Field-scale simulation of methane emissions from coastal wetlands in China using an improved version of CH4MOD\textsubscript{wetland}

Tingting Li \textsuperscript{a}, Baohua Xie \textsuperscript{b,c}, Guocheng Wang \textsuperscript{a}, Wen Zhang \textsuperscript{a}, Qing Zhang \textsuperscript{a}, Timo Vesala \textsuperscript{d,e}, Maarit Raivonen \textsuperscript{d}

\textsuperscript{a} LAPC, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, PR China
\textsuperscript{b} Key Laboratory of Coastal Environmental Processes and Ecological Remediation, Yantai Institute of Coastal Zone Research (YIC), Chinese Academy of Sciences (CAS), Yantai, Shandong 264003, PR China
\textsuperscript{c} Shandong Provincial Key Laboratory of Coastal Environmental Processes, YICCAS, Yantai, Shandong 264003, PR China
\textsuperscript{d} Department of Physics, P.O. Box 48, FI-00014, University of Helsinki, Finland
\textsuperscript{e} Department of Forest Sciences, P.O. Box 27, FI-00014, University of Helsinki, Finland

**Highlights**

- Salinity effect on CH\textsubscript{4} emissions in coastal wetlands was added into CH4MOD\textsubscript{wetland}.
- Modeled seasonal CH\textsubscript{4} variations corresponded well with observations in tidal marshes.
- Modeled seasonal CH\textsubscript{4} emissions agreed well with observations in coastal wetlands.
- The improved model significantly increased model efficiency.
- Previous regional/global CH\textsubscript{4} estimations from wetlands may be overestimated.

**Abstract**

Coastal wetlands are important CH\textsubscript{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\textsubscript{4} source remains highly uncertain. We introduced the influence of salinity on CH\textsubscript{4} production and CH\textsubscript{4} diffusion into a biogeophysical model named CH4MOD\textsubscript{wetland} so that it can be used in coastal wetlands. The improved model can generally simulate seasonal CH\textsubscript{4} variations from tidal marshes dominated by Phragmites and Scirpus. However, the model underestimated winter CH\textsubscript{4} fluxes from tidal marshes in the Yellow River Delta and YanCheng Estuary. It also failed to capture the accurate timing of the CH\textsubscript{4} peaks in YanCheng Estuary and ChongMing Island in 2012. The improved model could generally simulate the difference between the annual mean CH\textsubscript{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\textsubscript{4} emissions simulated across all of the coastal wetlands ranged from 0.1 to 44.90 g m\textsuperscript{-2}, with an average value of 7.89 g m\textsuperscript{-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 49%, respectively, and improved the EF from −18.30 to 0.99.

**Keywords:** CH\textsubscript{4} emissions, Salinity, Coastal wetlands, CH4MOD\textsubscript{wetland} model, China

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\textsuperscript{*} Corresponding authors.
E-mail addresses: litingtting@mail.iap.ac.cn (T. Li), bhxie@yic.ac.cn (B. Xie).
Model sensitivity analysis showed that CH4 emissions were most sensitive to $P_{\text{m}}$ in the tidal marshes and salinity in the mangroves. The results show that previous studies may have overestimated CH4 emissions on a regional or global scale by neglecting the influence of salinity. In general, the CH4MODwetland model can simulate seasonal CH4 emissions from different types of coastal wetlands under various conditions. Further improvements of CH4MODwetland should include the specific characteristics of CH4 processes in mangroves to decrease the uncertainty in estimating regional or global CH4 emissions from natural wetlands.

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1. Introduction

Methane (CH4) 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 CO2 (Forster et al., 2007). Recently, it was reported that when the indirect global warming effects of CH4 on aeronsols and other chemical compounds (e.g., O3) was incorporated, previous estimates of the global warming potential of CH4 may be 10–40% too low (Shindell et al., 2009; IPCC, 2013). Natural wetlands are the single largest natural source of CH4, accounting for approximately one third of total global CH4 emissions, i.e., equivalent to 115 (Fung et al., 1991) to 237 Tg CH4 yr$^{-1}$ (Hein et al., 1997). CH4 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; Ringleval et al., 2011). Moreover, they appear to dominate the interannual variability of the global CH4 source (Bousquet et al., 2006; Ciais et al., 2013). Therefore, increased knowledge of CH4 emissions from natural wetlands is important to understand the global CH4 budget.

Efforts have been made to estimate regional CH4 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 CH4 production, oxidation and emission as well as the influence of complex climate, soil, vegetation and hydrology conditions on CH4 emissions (Cao et al., 1996; Li, 2000; Zhang et al., 2002). Some process-based models focus on the CH4 cycling mechanisms and only describe the processes of CH4 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 CH4 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 CH4 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 $660 \times 10^3$ 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 CH4 than less saline wetlands (Bartlett et al., 1987; Popenbarger et al., 2011). This has been explained by the presence of sulfate in the sea water, which allows sulfate-reducing bacteria to outcompete methane producers for energy sources, consequently inhibiting methane production (DeLaune et al., 1983; Bartlett et al., 1987; Wang et al., 1996; Popenbarger et al., 2011). However, most of the above estimates of regional or global CH4 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 CH4 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 CH4 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 CH4 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 CH4MODwetland, which includes the processes of CH4 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 CH4 production into CH4MODwetland to improve the applicability of the model for use in coastal wetlands. We also compiled a CH4 measurement dataset from latitudinally distributed coastal zone sites using the literature and measurements. The objectives of this study are to modify the CH4MODwetland model, and to assess the performance of the improved model with respect to CH4 fluxes 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

CH4MODwetland is a biogeophysical process-based model that was developed to describe the processes of CH4 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 CH4 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 CH4 production and emissions.

The main processes related to CH4 production, oxidation and emission in CH4MODwetland 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 CH4 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 CH4 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 CH4 emission.
CH4 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 CH4 to ensure that the model can be used at mangrove sites. CH4 oxidation occurs through plant and diffusion transportation.

CH4MODwetland has been validated against independent field measurements of CH4 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 CH4MODwetland are well documented in previous studies (Li et al., 2010a, 2012, 2015).

2.2. Model modification

As shown in Fig. 1, CH4 production is influenced by the soil temperature, soil texture and soil redox potential. When the influence of soil salinity is included, CH4 production is calculated as:

\[ P = 0.27 \times SI \times TI \times F_{SI} \times F_{S} \times (C_P + C_L + C_{SOM}) \]  

(1)

where \( P \) is CH4 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; \( SI, TI, F_{SI} \) and \( F_{S} \) are the influence of soil texture, soil temperature, soil index potential and soil salinity, respectively, on CH4 production; 0.27 is a factor (mole weight basis) to convert carbohydrates 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 CH4 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:

\[ F_{S} = 10^{a-s} \]  

(2)

where \( F_{S} \) represents the effect of salinity on CH4 production, \( s \) is the salinity (ppt), and \( a \) is an empirical constant.

The original version of CH4MODwetland was mainly used to simulate aerenchymous vascular plant-dominated sites. At these sites, plant-mediated transportation and ebullition contribute ~90% of CH4 emissions. However, if the model is applied at a wetland site without aerenchymous vascular plants, e.g., a mangrove site, diffusion cannot be neglected. This is because compared with vascular plants, woody plants are poor transporters of CH4 (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 CH4 diffusion rate:

\[ E_{D} = R \times (P \times h - P_{max}) \times f_{out} \]  

(3)

where \( E_{D} \) is the CH4 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 \( f_{out} \) represents the fraction of CH4 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 CH4 fluxes and the synchronous air temperature, precipitation, water table depth and salinity at the YR site from July 2012 to
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 CH₄ 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 CH₄ 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 CH₄ emissions were calculated by summing the daily fluxes. The absence of CH₄ flux measurements between two adjacent days of observation was linearly interpolated. The observed total seasonal CH₄ emissions from mangroves were simply calculated using the annual mean CH₄ fluxes.

### Table 1

<table>
<thead>
<tr>
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<tbody>
<tr>
<td>Location</td>
<td>40°02′N, 122°20′E</td>
<td>37°45′N, 118°59′E</td>
<td>33°22′N, 120°42′E</td>
<td>31°15′N, 121°30′E</td>
<td>23°01′N, 113°46′E</td>
<td>19°51′N, 110°24′E</td>
</tr>
<tr>
<td>Wetland type</td>
<td>Tidal marsh</td>
<td>Tidal marsh</td>
<td>Tidal marsh</td>
<td>Tidal marsh</td>
<td>Mangrove</td>
<td>Mangrove</td>
</tr>
<tr>
<td>Dominant plant species</td>
<td>Phragmites</td>
<td>Phragmites</td>
<td>Phragmites</td>
<td>Scirpus</td>
<td>Aegiceras corniculatum etc.</td>
<td>Bruguiera sexangula</td>
</tr>
<tr>
<td>Annual air temperature (°C)</td>
<td>9</td>
<td>12.9</td>
<td>12.6</td>
<td>15.3</td>
<td>21.9</td>
<td></td>
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<tr>
<td>Annual precipitation (mm)</td>
<td>625</td>
<td>590</td>
<td>1040</td>
<td>1117</td>
<td>1582</td>
<td></td>
</tr>
<tr>
<td>Water environment</td>
<td>Seasonally</td>
<td>Occasionally</td>
<td>Occasionally</td>
<td>Perennially</td>
<td>Perennially/Seasonally/Never</td>
<td></td>
</tr>
<tr>
<td>Average salinity (ppt)</td>
<td>7.2</td>
<td>6.7</td>
<td>3.2</td>
<td>6.9</td>
<td>12.5</td>
<td></td>
</tr>
<tr>
<td>References</td>
<td>Huang et al. (2005)</td>
<td>This study</td>
<td>Yuan et al. (2015)</td>
<td>Yang et al. (2007); Li et al. (2010b; 2014)</td>
<td>Kang et al. (2008)</td>
<td></td>
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</tbody>
</table>

2.4. Model parameterization

The description and values of the main CH₄MOD 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$_{\text{above}}$ and F$_{\text{root}}$) 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 (L$_G$ 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$_{\text{veg}}$) differs between grass species and mangroves. According to Walter and
Table 2
Model input and parameters.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Description (unit)</th>
<th>Value</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>NPP</td>
<td>Net primary productivity (g m⁻² yr⁻¹)</td>
<td>2640, 935, 1738, 1570, 522, 2000, 3000</td>
<td>Shao et al. (1995)¹; This study²; Yuan et al. (2015)³; Li et al. (2014)⁴; Lin et al. (1990)⁵; Kang et al. (2008)⁶; Shao et al. (1995)⁷; Chen et al. (2005)⁸; Lin et al. (1998)⁹; Li et al. (2013).</td>
</tr>
<tr>
<td>f_above</td>
<td>Proportion of above-ground/leaves to the total production (dimensionless)</td>
<td>0.45, 0.45, 0.45, 0.45, 0.35, 0.35</td>
<td>Shao et al. (1995)¹; Chen et al. (2005)⁸; Lin et al. (1998)⁹; Li et al. (2014)⁴</td>
</tr>
<tr>
<td>f_root</td>
<td>Proportion of below-ground to the total production (dimensionless)</td>
<td>0.55, 0.55, 0.55, 0.55, 0.17, 0.17</td>
<td>Shao et al. (1995)¹; Chen et al. (2005)⁸; Lin et al. (1998)⁹; Li et al. (2014)⁴</td>
</tr>
<tr>
<td>T_ave</td>
<td>The fraction of plant mediated transport (dimensionless)</td>
<td>1, 1, 1, 1, 0, 0</td>
<td>Walter and Heimann (2000)</td>
</tr>
<tr>
<td>P_ox</td>
<td>The fraction of CH₄ oxidized during plant-mediated transport (dimensionless)</td>
<td>0.9, 0.9, 0.9, 0.9</td>
<td>Li et al. (2015)</td>
</tr>
<tr>
<td>VI</td>
<td>Vegetation index (dimensionless)</td>
<td>1, 1, 1, 1, 1</td>
<td>Li et al. (2015)</td>
</tr>
<tr>
<td>SAND</td>
<td>Soil sand fraction (%)</td>
<td>30, 58, 72, 52, 50, 40</td>
<td>SSSC (1993)¹; 1994a)²; This study³; 1994b)⁶; This study²; Yuan et al. (2015)³; Zhang et al. (2020)⁵; Zhang et al. (2015)⁶</td>
</tr>
<tr>
<td>SDM</td>
<td>Concentration of soil organic matter (g kg⁻¹)</td>
<td>103, 7.3, 9.3, 5.5, 25.7, 24.1</td>
<td>SSSC (1996)¹; This study²; Yuan et al. (2015)³; Shao et al. (1995)¹; Chen et al. (2005)⁴; Lin et al. (1990)⁵; Kang et al. (2008)⁶</td>
</tr>
<tr>
<td>β</td>
<td>Soil bulk density (g cm⁻³)</td>
<td>0.9, 1.27, 1.41, 1.06, 1.49, 1.52</td>
<td>SSSC (1996)¹; This study²; Yuan et al. (2015)³; Shao et al. (1995)¹; Chen et al. (2005)⁴; Lin et al. (1990)⁵; Kang et al. (2008)⁶</td>
</tr>
<tr>
<td>N₀</td>
<td>Initial concentration of nitrogen in plant litter (g kg⁻¹)</td>
<td>12.2; 12.2; 12.2; 12.2; 15.5; 9.7; 9.7;</td>
<td>Yinan et al. (2015)¹; Chen et al. (2005)⁴; Gordon and Jackson (2003)¹⁵; White et al. (2002)¹⁶</td>
</tr>
<tr>
<td>LG₀</td>
<td>Initial concentration of lignin in plant litter (g kg⁻¹)</td>
<td>160²; 160²; 16²; 16²; 16²; 16²; 16²; 16²; 16²</td>
<td>Yinan et al. (2015)¹; Chen et al. (2005)⁴; Gordon and Jackson (2003)¹⁵; Benner and Hodson (1985)</td>
</tr>
<tr>
<td>Q₁₀</td>
<td>Temperature coefficient (dimensionless)</td>
<td>3.0, 3.0, 3.0, 3.0, 3.0, 3.0</td>
<td>Lin et al. (2010a)</td>
</tr>
<tr>
<td>R</td>
<td>The exchange coefficient between the air and the soil (m day⁻¹)</td>
<td>0.3, 0.3, 0.3, 0.3, 0.3, 0.3</td>
<td>Xu (2012)</td>
</tr>
</tbody>
</table>

¹ For the year 2004 and 2011.
² For the year 2012.
³ For the aboveground litter.
⁴ For the belowground litter.
⁵ For LR site.
⁶ For YC site.
⁷ For CM site.
⁸ For GZ site.
⁹ For HK site.

¹⁰ For the year 2012.
¹¹ For the year 2004.
¹² For the year 2011.

Heimann (2000), trees are poor transporters compared with grass with aerenchyma. Therefore, we did not consider CH₄ transportation via plants in mangroves. The vegetation index (VI) and the fraction of CH₄ 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 CH₄ fluxes was minimized. The values and sources of the main parameters used for simulating the CH₄ 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., Q₁₀ and P_ox) as well as the factors in the improved submodels (e.g., salinity and R). The sensitivity of the temperature coefficient (Q₁₀ 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 CH₄ emission in tidal marshes. According to previous studies, CH₄ 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 CH₄ 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 Q₁₀ was iteratively simulated within the Q₁₀ range of 1.0–16.0 (Table 3) while other factors were set to the baseline value (Table 3). The baseline values of Q₁₀, 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 CH₄ emissions from tidal marshes

The seasonal patterns of the simulated and observed CH₄ emissions from the tidal marshes dominated by Phragmites and Scirpus are shown in Fig. 3. The improved CH4MOD_wetland model could generally simulate the seasonal changes in the CH₄ fluxes from the tidal marshes of the LR
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 CH4 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 CH4 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 CH4 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 CH4 fluxes using the improved model shows a similar trend with the observed variations at the YC site (Fig. 3e). However, the improved model systematically

Table 3

<table>
<thead>
<tr>
<th>Factors</th>
<th>Maximum</th>
<th>Baseline</th>
<th>Minimum</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Salinity</td>
<td>19.5</td>
<td>6.0; 13.9</td>
<td>3.2</td>
<td>This study (Table 1)</td>
</tr>
<tr>
<td>Q10</td>
<td>16</td>
<td>3.0</td>
<td>1</td>
<td>Dunfield et al. (1993); Westerman and Abring (1987); Yavitt et al. (1997)</td>
</tr>
<tr>
<td>Pox</td>
<td>1.0</td>
<td>0.9</td>
<td>0.0</td>
<td>Ström et al. (2005); Popp et al. (2000); Calhoun and King (1997)</td>
</tr>
<tr>
<td>R</td>
<td>0.63</td>
<td>0.3</td>
<td>0.18</td>
<td>Happell et al. (1995)</td>
</tr>
</tbody>
</table>

*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.
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 Scirpus (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 ($\text{RMSE} < \text{RMSE}_{\text{RES}}$ in Table 4). The modified model describes the trend in the measured data better than the mean of the observations at the 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 CH4MODwetland 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 1% 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_O), 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, $\text{RMSE} > \text{RMSE}_{\text{RES}}$ 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 $P_{\text{ex}}$ for the tidal marshes (Fig. 5c) but increased linearly with increasing $R$ (Fig. 5f) for the mangroves. The most sensitive factor was $P_{\text{ex}}$ 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; Conrad and Klug, 1983). In addition to sulfate, sulfate produced during the reduction of acid sulfate in soils following flooding is also considered to inhibit

### Table 4

<table>
<thead>
<tr>
<th>Criteria</th>
<th>LR</th>
<th>YR</th>
<th>YC</th>
<th>CM</th>
<th>GZ</th>
<th>HK</th>
<th>All</th>
<th>Seasonal</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R_M$</td>
<td>0.88</td>
<td>0.73</td>
<td>0.89</td>
<td>0.76</td>
<td>0.99</td>
<td>1.00</td>
<td>0.99</td>
<td>0.99</td>
</tr>
<tr>
<td>$R_O$</td>
<td>0.71</td>
<td>0.65</td>
<td>0.89</td>
<td>0.68</td>
<td>0.98</td>
<td>1.00</td>
<td>0.99</td>
<td>0.99</td>
</tr>
<tr>
<td>$\text{RMSE(\text{RMSE}_{\text{RES}}), M}$</td>
<td>94(225)</td>
<td>180(505)</td>
<td>48(213)</td>
<td>86(270)</td>
<td>7(137)</td>
<td>17(67)</td>
<td>56(1102)</td>
<td>14(164)</td>
</tr>
<tr>
<td>$\text{RMSE(\text{RMSE}_{\text{RES}}), O}$</td>
<td>341(225)</td>
<td>261(505)</td>
<td>104(213)</td>
<td>262(270)</td>
<td>455(137)</td>
<td>483(67)</td>
<td>3128(1102)</td>
<td>424(164)</td>
</tr>
<tr>
<td>RMD, M</td>
<td>35</td>
<td>11</td>
<td>−3</td>
<td>−6</td>
<td>−1</td>
<td>−16</td>
<td>0.41</td>
<td>−2</td>
</tr>
<tr>
<td>RMD, O</td>
<td>243</td>
<td>125</td>
<td>45</td>
<td>135</td>
<td>365</td>
<td>482</td>
<td>324</td>
<td>314</td>
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<td>EF, M</td>
<td>0.34</td>
<td>0.49</td>
<td>0.23</td>
<td>0.56</td>
<td>0.99</td>
<td>0.72</td>
<td>1.00</td>
<td>0.99</td>
</tr>
<tr>
<td>EF, O</td>
<td>−7.68</td>
<td>−0.06</td>
<td>−2.44</td>
<td>−3.04</td>
<td>−48.72</td>
<td>−235.23</td>
<td>−13.77</td>
<td>−18.30</td>
</tr>
<tr>
<td>CD, M</td>
<td>0.83</td>
<td>2.52</td>
<td>0.35</td>
<td>1.06</td>
<td>0.82</td>
<td>0.76</td>
<td>0.95</td>
<td>0.92</td>
</tr>
<tr>
<td>CD, O</td>
<td>0.06</td>
<td>0.50</td>
<td>0.08</td>
<td>0.11</td>
<td>0.01</td>
<td>0.00</td>
<td>0.04</td>
<td>0.03</td>
</tr>
<tr>
<td>Number of samplings</td>
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<td>102</td>
<td>66</td>
<td>30</td>
<td>3</td>
<td>3</td>
<td>224</td>
<td>13</td>
</tr>
</tbody>
</table>

3.3.1. Statistical analysis of model performance on daily and seasonal CH$_4$ emissions for different sites.
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.

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 Commonwealth 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.

Table 5

<table>
<thead>
<tr>
<th>Site</th>
<th>Period of estimation</th>
<th>Observation</th>
<th>Modeled$_I^a$</th>
<th>Modeled$_O^b$</th>
</tr>
</thead>
<tbody>
<tr>
<td>YR</td>
<td>2012.7–2012.12</td>
<td>0.09</td>
<td>0.11</td>
<td>0.23</td>
</tr>
<tr>
<td>YR</td>
<td>2013.1–2013.12</td>
<td>0.13</td>
<td>0.12</td>
<td>0.24</td>
</tr>
<tr>
<td>YC</td>
<td>2012.1–2012.12</td>
<td>1.09</td>
<td>0.94</td>
<td>1.41</td>
</tr>
<tr>
<td>CM</td>
<td>2004.5–2004.12</td>
<td>6.52</td>
<td>8.00</td>
<td>21.88</td>
</tr>
<tr>
<td>CM</td>
<td>2012.1–2012.12</td>
<td>5.05</td>
<td>2.08</td>
<td>5.49</td>
</tr>
<tr>
<td>HK</td>
<td>1996.1–1997.12</td>
<td>0.42</td>
<td>0.37</td>
<td>1.76</td>
</tr>
<tr>
<td>HK</td>
<td>1996.1–1997.12</td>
<td>0.30</td>
<td>0.27</td>
<td>1.76</td>
</tr>
<tr>
<td>HK</td>
<td>1996.1–1997.12</td>
<td>0.19</td>
<td>0.13</td>
<td>1.76</td>
</tr>
<tr>
<td>GZ</td>
<td>2005.3–2005.12</td>
<td>0.48</td>
<td>0.49</td>
<td>2.33</td>
</tr>
<tr>
<td>GZ</td>
<td>2005.3–2005.12</td>
<td>43.44</td>
<td>44.90</td>
<td>211.21</td>
</tr>
</tbody>
</table>

$^a$ 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.

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.
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 CH$_4$MODwetland 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 CH4 fluxes (Fig. 4a and b).

In future, more experiments focused on both site-specific observations and the mechanisms of CH4 production, oxidation and emission in mangroves are needed. Process-based models should include the specific characteristics of CH4 processes in mangroves to accurately estimate CH4 emissions at a regional or global scale.

4. Conclusion

This study introduced the influence of soil salinity on CH4 emissions into a biogeochemical model, i.e., CH4MODwetlands, to make it appropriate for use in coastal wetlands. The improved model can reasonably describe the observed CH4 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 CH4 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 CH4 transportation and oxidation in mangroves, are required to predict methane emissions more accurately from coastal wetlands.

Acknowledgements

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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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2010GC003946.
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variation of CH4 emission from Chongming east intertidal flat of Yangtze River Estu-
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