Science of the Total Environment 559 (2016) 256–267

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Field-scale simulation of methane emissions from coastal wetlands in China using an improved version of $CH4MOD_{wetland}$

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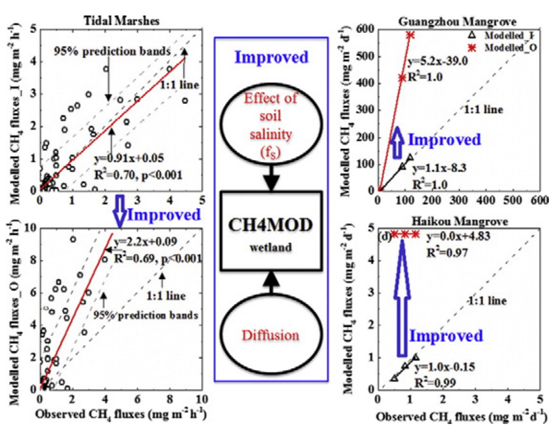
HIGHLIGHTS

- Salinity effect on CH4 emissions in coastal wetlands was added into $CH4MOD_{wetland}$.
- Modeled seasonal CH4 variations corresponded well with observations in tidal marshes.
- Modeled seasonal CH4 emissions agreed well with observations in coastal wetlands.
- The improved model significantly increased model efficiency.
- Previous regional/global CH4 estimations from wetlands may be overestimated.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 18 January 2016
Received in revised form 4 March 2016
Accepted 25 March 2016
Available online 8 April 2016

Editor: D. Barcelo

Keywords:
CH_4 emissions
Salinity
Coastal wetlands
$CH4MOD_{wetland}$ model
China

ABSTRACT

Coastal wetlands are important CH_4 sources to the atmosphere. Coastal wetlands account for ~10% of the total area of natural wetlands in China, but the size of this potential CH_4 source remains highly uncertain. We introduced the influence of salinity on CH_4 production and CH_4 diffusion into a biogeophysical model named $CH4MOD_{wetland}$ so that it can be used in coastal wetlands. The improved model can generally simulate seasonal CH_4 variations from tidal marshes dominated by *Phragmites* and *Scirpus*. However, the model underestimated winter CH_4 fluxes from tidal marshes in the Yellow River Delta and YanCheng Estuary. It also failed to capture the accurate timing of the CH_4 peaks in YanCheng Estuary and ChongMing Island in 2012. The improved model could generally simulate the difference between the annual mean CH_4 fluxes from mangrove sites in GuangZhou and HaiKou city under different salinity and water table depth conditions, although fluxes were systematically underestimated in the mangrove site of HaiKou city. Using the improved model, the seasonal CH_4 emissions simulated across all of the coastal wetlands ranged from 0.1 to 44.90 g m^{-2}, with an average value of 7.89 g m^{-2}, which is in good agreement with the observed values. The improved model significantly decreased the *RMSE* and *RMD* from 424% to 14% and 314% to −2%, respectively, and improved the *EF* from −18.30 to 0.99.

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http://dx.doi.org/10.1016/j.scitotenv.2016.03.186

Model sensitivity analysis showed that CH_4 emissions were most sensitive to P_{ox} in the tidal marshes and salinity in the mangroves. The results show that previous studies may have overestimated CH_4 emissions on a regional or global scale by neglecting the influence of salinity. In general, the $CH4MOD_{wetland}$ model can simulate seasonal CH_4 emissions from different types of coastal wetlands under various conditions. Further improvements of $CH4MOD_{wetland}$ should include the specific characteristics of CH_4 processes in mangroves to decrease the uncertainty in estimating regional or global CH_4 emissions from natural wetlands.

1. Introduction

Methane (CH_4) is an important greenhouse gas, with a 100-year global warming potential 25 times stronger than that of carbon dioxide (Lashof and Ahuja, 1990). Methane is responsible for approximately 18% of human-induced radiative forcing, making it the second most important greenhouse gas after CO_2 (Forster et al., 2007). Recently, it was reported that when the indirect global warming effects of CH_4 on aerosols and other chemical compounds (e.g., O_3) was incorporated, previous estimates of the global warming potential of CH_4 may be 10–40% too low (Shindell et al., 2009; IPCC, 2013). Natural wetlands are the single largest natural source of CH_4, accounting for approximately one third of total global CH_4 emissions, i.e., equivalent to 115 (Fung et al., 1991) to 237 Tg CH_4 yr^{-1} (Hein et al., 1997). CH_4 emissions from wetlands have been strongly responsive to climate in the past (Chappellaz et al., 1993a,b; Blunier et al., 1995; Loulergue et al., 2008), and will likely continue to be responsive to anthropogenic-driven climate change in the future (Gedney et al., 2004; Eliseev et al., 2008; Ringeval et al., 2011). Moreover, they appear to dominate the interannual variability of the global CH_4 source (Bousquet et al., 2006; Ciais et al., 2013). Therefore, increased knowledge of CH_4 emissions from natural wetlands is important to understand the global CH_4 budget.

Efforts have been made to estimate regional CH_4 emissions from natural wetlands by extrapolating field measurements to a given area (Bartlett and Harriss, 1993; Chen et al., 2013; Ding et al., 2004; Seiler and Conrad, 1987). However, this method is unreliable when scaling from site measurements to regional or global scales due to limitations in the spatial and temporal coverage of measurements (Cao et al., 1996). Compared with extrapolation, process-based models are regarded as an improved method for regional estimates because they can describe the processes of CH_4 production, oxidation and emission as well as the influence of complex climate, soil, vegetation and hydrology conditions on CH_4 emissions (Cao et al., 1996; Li, 2000; Zhang et al., 2002). Some process-based models focus on the CH_4 cycling mechanisms and only describe the processes of CH_4 production, consumption and transportation under different climatic and soil conditions (e.g., Potter, 1997; Walter and Heimann, 2000; Zhang et al., 2002; Li et al., 2010a). For use at a global scale, some modellers have developed or integrated a CH_4 emission module into a global land ecosystem model (e.g., Zhuang et al., 2004; Xu et al., 2010; Tian et al., 2010; Wania et al., 2010; Riley et al., 2011; Zhu et al., 2014). The popular way of estimating CH_4 emissions from natural wetlands is to first calibrate and validate the process-based model at the site-scale, and then upscale the results to a regional or global scale (e.g., Xu and Tian, 2012; Li et al., 2012; Wania et al., 2013; Bohn et al., 2015).

Coastal wetlands are characterized by high temporal and spatial variation related to topographic features, environmental factors and astronomic tidal fluctuation, and are very sensitive to global climate changes and human activities. The global coastal wetland area is 660×10^3 km^2, which accounts for ~10% of the total wetland area (Lehner and Döll, 2004). Previous studies have shown that coastal wetlands with high salinity usually emit less CH_4 than less saline wetlands (Bartlett et al., 1987; Poffenbarger et al., 2011). This has been explained by the presence of sulfate in the sea water, which allows sulfate-reducing bacteria to outcompete methanogens for energy sources, consequently inhibiting methane production (DeLaune et al., 1983; Bartlett et al., 1987; Wang et al., 1996; Poffenbarger et al., 2011). However, most of the above estimates of regional or global CH_4 emissions treated the coastal wetlands the same as freshwater wetlands. Moreover, most of the process-based models did not consider the influence of salinity on CH_4 fluxes, or these models were not validated at coastal wetland sites.

In China, coastal wetlands accounts for ~10% of the total area of natural wetlands (Niu et al., 2012). Although there are some sporadic measurements of CH_4 emissions from coastal wetlands (e.g., Ye et al., 2000; Huang et al., 2005; Kang et al., 2008; Yang et al., 2007; Yuan et al., 2015), most of the measurements were focused on the freshwater marshes in northeastern China and the Qinghai Tibet Plateau (Song et al., 2007; Ding et al., 2004; Jin et al., 1999; Wei et al., 2015). Moreover, the national CH_4 emissions have been estimated to range from 1.7 to 10.5 Tg yr^{-1}, using models or extrapolating measurements to a regional scale, but most of the studies were based on measurements from freshwater wetlands (Wang et al., 1993; Khalil et al., 1993; Jin et al., 1999; Ding et al., 2004; Wang et al., 2012).

Recently, we established a process-based model named $CH4MOD_{wetland}$, which includes the processes of CH_4 production, oxidation and emission (Li et al., 2010a). This model has been validated in various types of freshwater wetlands in China and North America (Li et al., 2010a, 2012). In this study, we integrated a relationship between salinity and CH_4 production into $CH4MOD_{wetland}$ to improve the applicability of the model for use in coastal wetlands. We also compiled a CH_4 measurement dataset from latitudinally distributed coastal zone sites using the literature and measurements. The objectives of this study are to modify the $CH4MOD_{wetland}$ model, and to assess the performance of the improved model with respect to CH_4 flux estimates by validating the model outputs against measurements collected from coastal wetland sites across different latitudes in China.

2. Materials and methods

2.1. Model overview

$CH4MOD_{wetland}$ is a biogeophysical process-based model that was developed to describe the processes of CH_4 production, oxidation and emission from natural freshwater wetlands (Li et al., 2010a, 2012). This model adopted the hypothesis of the CH4MOD model, which is used to simulate CH_4 emissions from rice paddies (Huang et al., 1998). We made modifications based on the supply of methanogenic substrates in natural wetlands, which differs significantly from that in rice paddies. The model inputs include daily air or soil temperature, water table depth, annual net primary productivity (*NPP*), soil sand fraction, soil organic matter and bulk density. The outputs are daily and annual CH_4 production and emissions.

The main processes related to CH_4 production, oxidation and emission in $CH4MOD_{wetland}$ are shown in Fig. 1. The methanogenic substrates are derived from root exudates as well as the anaerobic decomposition of plant litter and soil organic matter. In the original model, the environmental factors that influence CH_4 production include soil temperature, soil texture and soil redox potential. If the soil temperature was unavailable, we used the air temperature to calculate this variable. The soil redox potential is controlled by the water table depth. In this study, we added the influence of soil salinity on CH_4 production to improve the model for use in coastal wetlands. Because the model was formerly used at sites dominated by arenchymous vascular plants, plant-mediated transport is the primary mechanism of CH_4 emission.

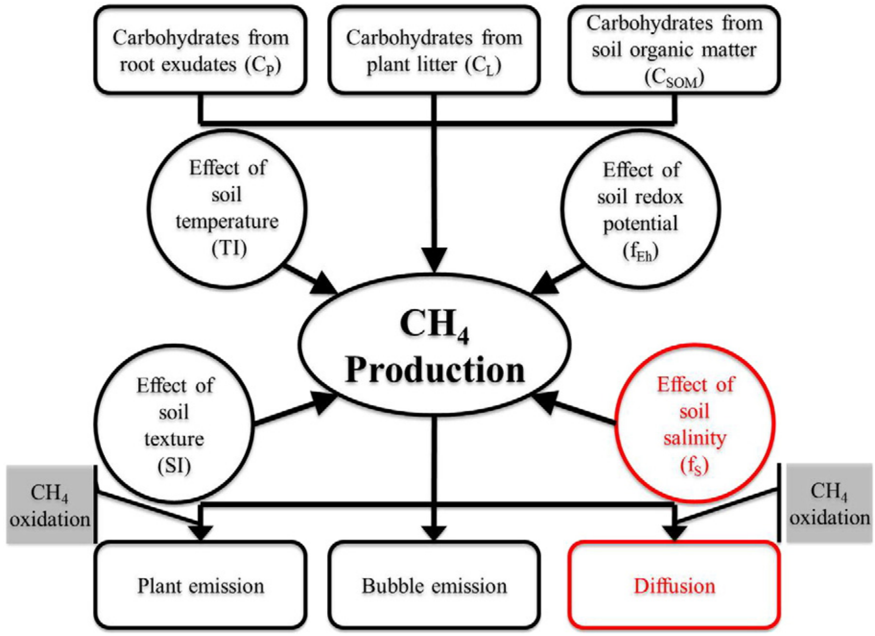

Fig. 1. Conceptual explanation of $CH4MOD_{wetland}$. The red frames are the modification to the model.

CH_4 emission via ebullition is significant in the early stages of plant growth. Diffusion was not considered in the original model. In this study, we added the diffusion process of CH_4 to ensure that the model can be used at mangrove sites. CH_4 oxidation occurs through plant and diffusion transportation.

$CH4MOD_{wetland}$ has been validated against independent field measurements of CH_4 fluxes from several types of freshwater wetland sites, including a freshwater marsh in northeastern China, peatland in the Ruoergai Plateau in China, as well as fen in Canada and USA (Li et al., 2010a). Additional details on $CH4MOD_{wetland}$ are well documented in previous studies (Li et al., 2010a, 2012, 2015).

2.2. Model modification

As shown in Fig. 1, CH_4 production is influenced by the soil temperature, soil texture and soil redox potential. When the influence of soil salinity is included, CH_4 production is calculated as:

$$P = 0.27 \times SI \times TI \times F_{EH} \times F_S \times (C_P + C_L + C_{SOM}) \quad (1)$$

where P is CH_4 production ($g\ m^{-2}\ d^{-1}$), C_P, C_L and C_{SOM} are the carbohydrates derived from plant root exudates, plant litter and soil organic matter, respectively; SI, TI, F_{EH} and F_S are the influence of soil texture, soil temperature, soil index potential and soil salinity, respectively, on CH_4 production; 0.27 is a factor (mole weight basis) to convert carbohydrate into methane.

Previous studies (Atkinson and Hall, 1976; King and Wiebe, 1978; Bartlett et al., 1985; 1987; Magenheimer et al., 1996) indicated that methane emissions from various coastal salt marshes in the temperate zones varied with salinity. This is because electron acceptors such as NO_3^- and SO_4^{2-} in coastal wetlands can compete with the methanogens for electrons. Poffenbarger et al. (2011) found a significant linear relationship between salinity and log-transformed CH_4 from 36 field cases where the salinity ranged from 0.4 to 35.1 ppt. In this study, we adopted this relationship and estimated the influence of salinity F_S in Eq. (2) as:

$$F_S = 10^{a \times s} \quad (2)$$

where F_S represents the effect of salinity on CH_4 production, s is the salinity (ppt), and a is an empirical constant.

The original version of $CH4MOD_{wetland}$ was mainly used to simulate arenchymous vascular plant-dominated sites. At these sites, plant-mediated transportation and ebullition contribute >90% of CH_4 emissions. However, if the model is applied at a wetland site without arenchymous vascular plants, e.g., a mangrove site, diffusion cannot be neglected. This is because compared with vascular plants, woody plants are poor transporters of CH_4 (Hook et al., 1971; Grosse et al., 1992; Walter et al., 1996; Walter and Heimann, 2000). We adopted the DLEM equation (Xu et al., 2010; Tian et al., 2010) to calculate the CH_4 diffusion rate:

$$E_D = R \times (P \times h - P_{max}) \times f_{oxi} \quad (3)$$

where E_D is the CH_4 emission rate via diffusion ($g\ m^{-2}\ d^{-1}$), P is methane production in Eq. (1) ($g\ m^{-2}\ d^{-1}$), h is the soil depth, for which we used 0.5 m in this study, P_{max} is the critical value at which diffusion occurs (Xu et al., 2010), which is 0.0012 $g\ m^{-3}$ according to the literature (Zhuang et al., 2004), R is the exchange coefficient between the air and soil, with a value of 0.3 $m\ day^{-1}$ (Xu and Tian, 2012; Happell et al., 1995), and f_{oxi} represents the fraction of CH_4 oxidized through diffusion, with a value of 85% (Whalen, 2005).

2.3. Site information and data sources

The coastal wetlands are distributed along the southeastern coast of China (Fig. 2). Tidal marshes and mangroves are the main types of coastal wetlands in this country. Tidal marshes are usually distributed in the north of HangZhou Bay, including the tidal marshes around LiaoTung Peninsula, BoHai Sea, ShanDong Province and JiangSu Province. In the south of HangZhou Bay, e.g., FuJian Province, GuangDong Province, and HaiNan Province, the coastal wetlands are dominated by mangroves (An et al., 2007). In this study, six coastal wetlands with different latitudes from north to south were chosen, e.g., the tidal marshes from the Liao River Estuary (LR), the Yellow River Delta (YR), the YanCheng Estuary (YC), and ChongMing Island (CM), as well as the mangroves from GuangZhou city (GZ) and HaiKou city (HK) (Fig. 1 and Table 1). We measured CH_4 fluxes and the synchronous air temperature, precipitation, water table depth and salinity at the YR site from July 2012 to

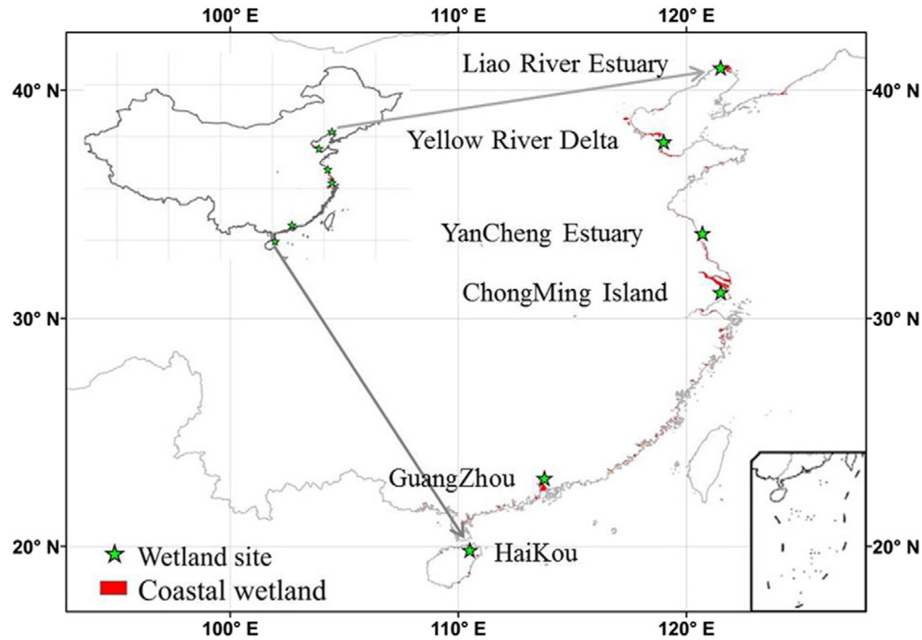

Fig. 2. Locations of the coastal wetland sites, the coastal wetland distribution map is from Niu et al. (2012).

December 2013. More details about the measurements were described in Supplementary material S1. The data for the five other coastal wetlands were obtained from previous studies and did not include synchronous measurements of salinity combined with the CH_4 sampling. For these sites, we only used the average salinity to drive the model (Table 1). For the mangrove observations, the accurate timing of the measurements was not available. We could only obtain the average yearly mean CH_4 emissions from the observations. For further details on the sites and measurements, please refer to Table 1 and Supplementary material S1.

At the tidal marsh sites, the total observed seasonal CH_4 emissions were calculated by summing the daily fluxes. The absence of CH_4 flux measurements between two adjacent days of observation was linearly interpolated. The observed total seasonal CH_4 emissions from mangroves were simply calculated using the annual mean CH_4 fluxes.

2.4. Model parameterization

The description and values of the main $CH4MOD_{wetland}$ parameters and inputs are shown in Table 2. Most of the parameters related to the plant species were obtained from the measurements of previous studies. For example, the fractions of aboveground and belowground to the total net primary productivity (F_{above} and F_{root}) of *Phragmites*, *Scirpus* and the mangroves were sourced from Shao et al. (1995), Chen et al. (2005) and Lin et al. (1990), respectively. Yuan et al. (2015) measured the initial concentrations of lignin and nitrogen in the plant litter (LG_0 and N_0) of *Phragmites* and *Scirpus.* These two parameters were used to calculate the non-structural and structural proportion of the plant litter. Most of the soil parameters were obtained from the State Soil Survey Service of China. The fraction of plant-mediated transport (T_{veg}) differs between grass species and mangroves. According to Walter and

Table 1
Site description.

Items	Liao River Estuary (LR)	Yellow River Delta (YR)	YanCheng Estuary (YC)	ChongMing Island (CM)	GuangZhou (GZ_P, GZ_N, GZ_S)[a]	HaiKou (HK_O, HK_M, HK_I)[b]
Location	40°02′N, 122°20′	37°45′ N, 118°59′ E	33°22′ N, 120°42′E	31°15′N, 121°30′E	23°01′N, 113°46′E	19°51′ N, 110°24′E
Wetland type	Tidal marsh	Tidal marsh	Tidal marsh	Tidal marsh	Mangrove	Mangrove
Year(s) of the experiment	1997.4–1997.10	2012.7–2013.12	2012.1–2012.12	2004. 5–2004.12 & 2011.2–2012.12	2005.3–2005.12	1996.1–1997.12
Dominant plant species	*Phragmites*	*Phragmites*	*Phragmites*	*Scirpus*	*Aegiceras corniculatum* etc.	*Bruguiera sexangula*
Annual air temperature (°C)	9	12.9	12.6	15.3	21.9	23.7
Annual precipitation (mm)	625	590	1040	1117	1582	1940
Water environment[c]	Seasonally	Occasionally	Occasionally	Perennially	Perennially/Seasonally/Never	Never
Average salinity (ppt)	7.2	6.7	3.2	6.9	12.5	11.9[d];14.2[e];19.5[f]
References	Huang et al. (2005)	This study	Yuan et al. (2015)	Yang et al. (2007); Li et al. (2010b; 2014)	Kang et al. (2008)	Ye et al. (2000)

[a] GZ_P, GZ_S and GZ_N are the perennially flooded microsite, seasonally flooded microsite and never flooded microsite, respectively.
[b] HK_O, HK_M and HK_I are the outer flat microsite, middle flat microsite and inner flat microsite, respectively.
[c] Perennially means always flooded during the observed period. Seasonally means that the sites are flooded during most of the observed periods, but drained for only one or two months; The LR site was flooded from May to September 1997; The GZ_P site was flooded from April to October 2005. Occasionally flooded means the site was always drained, and occasionally flooded after the heavy rainfall events. Never flooded means that the water table was never above the ground.
[d] For HK_O.
[e] For HK_M.
[f] For HK_I.

Table 2
Model input and parameters.

Parameters	Description (unit)	Value						References
		LR	YR	YC	CM	GZ	HN	
NPP	Net primary productivity ($g\ m^{-2}\ yr^{-1}$)	2640	935	1738	1570[$], 522[^]	2000	3000	Shao et al. (1995)[1]; This study[2]; Yuan et al. (2015)[3]; Li et al. (2014)[4]; Lin et al. (1990)[5]; Kang et al. (2008)[6]
F_{above}[*]	Proportion of above-ground/leaves to the total production (dimensionless)	0.45	0.45	0.45	0.45	0.35	0.35	Shao et al. (1995)[1,2,3]; Chen et al. (2005)[4]; Lin et al. (1990)[5,6]
f_{root}	Proportion of below-ground to the total production (dimensionless)	0.55	0.55	0.55	0.55	0.17	0.17	Shao et al. (1995)[1,3,4]; Chen et al. (2005)[2]; Lin et al. (1990)[5,6]
T_{veg}	The fraction of plant mediated transport was available (dimensionless)	1	1	1	1	0	0	Walter and Heimann (2000)
P_{ox}	The fraction of CH_4 oxidized during plant mediated transport (dimensionless)	0.9	0.9	0.9	0.9	–	–	Li et al. (2015)
VI	Vegetation index (dimensionless)	1	1	1	1	1	1	Li et al. (2015)
SAND	Soil sand fraction (%)	30	58	72	52	50	40	SSSC (1993[4]; 1994a[1]; 1994b[6]); This study[2]; Gong and Zhang (2015)[3]; SSSGD (1996)[5]
SOM	Concentration of soil organic matter ($g\ kg^{-1}$)	103	7.34	9.3	5.5	25.7	24.1	SSSC (1996)[1,5,6]; This study[2]; Yuan et al. (2015)[3]; Zhang et al. (2015)[4]
ρ	Soil bulk density ($g\ cm^{-3}$)	0.9	1.27	1.41	1.06	1.49	1.52	
N_0	Initial concentration of nitrogen in plant litter ($g\ kg^{-1}$)	12.2; 5.6[b]	12.2[a]; 5.6[b]	12.2[a]; 5.6[b]	15.5; 11.3[b]	9.7[a]; 1.0[b]	9.7[a]; 1.0[b]	Yuan et al. (2015)[1,2,3,4]; Gordon and Jackson (2003)[5,6]; White et al. (2002)[5,6]
LG_0	Initial concentration of lignin in plant litter ($g\ kg^{-1}$)	160[a]; 166[b]	160[a]; 166[b]	16[a]; 166[b]	207[a]; 218[b]	166[a]; 246[b]	9.7[a]; 1.0[b]	Yuan et al. (2015)[1,2,3,4]; Gordon and Jackson (2003)[5,6]; Benner and Hodson (1985)
Q_{10}	Temperature coefficient (dimensionless)	3.0	3.0	3.0	3.0	3.0	3.0	Li et al. (2010a)
R	The exchange coefficient between the air and the soil ($m\ day^{-1}$)	0.3	0.3	0.3	0.3	0.3	0.3	Xu (2012)

[$] For the year 2004 and 2011.
[^] For the year 2012.
[a] For the aboveground litter.
[b] For the belowground litter.
[1] For LR site.
[2] For YR site.
[3] For YC site.
[4] For CM site.
[5] For GZ site.
[6] For HK site.
[*] f_{above} represents the proportion of above-ground and leaves to the total production for the herbaceous plants and trees, respectively.

Heimann (2000), trees are poor transporters compared with grass with aerenchyma. Therefore, we did not consider CH_4 transportation via plants in mangroves. The vegetation index (*VI*) and the fraction of CH_4 oxidized during plant-mediated transport (P_{ox}) have been calibrated for freshwater marshlands (Li et al., 2015); therefore, the same values were used in this study. The constant *a* in Eq. (1) was calibrated as −0.056 by minimizing the root mean square error (*RMSE*) between the observed and simulated fluxes at a tidal marsh site in the Yellow River Delta (YR) from July to December 2012 (Table 1). By setting an increment of 0.001 for *a*, the model was run for all values of *a* within the range −0.2 to 0.0 until the *RMSE* between the simulated and observed daily CH_4 fluxes was minimized. The values and sources of the main parameters used for simulating the CH_4 emissions from the different sites are presented in Table 2.

2.5. Statistics used for model validation

We used a set of statistical methods (Smith et al., 1996, 1997) to evaluate the improvements of the model. In addition to the linear regressions between the simulations and observations (*R*), the *RMSE*, root mean deviation (*RMD*), model efficiency (*EF*) and coefficient of determination (*CD*) were used to quantify the performance of the model at the coastal wetland sites. The implications and the equations of the above statistical elements are described in Supplementary material S2.

2.6. Model sensitivity analysis

A sensitivity analysis was conducted to better understand the response of $CH4MOD_{wetland}$ to the drivers in the tidal marshes and mangroves. We tested the sensitivity of a subset of the parameters in a previous study (Li et al., 2010a). In this study, the sensitivity analysis was focused on the other factors (e.g., Q_{10} and P_{ox}) as well as the factors in the improved submodels (e.g., salinity and *R*). The sensitivity of the temperature coefficient (Q_{10} in Table 2) and the salinity (*s* in Eq. (2) and Table 2) was tested in both the tidal marshes and the mangroves. Plant-mediated transport is the primary mechanism of CH_4 emission in tidal marshes. According to previous studies, CH_4 oxidation through plant transportation differs between plant species (Ström et al., 2005; Popp et al., 2000; Calhoun and King, 1997). The sensitivity of the fraction of CH_4 oxidized during plant-mediated transport (P_{ox} in Table 2) was also tested for the tidal marshes. In the mangroves, we focused on the sensitivity of the exchange coefficient between the air and soil (*R* in Eq. (3) and Table 2). To determine model sensitivity, we ran $CH4MOD_{wetland}$ by changing the value of one factor while holding the remaining factors constant. For example, the response of the model to Q_{10} was iteratively simulated within the Q_{10} range of 1.0–16.0 (Table 3) while other factors were set to the baseline value (Table 3). The baseline values of Q_{10}, P_{ox} and *R* were obtained from Table 2. We used the average value of the tidal marshes and mangroves (Table 1) as the baseline salinity for each wetland type (Table 3). The average daily air temperature, water table depth, soil sand fraction and soil organic carbon of the tidal marshes and mangroves were used when running the model for each wetland type.

3. Results and discussion

3.1. Model validation

3.1.1. Validation of CH_4 emissions from tidal marshes

The seasonal patterns of the simulated and observed CH_4 emissions from the tidal marshes dominated by *Phragmites and Scripus* are shown in Fig. 3. The improved $CH4MOD_{wetland}$ model could generally simulate the seasonal changes in the CH_4 fluxes from the tidal marshes of the LR

Table 3
Range and baseline of the factors for model sensitivity analysis.

Factors	Maximum	Baseline	Minimum	Reference
Salinity	19.5	6.0[a]; 13.9[b]	3.2	This study (Table 1)
Q_{10}	16	3.0	1	Dunfield et al. (1993); Westerman and Ahring (1987); Yavitt et al. (1997)
P_{ox}	1.0	0.9[a]	0.0	Ström et al. (2005); Popp et al. (2000); Calhoun and King (1997)
R	0.63	0.3[b]	0.18	Happell et al. (1995)

[a] The baseline of the salinity for the herbaceous wetlands. The average salinity of the herbaceous site was used as the baseline salinity.
[b] The baseline of the salinity for the mangroves. The average salinity of the mangrove site was used as the baseline salinity.

site (Fig. 3a, $EF = 0.34$ in Table 4), YR site (Fig. 3c, $EF = 0.49$), YC site (Fig. 3e, $EF = 0.23$) and CM site (Fig. 3g, $EF = 0.56$).

At the LR site, peak CH_4 emission occurred in late August 1997 (Fig. 3a), in accordance with a relatively higher temperature as well as the highest water table depth (Fig. 3b). However, the model did not catch the low CH_4 fluxes in August and mid-September 1997 (Fig. 3a).

At the YR site, the improved model basically caught the peak emissions, i.e., in September 2012 and July 2013 (Fig. 3c), but some discrepancies were evident ($RMSE = 180\%$ in Table 4). Positive discrepancies between the modeled and observed CH_4 fluxes during the two years were evident in October. In addition, the improved model did not catch some small peaks during winter, especially in December (Fig. 3c).

The simulated seasonal variation of CH_4 fluxes using the improved model shows a similar trend with the observed variations at the YC site (Fig. 3e). However, the improved model systematically

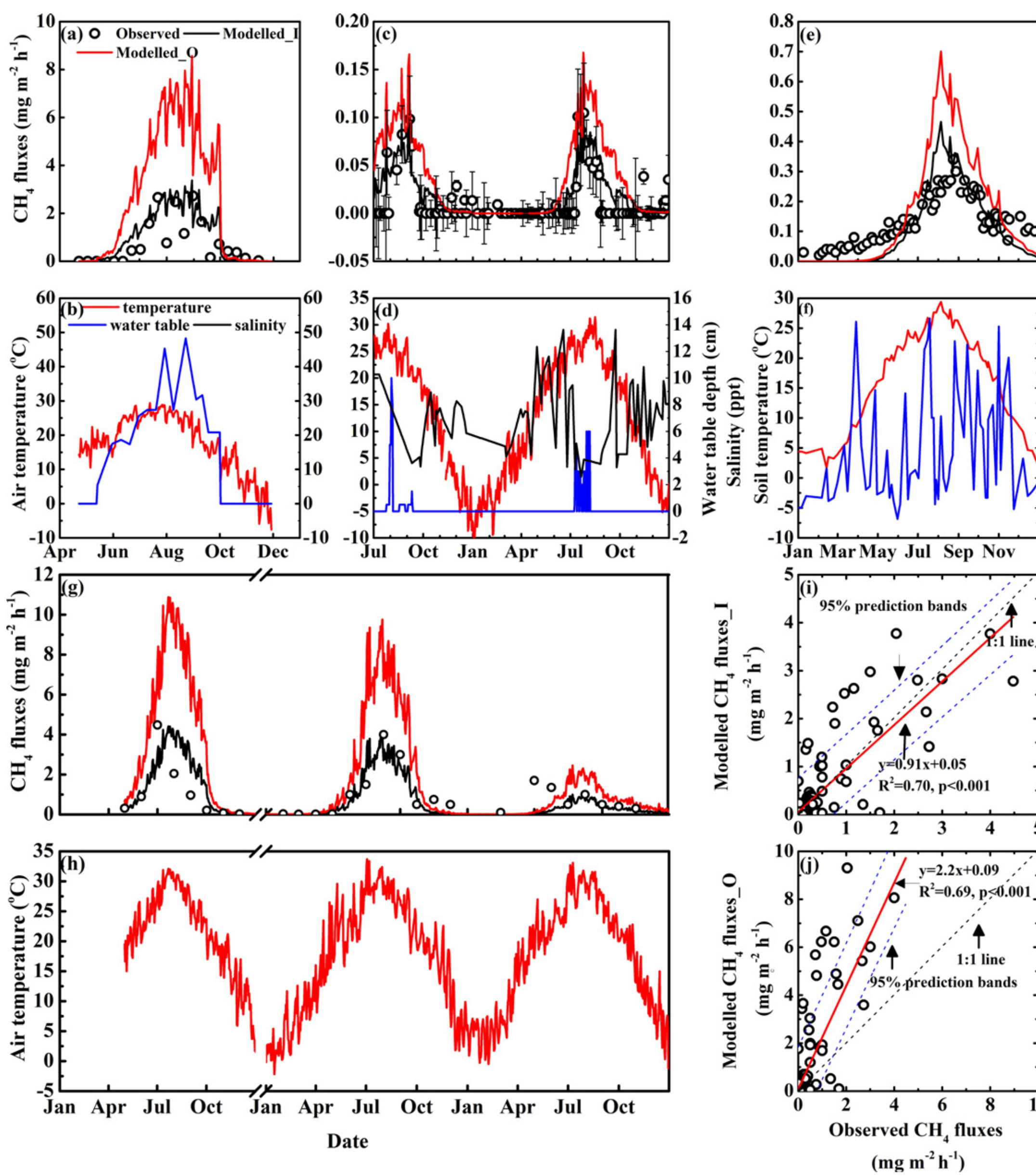

Fig. 3. Simulated and observed seasonal variations of CH_4 emissions and the environment factors from tidal marshes: (a) and (b) Liao River Estuary site; (c) and (d) Yellow River Delta site; (e) and (f) YanCheng Estuary site; (g) and (h) ChongMing Island site; The black and red lines represent the simulated CH_4 emissions by the improved and original model, respectively; Regression of simulated against the observed CH_4 fluxes from tidal marshes by the improved model (i) and the original model (j).

Table 4
Statistical analysis of model performance on daily and seasonal CH_4 emissions for different sites.

Criteria	LR	YR	YC	CM	GZ	HK	All	Seasonal
R_M	0.88	0.73	0.89	0.76	0.99	1.00	0.99	0.99
R_O	0.71	0.65	0.89	0.68	0.98	1.00	0.99	0.99
$RMSE(RMSE_{95\%})$_M %	94(225)	180(505)	48(213)	86(270)	7(137)	17(67)	56(1102)	14(164)
$RMSE(RMSE_{95\%})$_O %	341(225)	261(505)	104(213)	262(270)	455(137)	483(67)	3128(1102)	424(164)
RMD_M %	35	11	−3	−6	−1	−16	0.41	−2
RMD_O %	243	125	45	135	365	482	324	314
EF_M	0.34	0.49	0.23	0.56	0.99	0.72	1.00	0.99
EF_O	−7.68	−0.06	−2.44	−3.04	−48.72	−235.23	−13.77	−18.30
CD_M	0.83	2.52	0.35	1.06	0.82	0.76	0.95	0.92
CD_O	0.06	0.50	0.08	0.11	0.01	0.00	0.04	0.03
Number of samplings	20	102	66	30	3	3	224	13

underestimated the CH_4 fluxes during both the early and late growing season, and overestimated the CH_4 peak (Fig. 3e). The peak emission simulated by both the improved and original model corresponded to the highest soil temperature (Fig. 3f). However, the observed peak emission occurred approximately one month later than the simulated peaks (Fig. 3e).

The pattern of the simulated variations using the improved model generally matches the observed variations in different years at *CM* site dominated with *Scripus* (Fig. 3g). However, there was an approximate one- and two-month delay in the simulated peak emission rate when compared with the observed data from 2004 and 2012, respectively (Fig. 3g).

The regression of the simulated versus observed CH_4 fluxes (Fig. 3i) resulted in an R^2 value of 0.70, a slope of 0.91 and an intercept of 0.05 ($n = 221$, $p < 0.001$). Although some simulated points lay outside the standard errors of the individual measured values, they fell within the 95% confidence interval for the whole dataset ($RMSE < RMSE_{95\%}$ in Table 4). The modified model describes the trend in the measured data better than the mean of the observations at YR site and CM site ($CD > 1$ in Table 4). However, if the influence of salinity was not considered, the original model could not well simulate the observations from these sites (negative *EF* values in Table 4). The original model significantly overestimated the CH_4 fluxes (Fig. 3a, c, e and g). The regression of the simulated versus observed CH_4 fluxes (Fig. 3j) resulted in an R^2 value of 0.69, a slope of 2.2 and an intercept of 0.09 ($n = 221$, $p < 0.001$).

3.1.2. Validation of CH_4 emissions from mangroves

The $CH4MOD_{wetland}$ model could generally simulate the annual mean CH_4 fluxes between different mangrove sites under different water table depth and salinity conditions (Fig. 4, $EF > 1$ in Table 4). The simulated annual mean CH_4 fluxes increased in the sequence of the GZ_N, GZ_S and GZ_P microsites, which is in accordance with the observations (Fig. 4a). The annual mean CH_4 fluxes simulated using the original model are approximately four times higher than the observed data from the GZ_S and GZ_P microsites (Fig. 4a), with a linear regression slope of 5.2 (Fig. 4c). The *RMSE* and *RMD* were significantly decreased by the improved model, i.e., from 455% to 7% and from 365% to 1%, respectively (Table 4). Model efficiency was improved from −48.72 to 0.99 (Table 4).

At the HK site, the simulated CH_4 fluxes increased as the salinity decreased, i.e., in the following order: inner flat (HK_I), middle flat (HK_M) and outer flat (HK_O) (Fig. 4b, Table 4). This is in accordance with the observed data (Fig. 4b). There were small negative discrepancies between the simulated and observed CH_4 fluxes at the HK site when the improved model was used ($RMD = -16\%$ in Table 4). The original model could not accurately simulate the annual mean CH_4 fluxes at the HK site (negative *EF* value, $RMSE > RMSE_{95\%}$ in Table 4). This model significantly overestimated the annual mean CH_4 fluxes (Fig. 4d, $RMD = 482\%$), especially in the inner flat with the highest salinity (Fig. 4c). However, the improved model significantly increased the model *EF*, i.e., from −235.23 to 0.72 (Table 4).

3.1.3. Comparison of simulated and observed seasonal CH_4 emissions

A comparison of the observed and modeled seasonal CH_4 emissions is shown in Table 5. The improved model was able to simulate the differences in the CH_4 emissions between the sites and in different years ($EF = 0.99$ in Table 4). The observed seasonal CH_4 emissions ranged from 0.09 to 43.44 g m^{-2}, with an average value of 7.85 g m^{-2}. The simulated values using the improved model ranged from 0.1 to 44.90 g m^{-2}, with an average of 7.89 g m^{-2}. The seasonal CH_4 emissions simulated using the original model were much higher than the observed data ($RMD = 314\%$ in Table 4), especially at the mangrove sites, which were 4–10 times higher than the observed emissions (Table 5). Overall, the improved model significantly decreased the *RMSE* and *RMD*, i.e., from 424% to 14% and from 314% to −2%, and increased the *EF* and *CD*, i.e., from −18.3 to 0.99 and from 0.03 to 0.92, respectively (Table 4).

3.2. Model sensitivity analysis

The results of the sensitivity analysis are shown in Fig. 5. The model sensitivity analysis suggested that the CH_4 emissions decreased exponentially with increasing Q_{10} and salinity but levelled off when the Q_{10} exceeded 10 (Fig. 5a and d) and the salinity exceeded 12 (Fig. 5b and e). The CH_4 emissions decreased linearly with decreasing P_{ox} for the tidal marshes (Fig. 5c) but increased linearly with increasing R (Fig. 5f) for the mangroves. The most sensitive factor was P_{ox} for the tidal marshes (Fig. 5c). The CH_4 emissions from the mangroves were the most sensitive to salinity (Fig. 5e). The parameter R was of minor importance for the magnitude of the simulated CH_4 emissions (Fig. 5f). As Q_{10} increased from 1 to 16, the CH_4 emissions decreased by 85% and 62% for the tidal marshes (Fig. 5a) and mangroves (Fig. 5d), respectively. This suggests that CH_4 emissions in cold areas (Fig. 5a) are more sensitive to Q_{10} than in warm areas (Fig. 5d).

3.3. Salinity impacts on CH_4 simulation: importance and knowledge gap

The influence of salinity on CH_4 emissions happens widely not only in the coastal wetlands (Bartlett et al., 1987; Poffenbarger et al., 2011), but also in the saline and alkaline lakes (Joye et al., 1999; Bergier et al., 2014). The global coastal wetlands and the saline wetlands area accounts for ~13% of natural wetlands (Lehner and Döll, 2004). So it is important on studies concerning the salinity impacts on CH_4 emissions. The salinity may decrease CH_4 emissions by both limiting CH_4 production and promoting CH_4 oxidation. For one thing, salinity can inhibit the activities of or cause harm to methanogens, which reduces CH_4 emissions (Chidthaisong and Conrad, 2000; Baldwin et al., 2006). Reducing bacteria in the sediment that accept electrons, e.g., sulfate-reducing bacteria, compete with the methanogens for substrates and therefore inhibit CH_4 production (van der Gon et al. 2001; Lovley and Klug, 1983). In addition to sulfate, sulfide produced during the reduction of acid sulfate in soils following flooding is also considered to inhibit

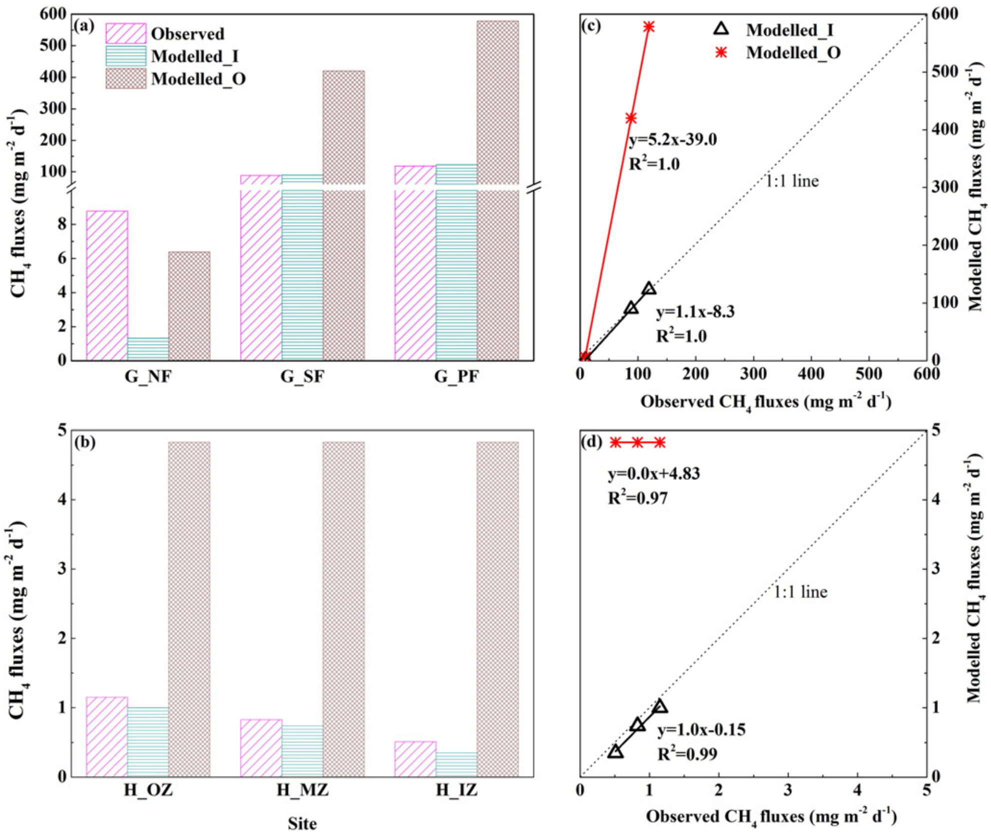

Fig. 4. Comparison and regression of simulated and observed annual mean CH_4 fluxes from mangrove sites in Guangzhou (a and c) and Haikou city (b and d). Modeled_I and Modeled_O represent the simulated CH_4 emissions by the modified and original model, respectively.

methanogenesis (Ramakrishnan et al., 1995). For another thing, the anaerobic oxidation of CH_4 (AOM) was regarded consequential in the wetland ecosystems in recent studies (Gupta et al., 2013; Segarra et al., 2015). Evidence suggested the sulfate may stimulate AOM then reduce CH_4 emissions (Eller et al., 2005; Schubert et al., 2011; Segarra et al., 2015).

The salinity impact on CH_4 emissions was so important but largely regarded as in consequential in the present simulations. First, estimating regional or global CH_4 emissions using a process-based model is prevalent in recent studies. However, most of the present process-based models (Cao et al., 1996; Walter et al., 1996; Potter, 1997; Zhang et al., 2002; Zhu et al., 2014; Riley et al., 2011; Wania et al., 2010; Tian et al., 2010; Xu et al., 2010) consider the influence of soil temperature, soil texture, redox potential and soil pH, but ignore the influence of salinity on CH_4 emissions. According to our results (Fig. 3j; Fig. 4c and d), these models may inevitably overestimate regional or global CH_4 emissions (Cao et al., 1998; Petrescu et al., 2010; Melton et al., 2013; Zhu et al., 2015). For example, WETCHIMP (Wetland CH_4 Inter-comparison of Models Project) determined that the global CH_4 emissions from natural wetlands were ~190 Tg using the above models (Melton et al., 2013). On a global scale, the coastal wetland area accounts for ~10% of natural wetlands (Lehner and Döll, 2004). If we simply assume that coastal wetlands are uniformly distributed across climatic zones, the CH_4 emissions from coastal wetlands account for 19 Tg. This study shows that if a model does not consider the influence of salinity, annual mean CH_4 emissions from coastal wetlands will be overestimated by ~280% (Table 5). WETCHIMP may overestimate CH_4 emissions by 14 Tg.

Table 5
Comparison of observed and modeled seasonal CH_4 emissions ($g\ m^{-2}\ yr^{-1}$).

Site	Period of estimation	Observation	Modeled_I[a]	Modeled_O[b]
LR	1997.4–1997.10	3.81	4.70	12.97
YR	2012.7–2012.12	0.09	0.11	0.23
YR	2013.1–2013.12	0.13	0.12	0.24
YC	2012.1–2012.12	1.09	0.94	1.41
CM	2004.5–2004.12	6.52	8.80	21.88
CM	2011.1–2011.12	8.29	7.07	19.50
CM	2012.1–2012.12	5.05	2.08	5.49
HK_O	1996.1–1997.12	0.42	0.37	1.76
HK_M	1996.1–1997.12	0.30	0.27	1.76
HK_I	1996.1–1997.12	0.19	0.13	1.76
GZ_N	2005.3–2005.12	0.48	0.49	2.33
GZ_S	2005.3–2005.12	32.19	32.63	153.39
GZ_P	2005.3–2005.12	43.44	44.90	211.21

[a] Modeled_I represents the modeled seasonal CH_4 emissions by the improved model.
[b] Modeled_O represents the modeled seasonal CH_4 emissions by the original model.

In addition, sea-level rise may result in greater saltwater intrusion and increased salinity in coastal wetlands (Titus, 1988), and even transform freshwater marshes near estuaries into tidal marshes in the near future (Weston et al., 2006, 2011; Herbert et al., 2015). Salinization will threaten numerous countries, including Australia, Argentina, China, the Common wealth of Independent States (former Soviet Union), India, Iran, Iraq, South Africa, Thailand and the United States of America, as well as those in Northern Africa (Bailey and James, 2000). Along with the salinity intrusion, the available sulfate radical in seawater (approximately 28 mmol L^{-1}) will replace methanogenesis as a dominant anaerobic terminal C mineralization process in marine sediments and salt marsh soils (Jørgensen, 1982; Capone and Kiene, 1988). In addition, increased sulfate loading may further reduce methane fluxes by stimulating AOM (Dise and Verry, 2001; Segarra et al., 2015). Predictions of CH_4 emissions from wetlands are important for the government to make policies for wetland restoration and address future climate change. The influence of salinity on CH_4 emissions needs to be considered to improve the prediction of CH_4 emissions.

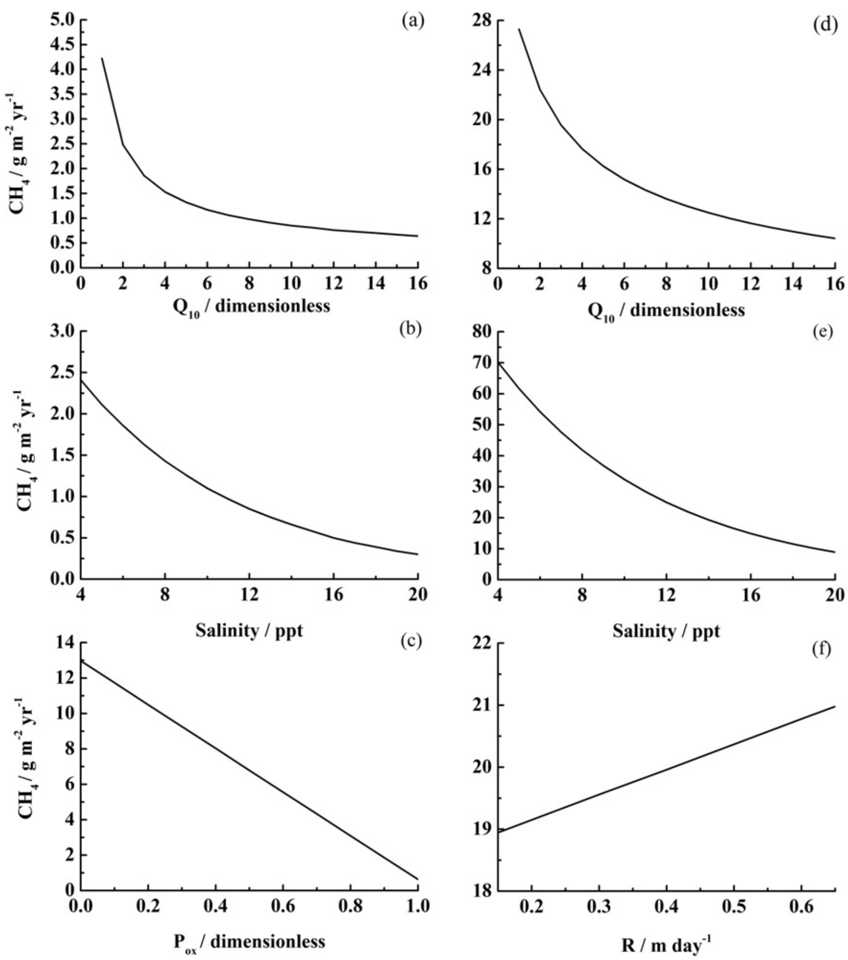

Fig. 5. Model sensitivity to model inputs and parameters. (a), (b) and (c) show the CH_4 sensitivity to Q_{10}, salinity and R in tidal marshes; (c), (d) and (e) show the CH_4 sensitivity to Q_{10}, salinity and P_{ox} in the mangroves, respectively.

Previous studies reported a negative relationship between CH_4 emissions and salinity (Delaune et al. 1983; Bartlett et al. 1987). Recently, Poffenbarger et al. (2011) extended this relationship to a wider range of salinity conditions (0.4 to 35.1 ppt). In China, the salinity is within a range of 2–7 ppt for tidal marshes and 10–20 ppt for mangroves (Song et al., 1996). Therefore, it seems reasonable to adopt the relationship of Poffenbarger et al. (2011) for $CH4MOD_{wetland}$ to simulate CH_4 emissions from China. However, the importance of salinity on methanogenesis and the methanogenic microbial population has not been studied in detail (Pattnaik et al., 2000). Salinity can influence the microbial processes of carbon cycling, but a simple non-linear regression between CH_4 production and salinity cannot express this complex process. This may have resulted in the discrepancies between the simulated and observed CH_4 emissions (Fig. 3, Fig. 4). A lack of observed salinity may also induce uncertainties in the CH_4 simulations. For example, we used the annual mean salinity to simulate the CH_4 emissions, which may induce bias between the simulated and observed values (Fig. 3a, e, g, a and b). A more detailed submodel with a mechanism to introduce the influence of different anions on methanogenesis and carbon cycling is needed in future to reduce the uncertainty in regional estimations.

3.4. CH_4 simulations in mangroves: uncertainties and future needs

Compared with freshwater marshes, methanogenesis in and methane emissions from mangroves have been regarded as minor, both in a particular ecosystem and on a regional scale (Sotomayor et al., 1994; Giani et al., 1996; Alongi et al., 2000, 2001). In China, mangroves are estimated to cover an area of 352 km^2, and are mainly distributed along the coast in Fujian, Guangdong, Guangxi, Taiwan and Hainan provinces (Zhang and Jiang, 2014). To the best of our knowledge, only Zhang and Jiang (2014) estimated 0.25 Gg of CH_4 emissions from mangroves in China by extrapolating the annual CH_4 flux from the HK site (Fig. 4b). However, high CH_4 emissions have also been observed in mangroves, e.g., in Guangzhou (Fig. 4a) and India (Purvaja and Ramesh, 2001). Compared with extrapolation methods, process-based models can express the complex physiological processes of plants and microorganisms that are regulated by climatic and edaphic factors. However, the mechanisms of CH_4 emissions from mangroves are notoriously uncertain in the present process-based models.

First, the mechanisms of CH_4 transportation and oxidation in mangroves are not as clear as in herbaceous plants. It is generally recognized that trees are poor transporters compared with herbaceous plants with aerenchyma (Hook et al., 1971; Grosse et al., 1992; Walter et al., 1996; Walter and Heimann, 2000). However, some studies have shown that mangrove pneumatophores may increase CH_4 fluxes by transport through vascular channels, but this may be counter-balanced to some degree by root releases of oxygen into the rhizosphere, thereby increasing CH_4 oxidation (Purvaja et al., 2004; Kitaya et al., 2002; Purnobasuki and Suzuki, 2005). Because it is difficult to quantify transportation and oxidation that occurs via pneumatophores, these processes are usually ignored in the present models. Second, according to the sensitivity analysis, CH_4 emission was most sensitive to salinity in the mangroves (Fig. 5a). This relationship (Eq. (2)) was based on observed data from tidal marshes (Poffenbarger et al., 2011). A negative relationship was found for mangroves at the HK site in China (Fig. 4b); however, few researchers have reported a similar relationship for other mangrove studies.

In addition to the model structure, uncertainties may have arisen from the limited input data. For example, at the HN site in our study, we used the *NPP* of *Bruguiera sexangula* from another study (Lin et al., 1990) because the author did not collect *NPP* data (Ye et al., 2000). At the GZ site, we used the same *NPP* value for all of the micro sites. However, there were spatial heterogeneities in the *NPP* of the mangroves. This may have caused the differences between the simulated and observed CH_4 fluxes (Fig. 4a and b).

In future, more experiments focused on both site-specific observations and the mechanisms of CH_4 production, oxidation and emission in mangroves are needed. Process-based models should include the specific characteristics of CH_4 processes in mangroves to accurately estimate CH_4 emissions at a regional or global scale.

4. Conclusion

This study introduced the influence of soil salinity on CH_4 emissions into a biogeophysical model, i.e., $CH4MOD_{wetland}$, to make it appropriate for use in coastal wetlands. The improved model can reasonably describe the observed CH_4 emission variations from the tidal marshes in the Liao River Estuary, Yellow River Delta, YanCheng Estuary and ChongMing Island. In addition, the model generally simulated the difference between the annual mean CH_4 fluxes from mangrove sites in GuangZhou and HaiKou city under different water table depth and salinity conditions. The improved model had a significantly lower *RMSE* and *RMD* and an improved efficiency compared with the original model. However, there was bias between the simulation and observations, which may have resulted from the model structure as well as the limited observations of environmental drivers. Further improvements in its descriptive power, i.e., the detailed microbial processes influenced by salinity and the specific characteristics of CH_4 transportation and oxidation in mangroves, are required to predict methane emissions more accurately from coastal wetlands.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (Grant No. 31000234, 41321064 and 41175132), the Chinese Academy of Sciences (CAS) strategic pilot technology special funds (Grant No. XDA05020204 and XDA05050507) and the Climate Change Special Foundation of China Meteorological Administration (CCSF201604). We could thank the National Meteorological Information Center of the China Meteorological Administration for providing the data.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2016.03.186.

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