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Peripheral freshwater deltaic wetlands are hotspots of methane flux in the coastal zone

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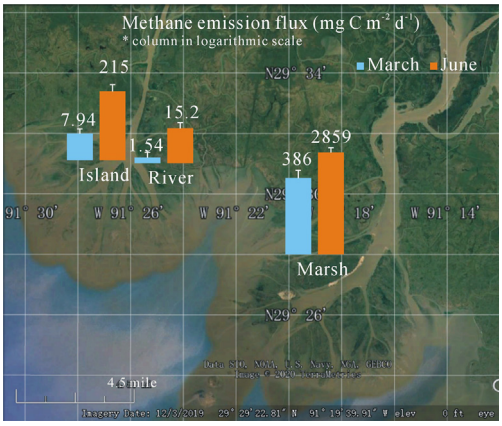
HIGHLIGHTS

- Methane (CH_4) fluxes were one-two order of magnitude greater in peripheral freshwater marshes than those in the active delta.
- High mineral content delta marshes and river channels had low CH_4 emissions.
- Marshes within the freshwater halo of deltas but outside of active sedimentation can be hotspots of CH_4 production in coast.
- Deltaic systems have a high spatial heterogeneity of CH_4 fluxes.

GRAPHICAL ABSTRACT



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ABSTRACT

Methane (CH_4) emissions are low in the coastal zone due to a higher redox poise, related to sulfate reduction. However, river deltas are a potential source of CH_4 flux in coastal zones globally, due to fresh condition and high primary production. The goal of this study was to seasonally measure CH_4flux at three different geomorphic settings (newly forming island, river channel bottom and established freshwater marsh) within the Wax Lake Delta, Louisiana, USA. CH_4 flux rates were 386 ± 327 mg C m^{-2} d^{-1} in March and 2859 ± 1286 mg C m^{-2} d^{-1} in June at the freshwater marsh site. At the island site, CH_4 flux was significantly smaller at 7.94 ± 3.57 mg C m^{-2} d^{-1} in March and 215 ± 153 mg C m^{-2} d^{-1} in June while at adjacent river channel bottom site, CH_4 flux was lowest at 2.49 ± 3.38 mg C m^{-2} d^{-1} in March and 19.5 ± 1.12 mg C m^{-2} d^{-1} in June at the air-water interface. CH_4 emission rates show significant spatial heterogeneity with rates up to two orders of

Abbreviations: CH_4, methane; C, carbon; CO_2, carbon dioxide; WMO, World Metrological Organization; GAW, Global Atmosphere Watch; SOC, sediment/soil organic carbon; SO_4^{2-}, sulfate radical; WLD, Wax Lake deltas; PVC, polyvinyl chloride; N_2, nitrogen gas; DOC, dissolved organic carbon; TOC, total organic carbon; TC, total carbon; BD, bulk density; MC, moisture content; C_w, CH_4 equilibrium concentration in water; C_a, corresponding air CH_4 concentration; β, Bunsen Solubility Coefficient; $F_{a\text{-}w}$, CH_4 flux; $C_{R\text{-}w}$, measured CH_4 concentration in the river water; $C_{S\text{-}a}$, saturation concentration of CH_4 in water; K, transfer coefficient for water turbulence; Sc, Schmidt value for CH_4 corrected for in situ water temperature; μ, long-term wind speed at 10 m height above the river; $F_{s\text{-}w}$, CH_4 flux at sediment-water interface; $C_{B\text{-}w}$, measured CH_4 concentration in the water at the beginning of incubation; $C_{E\text{-}w}$, measured CH_4 concentration in the water at the end of incubation; A, area of sediment core section; T, incubation time; $F_{Fick(s\text{-}w)}$, diffusion CH_4 flux; ∂C, CH_4 concentration gradient; ∂Z, diffusion distance; $C_{P\text{-}w}$, CH_4 concentration in sediment pore water; $C_{R\text{-}w}$, CH_4 concentration in the river water; Ds, effective diffusion coefficient of CH_4 in sediment pore water; Dw, measured diffusion coefficients of CH_4 in water; OM, organic matter; NEP, net ecosystem productivity.

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Carbon sequestration
Delta
Wetland succession
Marsh

magnitude greater at the marsh site at the periphery of the delta, related to greater soil total C. Therefore regions within the active delta do not provide a significant source of methane, due to a lack of soil C, despite freshwater conditions. However, marshes at the periphery within the halo of fresh water, populated with established plant communities can be significant hotspots of CH_4 emissions, despite their location within the coastal zone.

1. Introduction

Methane (CH_4) has significant global warming potential, which is ~28 times greater than carbon dioxide (CO_2) on a mass basis over a 100-year time frame [IPCC, 2013], and is also involved in a number of atmospheric chemical reactions [Cicerone and Oremland, 1988], playing a significant role in global climate change. A recent analysis of the World Metrological Organization -Global Atmosphere Watch (GAW) Program reports that globally averaged CH_4 concentration has reached 1869 ± 2 ppb, which is 259% of pre-industrial levels (~722 ppb) [WMO, 2019]. Although atmospheric CH_4 concentration increases are primarily due to increased emissions from anthropogenic sources, wetlands are the single largest, natural source of CH_4 with median emissions ~164 Tg yr^{-1}, which is about 1/3 of total global emissions [Bridgham et al., 2013]. Considerable research has been conducted on CH_4 fluxes for natural ecosystems around the world (e.g., northern peat [Moore et al., 1990; Roulet et al., 1993], estuarine/coastal wetland [DeLaune et al., 1983; Alford et al., 1997], and shallow water bodies [DeLaune et al., 1983; Wang et al., 2009a]). However, CH_4 emissions from global aquatic ecosystems and deltaic systems, in particular, are still largely uncertain in space and time [Bridgham et al., 2013].

In wetlands, primary plant production is a key factor determining the rate of CH_4 emissions [Whiting and Chanton, 1993], as the plants supply readily available C substrates through litter production and root exudates for methanogenic bacteria [Whiting and Chanton, 1992; Joabsson et al., 1999; King et al., 2002; Öquist and Svensson, 2002; Ström et al., 2003]. Therefore, the sediment/soil organic carbon (SOC) has a critical stimulatory effect on CH_4 production [DeLaune et al., 1983; Crozier and DeLaune, 1996]. In coastal deltaic regions, established vegetative cover can promote sediment accumulation through wetland vertical accretion [Elsey-Quirk et al., 2019]. In recent years, researchers have emphasized the importance of coastal vegetated regions acting as highly efficient C sinks [Chmura et al., 2003; Duarte et al., 2005; Bouillon et al., 2008; Lo Iacono et al., 2008; Duarte et al., 2010; Kennedy et al., 2010; McLeod et al., 2011; Sapkota and White, 2019) which has been described as "blue carbon" by Nellemann et al. (2009) with potential economic value (Sapkota and White, 2020).

Coastal systems are generally considered to be very low contributors to the global atmospheric pool of CH_4 due to the predominance of SO_4^{2-} reducing bacteria, whose activity poises the redox level above what is needed to promote methanogenesis (Poffenbarger et al., 2011; Steinmuller et al., 2019; Steinmuller et al., 2020). With tidal action, higher salinity water containing ample $SO_4^{2-}$is continually replaced to the marsh over time depressing methanogenesis. Deltaic systems, despite their proximity to the coastal ocean, and in some cases extending well out into the coastal ocean, are primarily comprised of fresh or oligohaline wetlands due to high discharges of fresh water through the delta channel network [Roberts et al., 2015]. While deltaic systems are renowned for high C accretion rates and land-building potential [DeLaune et al., 2016], both important functions in face of rising sea level and climate change, these systems are underrepresented in the literature with respect to CH_4 emission rates. Due to their prevailing freshwater influence, the implication is that it would be possible for these systems to be hotspots for methane production and flux despite their position within the coastal zone. To test this hypothesis, CH_4 emissions were measured from three geomorphic setting within an actively forming delta which represent stages of deltaic geomorphic evolution. [Roberts et al., 2015]. The objectives of the study were to 1) quantify the flux of CH_4 emission from the sediment/soil in the aforementioned geomorphic settings in March and June 2) compare the CH_4 fluxes with literature values of those comparably measured wetland units from abandoned lobes of the Mississippi River delta and 3) briefly discuss the net carbon sink function of the Mississippi River delta in context of measured and literature CH_4 emissions within the coastal deltaic complex.

2. Methods and materials

2.1. Study area

The Atchafalaya River is the largest distributary of the Mississippi River, which has 2 notable prograding sub-deltas within the Atchafalaya River Delta complex; the Atchafalaya River and Wax Lake deltas (WLD). Both of these two deltas are building land into the shallow, low energy (mean wave height ~ 0.5 m), microtidal (mean tidal range 0.35 m) Atchafalaya Bay [Rosen and Xu, 2013] and display typical lobate delta growth representative of a river- dominated delta, based on Galloway [1975]. From 2015 to 2020, the average monthly air temperature in March and June were 13.5 ± 2.1 °C and 27.1 ± 1.1 °C, and the River gage height were 5.9 ± 2.1 ft and 6.2 ± 2.6 ft respectively at the USGS 07381600 Lower Atchafalaya River at Morgan City Station (U.S. Geological Survey database). The Wax Lake delta is located roughly 32 km southwest of Morgan City, LA, west and adjacent to the Atchafalaya delta (Fig. 1). Three sampling sites were established in this area. Two sampling sites were located in the active delta region; on an actively forming island (site 11, 29° 30′ 29.68 N, 91° 25′ 35.22 W) and adjacent river channel bottom (site 12, 29° 30′ 30.03 N, 91° 25′ 34.24 W). The other site (10, 29° 31′ 11.32 N, 91° 20′ 52.15 W) is a more established marsh which receives sediment and freshwater input located 9 km from the first site but within the freshwater influence of the river (Fig. 2). We compare these rates with literature data from abandoned delta lobes of the Mississippi River delta complex to determine the status of these wetlands as carbon sinks from a climate forcing perspective. Research methodology is showed in Fig. 3.

2.2. Field sampling

Static diffusion chambers were used to collect CH_4 samples at the established marsh site and newly forming island site. Intact core incubations were used for measuring CH_4 flux at the sediment-water interface at the flooded river channel bottom site. In order to determine flux at the submerged site, sediment pore water and overlying river water CH_4 concentrations were used to calculate the CH_4 flux at sediment-water interface using Fick's law. From this, a two-layer model of diffusive gas exchange was also used to calculate the flux at air-water interface to provide a meaningful comparison to the other two sites.

Flux measurement using static diffusion chambers were taken twice in 2012 (March and June, air temperature and temperature at 5 cm soil depth in Table 2). The static chamber was composed of a removable chamber cover and the permanently installed base containing a U-shape groove (30 cm × 30 cm × 30 cm). At the start of the field campaign, triplicate bases were inserted into the soil at both the island and established marsh sites two weeks prior to any flux measurement to remove any artifacts in gas flux due to physical disturbance. At

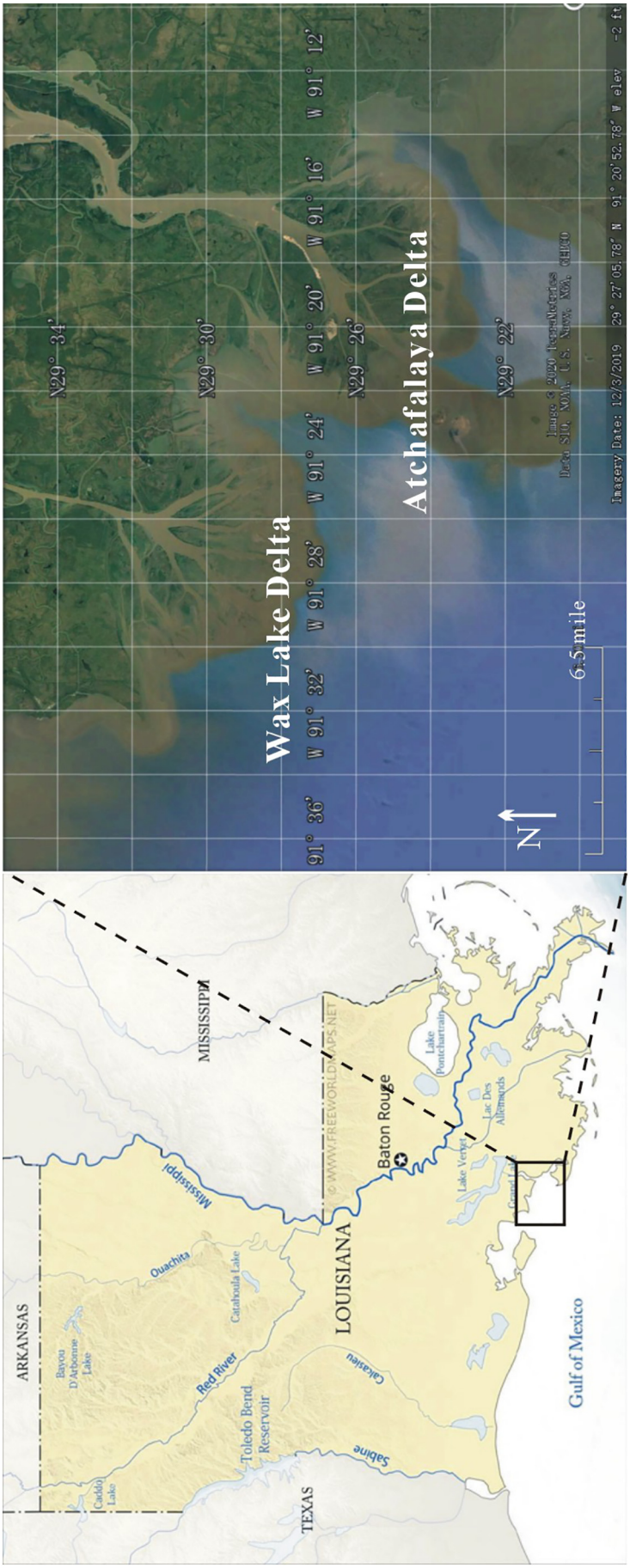

Fig. 1. Location of the Wax Lake and Atchafalaya Deltas along Louisiana's Gulf Coast.

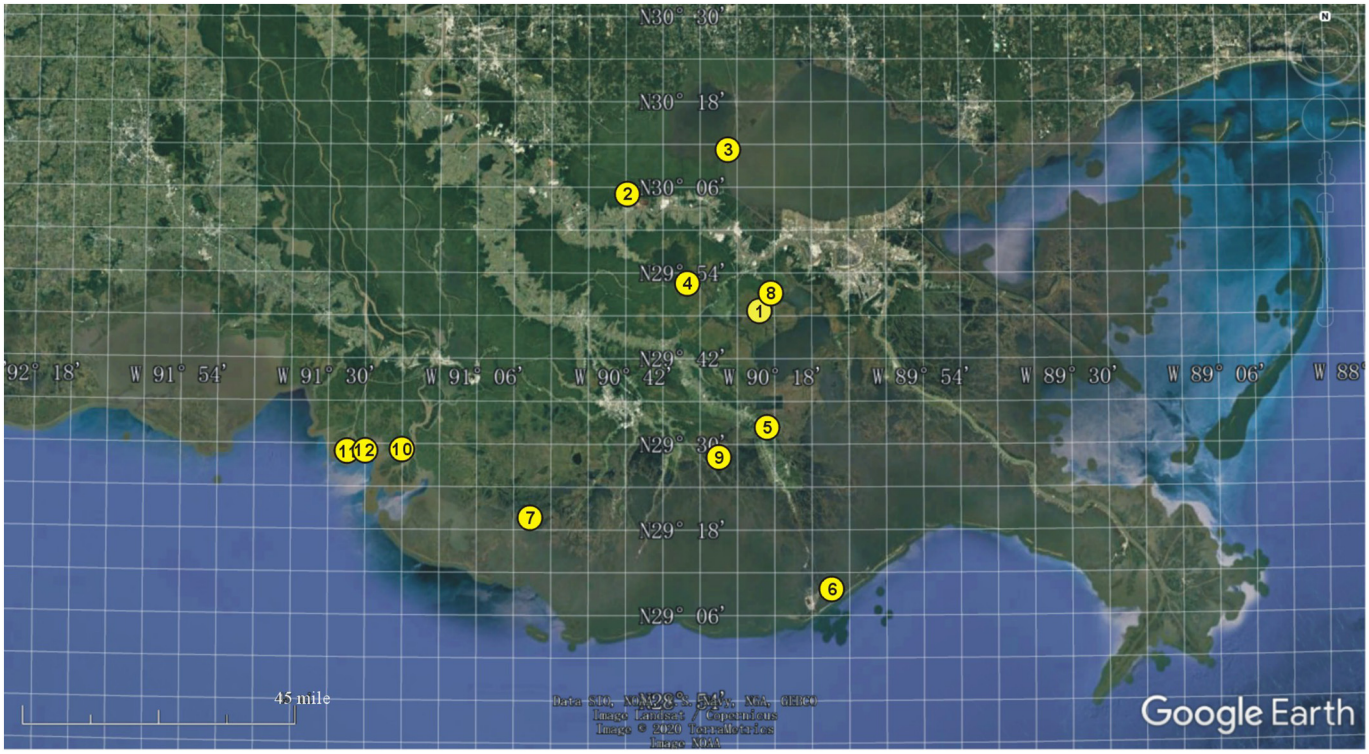

Fig. 2. Location of published research on methane emission rates within the Mississippi River deltaic plain (sites 1-9) for comparison with data from this study (sites 10-12). (google maps)

sampling, chambers were placed over the bases which were sealed with water added to the groove and then a 20 cm^3 headspace sample was drawn out from each chamber after 0, 20, 40 and 60 min to determine the CH_4 flux. At the river channel bottom site, which was submerged to 1 m water depth, four PVC diffusion cells (Ø: 3.4 cm, long: 50 cm) were inserted 30 cm into the sediment, and then sealed by rubber stoppers fitted with a septum. Replicate, five mls river water samples were injected into 12 ml sealed vials by syringe for determination of dissolved aqueous CH_4 concentrations. The vials were pre-vacuumed and flushed by high purity (99.99%) N_2 gas prior to sampling. Another three, 240 ml water samples were collected for measurement of water quality parameters. Three polycarbonate cores (~20 cm) were collected (Ø: 8 cm, long: 50 cm) from the established marsh and newly forming island sites for sediment characterization. Air temperature, water temperature and soil temperature at the 5 cm depth were measured in field. Water and gas samples were stored on ice and transported immediately back to lab with the intact sediment cores.

2.3. Laboratory incubation, sampling and analyses

After return to laboratory, three diffusion cells were immediately pre-incubated in a temperature-controlled incubator for 5 h to mimic

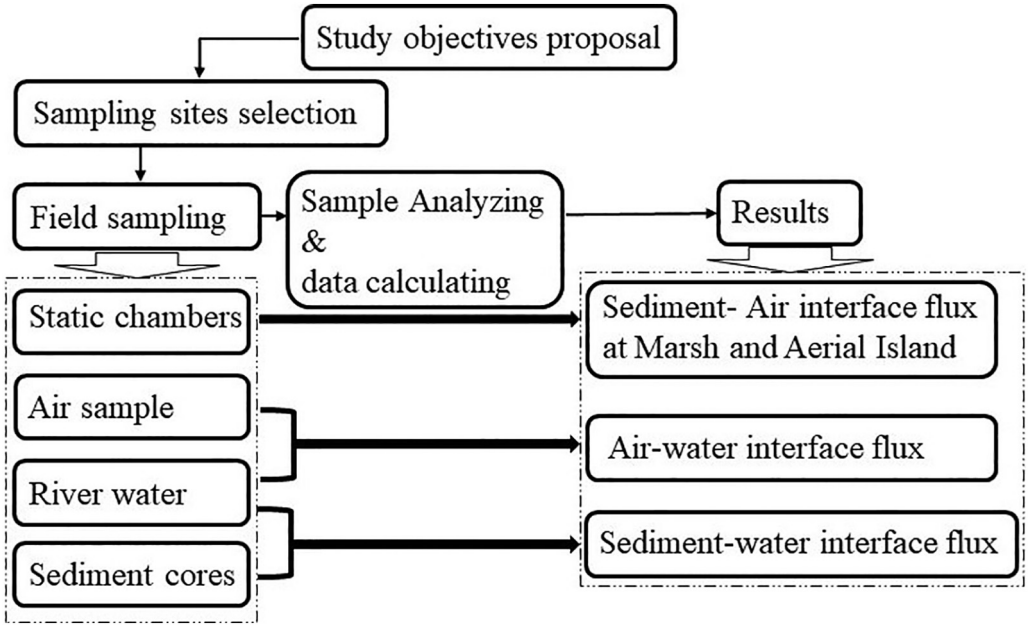

Fig. 3. Research methodology flowchart.

the air temperature from the field. After pre-incubation, 5 ml of overlying water in the incubation cells were withdrawn and injected into 12 ml vials for determination of dissolved CH_4 concentration, after vigorous shaking for 1 min to equilibrate the water and gas. Then, the top 25 cm of the sediment were sectioned into 1 cm slices and immediately placed into 240 ml bottles containing 60 ml deionized water. After vigorously shaking to release the trapped porewater and gas for 1 min, a headspace sample was draw out and injected into the GC for CH_4 concentration determination. After the air sample measurements, the slurry in the bottles were centrifuging at 2200*g* by a high speed, refrigerated centrifuge (Sorval Model RC 5C Pus) and vacuum filtered through 0.45 μm filter paper and stored at 4 °C for pore water dissolved organic carbon (DOC) measurement on a TOC analyzer (Shimadzu TOC-V). The remaining diffusion cell was sectioned into 1 cm subsamples to determine the moisture content of each layer and sediment characterization and the intact cores from established marsh and newly forming island sites were sliced into 5 cm increments for sediment properties characterization. Methods of CH_4 fluxes determination for each sampling site are in Table 1.

The CH_4 concentration in all air samples were analyzed by a GC equipped FID (Shimazu, GC-2014). The total carbon (TC) of sediment sample was analyzed by elemental analyzer (Costech Elemental Combustion System). The entire wet sediment sample slices were weighed, and a subsample was then dried in a forced air over at 70 °C until constant weight to get the moisture content (MC). The moisture content was used to calculate the dry-weight bulk density (BD) of each sediment interval. River water salinity was measured by a calibrated, portable YSI meter (Yellow Springs, model 556 MPS).

2.4. Data calculations

Gaseous flux in the diffusion chambers were calculated as the change in concentration of CH_4 in the chamber headspace over time. The CH_4 equilibrium concentration in water (C_w) was calculated using the corresponding air CH_4 concentration (C_a) in the head space of vial and bottle and in the field, and the Bunsen Solubility Coefficient (β) given by Wiesenburg and Guinasso (1979):

$$C_w = \beta \times C_a \quad (1)$$

The air-water interface CH_4 flux was calculated by using the two-layer model of diffusive gas exchange [Liss and Salter, 1974] using the following formula:

$$F_{a-w} = k \times (C_{R-w} - C_{S-a}) \quad (2)$$

where $F_{a\text{-}w}$ is the CH_4 flux, and a positive number means net CH_4 emission into the air; $C_{R\text{-}w}$ is measured CH_4 concentration in the river water; $C_{S\text{-}a}$ is the saturation concentration of CH_4 in water calculated from the CH_4 concentration in ambient air of the river sampling site; k is the transfer coefficient for water turbulence. Here, k was calculated from a wind-dependent model derived from field floating-dome, natural tracer, and tracer addition measurements specifically for estuarine rivers [Raymond and Cole, 2001]:

$$k = 1.91e^{0.35\mu} (Sc/600)^{-1/2} \quad (3)$$

Table 1
Method of determination of CH_4 for each sampling site in Wax Lake Delta.

Site	Method	Flux determination
Established marsh	Static chamber,	Soil-air interface flux
Newly forming Island	Static chamber,	Soil-air interface flux
River channel bottom	Sediment incubation	Sediment-water interface flux
	Head space,	Air-water interface flux

where Sc is the Schmidt value for CH_4 corrected for in situ water temperature [Wanninkhof, 1992]; μ is long-term wind speed at 10 m height above the river (m s^{-1}). To precisely quantify k, half hourly averaged 10 m height wind speeds at WLD area were collected from the USGS 07381654 Atchafalaya Bay, Eugene Island station (U.S. Geological Survey database) and used to calculate the averaged wind speed for CH_4 flux calculation in order to avoid the deviation from short-term, extreme values. The average wind speed at WLD was 5.53 ± 2.85 m s^{-1} in spring, (from February 15, 2012 to April 14, 2012, $n = 2916$), and the seasonal median, 5.14 m s^{-1} of wind speed ranged from 4.92 m s^{-1} (lower bound of 95% confidence interval) to 5.36 m s^{-1} (upper bound of 95% confidence interval). The average wind speed in summer was 4.96 ± 2.29 m s^{-1} (the average of every half hour wind speed from June 1, 2012 to July 31, 2012, $n = 2438$), and the seasonal median, 4.70 m s^{-1} of wind speed ranged from 4.47 m s^{-1} (lower bound of 95% confidence interval) to 4.92 m s^{-1} (upper bound of 95% confidence interval).

Diffusive flux of CH_4 at sediment-water interface was calculated as follow:

$$F_{s-w} = (C_{E-w} - C_{B-w})/(A \times T) \quad (4)$$

where $F_{s\text{-}w}$ is the CH_4 flux at sediment-water interface, and positive number means CH_4 diffusing from sediment to water; $C_{B\text{-}w}$ is measured CH_4 concentration in the water at the beginning of incubation; $C_{E\text{-}w}$ is measured CH_4 concentration in the water at the end of incubation; A is the area of sediment core section; T is the incubation time.

At the same time, sediment-water interface flux of CH_4 was also calculated by Fick's first law. The diffusion equation was described as Eq. (5):

$$F_{Fick(s-w)} = D_S \times (\partial C/\partial Z) = D_S \times ((C_{P-w} - C_{R-w})/\partial Z) \quad (5)$$

where $F_{Fick(s\text{-}w)}$ is the diffusion CH_4 flux calculated by Fick's law; ∂C is the CH_4 concentration gradient between the top 1 cm sediment porewater and overlying water; ∂Z is the diffusion distance; $C_{P\text{-}w}$ is CH_4 concentration in sediment pore water; $C_{R\text{-}w}$ is CH_4 concentration in the river water. Ds is the effective diffusion coefficient of CH_4 in sediment pore water. Ds was calculated by the polynomial regression equation that was obtained by the measured diffusion coefficients of CH_4 in water (Dw) in the range 0 °C to 35 °C [CRC, n.d. Handbook of Physics and Chemistry, 83rd Edition]:

$$Dw = 8.889 \times 10^{-11} T^3 - 1.714 \times 10^{-9} T^2 + 3.721 \times 10^{-7} T + 8.771 \times 10^{-6} \quad (6)$$

and then corrected by sediment porosity according to equation from Lerman (1979) and the modification by Ullman and Aller (1982), in which the empirical relationships between diffusion flux and φ was quadratic for unlithified marine sands or muds when φ is less than 0.7, appropriate for the mud sediment at the river channel bottom site.

$$Ds = Dw \times \varphi^2 \quad (7)$$

3. Results

3.1. Environmental parameters

There was a significant difference in sediment properties among the marsh, island and river channel bottom sites (Table 2). The moisture content and total C in the marsh soil were significantly higher than the island and river channel bottom site, while bulk density was greatest in river channel bottom. At the same time, surface soil of established marsh site of WLD has a significantly higher bulk density, compared to the more organic, inland marshes of the Mississippi deltaic plain not under river influence [Delaune et al., 1983; Alford et al., 1997].

Table 2
Properties of sediment and temperatures (average ± standard deviation of triple cores).

Sampling month in 2012	Site	Depth (cm)	Bulk density (g cm^{-3})	Moisture content (wt%)	Total nitrogen (wt%)	Total carbon (wt%)	Temp at 5 cm soil depth (°C)	Air temp (°C)
March	Marsh	0–5	0.32 ± 0.07	242 ± 28	0.64 ± 0.01	10.0 ± 0.27	22.0	26.0
		5–10	0.43 ± 0.16	183 ± 44	0.47 ± 0.17	6.50 ± 2.61		
		10–15	0.34 ± 0.17	273 ± 144	0.66 ± 0.38	9.08 ± 6.29		
	Island	0–5	1.25 ± 0.02	48.0 ± 3.4	0.10 ± 0.00	1.04 ± 0.06	18.0	18.0
		5–10	1.27 ± 0.06	41.4 ± 3.1	0.05 ± 0.04	0.76 ± 0.06		
		10–15	1.26 ± 0.11	40.7 ± 6.9	0.03 ± 0.05	0.78 ± 0.14		
	River Channel bottom	0–5	1.46 ± 0.13	33.6 ± 3.3	ND	0.66 ± 0.14	24.4[a]	18.0
		5–10	1.49 ± 0.03	28.9 ± 1.1	ND	0.36 ± 0.09		
		10–15	1.47 ± 0.05	29.1 ± 1.6	0.02 ± 0.00	0.40 ± 0.09		
June	Marsh	0–5	0.65 ± 0.11	108 ± 12	0.31 ± 0.05	3.80 ± 0.75	33.5	32.5
		5–10	0.73 ± 0.12	94.6 ± 17.9	0.25 ± 0.05	2.77 ± 0.68		
		10–15	0.79 ± 0.04	81.3 ± 9.7	0.19 ± 0.03	2.20 ± 0.35		
	Island	0–5	0.99 ± 0.17	67.0 ± 8.0	0.11 ± 0.01	1.40 ± 0.21	32.0	31.0
		5–10	1.13 ± 0.12	49.0 ± 6.7	0.09 ± 0.02	1.08 ± 0.23		
		10–15	1.22 ± 0.08	44.0 ± 2.4	0.08 ± 0.01	1.02 ± 0.13		
	River Channel bottom	0–5	1.65 ± 0.02	25.4 ± 0.7	ND	0.22 ± 0.03	30.0[a]	32.5
		5–10	1.37 ± 0.15	35.5 ± 5.5	0.06 ± 0.01	0.69 ± 0.14		
		10–15	1.44 ± 0.03	29.9 ± 0.6	0.05 ± 0.00	0.59 ± 0.02		

[a] River water temperature. TN: total nitrogen; TC: total carbon; 5 cm GT: 5 cm depth ground temperature; AT: air temperature; ND: not detected.

Typically, river connection can more than double the bulk density of marsh soil through sediment addition (Spera et al., 2020). The high inorganic sediment load of the Atchafalaya River has a significant effect on the structure and chemical makeup of the wetland soils (Roberts et al., 2015).

3.2. Methane emissions from Wax Lake Delta

Concentration of CH_4 in the surface water at the river channel bottom site was 0.72 ± 0.96 μg C L^{-1} in March and 6.0 ± 0.36 μg C L^{-1} in June. The % saturation of the surface water was calculated based on the ambient, atmospheric CH_4 concentration (~0.05 μg C L^{-1} corrected for temperature). The % saturation of CH_4 for the surface water was 1641 ± 2090% in March and 12,445 ± 707% in June, clearly indicative of CH_4 production from the sediments.

By using a two-layer model [Liss and Salter, 1974] and Eq. (3) [Raymond and Cole, 2001], the calculated diffusive CH_4 flux air-water interface was 2.49 ± 3.38 mg C m^{-2} d^{-1} in March and 19.5 ± 1.12 mg C m^{-2} d^{-1} in June. Results were comparable with the previous research at other sites in the Mississippi Delta plain [Delaune et al., 1983], in which CH_4 from the open water body at fresh marsh was 37 mg C m^{-2} d^{-1}, and 13 mg C m^{-2} d^{-1} at brackish marsh (Table 3). Considering the large deviation of wind speed over time, lower and upper bound (95% confidence interval) of the median of wind speed were used to calculate the deviation of the air-water CH_4 flux, which ranged from 2.01 ± 2.73 mg C m^{-2} d^{-1} and 2.35 ± 3.18 mg C m^{-2} d^{-1} in March and from 16.4 ± 0.94 mg C m^{-2} d^{-1} to 19.2 ± 1.10 mg C m^{-2} d^{-1} in June. If you consider that no oxidization of CH_4 occurs in the water column, the flux at air-water interface should be roughly equal to the flux at sediment-water interface because no appreciable methanogenesis occurs in overlying river water under high dissolved oxygen condition, typical of this system [Roberts et al., 2015]. In the laboratory incubation, the measured sediment-water interface flux of CH_4 was 1.54 ± 0.78 mg C m^{-2} d^{-1} in March and 15.2 ± 8.31 mg C m^{-2} d^{-1} in June. At the same time, calculated sediment-water interface CH_4 flux based on the gradient of CH_4 concentration in pore water and Eq. (5)

Table 3
Comparison of methane emissions from Mississippi deltaic wetlands.

Site	Wetland type	CH_4 flux (mg C m^{-2} d^{-1})[a]	Range (Min-Max) (mg C m^{-2} d^{-1})	Reference
1	Freshwater marsh (*Sagittaria lancifolia*)	188 ± 141	36–679	Alford et al., 1997[b]
2	Swamp forest (*Taxodium distichum*/*Nyssa aquatic*)	110 ± 149	BD-444	
3	Intermediate marsh (*Sagittaria lancifolia*/*Spartina patens*)	684 ± 692	9.8–2933	
4	Fresh marsh (*Spartina alterniyora*)	440		Delaune et al., 1983[c]
	Adjacent open water body	37		
5	Brackish marsh (*Spartina patens*)	200		
	Adjacent open water body	13		
6	Salt marsh (*Panicum hemitornon*)	11.8		
	Adjacent open water body	3.6		
7	Brackish marshes (Spartina patens)	423		Leventhal and Guntenspergen, 2004
8	Freshwater tidal wetlands (*Sagittaria lancifolia*, *Leersia oryzoides*, with patchy areas of *Typha domingensis*)	128	28.3–254	Holm Jr et al., 2016[d]
9	Brackish tidal wetlands (*Spartina patens* and *Schoenoplectus americanus*)	28.4	1.5–108	
10	Fresh marsh (Marsh site)	571.8	386–2859	This study[e]
11	Fresh marsh (Island site)	43.2	7.94–216	
12	Adjacent open water body (Channels)	3.90	2.49–19.5	

[a] CH_4 flux unit is unified to mg C m^{-2} d^{-1} based on the unit of the flux data in reference.
[b] The average data and range is from data of each months (12 measurements) of a whole year.
[c] The average data and range is from data of 17 time monthly measurements of two years.
[d] The average data and range is from data of 24 time monthly measurements from Dec. 2011 to Nov 2013 at Freshwater tidal wetland site and 15 time monthly measurements from Oct. 2011 to Dec. 2012 at Brackish tidal wetland site.
[e] The range is two time measurement in March and June. Average data is 20% of the data in June.

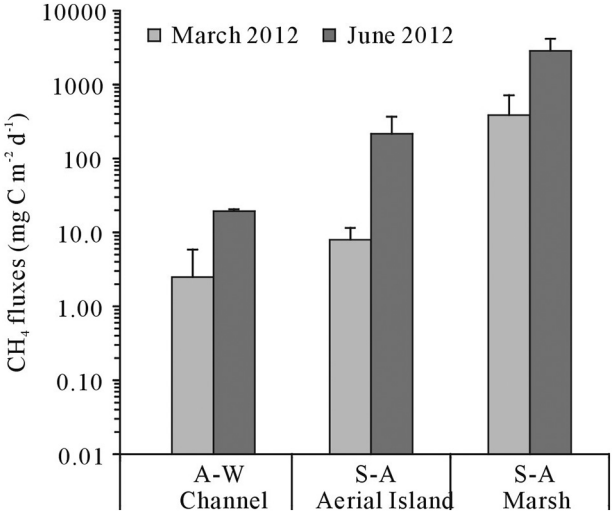

Fig. 4. CH_4 fluxes at the air-water (A-W) interface of the channel and the sediment-air (A-S) interfaces at the island and marsh sites.

was 1.99 ± 0.92 mg C m^{-2} d^{-1} in March and 23.6 ± 3.44 mg C m^{-2} d^{-1} in June. Flux of CH_4 at air-water interface was approximately equal to the flux at sediment-water interface, which suggests that very little of the CH_4 produced in the channel sediment is reduced in the water column by oxidation before being released to the atmosphere.

The CH_4 flux at the marsh site was 386 ± 327 mg C m^{-2} d^{-1} in March and 2859 ± 1286 mg C m^{-2} d^{-1} in June, and much greater than at the island site (7.94 ± 3.57 mg C m^{-2} d^{-1} in March and 215 ± 153 mg C m^{-2} d^{-1} in June). In general, the CH_4 flux rates at the marsh site were one order of magnitude greater than at island site, and the island site was one order of magnitude greater than the river channel bottom site (Fig. 4). Clearly, these results show a marked heterogeneity in methane flux rates within an active delta complex which are correlated to the soil C content. In the active delta region with high inorganic sedimentation, flux rates are low while in the peripheral established marshes with greater organic matter, the methane flux rates are an order of magnitude higher.

3.3. Comparison of CH_4 emission with Mississippi river deltaic plain wetland marshes

Methane flux data has indicated that wetlands are a significant source of CH_4 to the atmosphere throughout the year in previously reported research in the Mississippi River deltaic plain (Table 3). Delaune et al. (1983) measured CH_4 emissions from three vegetated marshes along the salinity gradient; average CH_4 flux at fresh marsh was 440 mg C m^{-2} d^{-1}, and 200 mg C m^{-2} d^{-1} at a brackish marsh and 11.8 mg C m^{-2} d^{-1} at salt marsh (Site 4, 5 and 6 in Fig. 2). Alford et al. (1997) measured CH_4 emissions from three wetland habitats. Average emission from swamp forest was 110 ± 149 mg C m^{-2} d^{-1} and emissions from freshwater marsh averaged 188 ± 141 mg C m^{-2} d^{-1}, while flux from an oligohaline marsh was significantly higher, 684 ± 692 mg C m^{-2} d^{-1} (Site 1, 2 and 3 in Fig. 2). Reported by Leventhal and Guntenspergen (2004), the CH_4 flux was 57.8 mg C m^{-2} d^{-1} at a brackish marsh (Site 7 in Fig. 2). Seasonal variation was significant with emissions being higher in the late summer and early fall, and temperature was found to be significantly correlated with CH_4 emission [Delaune et al., 1983; Alford et al., 1997]. By using eddy covariance techniques, Holm Jr et al. (2016) calculated CH_4 emissions from natural, freshwater and brackish wetlands in Louisiana not within the influence of the river. Annual estimates of methane emissions were 62.3 g CH_4 m^{-2} yr^{-1} and 13.8 g CH_4 m^{-2} yr^{-1} for the freshwater and brackish sites (Site 8 and 9 in Fig. 2), respectively. In this study at WLD, CH_4 flux was measured two times, once in March representing the end of the cooler period and once in June to represent the warmer summer months. According to the previous research, the maximum flux in summer was about 4 to 5 times greater than annual average based on monthly measurement for the Mississippi River delta [Delaune et al., 1983; Alford et al., 1997; Holm Jr et al., 2016]. Using this ratio, annual mean CH_4 flux was determined to be as 571.8 mg C m^{-2} d^{-1} at the marsh site, 43.0 mg C m^{-2} d^{-1} at island site and 3.9 mg C m^{-2} d^{-1} at the river channel bottom site (Site 10, 11 and 12 in Fig. 2). Again, these results when compared with the literature values highlight the high seasonal and spatial variability of CH_4 flux within a deltaic setting.

4. Discussion

4.1. Carbon sequestration and storage in estuarine and coastal wetland

Wetland ecosystems couple high rates of net primary production coupled with a slow rate of organic matter (OM) decomposition, due to prevailing anaerobic conditions in the flooded soils [White and Reddy, 2000; Hayes et al., 2021], to produce soil with the highest C content of all the soil orders [Bridgham et al., 2006] and creating an important sink for atmospheric CO_2 [DeLaune and White, 2012; White et al., 2019; Sapkota and White, 2021]. At the same time, deltaic systems provide significant inorganic sediment which when combined with strong trapping efficiency, can lead to high accretion rates in tidal freshwater marshes [Neubauer, 2008]. In this fashion, vegetated coastal habitats have the potential for greater long-term C sequestration capability of per unit area than that of most terrestrial ecosystems [McLeod et al., 2011].

Despite rising sea level, the high sediment supply of major deltaic systems leads to land building and carbon sequestration along the coastal margins [Roberts et al., 2015]. The mean growth rate of the entire Atchafalaya River Delta has been reported to be 3.1 km^2 y^{-1} (1984–2004) [Xu, 2010], 3.2 km^2 y^{-1} (1985–2010) [Couvillion et al., 2011] and 2.8 km^2 y^{-1} (1989–2010) [Rosen and Xu, 2013], in which the vegetated land average growth rate is 2.57 km^2 y^{-1} (1989–2010). Reported vertical accretion rate in the Mississippi river deltaic plain wetlands ranged from 0.59 to 1.35 cm y^{-1} with an average of ~1 cm y^{-1} [Delaune and White, 2012]. While in Yangtze estuary, vertical accretion rate at the intertidal marsh of east Chongming Island was from 1.9 cm y^{-1} to 5.18 cm y^{-1} and 1.63 cm y^{-1} at mudflat due to high suspended sediment river concentrations [Jiang et al., 2012]. Annual C accumulation rates reported for Louisiana coastal marsh soils were 261.2 ± 38.6 g C m^{-2} $year^{-1}$at fresh marsh and 261.8 ± 65.3 g C m^{-2} $year^{-1}$ at saline marsh [DeLaune and White, 2012, and references therein]. Carbon accumulation rates was 136 g C m^{-2} yr^{-1} at Chongming Island east intertidal flat in Yangtze estuary [unpublished data]. In tidal marshes of the continental United States (northeast (NE) and southeast Atlantic, Gulf coast, and West coast) Organic C accumulation rates were 140 ± 20 g C m^{-2} yr^{-1} at freshwater marsh (n = 9), 240 ± 30 g C m^{-2} yr^{-1} brackish marsh (n = 18) and 190 ± 40 g C m^{-2} yr^{-1} salt marsh (n = 21) [Craft, 2007].

4.2. Enhanced CH_4 flux resulting from increasing organic carbon in soil/sediment

Soil organic C content of wetland soil has an important effect on CH_4 production [DeLaune et al., 1983; Crozier and DeLaune, 1996] and emissions from wetland ecosystems are positively correlated with net ecosystem productivity (NEP) [Whiting and Chanton, 1993], where higher NEP leads to a higher input of substrates (litter production and root exudates) required for methanogenesis [Whiting and Chanton, 1992; Joabsson et al., 1999; King et al., 2002; Öquist and Svensson, 2002; Ström et al., 2003]. Substrate supply is the primary control on CH_4 production once anaerobic conditions have been achieved [Whalen, 2005]. For example, in the Yangtze River delta CH_4 flux rates at intertidal brackish vegetated marsh were higher than them at lower

Table 4
Methane emissions from Yangtze delta estuarine wetlands.

Site	CH4 flux ($mg\ C\ m^{-2}\ d^{-1}$)	Range (Min-Max) ($mg\ C\ m^{-2}\ d^{-1}$)	Reference
Yangtze estuarine intertidal brackish marsh (*Scirpus mariqueter*)	37.1	0.468–179.3	Wang et al., 2009b
Yangtze estuarine intertidal mudflat	0.72	0.036–1.62	
Yangtze estuarine intertidal brackish marsh (*Phragmites australis*)	116.9	23.8–210	Ma et al., 2012
Yangtze estuarine intertidal mudflats	0.07 ± 6.3		

carbon content submerged mudflat one to several orders of magnitude (Table 4) [Wang et al., 2009b; Ma et al., 2012]. In our research at WLD, sediment/soil total carbon (TC) at three sampling sites increased from river channel bottom, to newly formed island to established marsh site related to differences in delta development stage. This sequence was well correlated to CH_4 emissions for both sampling periods (Fig. 5).

4.3. Methane flux vs. carbon burial

Although wetlands could act as a significant sink for atmospheric CO_2 globally due to high primary productivity and subsequent long-term storage of organic carbon in saturated anoxic soil and sediments [e.g. Chmura et al., 2003; Bridgham et al., 2006; Mcleod et al., 2011], the anaerobic, organic-rich soil/sediment can also produce substantial CH_4 in wetlands, which contribute about a third of total global CH_4 emissions [Bridgham et al., 2013]. Researchers have pointed out that CH_4 emissions from wetlands can offset some or all of the reductions in radiative forcing attributed to carbon sequestration considering the greater "warming potential" of CH_4 relative to CO_2 [e.g. Whiting and Chanton, 2001; Sapkota and White, 2020].

At Great Sippewissett Salt Marsh, the loss of carbon as CH_4 was 1.2–3.6 g C m^{-2} yr^{-1}, which is a CO_2 equivalent of 14–41% of the C burial (88.8 g C m^{-2} yr^{-1}) [Howes et al., 1985]. This value is low because salt marshes generally produce little methane. About 42% of annual carbon delivered to the Scheldt Estuary intertidal mud flat sediment becomes buried (211.7 g C m^{-2} yr^{-1}), 7% of the remaining being emitted as CH_4 (20.5 g C m^{-2} yr^{-1}) [Middelburg et al., 1995], which is a CO_2 equivalent of 208.7 g C m^{-2} yr^{-1} almost completely offsetting the total C burial. This difference is likely related to the freshwater delivery to the estuary, lowering salinity, depressing redox, and allowing greater CH_4 production. Therefore, it is critical to determine CH4 production in a range of geomorphic settings within the coastal zone in order to more accurately predict the C credit potential (Sapkota and White, 2020).

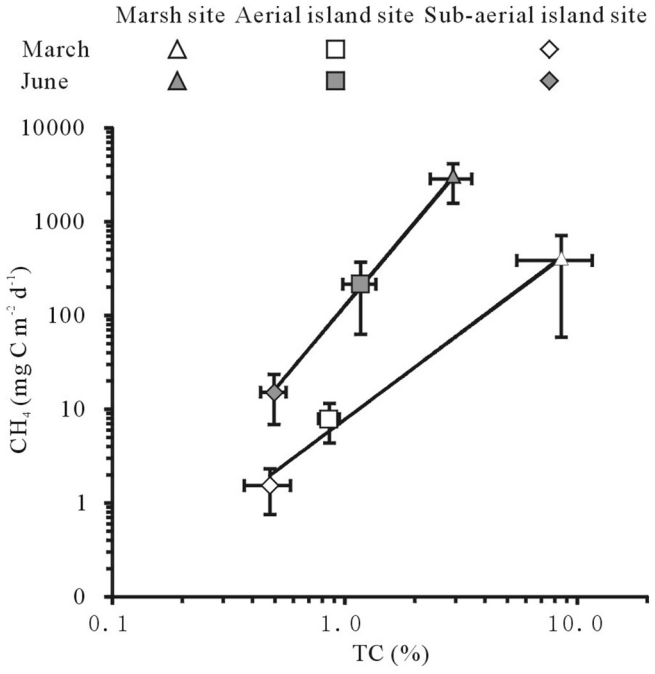

Fig. 5. Relationship between soil/sediment total carbon (TC) content and CH_4 flux at WLD for both the March and June sampling events.

Deltaic sediments are generally dominated by mineral material and attempts to determine the decadal accretion rates using ^{137}Cs were unsuccessful at the active delta sites. However, a simple mean was calculated by combining data at our three sites with previously published marsh data. This results in an average CH_4 emission in the Mississippi river deltaic plain of 108 ± 87.9 g CH_4-C m^{-2} yr^{-1} [Delaune et al., 1983; Alford et al., 1997; Leventhal and Guntenspergen, 2004; this study], which corresponds to CO_2-C equivalents of 1103 g CO_2-C m^{-2} yr^{-1}. This number is 4.2 times the mean carbon burial rate for these wetland areas [DeLaune and White, 2012, and reference therein] which could suggests that the mean Mississippi deltaic plain wetlands are not a carbon sink in terms of climate forcing. However, our results demonstrate that taking a simple mean and applying it across the coastal zone is not correct due to the high heterogeneity in flux rates in deltaic systems. Ideally, any attempt at spatial modeling would need to consider both the salinity and the organic matter content of the wetland soil. There is a bias toward fresher systems for CH_4 flux measurements in the literature while much of the Mississippi River coast plain has elevated salinity due to little interaction with the freshwater river due to levee construction during the past century (Peyronnin et al., 2017). Hence, there a skewness in our small data set toward more brackish and fresh marsh systems, which produce orders of magnitude greater more CH_4. More measurements are needed over a range of spatial scales to capture the variance in salinity and vegetation type with measurements distributed more appropriately to the occurrence of marsh type to accurately determine if the coastal zone marshes are indeed C sinks or sources from a climatic forcing perspective.

5. Conclusions

Coastal marshes are well documented for low flux levels of CH_4 due to high salinity. However, river deltas provide freshwater foci at the otherwise saline coastal margins. This study found that, in the two major geomorphic areas within the active delta area, both newly forming island and river channel bottom sediment have relatively low CH_4 flux rates, correlated to low total C despite freshwater conditions. However, the more established fresh marshes located proximal from the active delta, but within the halo of freshwater influence, produced substantially greater CH_4 correlated to greater soil C. Consequently, CH_4 flux in this coastal river delta appears to be primarily constrained by soil C, with an active delta zone with very low CH_4 output due to high inorganic sediment inputs. Consequently, the heterogeneity of CH_4 flux within the delta all within freshwater conditions, spanning three orders of magnitude, demonstrates the need for more spatially explicit measurements to determine a reliable mean CH_4 flux rate. The research has implications for determining coastal C balance and C credits in the context of climate forcing.

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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Credit author statement

Dongqi Wang: Conceptualization, Methodology, Investigation, Writing - Original draft preparation. **John R. White:** Writing - Reviewing and Editing. **Ronald D. Delaune:** Supervision. **Zhongjie Yu:** Data curation, Writing - Original draft preparation. **Yujie Hu:** Figure making.

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