Article

Methane Emissions from Surface of Mangrove River on Hainan Island, China

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Abstract: The surfaces of rivers are considered important sources of atmospheric methane (CH₄), however research on this topic is still constrained, especially in freshwater rivers and with the consideration of spatial heterogeneity. Three regions (upper reaches, midstream and downstream) were selected to examine the CH₄ fluxes from a freshwater river surface in a mangrove forest wetland from 2012 to 2013, using floating chambers. Results showed that the CH₄ fluxes varied significantly among the three regions, with the lowest fluxes at downstream ($0.50 \pm 0.20 \text{ mg m}^{-2} \text{ h}^{-1}$), and highest at upper reaches ($1.19 \pm 0.36 \text{ mg m}^{-2} \text{ h}^{-1}$). The average emission rate at midstream was $0.95 \pm 0.37 \text{ mg m}^{-2} \text{ h}^{-1}$. The methane flux also varied with seasons, with higher flux in rain-abundant seasons. On average, the CH₄ flux in our research river was $0.88 \pm 0.31 \text{ mg m}^{-2} \text{ h}^{-1}$, which was less than other tropical rivers. In addition, we found that the CH₄ flux was significantly correlated with the water characteristics of temperature and atmospheric pressure. Thereby, this study quantified the methane emission from a freshwater river surface in a tropical mangrove forest, enriching the existing knowledge of river surface CH₄ flux.

Keywords: methane emissions; coastal river; mangrove; water surface flux; salinity

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1. Introduction

Methane (CH₄) is an important greenhouse gas, and is the second-most abundant after CO₂ [1]. Previous research has shown that rivers could be important sources of atmospheric methane (CH₄), but this remains uncertain due to a lack of global studies [2,3]. Global water CH₄ emissions into the atmosphere were estimated at about 1.2–2.1 Pg_C·year^{−1} [4], of which more than 20% originated from freshwaters [5,6]. Research has also estimated that the CH₄ emission from rivers is about 1.5 Tg_{CH₄}·year^{−1} [4]. Despite their small area, rivers play an important role in the global methane budget [4].

CH₄ is produced in anaerobic environments and the production process can be divided into the following three steps: (1) complex organic matter is transformed into monosaccharides by hydrolytic enzymes (synthesized by hydrolytic fermentation bacteria), and further fermented to synthesize fatty acids, carbon dioxide and hydrogen; (2) fatty acids, carbon dioxide and hydrogen are oxidized to acetic acid, CO₂, and H₂; (3) acetic acids are oxidized to CH₄, or the CO₂ is reduced by H₂ to produce CH₄ by acetoclastic methanogenesis (AM) and hydrogenotrophic methanogenesis (HM), respectively [7–9]. According to the pathway of CH₄ formation in step (3), CH₄ formation always divides into two pathways: (1) fermentation of acetate by AM; (2) acetate oxidation during which CO₂ is reduced with H₂ by HM. The dominant pathway of CH₄ production is mainly determined by the substance supplying the methanogens [10].

In general, tropical regions were shown to have the highest CH₄ emissions [11]. High content of organic matter, strong heat and strictly anaerobic environments were favorable

for methanogens and were the main causes of high methane emission [12]. Freshwater rivers in these regions also have a large amount of carbon substrates [10] which supply abundant substance for methanogens, leading to a higher CH_4 emission. Large amounts of CH_4 emission from waters and rivers, especially through the water-gas interface, is an important source of CH_4 emissions [12].

Rivers in mangrove wetlands are well adapted to the transition zone between land and sea and the river water body has a natural salinity difference [13,14]. At present, there are many studies on swamps and lakes, and relatively few studies on rivers. Some studies have shown that freshwater rivers, though small in proportion to entire wetlands, contribute more than their share of methane emissions [15,16].

At present the study of river methane emissions occurs mainly in the Amazon in South America, the Congo River basin in West Africa, the Mississippi River basin in North America, Costa Rica in Central America, and in some of Europe's rivers [3,11,17–22]. In China, the research surrounding river CH_4 flux is mainly concentrated in the Pearl River watershed, the Yangtze River basin, and in the Yellow River [23–26].

Rivers are an important source of atmospheric CH_4 emission, especially in tropical areas [27]. We speculate that river-flow through the mangrove wetland may have a high CH_4 emission rate due to the high organic carbon [13,14] and varied salinity, which could inhibit the activity of methanogens and their reduction in CH_4 emission [15,28].

In tropical areas, rivers are characterized by fast substance-exchange such as available carbon, higher rainfall and higher oxygen levels [29]. These characteristics have led to the CH_4 emissions in this region being more complex than those in temperate areas. Natural mangrove wetlands are the largest and earliest nature reserves in China. With the characteristics of tropical rivers, the rivers distributed throughout natural mangrove wetlands are also contaminated by human waste, which further increases the organic carbon in the water. Until now, the CH_4 flux in such rivers has remained unknown, especially with consideration of spatial-temporal heterogeneity. Thus, the objectives of this study are: (1) to investigate the seasonal and spatial dynamics of CH_4 flux from a typical river in mangrove wetland; (2) to examine the influencing factors of CH_4 flux.

2. Materials and Methods

2.1. Study Site

The river we studied was located in Dongzhaigang National Nature Reserve ($19^{\circ}57' - 20^{\circ}01' \text{ N}$, $110^{\circ}32' - 110^{\circ}37' \text{ E}$), in the north east of Hainan province (Figure 1). Dongzhaigang National Nature Reserve is the first-founded (1980) and the largest mangrove forest reserve in China. It accounts for 44.51% of the mangrove forests of Hainan Island and holds the most abundant mangrove species.

Figure 1. Meteorological data of the study area from April 2012 to December 2013 (data were from a weather station set by the Institute of Tropical Forestry, Chinese Academy of Forestry).

The climate of Dongzhaigang National Nature Reserve is typical of tropical monsoon marine regions with an average rainfall of 1700–1933 mm, in which 80% occurs during May to October, and a mean annual temperature of 23.3–23.8 °C [30–36]. During the sampling period of 2012 to 2013, the average monthly air temperature was 25.78 °C, with the warmest monthly temperature of 29.94 °C in July 2013 and the coldest monthly temperature of 18.67 °C in December 2013. The rainfall in 2012 was 1657.0 mm (the data from January to March is absent), with a maximum rainfall of 351.2 mm in June and minimum rainfall of 18.6 mm in December. The rainfall in 2013 was lower (898.8 mm) than in 2012, with maximum rainfall of 251.4 mm in August and minimum rainfall of 3.4 mm in December (Figure 1).

The dominant plant species consisted of 26 mangrove species, such as *Bruquiera gymnorrhiza*, *Kadelia candel*, *Bruquiera sexangula*, *Ceriops tagal*, *Aegiceras corniculatum*, *Sonneratia apetala*, etc.

3 regions of the Yanfeng River were chosen for our research: (1) upper reaches (UP; 110°33'789" E, 19°56'909" N); midstream (MID; 110°34'729" E, 19°57'076" N), and downstream (DOWN; 110°35'138" E, 19°57'326" N) (Figure 1). The UP region is distal to the estuary, is rarely influenced by tides and its salinity is lowest. The DOWN region is near the estuary, is severely influenced by tides, and its salinity is highest. The MID region is the transition area between UP and DOWN. The hydrological environments of the three regions also differed.

2.2. Gas Sampling

Three plots at each region of the river were established to measure the CH₄ flux of the water surface, using vented closed chambers (26 cm in diameter, 46 cm in height) which consisted of two parts. One was made out of a cylindrical polycarbonate (PC) pipe, and the other was made out of a gas filled floating chamber (Figure 2c,d) [37,38]. Three plots were selected with a distance of about 20 m, and the distance to riverbank was 4 m. The CH₄ was measured monthly from February 2012 to December 2013. The gas was sampled from 8:30 a.m. to 11:30 p.m., with 10 mL disposable vacuum tubes at 10 min intervals over a 30 min period after floating on the water. The air temperature inside the chamber was measured at 0 and 30 min. The concentration of CH₄ in the samples was determined by a gas chromatograph (Agilent 7890A, Agilent Co., Santa Clara, CA, USA) equipped with a flame ionization detector (FID), which operated at 350 °C. The Nickel conversion furnace temperature was maintained at 350 °C. The column temperature was maintained at 250 °C and the carrier gas was pure nitrogen at a flow rate of 20 mL min^{−1}, the combustion gas was air at a flow rate of 500 mL min^{−1}, the combustion supporting gas was pure hydrogen with a flow rate of 50 mL min^{−1}.

The flux (mg CH₄ m^{−2} h^{−1}) of CH₄ was calculated as:

$$J = \frac{dc}{dt} \frac{M}{V_0} \frac{P}{P_0} \frac{M}{V_0} \frac{T_0}{T} H$$

where d_C/d_t (mol·h^{−1}) is the rate of concentration change; M (mg mol^{−1}) is the molar mass of CH₄; P (Pa) is the atmospheric pressure of the sampling site; T (K) is the absolute temperature at the sampling time; V_0 (m³), P_0 (Pa), T_0 (K) are the molar volume, atmospheric pressure, and absolute temperature, respectively, under the standard condition; H (m) is the chamber height over the water surface.

Figure 2. Sampling site—Dongzhaigang National Natural Reserve in Hainan Province (a,b) and CH_4 sampling device (c) and process (d) ((c,d) were photographed by Wei Guan).

2.3. Water Sampling and Analysis

Water was collected in 10 mL glass bottles monthly and randomly at our sampling site, and was transported to the lab at 4 °C and analyzed within one week. The characteristics of water temperature, salinity, pH, oxidation–reduction potential (ORP) and conductivity were measured during sampling using a portable digital meter (YSI ProPlus Portable multi-parameter water quality analyzer, USA). The water and air temperature were also measured using a handheld digital thermometer, and air pressure was measured monthly. In the lab, water DOC, NO_3^- -N, NH_4^+ -N and PO_4^{3-} -P concentrations were analyzed using a continuous flow analytical system (SKALAR San⁺⁺, SKALAR Co., Breda, The Netherlands). Before DOC measurement, the sampled water was filtered through a 0.45 μm cellulose acetate membrane filter, and before other index measurements the sampled water was filtered through filter paper to eliminate the impurities.

2.4. Statistical Analysis

The difference in CH_4 emissions from different regions of the river was analyzed by one-way analysis of variance (ANOVA). ANOVA was also used to compare the differences in water temperature, salinity, pH, oxidation–reduction potential, conductivity, DOC, NO_3^- -N, NH_4^+ -N and PO_4^{3-} -P among the three regions. The relationships between CH_4 flux and CH_4 concentration in water, air and water temperatures and DOC concentration were detected by regression analysis. All data were analyzed using SPSS 22.0 for Windows (SPSS Inc., Chicago, IL, USA) and all graphs were drawn using Origin Pro 8.1.

3. Results

3.1. Water Characteristics and Environmental Factors

Of the environmental factors, water salinity and conductivity significantly increased from UP to DOWN ($p < 0.05$ for both). The same was true for pH and PO_4^{3-} -P ($p < 0.05$ for both). Other characteristics did not show any significant difference among the three regions

(Table 1). In addition, salinity, water temperature, conductivity, ORP, and air temperature also varied seasonally (Figure 3a–g).

Table 1. Environmental factors and properties of water in Yanfeng river.

Environmental Factors	Upper Reaches	Midstream	Downstream
Air Temperature (°C)	30.11 ± 3.95	29.3 ± 4.77	30.75 ± 6.09
Salinity (‰)	6.95 ± 6.16 b	12.23 ± 8.65 ab	15.52 ± 8.20 a
Water Temperature (°C)	26.86 ± 4.74	27.27 ± 4.22	27.13 ± 4.35
Atmospheric pressure (mbar)	1008.94 ± 5.87	1008.72 ± 5.76	1008.88 ± 6.52
Conductivity (µS/cm)	1.31 ± 1.29 b	2.16 ± 1.30 ab	2.78 ± 1.20 a
pH	7.30 ± 0.26 b	7.44 ± 0.27 b	7.72 ± 0.34 a
ORP (mv)	71.39 ± 47.74	74.58 ± 45.36	84.84 ± 49.26
DOC (mg/L)	2.98 ± 2.60	3.63 ± 2.34	2.61 ± 2.38
NH ₄ ⁺ -N (mg/L)	0.48 ± 0.44	0.74 ± 0.73	0.35 ± 0.32
NO ₃ ⁻ -N (mg/L)	0.27 ± 0.26	0.24 ± 0.23	0.23 ± 0.22
PO ₄ ³⁻ (mg/L)	0.15 ± 0.14 b	0.23 ± 0.16 b	0.57 ± 0.56 a

Values are means ± standard error. Letters indicate significant difference (LSD test, lowercase: $p < 0.05$).

Figure 3. Variations of water variables during sampling months. (a) Salinity; (b) WT: water temperature; (c) AP: Atmospheric pressure; (d) Cond: Conductivity; (e) pH; (f) ORP: Oxidation-Reduction Potential; (g) AT: air temperature.

3.2. CH₄ Fluxes

The CH₄ fluxes of the three regions varied by month (Figure 3). At the UP region, higher CH₄ flux was observed during June to September 2012 and May and August 2013. At the MID region, higher CH₄ flux was observed during Feb. to Apr. and June to Aug. 2013. At the DOWN region, higher CH₄ flux was observed during the later months of each year. In summary, higher CH₄ fluxes were observed in rain-abundant seasons at the UP and MID regions and later in the rainy season at the DOWN region.

Of the three regions, the UP region had significantly higher CH₄ fluxes than the MID and DOWN regions in 2012 ($p < 0.01$). Additionally, the UP and MID regions had relatively higher CH₄ fluxes than DOWN regions in 2013 ($p > 0.05$). On average, the CH₄ fluxes were higher at the UP (1.19 ± 0.36 mg CH₄ m⁻² h⁻¹) and MID (0.95 ± 0.37 mg CH₄ m⁻² h⁻¹) regions than at the DOWN region (0.49 ± 0.20 mg CH₄ m⁻² h⁻¹; $p < 0.01$; Figures 4 and 5).

Figure 4. Seasonal variations in CH₄ flux in the three regions.

Figure 5. The average CH₄ fluxes of the three regions (LSD test, uppercase: $p < 0.01$).

3.3. Relationship between Methane Fluxes and Factors

Correlation tests were conducted among CH₄ fluxes and water environment factors. The results showed that the significantly correlated variables were different among the three regions. At the UP region, water temperature was significantly positively correlated with CH₄ fluxes and atmospheric pressure was significantly negatively correlated with CH₄ fluxes. The MID and DOWN regions did show any significant correlations with water variables (Table 2).

Table 2. Correlations among CH₄ fluxes and environment factors.

Regions		Salinity	WT	AP	pH	Cond	ORP mV	AT
UP	<i>r</i>	−0.29	0.490 *	−0.505 *	−0.06	−0.22	0.06	0.26
	<i>p</i>	0.24	0.04	0.03	0.81	0.40	0.85	0.27
	<i>n</i>	18	18	18	18	170	15	20
MID	<i>r</i>	0.08	0.25	−0.39	0.01	0.10	0.28	−0.11
	<i>p</i>	0.73	0.30	0.09	0.97	0.68	0.30	0.63
	<i>n</i>	19	19	19	19	18	16	20
DOWN	<i>r</i>	−0.38	0.41	−0.29	−0.06	−0.32	−0.50	0.41
	<i>p</i>	0.11	0.08	0.23	0.81	0.20	0.05	0.07
	<i>n</i>	19	19	19	19	18	16	20

WT: water temperature; AP: Atmospheric pressure; Cond: Conductivity; AT: air temperature (*: $p < 0.05$).

4. Discussion

4.1. Comparisons with Other Studies

The CH₄ flux in our study ranged from 0.47 to 1.30 mg CH₄ m^{−2} h^{−1}, which was severely lower than in other research [3,39–41] (Table 3). The possible reason was that we neglected the pathway of ebullition of CH₄ emission, resulting in relatively lower CH₄ flux. With deeper research about CH₄ flux in waters and lakes, results have found that ebullition is one of the dominant pathways in open water CH₄ flux [4]. In general, the CH₄ flux in tropical and subtropical open water/water bodies was relatively higher, such as in the open water in India and South America [40,41]. Polluted water CH₄ fluxes were also higher than other water bodies due to their relatively high organic carbon, such as one open water-source in India [3]. On the contrary, the CH₄ flux in water bodies with lower organic carbon in sediments was lower than natural water bodies [42]. In addition, macrophytes were also one of critical factors enhancing water bodies CH₄ flux, as was observed in South America [40]. Macrophytes could supply an abundance of organic matter into waters, which is the original substance during CH₄ production, and may therefore lead to higher CH₄ flux [43].

Table 3. Comparison of CH₄ flux of rivers in other regions globally.

Name of River	Country	Type	CH ₄ Flux (mg m ^{−2} h ^{−1})			Reference
			Minimum	Maximum	Mean	
Yennisei River	Russia	Mires	ND	ND	1.04	[39]
Miranda River	Brazil	Open water	0.067	91.1	5.93	[40]
Adyar River	India	Open water	0.0013	76.1	0.53–15.3 *	[41]
Andhra Pradesh	India	Water bodies	0.006	34.73	5.2	[3]
Sundarbans	Bangladesh	Mangrove	−0.66	1.33	0.67	[44]
Yanfeng river	China	Open water	0.011	5.156	0.96	This study

*: mean fluxes at different locations of the river; ND: no data.

4.2. Spatial and Seasonal Variations of CH₄ Fluxes

Without four seasons, time frames in our research were divided into rainy season (end of March to the beginning of November) and dry season (beginning of November to the end of March). Some research has showed that heavy rain enhances CH₄ emission in North India [42] with obviously seasonal dynamics. Some research has showed that the CH₄ fluxes did not vary with seasons, such as in Amazon River basin [45]. Research in Pantanal also showed that the emission of CH₄ was slightly higher during the rainy season than the dry season [40]. The seasonal dynamic of CH₄ fluxes in tropical rivers is still controversial. In our research, we found that the CH₄ fluxes were higher in the rainy season than in the dry season.

The CH₄ fluxes in our research also had significant spatial variability, with the highest flux at the UP region in 2012, and relatively higher flux at the UP and MID regions in

2013. Taken together, a relatively higher emission rate was observed at the UP region. The abnormal or extreme value of the middle reaches is mainly due to the presence of the town and severe human activity.

4.3. Effect of Environmental Factors on CH₄ Fluxes

The heterogeneity of CH₄ throughout seasons and sites mainly resulted from the variation of water characteristics [17]. Of the water characteristics, salinity is one of the most critical factors. Numerous studies have shown that higher salinity consistently reduces CH₄ emission in waters by inhibiting methanogen activities [46]. The inhibiting effect of salinity on CH₄ emission could explain why CH₄ flux was higher in the rainy season. In the rainy season, an abundance of water reduces water salinity and its inhibiting effect on methanogens, together resulting in a higher CH₄ flux [13,38]. The inhibiting effect of salinity on methanogens also could explain the higher CH₄ flux in the UP region. The UP region, which is far from the bay and was little-affected by tides, has a relatively low salinity and higher CH₄ flux.

Water temperature and air pressure were also critical factors in influencing CH₄ flux, as observed in our study. Research has showed that water temperature affects CH₄ flux through regulating methanogen activity. Atmospheric pressure is always negatively correlated with CH₄ flux, for higher air pressure inhibits the emission of CH₄ from water to air. Higher air pressure also reduces the transportation of bubbles to air, reducing CH₄ flux. At our research site, extreme weather such as typhoons always causes low pressure and an increase in CH₄ flux.

5. Conclusions

This research quantified the CH₄ flux of a tropical river in a mangrove wetland. The results showed that CH₄ flux varied significantly during rainy and dry seasons, and varied among regions of the river. In our river, the UP region always showed higher CH₄ flux than the MIN and DOWN regions due to higher salinity which inhibits the activity of methanogens and reduces CH₄ production. Water temperature also affected CH₄ flux through regulating methanogen activity. Air pressure physically regulated CH₄ flux through control of the transportation of CH₄ from water to air. Our research could enrich the available data on river CH₄ flux in tropical mangrove wetlands.

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