temperature (14–30 °C). On Days 1, 3, 5, 7, 9, 11, 13 and 15, a 5-mL headspace gas sample was taken with a syringe from each bottle and 5 mL pure N_2 gas was added back to maintain the pressure. CH_4 concentrations in the extracted gas samples were analyzed on a gas chromatograph (GC-2010 with FID, Shimadzu, Kyoto, Japan). Sediment CH_4 production potential was calculated from the slope of linear regression between headspace CH_4 concentration and incubation time for each sample bottle (Liu et al., 2019; Wassmann et al., 1998).

2.5. Ancillary environmental variables

Meteorological parameters, including wind speed (W_S), air temperature (T_A) and air pressure (A_P), were recorded by an automated meteorological station in the Shanyutan Wetland. Sediment temperature (T_S) and water temperature (T_W) were measured *in situ* using a portable instrument (IQ150, IQ Scientific Instruments, USA). Dissolved oxygen concentration (DO) in the aquaculture pond water was measured with a multiparameter probe (550A YSI sonde, USA) at a 20-cm water depth. Subsamples of the sediment were freeze-dried, homogenized and ground to a fine powder for analysis of physicochemical properties. Sediment was diluted with deionized water before measuring pH (Orion 868 pH meter, USA; sediment-to-water ratio 1:2.5 w/v) and salinity (Eutech Instruments-Salt6 salinity meter, USA; sediment-to-water ratio 1:5 w/v). Sediment TC (total carbon) and TN

(total nitrogen) were measured on a combustion analyzer (Elementar Vario MAX CN, Germany). Porewater was analyzed for NH_4^+-N concentration (flow injection analyzer; Skalar Analytical SAN^{++}, Netherlands) and Cl^- and SO_4^{2-} concentrations (Dionex 2100 ion chromatograph).

2.6. Statistical analysis

All data were checked for normality and homogeneity of variance. Significant differences in environmental variables, CH_4 production potential and CH_4 flux between habitat types were tested by analysis of variance (ANOVA) using the SPSS version 22.0 (IBM, Armonk, NY, USA). Spearman correlation analysis was done to evaluate the relationships between CH_4 flux (or CH_4 production potential) and various environmental variables, using the corrplot and Hmisc packages in R software (Version 4.1.0). The weighted response ratios (RR_{++}) were calculated to assess the responses of sediment CH_4 production potential and CH_4 flux to habitat modification, following Hedges et al. (1999) and Tan et al. (2019). Redundancy analysis (RDA) was performed to assess which environmental variables best explained the temporal variations in CH_4 flux (or CH_4 production potential), using CANOCO 5.0 for Windows (Microcomputer Power, Ithaca, USA). All results were presented as mean ± 1 standard error, unless otherwise stated. To ease comparison, data were grouped by seasons where appropriate.

3. Results

3.1. Water and sediment properties

The physicochemical properties of sediment and porewater in the marsh and

aquaculture ponds were shown in Table S1. Overall, there were no appreciable differences in sediment pH and porewater SO_4^{2-} between the two habitat types, but T_S, salinity, TC, TN, and porewater NH_4^+-N and Cl^- were all significantly lower in aquaculture ponds than in the marsh. When comparing across seasons (Table 1), we observed generally higher T_S, T_W, TC and TN in the summer/autumn, but higher Cl^-, SO_4^{2-} and DO in the winter (Table S1).

3.2. Differences in CH_4 production and emission between habitat types

The sediment CH_4 production potential in the marsh (45.21±12.73 ng g^{-1} d^{-1}) was significantly higher (p<0.01; Figure S1a), by 124.5% on average, than in the ponds (20.14±5.59 ng g^{-1} d^{-1}). CH_4 emission flux in the SCT marsh plots averaged 0.31±0.05 mg m^{-2} h^{-1} (ranged 0.03–1.29 mg m^{-2} h^{-1}), which was less than the UCT plots (mean 1.34±0.24 mg m^{-2} h^{-1}; range 0.25–8.78 mg m^{-2} h^{-1}). The mean CH_4 emission flux in the ponds was more than 10-fold higher, at 17.4±2.7 mg m^{-2} h^{-1} ($F_{df=1}$=31.419, p<0.0001; Figure S1b).

CH_4 emission flux showed strong seasonal differences in both habitat types (Figure S2). Overall, CH_4 emission flux in the marsh was highest in the summer, followed by spring, autumn and winter (Figure S2a), whereas CH_4 emission in the ponds followed the order of summer > autumn > spring > winter (Figure S2b).

3.3. Transport pathways for CH_4 emission

Total CH_4 emission flux in the marsh varied between 0.2±0.1 and 8.8±7.5 mg m^{-2} h^{-1} across all sampling dates (Figure 2). Plant-mediated CH_4 emission flux, derived from the difference between UCT plots and SCT plots, was 0.1-8.1 mg m^{-2} h^{-1} (average 1.1±0.2 mg m^{-2} h^{-1}), which accounted for 21–96 % (Figure 3a), average 75%, of the total emission in

228 the marsh. The rest of the emission was likely a combination of diffusion and ebullition.

229 Total CH_4 emission flux in the aquaculture ponds ranged from 0.1±0.4 to 127.1±13.3
230 mg m^{-2} h^{-1} over the study period (Figure 2). Ebullition was the dominant transport pathway,
231 accounting for 0.1–126.9 mg m^{-2} h^{-1}, or 45.6–99.8 % (average 85%) of the CH_4 emission
232 flux in the ponds (Figure 3b). The remaining emission flux came from diffusion.

233 *3.4. Responses of CH_4 production and emission to habitat modification*

234 The Weighted response ratios (RR_{++}) of sediment CH_4 production potential (PP_{CH4})
235 and CH_4 emission flux (F_{CH4}) are shown in Figure 4. Conversion of the marsh to aquaculture
236 ponds decreased sediment CH_4 production potential significantly (p<0.05) by 6.9% (Figure
237 4a), but increased CH_4 emission flux by 52.4% (Figure 4b).

238 *3.5. Relationships between CH_4 emission and environmental variables*

239 Spearman correlation analysis showed that CH_4 emission flux was positively
240 correlated with T_A, T_S and TC in both habitats (p<0.01 or p<0.001), but negatively
241 correlated with A_P, sediment salinity, porewater Cl^- and SO_4^{2-} concentrations (p<0.01 or
242 p<0.001) (Figure 5). Moreover, CH_4 emission flux in the ponds was positively correlated
243 with T_W but negatively with DO (p<0.001; Figure 5b). Sediment CH_4 production potential
244 and CH_4 emission flux were positively correlated with each other in both habitats (p<0.001).

245 Based on the result of RDA, T_S, NH_4^+-N, SO_4^{2-} and TC were the variables that best
246 explained the variation in CH_4 emission flux in the marsh (Figure 6a), of which T_S
247 accounted for the highest percentage (38.9%), followed by NH_4^+-N (21.3%), SO_4^{2-} (13.6%)
248 and TC (11.7%). The CH_4 emission flux the aquaculture ponds was mostly driven by TC,

249 which explained 63.8% of the variation, followed by NH_4^+-N (15%) and T_S (5.5%) (Figure
250 6b).

251 # 4. Discussion

252 *4.1. Land conversion effects on CH_4 dynamics*

253 CH_4 is produced in the sediment by methanogenic archaea through the stepwise
254 degradation of organic matter under anoxic condition (Segers, 1998; Villa et al., 2020).
255 Once produced, CH_4 can be exported from the sediment to the atmosphere via different
256 pathways (Bastviken et al., 2004; Jeffrey et al., 2019; Villa et al., 2020). The presence of
257 marsh plants may affect CH_4 dynamics in several ways. Deposition of organic carbon by
258 the plants may increase sediment methanogenesis. As plants use their vascular systems for
259 gaseous exchange between tissues, the process brings O_2 to the below-ground biomass but
260 may also accelerate the transport and release of sediment CH_4 to air (Sorrell and Boon,
261 1994). The overall effect may depend on the plant species, leading to different degrees of
262 increase or decrease in CH_4 emission relative to bare sediment (Gurbir et al., 2013). The
263 contributions of plant-mediated transport to total CH_4 emission vary widely between
264 different wetlands, ranging from 8% to 98% (Jeffrey et al., 2019; Knoblauch et al., 2015;
265 Korrensalo et al., 2022; Morrissey and Livingston, 1992). In the present study, we found
266 that the marsh plots with clipped above-ground vegetation (0.3±0.1 mg m^{-2} h^{-1}) had lower
267 CH_4 emission flux than the plots with intact vegetation (1.3±0.2 mg m^{-2} h^{-1}), indicating that
268 the presence of *C. malaccensis* increased CH_4 emission relative to bare marsh sediment,
269 and plant-mediated transport accounted for on average 75%, at times close to 100%, of the

CH_4 emission in the marsh (Figure 3a).

A notable observation in this study was that CH_4 emission flux in the aquaculture ponds on average was much higher than the marsh (Figure S1b), suggesting that conversion of marsh stand to aquaculture ponds would increase the areal output of CH_4 from the Shanyutan Wetland. These results are consistent with an earlier study (Yang et al. 2017) and are in line with a meta-analysis showing that CH_4 emission fluxes increased by 347% following the conversion of coastal wetlands to aquaculture ponds (Tan et al., 2020). However, CH_4 production potential independently measured by incubation experiment was actually lower in the aquaculture pond sediment than in the marsh sediment (Figure S1), confirming the observations in an earlier study and which could be attributed to the lower organic content, lower methanogen diversity and weaker interactions among the methanogens in the pond sediment (Yang et al., 2022). It should be noted that the CH_4 production was measured by anoxic incubation of slurry and therefore it reflected the potential capacity of CH_4 production, rather than actual in situ CH_4 production. Nevertheless, the paradoxical observations suggest that a higher CH_4 emission was not necessarily an indication of a higher capacity of the sediment to produce CH_4; rather, other factors need to be taken into consideration that might affect the fate of CH_4 in the system.

Net CH_4 emission is determined by the balance between CH_4 production and CH_4 consumption (Schimel, 1995; Korrensalo et al., 2022; Yang et al., 2019a). Methanogenesis in the sediment is driven by organic carbon substrates and activity of methanogens under anoxic condition. CH_4 from the sediment may be consumed by methanotrophs in the

291 overlying oxic water before reaching the atmosphere (Bastviken et al., 2008). The vascular
292 systems of the marsh plants that facilitate CH_4 transport can also bring atmospheric oxygen
293 belowground (Blossfeld et al., 2011; Korrensalo et al., 2022; Turner et al., 2020), potentially
294 increasing CH_4 oxidation around the roots (Laanbroek, 2010). Some studies have reported
295 that rhizospheric oxidation removes most of the CH_4 produced in the sediment (e.g.,
296 Calhoun and King, 1997; Popp et al., 2000; Fritz et al., 2011). In addition, river flow and
297 periodic tidal flushing within the marsh would increase DO level in surface sediment (Well
298 et al., 2018), thereby enhancing CH_4 oxidation and minimizing CH_4 accumulation within
299 the sediment (Tan et al., 2020; Yamamoto et al., 2011). This altogether perhaps explains the
300 low CH_4 emission flux in our marsh stand despite the higher sediment CH_4 production
301 potential.

302 Marsh vegetation was removed to create the aquaculture ponds within the Shanyutan
303 Wetland; therefore, plant-mediated CH_4 emission and rhizospheric oxidation were absent
304 in the ponds. Instead, CH_4 diffusing out of the sediment could be consumed by CH_4
305 oxidation within the water column before reaching the air. Indeed, our results showed that
306 CH_4 emission flux in the ponds was strongly and negatively correlated with dissolved
307 oxygen concentration (Figures 5 and 6). On the other hand, the stagnant nature of the pond
308 water may allow CH_4 to accumulate and form bubbles in the sediment, and subsequent
309 ebullition would allow the CH_4 to bypass oxidation within the water column (Yuan et al.,
310 2021). Earlier studies have shown that porewater CH_4 in the pond sediment accumulated to
311 a much higher level (51.5 μM; Yang et al., 2019b) than the marsh sediment (17.1 μM; Tong

et al., 2018). Consequently, despite the lower sediment CH_4 production potential in the ponds (Figure S1a), the CH_4 emission flux was more than 10-fold higher than the marsh, with ebullition accounting for on average 85% of the total CH_4 emission (Figure 3b). This percentage is similar to observations in freshwater aquaculture ponds (Yuan et al., 2021) and it falls within the range (50–95 % via ebullition) reported for organic-rich shallow aquatic systems (Natchimuthu et al., 2014; Wang et al., 2021a; Wu et al., 2019; Zhang et al., 2020).

4.2. Environmental drivers of temporal variations in CH_4 emission

Our results show that both the brackish marsh and the aquaculture ponds in the Shanyutan Wetland were atmospheric CH_4 sources with similar seasonal variabilities (Figure 2). Higher CH_4 emissions occurred in the summer and lower emissions in the winter in both habitats (Figure S2), which is consistent with observations in others coastal wetlands (Jacotot et al., 2019; Olsson et al., 2015; Yang et al., 2021) and aquatic systems (Borges, et al., 2018; Praetzel et al., 2021; Sierra et al., 2017; Wu et al., 2019). Despite the similar seasonal patterns, we suggest that the underlying causes differed when considering the respective dominant transport pathway and environmental drivers in each habitat type.

In the marsh stand, sediment carbon content (TC) was only moderately important in driving CH_4 emission flux (explaining 11.7% of the variation) (Figure 6), suggesting that CH_4 emission flux in the marsh was weakly coupled to methanogenesis in the sediment. Rather, sediment temperature (T_S) was the strongest factor driving CH_4 emission flux, explaining 39.8% of the variation. Given that plant-mediated transport was the dominant

CH_4 emission pathway in the marsh, higher T_S in the summer might increase evapotranspiration rate leading to higher CH_4 emission, as has been observed in other vegetated wetlands (MacDonald et al., 1998). However, plant-mediated emission was irrelevant in the aquaculture ponds. Instead, the rise and fall of CH_4 emission flux followed the farming cycle: the start of farming activity in the summer would have increased organic substrate availability (from feeds and animal wastes) in the sediment and driven up CH_4 emission. Indeed, RDA analysis showed that sediment carbon content (TC) was the overwhelming factor that determined CH_4 emission flux in the aquaculture ponds (63.8%). As the farming season came to an end in the winter, CH_4 emission decreased accordingly.

In the present study, CH_4 emissions from the marsh and ponds also exhibited substantial inter-annual variability, with the coefficients of variation of 58% and 39%, respectively. This may be attributed to the differences in salinity ($F_{df=2}$=452.362, p<0.001) as a result of interannual variations in precipitation (1807 mm in 2019, 1516 mm in 2020 and 1439 mm in 2021; the MRE weather station), which affected competition between sulfate reducing bacteria and methanogens (Chambers et al., 2013; Neubauer et al., 2013; Vizza et al., 2017). This was further confirmed by the significant and negative relationship between CH_4 flux and sediment salinity in both habitat types (p<0.01; Table S2).

4.3. Limitations and caveats

There are some limitations and caveats in this study. Firstly, sediment CH_4 production potential was measured by incubating slurry in anoxic condition, which may not reflect the actual CH_4 production rate where the sediment oxygen level varies. In situ measurements

using tracer technique, without the need for incubation, may give more accurate CH_4 production rates (Ashley et al., 2021; Wang et al., 2021b). The conversion of coastal wetland to aquaculture ponds can exert significant impacts on sediment physicochemical properties and various microorganisms, including methanotrophs (Gao et al., 2019). However, we did not investigate CH_4 oxidizers or oxidation rate in this study, a knowledge gap in a fuller understanding of CH_4 dynamics in the system. In the aquaculture ponds, the shrimp may increase the release of sediment CH_4 via bioturbation (Bezerra et al., 2020; Yuan et al., 2021), but the effect can be patchy and difficult to quantify with the floating chamber method. Lastly, land conversion is widespread along the coast of China and the Asian Pacific, but different wetlands can be dominated by different types of vegetation and likewise, different aquaculture systems have different farmed species and management practices (e.g., feed applications, aeration). Because of the highly variable species-specific effects of vegetation on sediment CH_4 dynamics (Bridgham et al., 2013; Laanbroek, 2010), a more detailed comparative study across regions and system types will improve our understanding of the climate impact of aquaculture-related landscape modification.

5. Conclusions

Overall, the conversion of marsh stand to aquaculture ponds in the Shanyutan Wetland changed not only the sediment capacity to produce CH_4, but also the main CH_4 emission pathway. Despite the lower CH_4 production potential in the pond sediment, CH_4 emission flux was higher thanks to the effective transport pathway through ebullition. Plant-mediated

transport was the main pathway for CH_4 emission in the marsh, but rhizospheric oxidation by the rooted vegetation likely had resulted in the much lower CH_4 emission flux despite the higher sediment CH_4 production potential. Therefore, the overall effect of land conversion was to increase the areal CH_4 output to air, which raises concerns of the climate impact of the fast-growing small-hold aquaculture sector in China.

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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Table 1

Summary of two-way ANOVAs examining the effect of habitat types (*HT*), sampling season (*SS*) and their interactions on sediment and porewater properties.

		Sediment properties								**Porewater properties**					
	df	**Temperature (*T*S)**		**Salinity**		**Total carbon (TC)**		**Total nitrogen (TC)**		**NH_4^+-N**		**Cl^-**		**SO_4^{2-}**	
		F values	*P values*	*F values*	*P values*	*F values*	*P values*	*F values*	*P values*	*F values*	*P values*	*F values*	*P values*	*F values*	*P values*
HT	**1**	70.663	<0.001	41.227	<0.001	2.322	=0.043	51.253	<0.001	20.102	<0.001	14.847	<0.001	0.245	0.622
SS	**3**	74.999	<0.001	14.611	<0.001	12.755	<0.001	2.489	=0.001	2.145	=0.106	13.108	<0.001	15.660	<0.001
HT*×*SS	**3**	5.194	=0.003	12.394	<0.001	2.289	=0.089	6.679	=0.001	26.583	<0.001	7.376	<0.001	6.265	=0.001

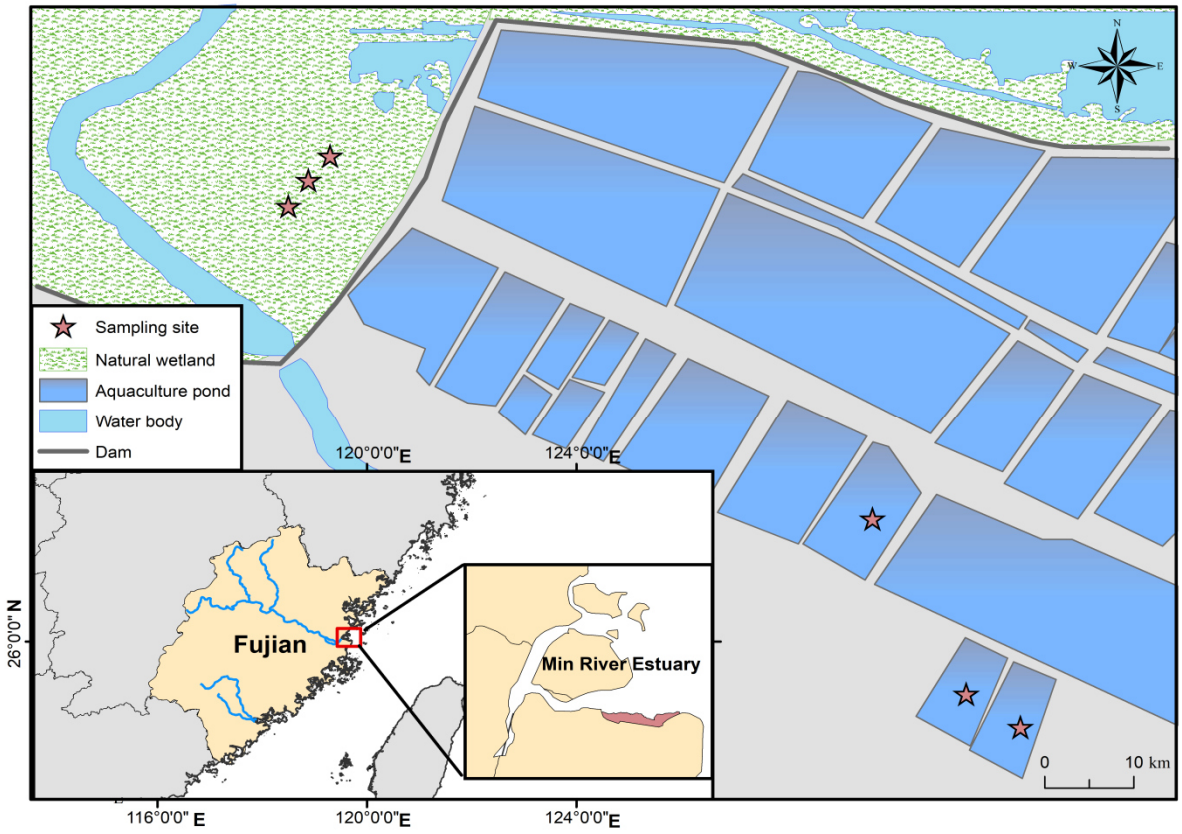

Figure 1. Location of sampling sites in a *Cyperus malaccensis* marsh and nearby aquaculture ponds in the Shanyutan Wetland, southeastern China.

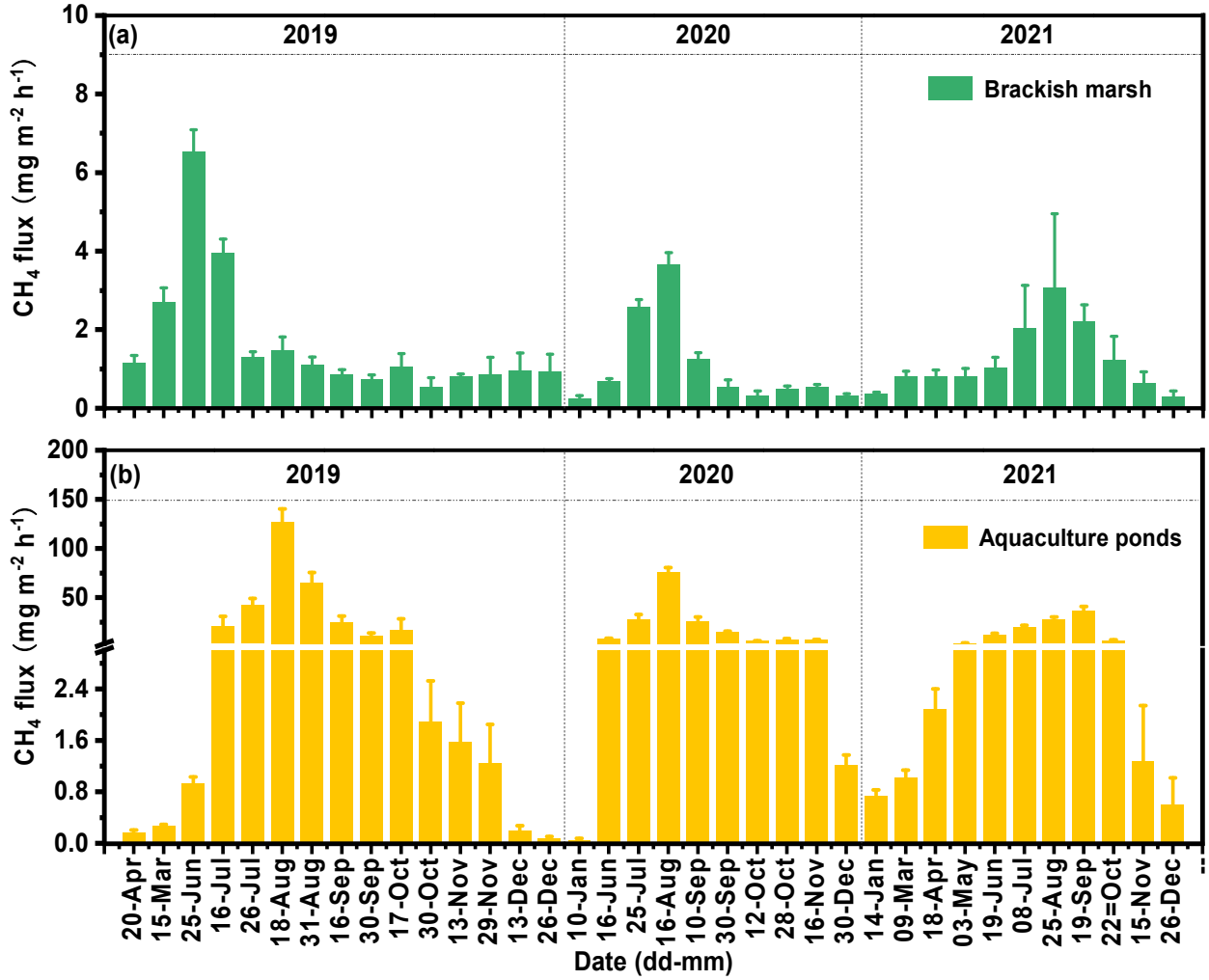

Figure 2. Temporal data of CH_4 emission flux in the marsh and aquaculture ponds. Bars represent mean ± SE (n = 3).

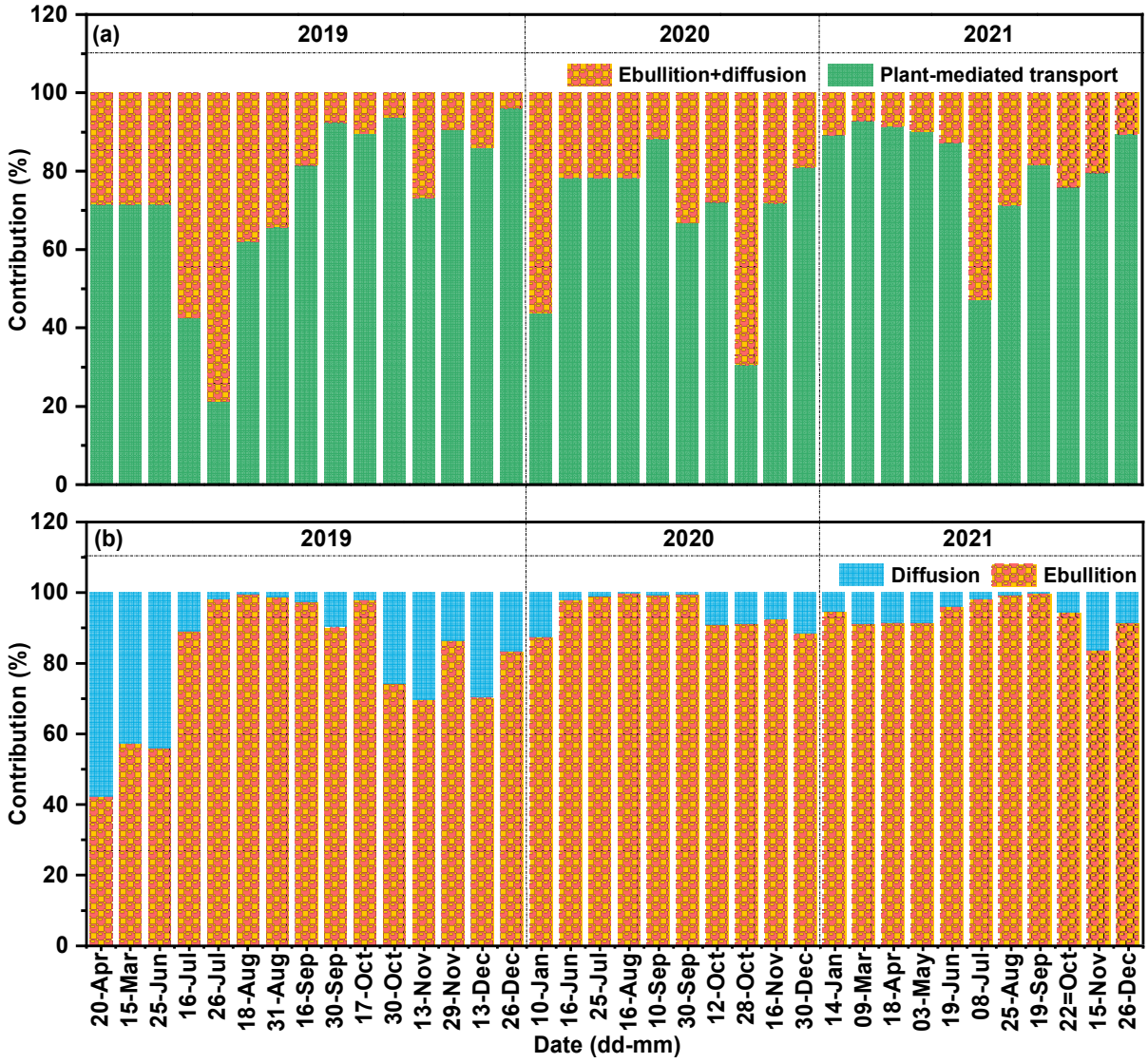

Figure 3. Temporal data of contributions of different transport pathways to CH_4 emission in (a) the marsh and (b) the aquaculture ponds.

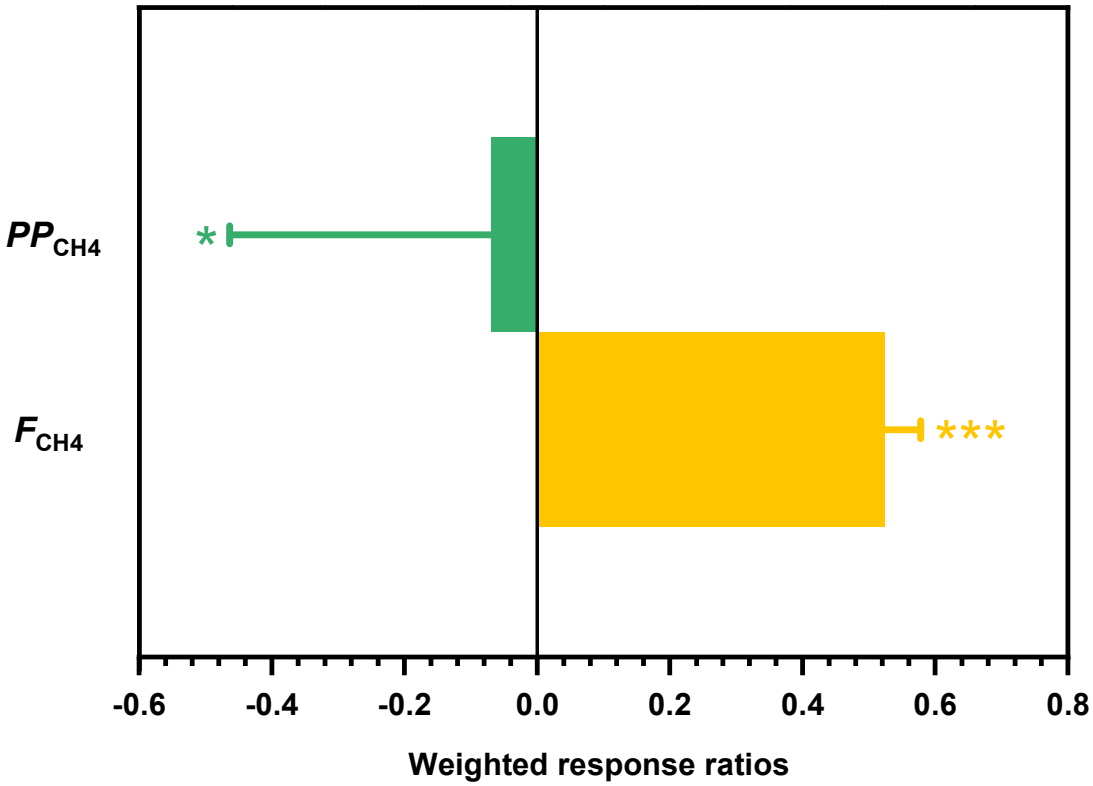

Figure 4. Weighted response ratios (RR_{++}) of (a) sediment CH_4 production potential (PP_{CH4}) and (b) CH_4 emission flux (F_{CH4}) for the conversion of brackish marsh to aquaculture ponds. Bars represent the RR_{++} values and 95% CIs (n = 63). Effect of habitat modification was significant for both parameters (* $p < 0.05$; *** $p < 0.001$).

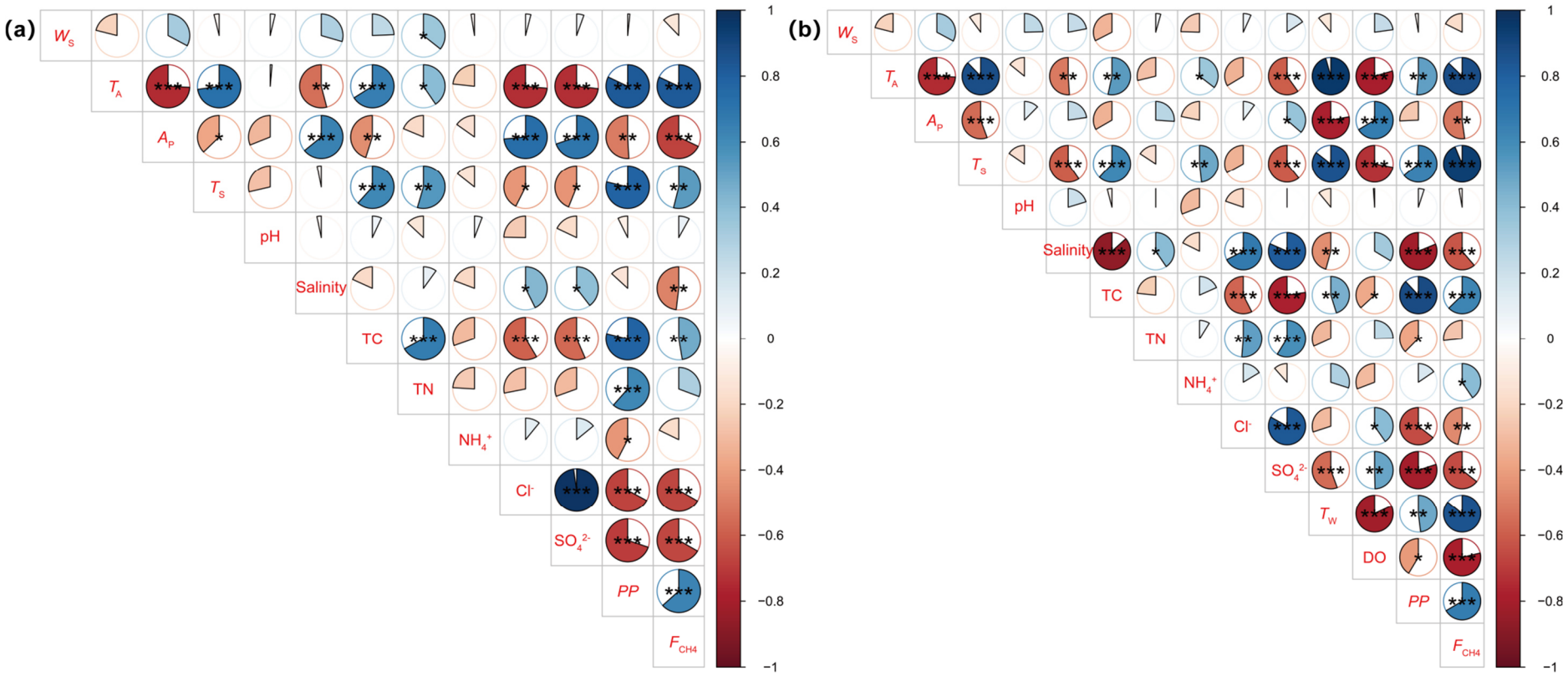

Figure 5. Correlation matrix for (a) the brackish marsh, and (b) the aquaculture ponds. Colors of the circle segments indicate the direction of correlation (blue = positive; red = negative); size of the colored segment is proportional to the *r* value (between -1 and 1). Asterisks within each circle indicate level of significance (*$p < 0.05$; **$p < 0.01$; ***$p < 0.001$). W_S, T_A, A_P, T_s, TC, TN, T_W, DO, *PP* and F_{CH4} represent wind speed, air temperature, atmospheric pressure, sediment temperature, total carbon, total nitrogen, water temperature, dissolved oxygen, CH_4 production potential and CH_4 emission flux, respectively.

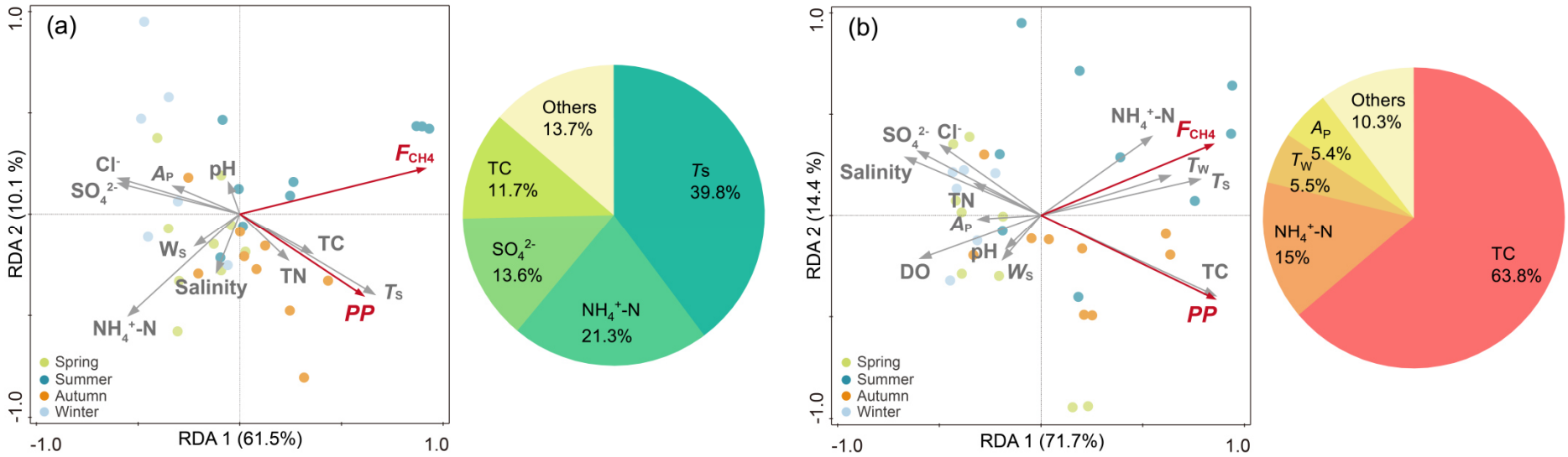

Figure 6. Results of redundancy analysis (RDA) between CH_4 emission flux (F_{CH4}) [or sediment CH_4 production potential (PP)] and environmental variables in (a) the marsh, and (b) the aquaculture ponds, showing the loadings of the different environmental variables. W_S, A_P, T_S, TC, TN, T_W and DO represent wind speed, atmospheric pressure, sediment temperature, total carbon, total nitrogen, water temperature and dissolved oxygen, respectively. The pie charts show the percentages of the variance of F_{CH4} explained by the different variables.

Supporting Information

Large increase in CH_4 emission following conversion of coastal marsh to aquaculture ponds explained by gas transport pathways

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Supporting Information Summary

No. of pages: 7 No. of figures: 2 No. of tables: 2

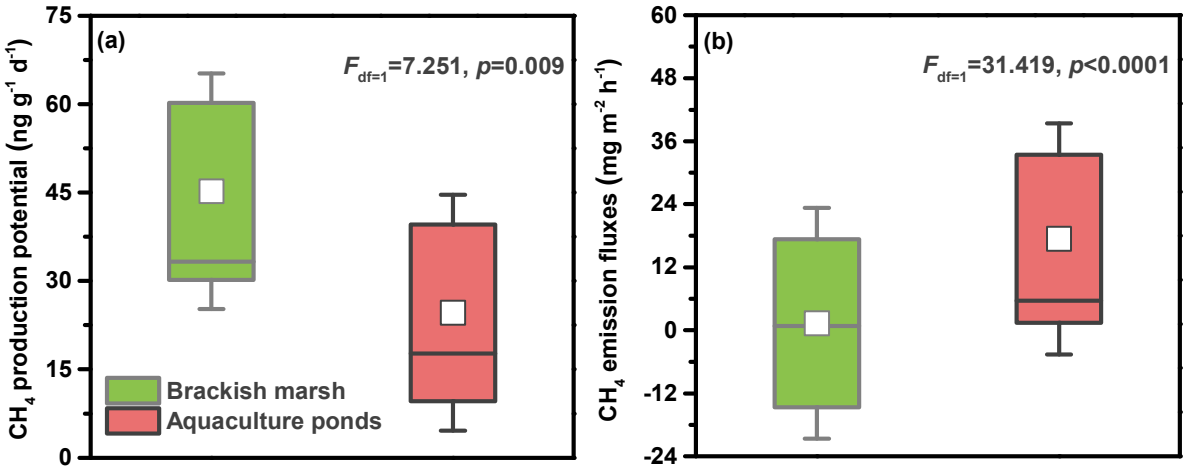

Figure S1. Boxplots of (a) sediment CH_4 production potential (n = 60) and (b) CH_4 emission flux (n = 216) in the brackish marsh and aquaculture ponds.

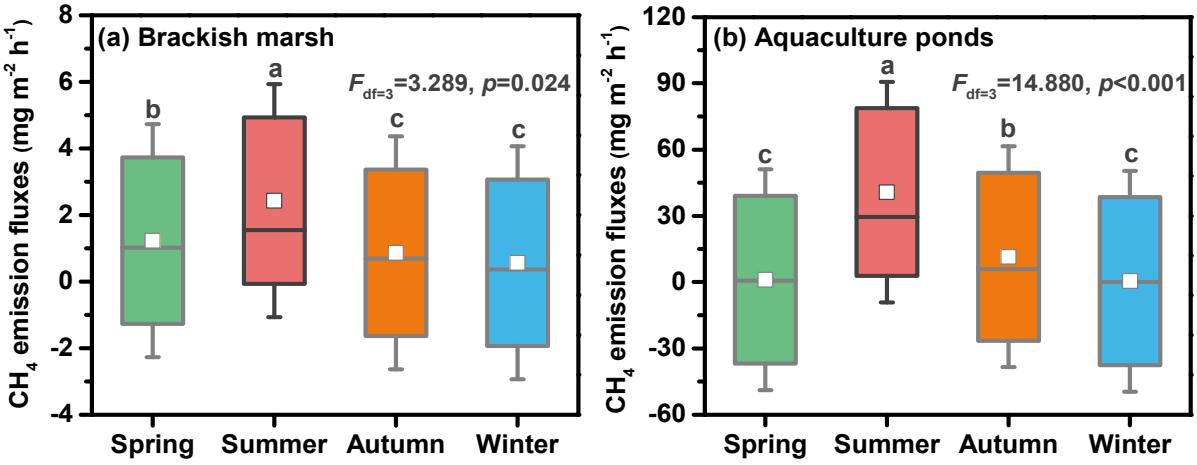

Figure S2. Boxplots of seasonal CH_4 emission flux in (a) the brackish marsh, and (b) the aquaculture ponds. Different lowercase letters within each panel indicate significant differences at the $p < 0.05$ level between seasons.

Table S1

Physicochemical properties of sediment and porewater in the brackish marsh and aquaculture ponds (mean±SE). Different lowercase letters within the same row indicate significant differences at $p < 0.05$ level between the two habitats.

Environmental variables	Brackish marsh					Aquaculture ponds				
	Spring	Summer	Autumn	Winter	Mean	Spring	Summer	Autumn	Winter	Mean
Sediment										
Temperature (°C)	23.6±0.3	28.2±0.1	27.8±0.6	17.6±1.2	24.7±0.8**a**	18.5±0.7	24.6±1.2	21.5±2.6	16.4±0.8	20.4±0.6**b**
pH	6.7±0.1	7.0±0.4	6.6±0.1	6.6±0.1	6.7±0.1**a**	6.8±0.3	6.7±0.1	6.9±0.1	6.7±0.1	6.7±0.1**a**
Salinity (‰)	2.6±0.1	2.8±0.3	7.8±0.5	10.8±0.4	6.0±1.9**a**	2.7±0.4	2.2±0.2	2.2±0.2	3.25±0.4	2.5±0.2**b**
TC (g kg^{-1})	13.1±2.9	27.8±2.2	22.2±1.4	10.6±2.9	18.4±3.9a	15.7±3.4	18.6±2.3	19.6±2.1	9.6±1.4	15.9±2.3**b**
TN (g kg^{-1})	1.3±0.1	1.9±0.2	1.0±0.1	0.7±0.3	1.3±0.1**a**	0.6±0.1	0.7±0.1	0.7±0.1	0.5±0.2	0.7±0.1**b**
Porewater										
NH_4^+-N (mg L^{-1})	0.9±0.1	0.3±0.1	0.5±0.1	0.4±0.1	0.5±0.1**a**	0.1±0.1	0.2±0.1	0.3±0.1	0.4±0.1	0.3±0.1**b**
NO_3^--N (mg L^{-1})	0.9±0.1	0.5±0.1	0.3±0.1	0.3±0.1	0.5±0.1**a**	0.1±0.1	0.4±0.1	0.1±0.1	0.2±0.1	0.2±0.1**b**
Cl^- (mg L^{-1})	5405±2528	135±29	6639±620	9090±932	5317±1889**a**	2090±671	2406±1470	1967±483	4803±858	2816±668**b**
SO_4^{2-} (mg L^{-1})	874±393	48±9	1099±90	1462±136	871±299**a**	664±150	678±133	581±168	1347±154	817±177**a**
Surface water										
Temperature (°C)	ND	ND	ND	ND	ND	22.9±0.6	29.9±0.3	23.2±0.8	13.8±0.8	22.4±3.3
DO (mg L^{-1})	ND	ND	ND	ND	ND	8.6±0.4	5.4±0.2	7.9±1.1	11.2±0.3	8.3±1.2

TC, Total carbon; TN, Total nitrogen; DO, Dissolved oxygen; ND, No data. Data are after *Yang et al.* [2022] for reference and review only.

Table S2

Linear relationship between CH_4 emission flux (F_{CH4}, mg m^{-2} h^{-1}) and sediment salinity (Sal_{sed}, ‰) in the brackish marsh and aquaculture ponds.

Habitat type	Linear regression equation	r^2	*p* value	*n*
Brackish marsh	F_{CH4} = -0.3635Sal_{sed}+3.7389	0.53	<0.01	9
Aquaculture ponds	F_{CH4} = -2.5831Sal_{sed}+29.391	0.43	<0.01	9

Reference

Yang, P., Tang, K. W., Tong, C., Lai, D. Y. F., Wu, L. Z., Yang, H., Zhang, L. H., Tang, C., Hong, Y., Zhao, G. H., 2022. Changes in sediment methanogenic archaea community structure and methane production potential following conversion of coastal marsh to aquaculture ponds. Environm. Pollut. 305, 119276. https://doi.org/10.1016/j.envpol.2022.119276