

Diffusive CH₄ fluxes from aquaculture ponds using floating chambers and thin boundary layer equations

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1 **Diffusive CH₄ fluxes from aquaculture ponds using floating chambers**
2 **and thin boundary layer equations**

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29 HIGHLIGHTS

- 30 ● Aquaculture ponds emit CH₄.
- 31 ● Large variations in diffusive CH₄ fluxes are estimated by different thin boundary layer (TBL) models.
- 32 ● Methane fluxes measured by chambers and match those estimated by only some TBL models.

33 A B S T R A C T

34 Static floating chambers (*FCs*) are the conventional method to measure CH₄ fluxes across the
35 water-air interface in ponds, while thin boundary layer (*TBL*) modelling is increasingly used
36 to estimate CH₄ fluxes. In this study, both *FCs* measurements and *TBL* models of gas transfer
37 velocity were used to determine CH₄ evasion from aquaculture ponds in southeastern China.
38 The surface water CH₄ concentrations ranged from 0.4 to 9.1 μmol L⁻¹ with an average of
39 4.8 ± 0.8 μmol L⁻¹. CH₄ flux was always positive, indicating the ponds as a persistent
40 CH₄ source to air. Mean CH₄ flux based on different *TBL* models showed large variations,
41 ranging between 19 and 316 μmol m⁻² h⁻¹. Compared against the direct measurement *FCs*,
42 three *TBL* models developed for the open sea, flowing estuarine system and lentic ecosystem
43 (*TBLW92a*, *TBLRC01*, and *TBLCL98*, respectively) overestimated CH₄ emission by
44 40–200%, while the wind tunnel-based *TBL* model (*TBLLM86*) underestimated
45 CH₄ emission. Two *TBL* models developed for lakes (*TBLW92b* and *TBLCW03*) gave
46 estimates similar to *FCs*.

47 Keywords: Methane fluxes; Thin boundary layer models; Floating chambers; Water-air interface; Shallow
48 aquaculture pond; Subtropical estuary

49 1. Introduction

50 Methane (CH₄) emissions from inland and coastal aquatic systems are potentially significant sources of
51 atmospheric CH₄ (Bastviken et al., 2011; Musenze et al., 2014; Yang et al., 2011). CH₄ release from open
52 water surfaces can take place via diffusion and/or ebullition (bubbling) (Bastviken et al., 2004). Diffusive
53 fluxes across the water-air interface are traditionally measured by using static floating chambers (FCs) or
54 thin boundary layer (TBL) model. The FCs approach determines CH₄ fluxes based on the change in CH₄
55 concentrations in the chamber headspace over time. The TBL approach calculates the CH₄ flux using a
56 piston velocity and gas concentration in the water (Natchimuthu et al., 2017; Zhao et al., 2019). *TBL*
57 modelling can be used to estimate CH₄ emissions from aquatic environment at a large-scale (Zhao et al.,
58 2015), while static FCs measurements are widely operated to quantify the small-scale spatial variation in
59 CH₄ fluxes over an area of < 1 m² (Denmead, 2008; Xiao et al., 2016). Previous studies have used either
60 one of the two approaches to quantify CH₄ fluxes from aquatic ecosystems (e.g., Musenze et al., 2014;
61 Natchimuthu et al., 2016; Wang et al., 2017; Welti et al., 2017). However, integrated comparative studies
62 of these two methods for determining CH₄ emissions from aquatic ecosystems remain scarce (e.g.,
63 Duchemin et al., 1999; Matthews et al., 2003), particularly in small pond ecosystems.

64 Recent studies have shown that very small ponds (area <0.001 km²) are hotspots of CH₄ emission
65 (Holgerson, 2015; Holgerson and Raymond, 2016; Wik et al., 2016; Yuan et al., 2019). However, the
66 accuracy of these estimates are largely constrained by the lack of rigorous quantifications of the area,
67 number, and spatial distribution of small ponds globally (Jonsson et al., 2008; Zhao et al., 2019) and the
68 large variations in flux measurement methods between different studies. In particular, the lack of
69 consensus between existing gas flux measurement methods remains a major source for the uncertainty of

70 GHGs accounting. This lack of agreement might be related to the variation in developing environment
71 between methods. For instance, the TBLLM86, TBLWan92a and TBLWan92b, TBLRC01, TBLCL98, and
72 TBLCW03 models which were developed by Liss and Merlivatt (1986), Wanninkhof (1992), Raymond
73 and Cole (2001), Cole and Caraco (1998), and Crusius and Wanninkhof (2003), respectively, are widely
74 accepted wind-based models to estimate CH₄ transfer velocities and fluxes. Among these TBL models,
75 the TBLLM86, TBLWan92a, and TBLRC01 models were developed in wind tunnels, open sea, and
76 flowing estuarine systems, respectively, while TBWan92b, TBLCL98 and TBLCW03 models were
77 established in the lentic ecosystem (e.g., lake). It is still unclear to what level of certainty these different
78 models can accurately calculate the gas transfer velocities in various aquatic ecosystems (Musenze et al.,
79 2014). Thus, a simple, low-cost, and standardized technique is still required for the accurate estimation of
80 CH₄ fluxes at the regional and global scales.

81 Aquaculture ponds form an important component of the global network of small ponds (FAO, 2017),
82 and the total surface area of freshwater and brackish aquaculture ponds is estimated to be around 110,000
83 km² (Verdegem and Bosma, 2009). Despite the importance of aquaculture ponds for CH₄ emission (Hu et
84 al., 2016; Wu et al., 2018; Yang et al., 2015, 2019a; Yuan et al., 2019), CH₄ flux data are
85 disproportionately scarce, and the published results were predominantly determined by FCs, rather than
86 TBL modelling (Hu et al., 2016; Wu et al., 2018; Yang et al., 2015, 2019a). Clearly, there is a paucity of
87 researches on comparing CH₄ fluxes obtained by using different approaches. In this study, FCs and six
88 TBL models were applied in aquacultural ponds in Southeast China, and the CH₄ fluxes were compared.
89 The primary research aims are: (1) to evaluate the performances of different wind-based TBL models for
90 the estimation of CH₄ fluxes; (2) to compare the diffusive CH₄ emissions from aquaculture ponds derived
91 from the FCs measurements and TBL modellings; and (3) to identify the TBL model(s) which can be

92 applied to replace the FCs for the measurement of CH₄ fluxes from ponds, with the minimal uncertainty.

93 2. Materials and Methods

94 2.1. Study area

95 Our study sites were located at the central-western Shanyutan Wetlands in the Min River Estuary
96 (MRE) in Southeast China (Figure S1, 26°00'36"–26°03'42" N, 119°34'12"–119°40'40" E). This area is
97 characterized by a subtropical monsoon climate, with a multi-year average annual temperature and
98 precipitation of 19.6 °C and 1,350 mm, respectively (Tong et al., 2010). The wetlands are dominated by a
99 semidiurnal tide with a large tidal range (2.5–6 m) that follows a spring-neap-spring tidal cycle (Luo et al.,
100 2014; Tong et al., 2010). The dominant vegetation in the wetland are the native *Cyperus malaccensis* and
101 *Phragmites australis*, and the invasive *Spartina alterniflora*. Over the past 10 years, much of the tidal
102 marshes have been converted to aquacultural ponds (Yang et al., 2017a).

103 2.2. Aquaculture pond management

104 Small and shallow aquaculture ponds (area of 0.8–2.5 ha and depth of 1.1–1.8 m) are a key feature in
105 the MRE, covering an area of around 234 ha in the Shanyutan Wetland (Yang et al., 2017b). Aquaculture
106 production, which is concentrated between June and November, yields a single annual crop of shrimps
107 from the semi-intensive earthen ponds, which are filled with salt water (average salinity of 2.0– 8.5‰)
108 from the MRE using a submerged pump. The shrimps are fed twice a day (at 07:00 and 16:00 hr) with
109 commercial aquatic feed pellets containing 42% protein. Three to five 1500 W paddlewheel aerators
110 operate four times a day (07:00–09:00, 12:00–14:00, 18:00–20:00, and 00:00–03:00 hr) to provide
111 sufficient oxygen. This study selected three replicate ponds that were separated by a distance of <10 m (see
112 Table S1 for basic characteristics) (Zhang et al., 2019) for the field measurements. Additional details about

113 the shrimp pond system and management can be found in Yang et al. (2017b).

114 2.3. Determination of dissolved CH₄ concentration

115 Field sampling campaigns were carried out at three aquaculture ponds between June and November
116 2017 following the main aquaculture practice. In each pond, a wooden bridge was built reaching ~10 m
117 from the pond embankment to the pond center in order to conveniently collect the water and gas samples at
118 three sites. Samples were collected two or three times each month in the three aquaculture ponds. Overall,
119 sampling was conducted for 15 different times. The total number of samples was 3 ponds × 3 sites × 15
120 times = 135. To measure the dissolved CH₄ concentrations, surface water (at a depth of ~20 cm) was
121 collected using a homemade water sampler and transferred into a 55-mL gas-tight glass serum bottle that
122 had been flushed with pond water for 2-3 times. After being completely filled, the glass bottles were
123 immediately sealed with a butyl rubber stopper using an aluminum screw cap, ensuring that all air bubbles
124 were excluded. To inhibit bacterial activity, 0.2 mL of saturated HgCl₂ solution was added to each bottle of
125 water sample (Borges et al., 2018; Hu et al., 2018). Samples were transported back to the laboratory in an
126 ice-packed cooler. Dissolved CH₄ concentrations of the samples were measured within 2 d of collection
127 following the headspace equilibration method. Approximately 25 mL headspace was created by injecting
128 ultra-high purity N₂ gas (>99.999%) into the glass bottle, while simultaneously 25 mL water sample was
129 withdrawn. The bottle was then shaken vigorously for 20 min and left at room temperature for 30 min to
130 form a complete equilibration between the air and water phases (Cotovicz et al., 2016). Approximately 10
131 mL of the equilibrated headspace air was subsequently extracted and injected into a gas chromatograph
132 (GC-2010, Shimadzu, Kyoto, Japan) equipped with a flame ionization detector (FID) to determine the CH₄
133 concentrations. Standard CH₄ gases at five concentrations, namely 2, 8, 500, 1000, and 10,000 ppm, were

134 used to calibrate the FID of gas chromatograph. Dissolved CH₄ concentrations of the sample were
135 calculated based on the volume of water, headspace air in the sampling glass bottle, and gas solubility
136 coefficient that was a function of water temperature and salinity (Farías et al., 2017; Wanninkhof, 1992;
137 Xiao et al., 2017).

138 2.4. Determination of diffusive CH₄ flux across the water-air interface

139 2.4.1. Measurement using floating chambers

140 The floating chamber methods (FCs) are one of the foremost techniques for directly measuring CH₄
141 emissions from aquatic ecosystems (e.g., Chuang et al., 2017; Gålfalk et al., 2013; Welti et al., 2017). In
142 order to measure the diffusive CH₄ effluxes from the aquaculture ponds, this study used a modified
143 chamber placed on a floating buoy (Figure S2). The opaque floating chambers were made from inverted
144 plastic basin (polyethylene/plexiglas®) with a volume and area of 5.2 L and 0.1 m², respectively. The
145 chambers were covered with aluminum tape to minimize internal heating by sunlight (Natchimuthu et al.,
146 2016; Yang et al., 2019). Thin gauze (bore diameter 0.001 mm) was used to cover the FCs aperture to
147 minimize the entry of bubbles into the chamber (Figure S2). A fan was installed inside the chamber to mix
148 the headspace air during the gas sampling. In order to quantify the potential contribution of CH₄ ebullition
149 flux from the aquaculture ponds, total CH₄ fluxes were also determined by using floating chamber without
150 gauze.

151 CH₄ fluxes were measured over a period of 45 min, with four headspace air samples being collected
152 inside the chamber at 15-min interval using 60-mL plastic syringes equipped with three-way stopcocks.
153 The gas samples were immediately transferred into pre-evacuated airtight gas sampling bags (Dalian Delin
154 Gas Packing Co., Ltd., China), transported to the laboratory, and analyzed within 48 h using a gas

155 chromatograph (GC-2010, Shimadzu, Kyoto, Japan) equipped with a FID, following the method of Tong et
156 al. (2010). The detection limits for CH₄ were 0.3 ppm, and the relative standard deviations of CH₄
157 analyses were $\leq 2.0\%$ in 24 h.

158 CH₄ emission fluxes (FCH₄, $\mu\text{mol m}^{-2} \text{hr}^{-1}$) were calculated based on the slope of the regression
159 between headspace CH₄ concentration and time (Yang et al., 2019). Generally, if r² of the correlation
160 between headspace CH₄ concentration and the elapsed time is larger than 0.90, the CH₄ emission is
161 considered as diffusion only (Bastviken et al., 2010; Zhu et al., 2016). If r² is below 0.90, the emission is
162 considered as the combination of ebullition and diffusion. The floating chambers with gauze (FCs-G) and
163 without gauze (FCs-NG) showed distinct linear (r²>0.9) and nonlinear (r²<0.9) increases in methane
164 concentration, and therefore the contribution of ebullition was calculated by the difference of the diffusion
165 flux measured between the FCs-G and the FCs-NG methods.

166 2.4.2. Estimation using thin boundary layer models

167 Saturation (S) of CH₄ in pond water was the ratio between the in situ dissolved concentration of CH₄
168 in pond water and the calculated saturated CH₄ concentration corresponding to ambient air CH₄
169 concentration (Hu et al., 2018) (Eq. 1):

$$170 S = C_{\text{water}}/C_{\text{Ws}} = C_{\text{water}}/(\alpha \times C_{\text{air}}) \times 100\% \quad (\text{Eq. 1})$$

171 where C_{water} is dissolved CH₄ concentration in pond water; C_{Ws} is the saturated CH₄ concentration
172 ($\mu\text{mol L}^{-1}$); C_{air} is the atmospheric concentration ($\mu\text{mol mol}^{-1}$) of the sampling sites; and α is the Bunsen
173 coefficient (Wanninkhof, 1992).

174 Diffusive fluxes of CH₄ (F, $\mu\text{mol m}^{-2} \text{hr}^{-1}$) across the water-air interface can be described by using
175 a theoretical diffusion model (Eq. 2) (Musenze et al., 2014):

176 $F = k \times (C_{\text{water}} - C_{\text{eq}})$ (Eq. 2)

177 where C_{water} ($\mu\text{mol L}^{-1}$) is the measured dissolved CH₄ concentration in surface water, C_{eq} ($\mu\text{mol L}^{-1}$) is
 178 the dissolved CH₄ concentration in equilibrium with the air above, and k is the gas transfer velocity (cm
 179 h⁻¹). The k value was parameterized as a function of wind speed and normalized for surface water
 180 temperature (T , °C) using a Schmidt number (Sc) derived from Eq. 3:

181 $Sc = 2039.20 - 120.31T + 3.4209T^2 - 0.040437T^3$ (Eq. 3)

182 This study evaluated the variations in CH₄ fluxes across the water-air interface estimated by eight widely
 183 used wind-based models developed in various conditions, including wind tunnels, open sea, estuarine
 184 systems, and lakes, as follows:

185 LM86 (Liss and Merlivatt 1986)

186 $F_{LM86} = 0.17U_{10}(Sc/600)^{-2/3}(C_{\text{water}} - C_{\text{eq}})$ $0 < U_{10} \leq 3.6$ (Eq. 4)

187 $F_{LM86} = (2.85U_{10} - 9.65)(Sc/600)^{-1/2}(C_{\text{water}} - C_{\text{eq}})$ $3.6 < U_{10} \leq 13$ (Eq. 5)

188 W92a (Wanninkhof, 1992)

189 $F_{W92a} = 0.31U_{10}^2(Sc/660)^{-1/2}(C_{\text{water}} - C_{\text{eq}})$ (Eq. 6)

190 RC01 (Raymond and Cole, 2001)

191 $F_{RC01} = 1.91 \exp(0.35U_{10})(Sc/600)^{-1/2}(C_{\text{water}} - C_{\text{eq}})$ (Eq. 7)

192 CL98 (Cole & Caraco, 1998)

193 $F_{CL98} = [2.07 + (0.215 \times U_{10}^{1.7})](Sc/600)^{-2/3}(C_{\text{water}} - C_{\text{eq}})$ (Eq. 8)

194 W92b (Wanninkhof, 1992)

195 $F_{W92b} = 0.45U_{10}^{1.64}(Sc/600)^{-1/2}(C_{water} - C_{eq})$ (Eq. 9)

196 CW03 (Crusius & Wanninkhof, 2003)

197 $F_{CW03} = 0.72U_{10}(Sc/600)^{-2/3}(C_{water} - C_{eq})$ $U_{10} < 3.7$ (Eq. 10)

198 $F_{CW03} = (4.33U_{10} - 13.3)(Sc/600)^{-1/2}(C_{water} - C_{eq})$ $U_{10} \geq 3.7$ (Eq. 11)

199 where U10 was determined according to the logarithmic wind profile relationship using Eq. 12 (Crusius
200 and Wanninkhof, 2003):

201 $U_{10} = U_z [1 + \frac{(C_{d10})^{1/2}}{K} \ln(\frac{10}{z})]$ (Eq. 12)

202 where U_z is the wind speed (m s⁻¹) at height z above the water surface (2.5 m in this study), C_{d10} is the
203 drag coefficient at 10 m above the water surface (0.0013 m s⁻¹), and K is the von Karman constant (0.41).

204 Generally, the stability of the atmosphere was an important factor influencing the calculation of U10 using

205 the wind-based equations. If the atmosphere over the aquatic systems is unstable, and the equation used to

206 calculate U10 needs to be adjusted. The air-water temperature difference can be used to determine if the

207 atmosphere over the aquatic systems is stable or not. If the air-water temperature difference is positive, the

208 atmosphere over the aquatic systems is stable. In the present study, the air temperature in ponds were

209 higher than water temperature during the study period, with the air-water temperature difference period

210 ranged from 0.1 to 3.8 oC, indicating that the atmosphere over the ponds is neutral stability regime.

211 Therefore, no adjustment is needed for U10, and the equation (12) was appropriate for the calculation of

212 U10. Some recent studies have applied surface renewal models that take into account both wind speed and

213 buoyancy to determine the k values (e.g., Czikowsky et al., 2018; MacIntyre et al., 2010; MacIntyre et al.,

214 2018).

215 2.5. Measurement of meteorological and environmental variables

216 Meteorological variables, including air temperature (AT), air pressure (AP), wind speed (WS), and
217 precipitation, were recorded at 30-min intervals using an automatic meteorological station (Vantage Pro 2,
218 China) installed at the MRE weather station in the China Wetland Ecosystem Research Network. The
219 distance between the automatic meteorological station and sampling ponds is about 75 m. The precision for
220 air temperature, atmospheric pressure, and precipitation were ± 0.2 °C, ± 1.5 hPa, and ± 0.4 mm min⁻¹,
221 respectively (Yang et al., 2020). The air temperature and wind speed were sampled at 1 Hz. WS were
222 determined using a cup anemometer that was connected to a Anemometer Sensors that registered the wind
223 speed in 1.0 m s⁻¹ bins at 1-min interval. The threshold for startup of the anemometer was 0.4 m s⁻¹.
224 Approximately 6% of wind speed measurements during the study period were below the threshold of 0.4 m
225 s⁻¹ at 2.5 m height.

226 Water temperature, electrical conductivity (EC), pH, dissolved oxygen (DO), total organic carbon
227 (TOC), and total dissolved nitrogen (TDN) content of surface water (~20 cm below the water surface) were
228 recorded at the three study sites in all 15 sampling campaigns. Water temperature and pH were measured
229 using a portable pH/mV/Temperature meter (IQ150, IQ Scientific Instruments, USA), and EC and DO
230 were determined using an electrical conductivity meter (2265FS EC, Spectrum Technologies, USA) and a
231 multiparameter water quality probe (550A YSI, USA), respectively. The relative standard deviations of EC,
232 pH, and DO analyses were $\leq 1.0\%$, $\leq 1.0\%$ and $\leq 2.0\%$, respectively

233 Water samples for TOC and TDN analyses were collected using a 5-L plexiglass hydrophore,
234 transferred to a 150-mL polyethylene bottle, and then transported to the laboratory in an ice-packed cooler.
235 TOC and N-NO_x- (NO₂- + NO₃-) concentrations were analyzed, after filtering through a 0.45- μ m

236 cellulose acetate filter (Biotrans nylon membranes), using a TOC analyzer (TOC-VCPH/CPN, Shimadzu,
237 Kyoto, Japan) and a flow injection analyzer (Skalar Analytical SAN++, The Netherlands), respectively.
238 The detection limits for N-NO_x⁻ and TOC were 6 µg L⁻¹ and 4 µg L⁻¹, respectively. The relative standard
239 deviations of N-NO_x⁻ and TOC analyses were $\leq 3.0\%$ and $\leq 1.0\%$, respectively.

240 2.6. Statistical analysis

241 Repeated-measures analysis of variance (RMANOVA) was conducted to test the differences in
242 diffusive CH₄ fluxes between the two approaches over the study period. Pearson correlation analyses were
243 conducted to examine the relationships between (1) dissolved CH₄ concentration or CH₄ fluxes and
244 environmental variables, and (2) diffusive CH₄ fluxes measured using *FCs* and estimated using the gas
245 transfer velocity models. The coefficient of variation (CV) for CH₄ fluxes on each sampling campaign was
246 determined by dividing the standard deviation by the mean value. Statistical analyses were conducted
247 using software SPSS (v. 17.0, SPSS Inc., USA) at a significance level of 0.05. Data are presented as mean
248 ± 1 standard error.

249 Generalized linear modelling was conducted to determine the variables that influenced CH₄ emission
250 fluxes from these seven different methods (i.e. *FCs* + 8 TBL models). The “*gls*” function from the “*nlme*”
251 R package (Pinheiro et al., 2018) with a saturated model was conducted for all variables (dissolved CH₄,
252 U₁₀, water temperature, dissolved oxygen, total dissolved carbon and dissolved nitrate). This model was
253 run using the *stepAIC* function in R “*MASS*” package that follows the Akaike Information Criterion (AIC)
254 (Venables and Ripley, 2002). It can identify the best model (lowest AIC value) in each case.

255 3. Results

256 3.1. Meteorological and environmental variables

257 The average air temperature (AT) and air pressure (AP) during the research period were 28.7 ± 0.4 °C
258 (range: 18.6–35.6 °C) and 1010.0 ± 0.5 hPa (range: 985–1025 hPa), respectively. Notably, the maximal AT
259 appeared in July and the minimal AP happened in August, very different from other months. The WS
260 during the study period ranged from 0.2 to 18.8 m s⁻¹, and varied between seasons, with a peak in July
261 (Figure S3a). Approximately 92% of WS fell within the range of 0.2–4.0 m s⁻¹ (Figure S3b).

262 There were also clear temporal variations in surface water characteristics during the study period. The
263 mean water temperature ranged from 18.1 °C (November) to 34.4 °C (August) (Figure S4a), while the
264 mean DO concentration changed between 9.4 mg L⁻¹ (August) and 19.9 mg L⁻¹ (November) (Figure S4).
265 The mean TOC concentrations varied between 9.9 mg L⁻¹ (July) and 57.3 mg L⁻¹ (November) (Figure S3),
266 while N-NO_x- concentrations ranged from 504 µg N L⁻¹ (June) to 10.7 µg N L⁻¹ (November) (Figure S4).

267 3.2. Model estimated k values and dissolved CH₄ concentrations

268 The mean k values showed considerable variations between different models and decreased in the
269 order: kRC01 (6.5 ± 0.8 cm h⁻¹) > kW92a (3.5 ± 0.7 cm h⁻¹) > kFCs (3.2 ± 0.4 cm h⁻¹) > kCL98 (2.9 ± 0.3 cm
270 h⁻¹) > kCW03 (2.5 ± 0.5 cm h⁻¹) > kW92b (2.4 ± 0.4 cm h⁻¹) > kLM86 (0.6 ± 0.1 cm h⁻¹) (Figure 1).

271 Dissolved CH₄ concentrations demonstrated large variations over the study period (0.1–31.1 µmol
272 L⁻¹), and they increased first and decreased to a valley later (Figure 2). CH₄ concentrations were
273 supersaturated across all ponds and all sampling dates, with an overall mean of 4.8 ± 0.8 µmol L⁻¹ (162.0
274 ± 18.4 ppmv), equivalent to 8700% saturation (range of 200–5.9 × 104% saturation).

275 3.3. CH₄ flux estimates by using TBL models and FCs method

276 There were considerable differences in the estimated diffusive CH₄ fluxes among

277 the *TBL* models (*TBLRC01*: $215.9 \pm 39.2 \mu\text{mol m}^{-2} \text{h}^{-1}$; *TBLCL98*:
278 $115.0 \pm 21.9 \mu\text{mol m}^{-2} \text{h}^{-1}$; *TBLW92a*: $102.9 \pm 19.5 \mu\text{mol m}^{-2} \text{h}^{-1}$; *TBLW92b*:
279 $78.3 \pm 13.9 \mu\text{mol m}^{-2} \text{h}^{-1}$; *TBLCW03*: $74.9 \pm 13.2 \mu\text{mol m}^{-2} \text{h}^{-1}$; and, *TBLLM86*:
280 $19.5 \pm 3.7 \mu\text{mol m}^{-2} \text{h}^{-1}$) (Table 1, Fig. 3 and Figure S5). Although there were marked
281 variations in the flux estimates among the various models, results from all models showed
282 similar temporal patterns (Fig. 3). The largest fluxes were generally recorded between August
283 and October, while the lowest fluxes were consistently recorded in June and November (Fig.
284 3).

285 Direct measurements using *FCs* with gauze (*FCs-G*) and without gauze (*FCs-NG*)
286 methods were 75.0 ± 12.5 (Fig. 3) and $231.3 \pm 681.3 \mu\text{mol m}^{-2} \text{h}^{-1}$ (Figure S6; Yang et al.,
287 unpublished data), showing significant difference between the two methods (Independent
288 Samples T-Test, $F = 118.190$, $p < 0.001$). On average, ebullitive CH_4 flux accounted for
289 33%–99% of the total CH_4 emissions during the study period.

290 3.4. Environmental influences on dissolved CH_4 concentrations and fluxes

291 Pearson correlation analysis showed that dissolved CH_4 concentrations in the shrimp ponds were
292 significantly positive correlated with air temperature and TOC concentration ($p < 0.01$), and negatively
293 correlated with N-NO_3^- concentration and EC ($p < 0.01$) (Table 2). CH_4 fluxes were found to be positively
294 correlated with air temperature ($p < 0.05$), TOC concentration and dissolved CH_4 concentration ($p < 0.01$),
295 and negatively correlated with water N-NO_3^- concentration ($p < 0.01$) and EC ($p < 0.05$) (Table 2 and Table
296 S3). This study also analyzed the relationships between the CH_4 fluxes derived from the seven different
297 methods and various environmental variables. N-NO_3^- concentration was consistently and negatively

298 correlated with CH₄ fluxes (Table S2). Environmental variables explained a larger proportion of variability
299 in CH₄ fluxes derived from the six TBL models (R²=0.46-0.54) than those from direct FCs measurements
300 (R²=0.35) (Table S2).

301 4. Discussion

302 4.1. CH₄ supersaturation and degassing from aquaculture ponds

303 There are few studies on CH₄ concentrations in small ponds, particularly, those created for
304 aquaculture purposes. In this study, the dissolved CH₄ concentration in surface water of the aquaculture
305 ponds ranged from 0.14 to 31.13 μmol L⁻¹ during the study period. The CH₄ concentration in our ponds
306 were higher than those observed in many small ponds in Florida (~2.2 μmol L⁻¹; Barber et al., 1988),
307 Colorado (~1.0 μmol L⁻¹; Bastviken et al., 2004), and Wisconsin and Minnesota (0.3–2.3 μmol L⁻¹; Smith
308 and Lewis, 1992) in the USA, in Sweden (~1.3 μmol L⁻¹; Natchimuthu et al., 2014), Canada (0.5–6.7
309 μmol L⁻¹; Pelletier et al., 2014), and Siberia (~2.6 μmol L⁻¹; Repo et al., 2007). In addition, CH₄
310 concentration in our researched aquaculture ponds were generally larger than those in some
311 nutrient-enriched rivers in China, i.e. Lixiahe River (0.2–0.81 μmol L⁻¹; Wu et al., 2019), and Beitang
312 Drainage River and Dagu Drainage River (0.3–1.7 μmol L⁻¹; Hu et al., 2018). Similar to inland aquatic
313 systems, such as lakes (e.g., Wen et al., 2016; Wik et al., 2016; Yan et al., 2018), reservoirs (e.g., Deemer
314 et al., 2016; Musenze et al., 2014; Wang et al., 2017), rivers (e.g., Barbosa et al., 2016; Striegl et al., 2012),
315 floodplains (Barbosa et al., 2020) and small ponds (e.g., Holgerson and Raymond, 2016; Wik et al., 2016),
316 aquaculture ponds were supersaturated for CH₄ (range of 2.71–599.81 folds supersaturation) with respect
317 to the atmospheric equilibrium (Figure 2b). The small temporary ponds in the Yale Myers Forest in
318 Connecticut, the USA, have, until now, the highest concentrations of CH₄, with the range of 21.0–58.9

319 $\mu\text{mol L}^{-1}$, equal to 119.0–2906.6 folds supersaturation (Holgerson, 2015). The CH_4 concentrations and
320 supersaturations in our aquaculture ponds fall well within the range reported previously by Holgerson
321 (2015). Our results indicated that aquaculture ponds in the subtropical estuaries were hotspots for CH_4
322 emission.

323 In inland aquatic ecosystems, the strong CH_4 release is likely a result of large organic matter input
324 from the catchment that sustains high methanogenic rates (Finlay et al., 2009; Lundin et al., 2013;
325 Venkiteswaran et al., 2013; Yan et al., 2018), which is supported by the significant relationship between
326 dissolved CH_4 and nutrient level (Huttunen et al., 2003; Kortelainen et al., 2001; Wen et al., 2016). In this
327 study, aquaculture shrimp ponds were semi-artificial ecosystems that were maintained through a daily feed
328 supply for the production of aquatic animals. However, only a small portion of the feed input was actually
329 converted into shrimp biomass, with the feed utilization efficiency of ~4.0–27.4% (Chen et al., 2016;
330 Molnar et al., 2013; Yang et al., 2017b). Surface sediments in the aquaculture systems typically retain a
331 large amount of organic matter from feces and residual feeds (Chen et al., 2016; Yang et al., 2017b) that
332 can support high levels of CH_4 production and its subsequent release to atmosphere. Although organic
333 matter content was not quantified in this study, our results confirmed the significantly positive correlation
334 between dissolved CH_4 and TOC concentration ($p < 0.01$; Table 2), which lent support to the idea that CH_4
335 supersaturation in the aquaculture ponds was related to the large input of organic matter.

336 4.2. Comparison of six different TBL modelled CH_4 fluxes

337 Although previous studies have compared the performance of different TBL models in estimating
338 diffusive CH_4 fluxes in inland waters (Amouroux et al., 2002; Li et al., 2015; Musenze et al., 2014; Xiao
339 et al., 2017; Zappa et al., 2007), such comparison is scarce for shallow ponds, particularly those created for

340 aquaculture. To the best of our knowledge, this study is the first attempt to compare the estimates of
341 diffusive CH₄ flux using different TBL models over the whole aquaculture period in aquaculture ponds.
342 Interestingly, although the patterns of temporal variations in diffusive CH₄ fluxes in the shrimp ponds were
343 largely consistent across the TBL models (Figure 3), there were differences in the magnitude of fluxes
344 estimated from different models (Table 1).

345 Notably, the mean flux estimate using the TBLRC01 model (215.6 $\mu\text{mol m}^{-2} \text{h}^{-1}$) was an order of
346 magnitude greater than that derived from the TBLLM86 model (19.4 $\mu\text{mol m}^{-2} \text{h}^{-1}$, Figure 3). Moreover,
347 CH₄ fluxes estimated using the TBLRC01 model were 2 - 3 folds larger than those using the TBLW92a,
348 TBLCL98, TBLW92b, and TBLCW03 models (Table 1 and Figure S5). However, there were no
349 significant differences in mean fluxes between the TBLW92a and TBLCL98 models ($p > 0.05$; Table 1 and
350 Figure S5), as well as between the TBLW92b and TBLCW03 models ($p > 0.05$; Table 1 and Figure S5).
351 Inland waters (river and reservoirs), similarly, Gao et al. (2014) and Musenze et al. (2014) found that the
352 estimated diffusive CH₄ fluxes derived from the TBLRC01 model were substantially greater than those
353 from other TBL models. These results indicated a potential bias in diffusive CH₄ flux estimation when
354 only a single TBL model was used. As Musenze et al. (2014) suggested, the lack of consensus among the
355 existing TBL models might be a major source for the uncertainty in GHGs accounting.

356 The difference of the estimated CH₄ fluxes between TBL models was likely a result of the variation
357 in weighting wind as a driver of gas transfer velocity (Musenze et al., 2014, Figure 1). Since these
358 wind-based models were developed using a range of techniques under different conditions in specific
359 systems (Gao et al., 2014; Musenze et al., 2014), their generalized applicability could be limited by the
360 local conditions (Bade, 2009; Musenze et al., 2014; Schilder et al., 2013). Therefore, the TBLCL98 and

361 TBLCW03 are more appropriate wind-based models for estimating k value and CH₄ fluxes in aquaculture
362 ponds, due to that their experiment environments (e.g., lentic ecosystem, a range of wind speeds) were
363 closest to the studied aquaculture ponds. Obviously, more in situ measurement is still needed to further
364 increase the accuracy of the estimate.

365 4.3. Comparison of CH₄ fluxes derived from FCs measurement and TBL models

366 Previous studies have shown that CH₄ fluxes estimated by *TBL* models tend to be lower
367 than those measured by *FCs* (Chuang et al., 2017; Duchemin et al., 1999; Li et al.,
368 2015; Matthews et al., 2003). This study also compared CH₄ fluxes measured by *FCs* and
369 those estimated by *TBL* models over the aquaculture season (Table 1 and Figure S5).

370 Although there were significant correlations between *TBL* model estimates
371 and *FCs* measurements ($p < 0.05$ in all cases), the agreement between the two methods varied
372 considerably between models (Fig. 4). The *TBLW92b* and *TBLCW03* models gave the
373 largest r^2 values (0.82 and 0.83, respectively) and good agreements with *FCs* measurements
374 (slope = 0.92 and 0.89, respectively), whereas *TBLCL98* yielded mean estimates virtually
375 identical to *FCs* measurements (slope = 1) but with larger variability around the mean
376 ($r^2 = 0.53$) (Fig. 4d–f). In contrast, *TBLLM86* vastly underestimated *FCs* fluxes
377 whereas *TBLRC01* grossly overestimated *FCs* fluxes (Fig. 4a and b). Approximately 80% of
378 the diffusive CH₄ fluxes estimated by the models fell within the range measured by
379 the *FC* method (see Fig. 5).

380 Balancing the consideration of overall agreement (regression slope) and estimate variability
381 (regression r^2), the *TBLW92b* and *TBLCW03* models appeared to give the best

382 approximations of *FCs* measurements. While previous studies showed that *FCs* were more
383 appropriate for determining greenhouse gas fluxes in heterogeneous environments such as
384 lakes and reservoirs (Cole et al., 2010; Duchemin et al., 1999; Murray et al., 2015; Vachon et
385 al., 2010; Wu et al., 2018), our results suggest that *TBLW92b* and *TBLCW03* models are
386 reliable alternatives for estimating CH₄ diffusive flux in shallow aquaculture ponds.

387 In addition to diffusive flux from the water column, bottom sediment could also contribute to
388 CH₄ emission via ebullition, especially in eutrophic, shallow aquaculture ponds. This is
389 illustrated by the differences in the measured CH₄ flux using *FCs* with and without gauze in
390 our aquaculture ponds (Figure S6). The CH₄ flux measured by *FCs* without gauze
391 ($2231.3 \pm 681.3 \mu\text{mol m}^{-2} \text{h}^{-1}$) were one to two orders of magnitude higher than that
392 by *FCs* with gauze ($75.0 \pm 12.5 \mu\text{mol m}^{-2} \text{h}^{-1}$) (Figure S6); from this ebullition was estimate
393 to contribute 96.6% to the total CH₄ emissions. Overall, our results showed that ebullition was
394 the primary path of CH₄ emission in aquaculture ponds, and that ebullitive flux vs. diffusive
395 flux could be easily resolved with a simple design of *FCs* with a detachable gauze.

396 4.4. Implications of the comparison between different methods

397 The *FCs* method is the popular technique for measuring CH₄ emissions due to its ability to
398 detect low fluxes and the simplicity of its operating principle (Bastviken et al., 2015; Lorke et
399 al., 2015; Musenze et al., 2014; Podgrajsek et al., 2014). However, the *FCs* method requires
400 time-consuming manual operation, which limits the frequency of measurements and can be
401 difficult to deploy in remote areas (Acosta et al., 2017; Morin et al., 2017). Improvement of the

402 global CH₄ budget would require high-resolution emission data covering large time and spatial
403 scales, which obviously is difficult to achieve with the *FCs* method.

404 Large-scale estimates of aquatic CH₄ emissions using *TBL* models has been gaining popularity
405 (Holgerson and Raymond, 2016; Martinez-Cruz et al., 2016; Musenze et al., 2014; Wang et al.,
406 2017) due to their simplicity, practicality and low cost. There are, however,
407 different *TBL* models to choose from, and the large differences in the model performances (Fig.
408 4) mean that selecting the appropriate model(s) would be critical, or otherwise large errors
409 would occur when upscaling the results from small ponds to the regional/global scale. Our
410 results suggest that *TBLW92b* and *TBLCW03* models could be used as effective and
411 convenient alternatives to *FCs* in shallow aquaculture ponds.

412 4.5. Limitation and future research

413 The *FCs* method is a common method to measure CH₄ fluxes from aquatic ecosystems.
414 However, *FCs* may create microenvironments that affect the boundary layer conditions
415 through, for instance, blockage of wind, change of atmospheric pressure at the measurement
416 point, and change in the gas transfer rate through pressure build-up (Duchemin et al.,
417 1999; Matthews et al., 2003; Musenze et al., 2014). For example, the turbulence resulted from
418 the chamber walls can enhance the efficiency of gas exchange and increase gas fluxes during
419 low wind conditions (Matthews et al., 2003; Xiao et al., 2016).

420 *TBL* models rely on the gas transfer velocity coefficient (kx), which itself is estimated from
421 some empirical wind-based models. Effects of artificial aeration, which is commonly done in
422 aquaculture ponds, on kx are unknown. More importantly, the *TBL* models ignore the effect of

423 buoyancy fluxes near the air-water interface on kx . An alternative is the surface renewal model
424 (SRM), which considers both wind speed and buoyancy (e.g., Czikowsky et al.,
425 2018; MacIntyre et al., 2010; MacIntyre et al., 2018).

426 The use of eddy covariance (EC) technique is increasingly popular as it can provide a better
427 characterization of the variation in CH₄ fluxes through quasi-continuous measurements
428 (Acosta et al., 2017; Morin et al., 2017; Xiao et al., 2014; Zhao et al., 2019). However, its
429 application in small water bodies (e.g., ponds) is limited by footprint contamination (Zhao et al.,
430 2019). Developing a practical and effective way to reduce the flux footprint and the
431 contamination from gaseous sources outside the water body will allow broader application of
432 EC method in the future.

433 Different methods have their own limitations; careful comparison and cross calibration would
434 be needed to increase the overall accuracy of these methods and to improve the global
435 CH₄ budget.

436 5. Conclusions

437 Despite the large CH₄ emission from ponds, there is limited information about the comparison
438 between different methods applied for the estimations of CH₄ fluxes across the water-air interface. In this
439 study, FCs and six TBL models were applied to estimate CH₄ fluxes from aquaculture ponds. Our results
440 indicate that dissolved CH₄ concentrations in the subtropical shallow aquaculture ponds were on average
441 ~87 times more oversaturated than the ambient air, and thus the pond surfaces acted as considerable
442 atmospheric CH₄ sources. The high organic matter loading contributed to the CH₄ supersaturated in the
443 aquaculture ponds. As the first attempt in aquaculture ponds, this study also compared the CH₄ fluxes

444 directly measured by using floating chambers (FCs) and estimated by six thin boundary layer (TBL)
445 models (TBLLM86, TBLW92a, TBLRC01, TBLCL98, TBLW92b, and TBLCW03). The diffusive CH₄
446 fluxes estimated by different TBL models were largely variable, but overall they were 27 - 300% larger
447 than those directly measured by FCs. The agreement between FCs-measured and model-estimated CH₄
448 fluxes was highest for the TBLW92b, TBLCW03 models. While our results suggested that the estimation
449 of CH₄ fluxes using a single TBL model could lead to high levels of uncertainty, the application of
450 TBLW92b and TBLCW03 models could provide a robust and simple way for characterizing CH₄ fluxes
451 over direct measurements using FCs. Our results suggest that the comparison of different methods and
452 selection of the most appropriate method(s) for determining CH₄ flux should be a top research priority to
453 improve the estimation accuracy of GHGs fluxes in aquaculture ponds and other aquatic ecosystems.

454 Declaration of competing interest

455 The authors declare that they have no known competing financial interests or personal relationships
456 that could have appeared to influence the work reported in this paper.

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739 **Table 1**

740 Summary of the *TBL* and *FCs* methods applied to measure CH₄ diffusive fluxes from the aquaculture ponds in Min River Estuary during the
 741 aquaculture period.

	<i>TBL</i> methods						<i>FCs</i> method
	<i>TBL</i> _{LM86}	<i>TBL</i> _{W92a}	<i>TBL</i> _{RC01}	<i>TBL</i> _{CL98}	<i>TBL</i> _{W92b}	<i>TBL</i> _{CW03}	
Minimum (μmol m ⁻² h ⁻¹)	0.6	1.3	5.6	1.9	1.9	1.3	1.3
Maximum (μmol m ⁻² h ⁻¹)	108.8	650.0	1079.4	527.5	454.4	428.8	476.3
Average (μmol m ⁻² h ⁻¹)	19.4	103.1	215.6	115.0	78.1	75.0	71.9
Standard deviation	23.1	130.6	236.3	122.5	91.3	86.9	88.8
Coefficient of variation	1.18	1.27	1.09	1.06	1.16	1.15	1.24

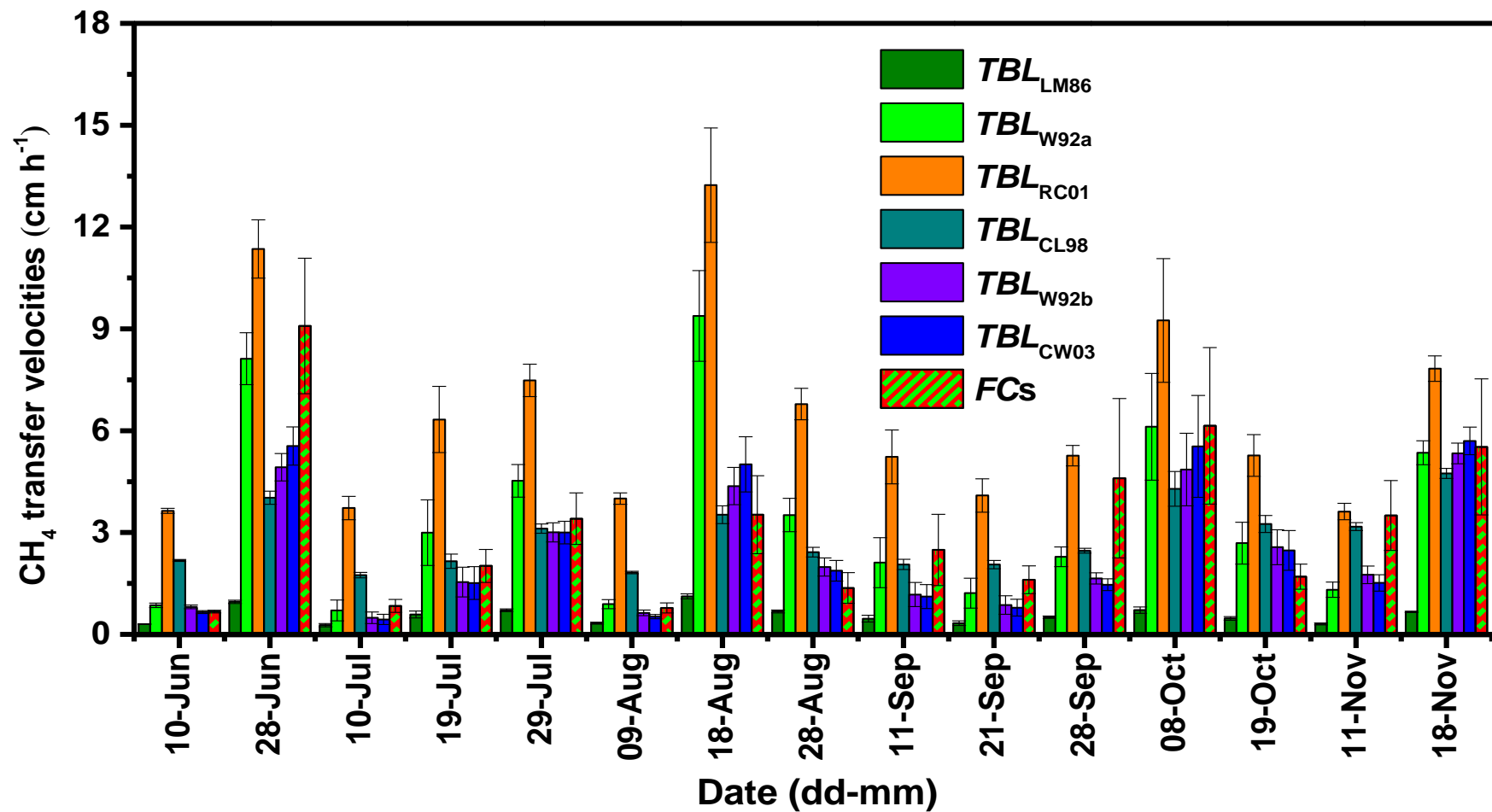
742 **Table 2**

743 Pearson correlation coefficients for dissolved CH₄ concentration, CH₄ diffusive fluxes and environmental variables from the aquaculture
744 ponds in Min River Estuary during the aquaculture period^a. Bold numbers denote correlation coefficients for significant relationships.

Environmental variables	Dissolved CH ₄ concentration	CH ₄ diffusive fluxes
Meteorological parameters		
Air temperature	0.214*	0.203*
Wind speed (<i>W_s</i>)	NS	0.281*
Atmospheric pressure	NS	NS
Water parameters		
Water temperature	NS	NS
Dissolved oxygen (DO)	NS	NS
TOC concentration	0.312**	0.296**
N-NO ₃ ⁻ concentration	-0.401**	-0.392**
Electrical conductivity (EC)	-0.361**	-0.185*

745 ^aThe symbols * and ** indicate significant correlations at the 0.05 and 0.01 levels, respectively. *n* = 135 for environmental variables and CH₄ diffusive fluxes from the aquaculture
746 ponds. CH₄ diffusive fluxes were directly measured using floating chambers method.

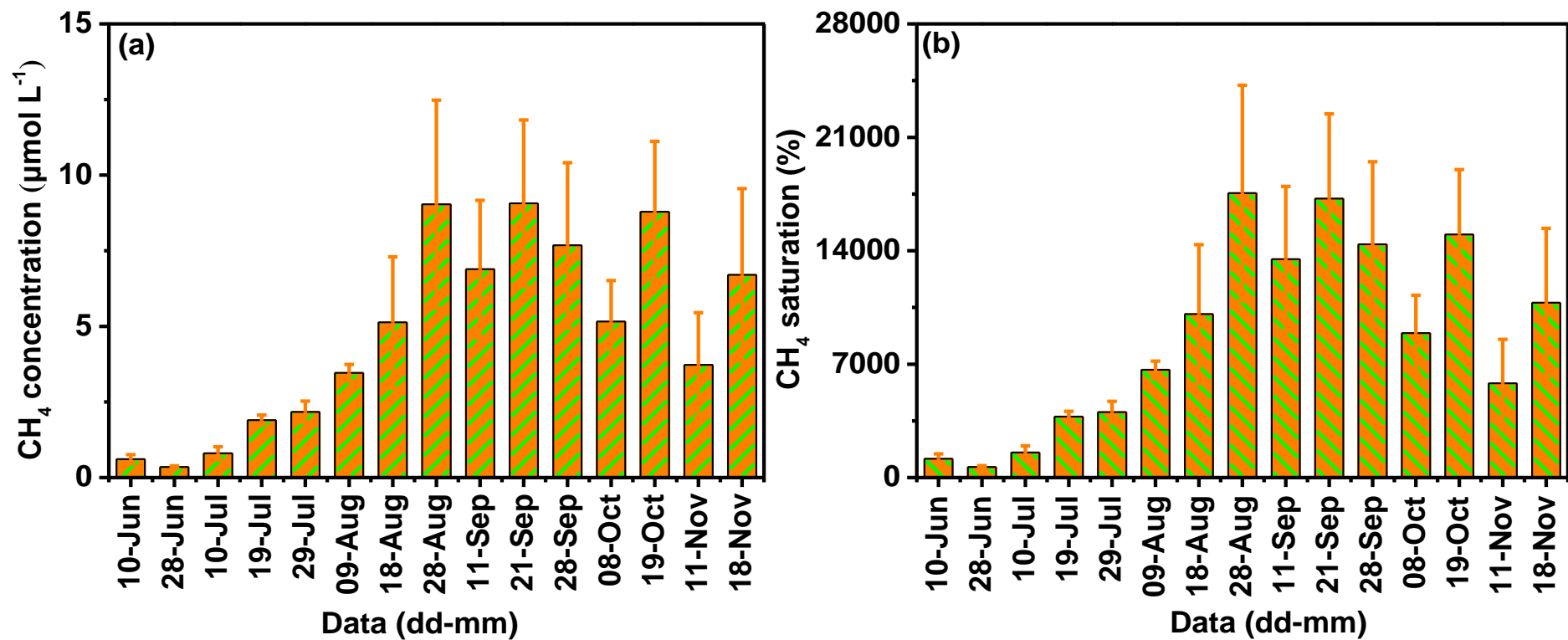
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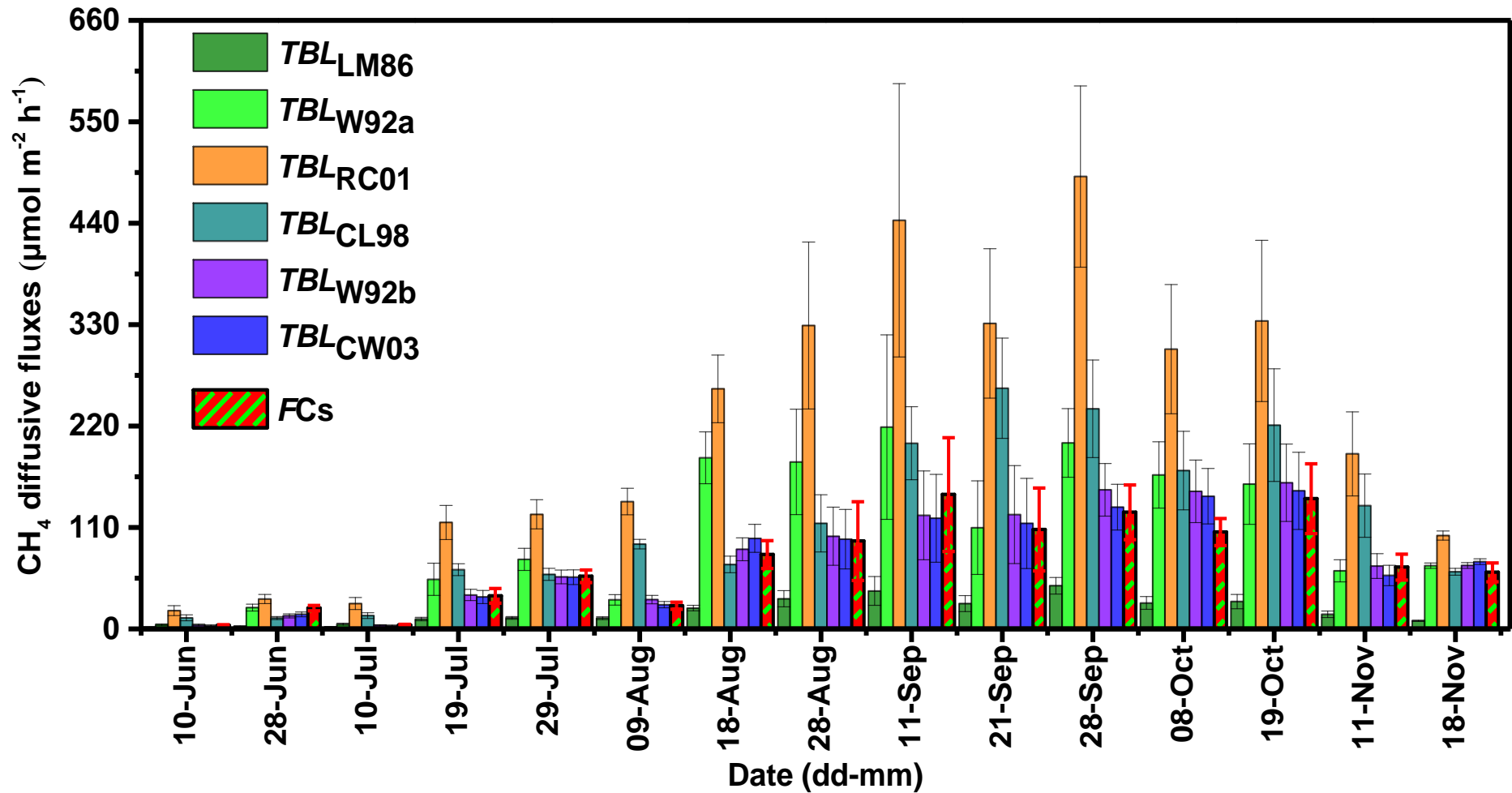
749 **Figure 1.** Temporal variation in CH₄ transfer velocities from the aquaculture ponds during the aquaculture period in the Min River

750 Estuary. Values represent the means of nine replicates samples, while the vertical lines indicate standard errors .



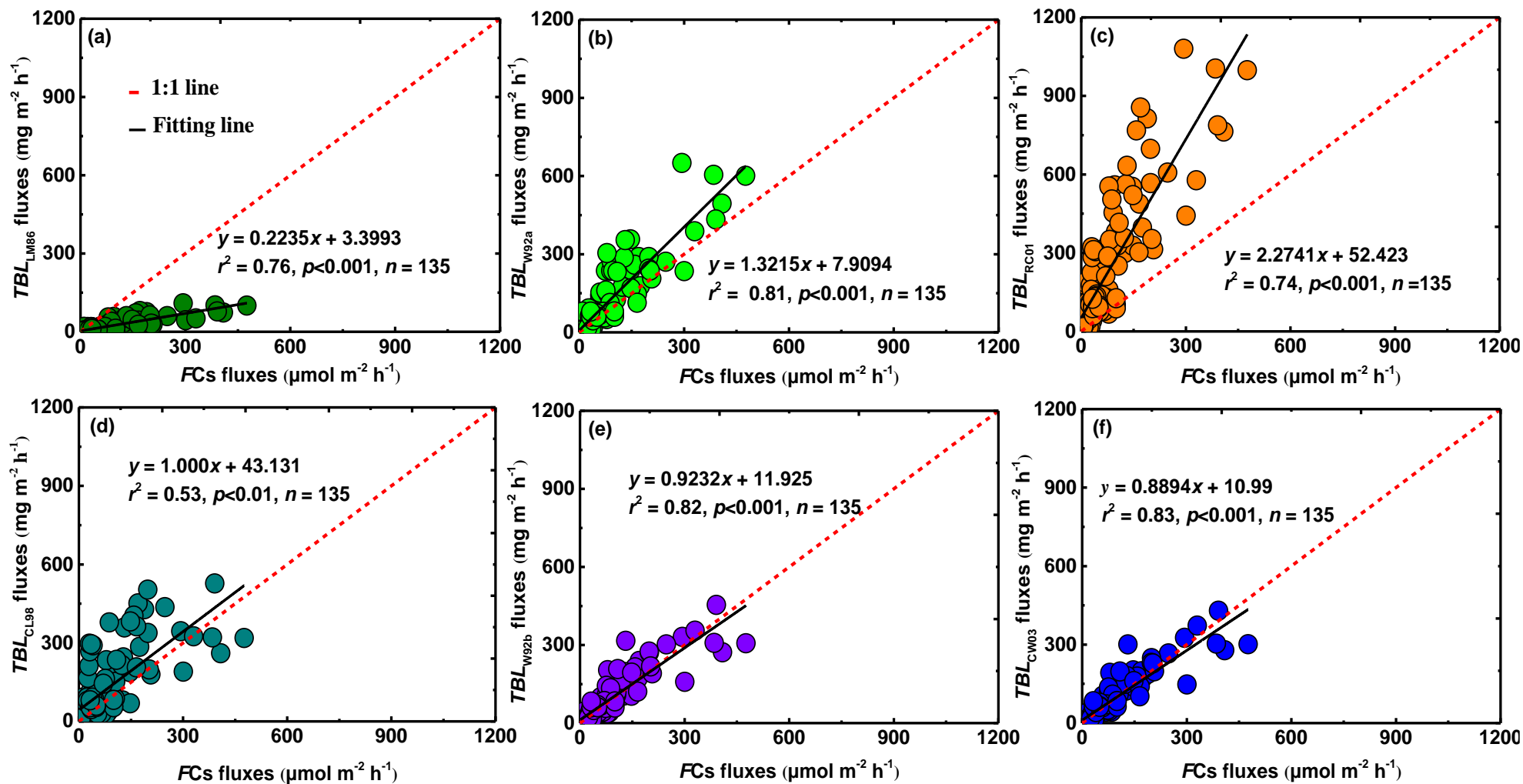
751

752 **Figure 2.** Temporal variation in (a) CH₄ concentration and (b) CH₄ saturation in the surface water (20 cm depth) of the aquaculture ponds in the
 753 Min River Estuary during the aquaculture period. Values represent the means of nine replicates samples, while the vertical lines indicate standard
 754 errors.



755

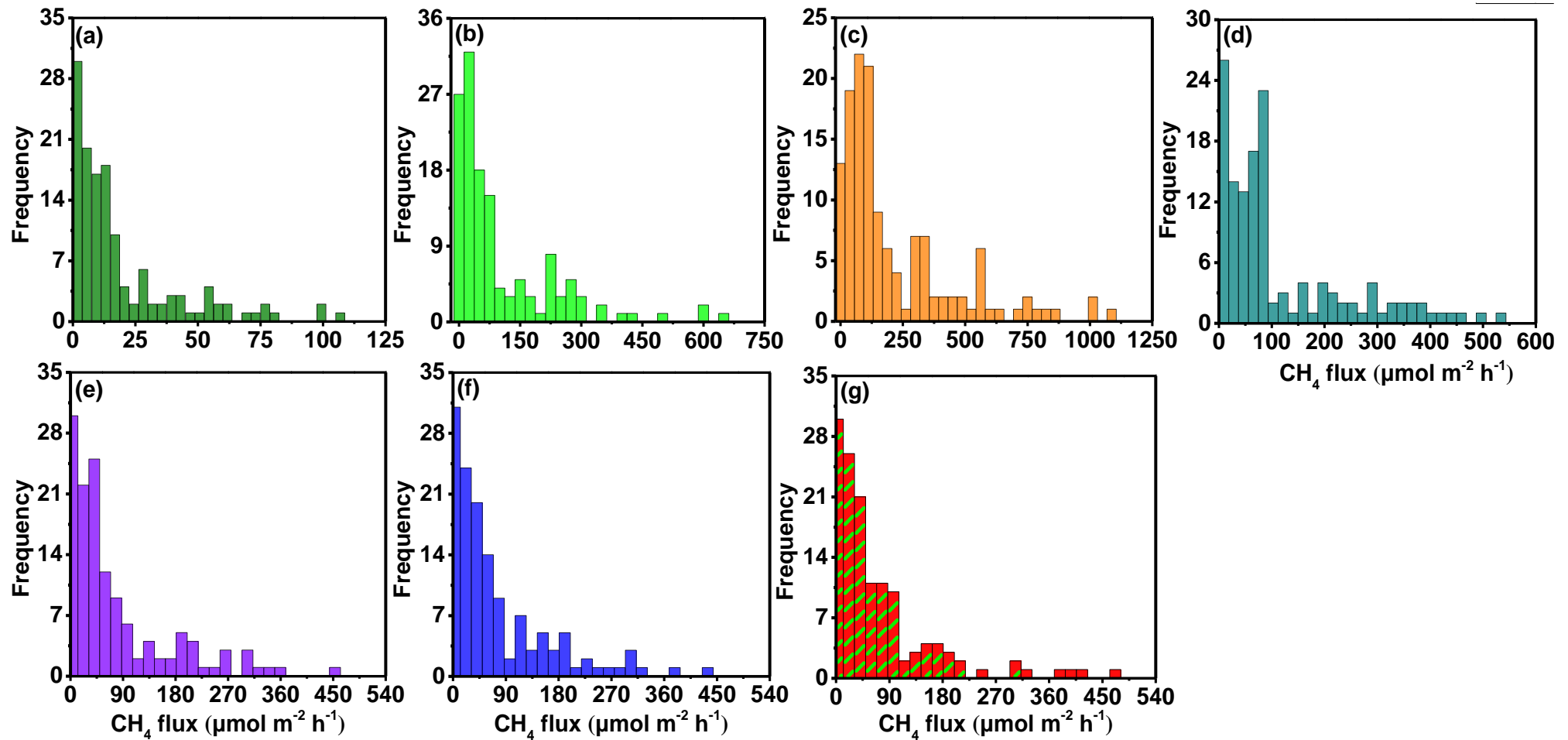
756 **Figure 3.** Temporal variation in CH₄ diffusive fluxes measured with the floating chamber method and the gas transfer velocity model methods
 757 during the aquaculture period from the aquaculture ponds in the Min River Estuary. Values represent the means of nine replicates samples, while
 758 the vertical lines indicate standard errors.



759

760 **Figure 4.** Comparison of CH_4 diffusive flux measured by using the *FCs* method and *TBL* models. Regression equation, linear correlation (r^2) and

761 significance (p) are also shown. Parameter bounds on the regression coefficients are 95% confidence intervals.



762

763 **Figure 5.** Frequency distribution of CH₄ diffusive fluxes from (a) *TBL_{LM86}*, (b) *TBL_{W92a}*, (c) *TBL_{RC01}*, (d) *TBL_{CL98}*, (e) *TBL_{W92b}*, (f) *TBL_{CW03}*, and

764 (g) FCs measurements at the aquaculture ponds in the Min River Estuary during the aquaculture period.

