

## Methane exchange between marshland and the atmosphere over China during 1949–2008

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[1] We used a process-based ecosystem model to examine methane (CH<sub>4</sub>) fluxes in the marshland across China as a result of multifactor global changes during 1949–2008. Our simulated results show a significant declining rate of 18.7 Gg C a<sup>-1</sup> (1Gg = 10<sup>9</sup> g) (with a 95% confidence boundary of 17.6 ~ 19.8 Gg C a<sup>-1</sup>) at national scale, but substantially varying from the maximum annual CH<sub>4</sub> emission of 2.4 Tg C a<sup>-1</sup> (1Tg = 10<sup>12</sup> g) (with a 95% confident boundary of 1.8 ~ 3.4 Tg C a<sup>-1</sup>) in 1952 to the minimum annual CH<sub>4</sub> emission of 1.3 Tg C a<sup>-1</sup> (with a 95% confident boundary of 1.0 ~ 1.9 Tg C a<sup>-1</sup>) in 2003. The marshland loss made the largest contribution to the CH<sub>4</sub> emission reduction with an cumulative effect of 37.9 Tg C (with a 95% confident boundary of 28.0 ~ 54.1 Tg C) for the past 60 years. Ozone pollution reduced CH<sub>4</sub> emission, while elevated atmospheric CO<sub>2</sub>, nitrogen deposition, climate change, and multiple-factor interaction, cumulatively stimulated CH<sub>4</sub> emission. Climate variability predominately controlled the inter-annual variations in CH<sub>4</sub> emissions. A substantial spatial variation in CH<sub>4</sub> emission was observed across China's marshland. At regional scale, the Northeast, followed by Northwest and Southeast, made the greatest contribution, while North and Southwest made minor contributions to the national CH<sub>4</sub> emission. This study suggests that it is necessary to consider multiple global change factors when estimating regional CH<sub>4</sub> fluxes in natural wetlands.

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### 1. Introduction

[2] Methane (CH<sub>4</sub>), the second potent greenhouse gas, contributes approximately 15% to the increase in radiative forcing caused by the anthropogenic release of greenhouse gases to the atmosphere [Forster *et al.*, 2007; Rodhe, 1990; Shindell *et al.*, 2005]. The concentration of atmospheric CH<sub>4</sub> has increased from 0.7 ppm before the pre-Industrial Revolution to 1.8 ppm in recent years [Dlugokencky *et al.*, 2009; Forster *et al.*, 2007]. Quantifying the sources and sinks of atmospheric CH<sub>4</sub> is of utmost importance for further mitigation of atmospheric CH<sub>4</sub> increase and climate warming [Denman *et al.*, 2007; Forster *et al.*, 2007]. Given the relatively constant of sink strength of atmospheric CH<sub>4</sub> [Denman *et al.*, 2007; Dlugokencky *et al.*, 2009], estimating the source strength of atmospheric CH<sub>4</sub> becomes one of the major issues of global change research [Bergamaschi *et al.*, 2009; Dlugokencky *et al.*, 2009].

[3] Apart from the non-biogenic sources such as fossil fuel combustion, natural gas, petroleum and coal, the terrestrial biogenic source contributed more than 70% to the global CH<sub>4</sub> source in total [Denman *et al.*, 2007]. Of the biogenic source, natural wetlands are one of the major contributors [Denman *et al.*, 2007; Potter *et al.*, 2006; Whalen, 2005]. Thus, quantifying the source strength of atmospheric CH<sub>4</sub> from natural wetlands is critically important [Bousquet *et al.*, 2006; Denman *et al.*, 2007]; the regional estimation of CH<sub>4</sub> emission from wetlands, however, is still lacking [Song *et al.*, 2009; Wofsy and Harriss, 2002; Zhuang *et al.*, 2004].

[4] The net flux of CH<sub>4</sub> from wetland ecosystems at large scale is generally estimated by three approaches: (1) the direct extrapolation from site-level field measurements and observations [Ding *et al.*, 2004b, 2007], (2) process-based modeling approach (bottom-up approach) [Zhuang *et al.*, 2004; Tian *et al.*, 2010a; Xu *et al.*, 2010], and (3) the inverse modeling approach (top-down approach) [Kort *et al.*, 2008; Denman *et al.*, 2007; Liu, 1996]. The process-based modeling approach provides the source strength of regional terrestrial CH<sub>4</sub> flux over long time period with consideration of the spatial heterogeneity of ecosystem properties and climate conditions, and is gaining popularity in estimating regional CH<sub>4</sub> flux from natural wetlands [Liu, 1996; Potter *et al.*, 2006; Tian *et al.*, 2010a]. A number of studies have been conducted to estimate the regional CH<sub>4</sub> source strength of natural wetlands by using the process-based modeling

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**Table 1.** The Area of Marshland Over China (km<sup>2</sup>)

	Sanjiang Plain Marshland	Marshland Area Across China
1949	53,400 <sup>a,b</sup>	
1950		178,000 <sup>c</sup>
1983	18,357 <sup>a,b</sup>	
1994	10,406 <sup>a,b</sup>	
1996	9,466.2 <sup>b</sup>	
2000		94,000 <sup>c</sup>

<sup>a</sup>Liu and Ma [2000].<sup>b</sup>Wang [2002].<sup>c</sup>An et al. [2007].

approaches [Cao et al., 1998; Zhuang et al., 2004]; however, most of the previous studies did not simultaneously take into consideration of the effects of multiple global change factors, such as climate variability, ozone (O<sub>3</sub>) pollution, marshland loss, and rising atmospheric carbon dioxide (CO<sub>2</sub>) concentration; thus they could not be used for factorial attribution. For example, Zhuang et al.'s studies only considered the effects of climate variability, elevated atmospheric CO<sub>2</sub>, and land cover type [Zhuang et al., 2004, 2007]; several other studies even solely simulated the effects of climate variability [Cao et al., 1998; Potter, 1997; Walter et al., 2001].

[5] All environmental factors have been confirmed yielding substantial effects on CH<sub>4</sub> production and/or consumption, individually or in combination [Bloom et al., 2010; Conrad, 1996; Frolking and Crill, 1994; Hutchin et al., 1995; Pancotto et al., 2010]. For example, elevated atmospheric CO<sub>2</sub> may enhance CH<sub>4</sub> emission from wetlands [Hutchin et al., 1995]; O<sub>3</sub> pollution might suppress CH<sub>4</sub> emission [Mörsky et al., 2008]; climate change may increase or decrease CH<sub>4</sub> emission [Cao et al., 1998; Frolking and Crill, 1994]; nitrogen (N) input might increase or decrease CH<sub>4</sub> oxidation [Bodelier et al., 2000; Ding et al., 2004a; Liu and Greaver, 2009]; the interaction between/among multiple factors might also alter the regime of CH<sub>4</sub> flux [Pancotto et al., 2010]. Thus, it is essential to simultaneously incorporate multiple factors, rather than one or two factors, in regional estimation of CH<sub>4</sub> emission from wetlands.

[6] China, the country hosts a large area of natural wetlands, has experienced a severe decline of wetland area during the second half of the 20th century [An et al., 2007; Mitsch and Gosselink, 2007; Zhao, 1999]. Meanwhile, China has also experienced dramatic changes in environmental conditions including atmospheric pollution (e.g., nitrogen deposition, ozone pollution), climate change, etc [Chen et al., 2006; Lu and Tian, 2007]. Marshland, one of the major wetland types in China [Zhao, 1999], is still lack of intensive study of CH<sub>4</sub> flux, especially at regional scale [Ding and Cai, 2007; Song et al., 2009]. How these environmental changes altered the source strength of CH<sub>4</sub> from marshland is really critical for regional study of terrestrial ecosystem feedback to climate system and for policy making targeting climate mitigation over China.

[7] The objectives of this study were: 1) to estimate the magnitude of CH<sub>4</sub> flux from marshland over China; 2) to examine the spatiotemporal patterns of CH<sub>4</sub> flux in China's marshland from 1949 through 2008; and 3) to further attribute the spatiotemporal variations in CH<sub>4</sub> flux to multiple global change factors including marshland loss, elevated

atmospheric CO<sub>2</sub>, ozone pollution, climate change, and increased nitrogen input through atmospheric deposition.

## 2. Data and Model

### 2.1. Data Development

[8] A series of spatially and temporally explicit data sets have been developed to characterize the driving forces for the ecosystem model; they are changes in climate (daily average, maximum, and minimum temperature, precipitation, shortwave radiation and humidity) [Chen et al., 2006], and atmospheric composition (O<sub>3</sub> and N deposition) [Lu and Tian, 2007; Lu et al., 2011; Ren et al., 2007] with a time step ranging from daily to yearly. The atmospheric CO<sub>2</sub> concentration data from 1900 to 2008 was obtained from the Carbon Dioxide Information Analysis Center (CDIAC, <http://cdiac.ornl.gov/>). The soil depth, pH, and texture maps were generated from the 1:1 million soil maps (Institute of Soil Science, Chinese Academy of Sciences) and the second national soil survey of China (National Soil Survey Office in China).

[9] The gridded time series climate data sets were developed by using the extrapolation algorithm of the mountain microclimate simulator developed by Thornton et al. [1997]. The daily climate data from 740 weather stations in mainland China, 6 stations in Taiwan, and 29 stations in neighbor countries were used for extrapolation. Based on the climate data from 1961 to 1990, we generated de-trended climate data for model spin-up and to represent climate conditions from 1901 to 1960. The ozone data set was derived from the global historical ozone data sets constructed by Felzer et al. [2005], as described by Ren et al. [2007]. The atmospheric nitrogen deposition data for China were developed on the basis of monitoring data and the results of a three-dimensional atmospheric chemical transport model [Dentener, 2006; Lu and Tian, 2007].

[10] To develop the gridded time series data set of historical marshland area for our study, we first collected the census data of historical wetland area in China since the 1940s [An et al., 2007; Liu and Ma, 2000; Wang, 2002]. Although the area of the Sanjiang Plain is approximately 1% of national land area, its highly dense distribution of wetland [Zhao, 1999] and dramatic decline of wetland area since the foundation of the Chinese government suggest its importance in examining the historical CH<sub>4</sub> emission [An et al., 2007; Huang et al., 2010; Song et al., 2009]. Thus, we separately processed the Sanjiang Plain from other area in China. Based on the summarized area for the Sanjiang Plain by other scientists (Table 1), we linearly interpolated wetland area over the study period on the basis of the known wetland area. For the wetland area in other areas over China, we extrapolated the area of several data points by using a negative correlation between wetland areas and national population. Given that a number of bills were issued for wetland protection and restoration, we assumed that marshland area was unchanged for the time period of 2000–2008. Finally, the time series of wetland area for the Sanjiang Plain and entire China during 1949–2008 were generated. Then we combined the time series of wetland area and national percentage distribution of wetland map in the year of 2000 [Liu et al., 2005; Liu and Tian, 2010], further generated a time series spatial data set of marshland distribution based

on the rule that less dense marshland is easier to be converted to other landscapes. For instance, our census data showed that the marshland area is 9466 km<sup>2</sup> in 1996, equivalent to 2366 grids at a spatial resolution of 2 km × 2 km; while the grids with ≥33.15% wetlands in the wetland fractional map sums up to 2350 grid, closest to census area. Thus we assumed that the grids with ≥33.15% wetlands are marshland, while the other grids are other biomes. From 1949 to 2008, the loss of marshland was converted to cropland. All the regional data were re-projected and re-sampled to be consistent at a spatial resolution of 2 km × 2 km. The marshland in this study includes almost all natural wetlands except river wetlands, lake and coastal wetlands [An *et al.*, 2007].

## 2.2. Model Description

[11] The dynamic land ecosystem model (DLEM) is applied in this study; it is a highly integrated process-based ecosystem model that aims at simulating the fluxes and storages of carbon, water and nitrogen among/within terrestrial ecosystem components while taking into consideration multiple natural and anthropogenic perturbations [Tian *et al.*, 2010b]. The DLEM is composed of five major sub-modules focusing on biophysics, plant physiology, soil biogeochemistry, vegetation dynamics, land use and management as well as disturbances such as hurricane, fire, insect etc. DLEM has been widely applied to a variety of terrestrial ecosystem types across the globe and the detailed assumptions and processes are well documented in our previous publications [Tian *et al.*, 2008, 2010a, 2010b, 2011a, 2011b; Chen *et al.*, 2006; Liu *et al.*, 2008; Lu *et al.*, 2011; Ren *et al.*, 2007, 2011; Xu *et al.*, 2010, 2011; Xu, 2010; C. Zhang *et al.*, 2007, 2010, 2012].

[12] The methane module in the DLEM model mainly simulates the production, consumption, and transport of CH<sub>4</sub> [Tian *et al.*, 2010a; Xu *et al.*, 2010]. Due to relatively small contribution from other substrates [Conrad, 1996; Le Mer and Roger, 2001], DLEM only considers the CH<sub>4</sub> production from dissolved organic carbon (DOC), which is indirectly controlled by environmental factors including soil pH, temperature and soil moisture content. The DOC was produced through three pathways, gross primary production allocation, and decomposition byproducts from soil organic matter and litter-fall. CH<sub>4</sub> oxidation, including the oxidation during CH<sub>4</sub> transport to the atmosphere, CH<sub>4</sub> oxidation in the soil water, and atmospheric CH<sub>4</sub> oxidation on the soil surface, is determined by CH<sub>4</sub> concentrations in the air or soil water, as well as soil moisture, pH, and temperature. Most CH<sub>4</sub>-related biogeochemical reactions in the DLEM are described as the Michaelis-Menten equation with two coefficients: maximum reaction rate and half-saturated coefficient. The CH<sub>4</sub> transport from soil water to the atmosphere includes three pathways: diffusion, ebullition, and plant-mediated transport through aerenchyma system [Tian *et al.*, 2010a; Xu *et al.*, 2010].

[13] Multiple global change factors yield direct and/or indirect impacts on CH<sub>4</sub> processes as simulated in the DLEM [Xu *et al.*, 2010]. The detailed information and equation could be referred to Xu *et al.* [2010]. The atmospheric CO<sub>2</sub> concentration, O<sub>3</sub> pollution, soil moisture, air temperature, and absorbed photosynthetically active radiation yield indirect effects on CH<sub>4</sub> production through their effects on

photosynthesis; the N input has indirect effects on CH<sub>4</sub> production through its impacts on photosynthesis and ecosystem respiration; the soil temperature and moisture exert direct effects on CH<sub>4</sub> oxidation through their direct control on CH<sub>4</sub> production and oxidation. Meanwhile, soil temperature, pH and moisture directly influence CH<sub>4</sub> production, while O<sub>3</sub> pollution and N input indirectly influence CH<sub>4</sub> oxidation through their impacts on physical processes in the ecosystem. The impacts of land conversion on CH<sub>4</sub> flux could be caused by land-conversion-induced alterations in either biological substrate or physiochemical processes. It should be noted that some other environmental factors which might influence CH<sub>4</sub> processes were not explicitly described here, for example, soil pH, soil texture, etc. [Xu *et al.*, 2010].

## 2.3. Model Parameterization

[14] A number of parameters directly control CH<sub>4</sub> production and consumption, and need to be determined before model simulations. Based on the parameters for our previous simulation for North America [Tian *et al.*, 2010a], we first fine-tuned the sensitive parameters to fit the simulated CH<sub>4</sub> flux to measured CH<sub>4</sub> flux from marshland in China. We try to keep most parameters unchanged based on the simulations for the North America [Tian *et al.*, 2010a]. Finally, one parameter, maximum rate of CH<sub>4</sub> production, was tuned to 0.675 g C m<sup>-3</sup> day<sup>-1</sup>, rather than 1.45 g C m<sup>-3</sup> day<sup>-1</sup> for herbaceous wetlands and 0.55 g C m<sup>-3</sup> day<sup>-1</sup> for woody wetlands in our North America studies [Tian *et al.*, 2010a; Xu *et al.*, 2010]. The calibrated parameters were used for regional simulations.

## 2.4. Model Simulation

[15] The model simulations were consisted of two stages: equilibrium run and transient run. First, the equilibrium run was set up to determine the initial conditions of January 1, 1901; the DLEM was fed with de-trended climate data during 1901–1930, the actual values of atmospheric CO<sub>2</sub> concentration, O<sub>3</sub> AOT40 index (accumulated ozone exposure over a threshold of 40 parts per billion), N deposition, and land use pattern in the year of 1900. The equilibrium state is defined as the absolute value of annual carbon storage change is less than 0.1 g C m<sup>-2</sup>, the change in soil water pool is less than 0.1 mm, and the difference in soil mineral nitrogen content and N uptake is less than 0.1 g N m<sup>-2</sup> among consecutive years. Then the model was run in transient mode, where the simulated CH<sub>4</sub> flux was determined by the time series of multiple environmental changes from 1901 to 2008. We started the simulation in the year 1901 in order to capture the legacy effects of changes in land conversion, climate variability, nitrogen deposition, O<sub>3</sub> pollution, and elevated atmospheric CO<sub>2</sub>, on CH<sub>4</sub> fluxes. This study focused on the simulation results from 1949, the foundation of the People's Republic of China. This period was selected because the great changes in both natural and anthropogenic perspectives were taking place across China.

[16] Totally, nine simulations were set up for this study (Table 2). One overall simulation (S6) considers all driving forces, and five more simulations were set up to simulate the effects of each global change factor: climate variability (temperature, precipitation, relative humidity, and solar radiation), elevated atmospheric CO<sub>2</sub>, O<sub>3</sub> pollution, marshland loss,

**Table 2.** Simulation Design<sup>a</sup>

Simulations	Elevated Atmospheric CO <sub>2</sub>	Nitrogen Deposition	O <sub>3</sub> Pollution	Climate Change	Land Conversion
S1	1901–2008	1900	1900	1900	1900
S2	1900	1901–2008	1900	1900	1900
S3	1900	1900	1901–2008	1900	1900
S4	1900	1900	1900	1901–2008	1900
S5	1900	1900	1900	1900	1901–2008
S6	1901–2008	1901–2008	1901–2008	1901–2008	1901–2008
S7	1901–2008	1901–2008	1900	1900	1900

Simulations	Elevated Atmospheric CO <sub>2</sub>	Nitrogen Deposition	O <sub>3</sub> Pollution	Climate Change		Land Conversion
				Temperature	Precipitation	
S8	1900	1900	1900	1901–2008	1900	1900
S9	1900	1900	1900	1900	1901–2008	1900

<sup>a</sup>1900 year climate data represents the 30-year (1961–1990) average.

and nitrogen deposition, on CH<sub>4</sub> emission (S1–S5). One more simulation was set up to evaluate the interactive effects of N deposition and elevated atmospheric CO<sub>2</sub> on CH<sub>4</sub> emission (S7). Two more simulations were set up to separate the contributions from temperature and precipitation in climate impacts (S8 and S9). The results for the time period of 1949–2008 were analyzed. Baseline flux was defined as the CH<sub>4</sub> flux in 1949, the changes thereafter comparing to baseline flux was assumed solely caused by global change factors, individually or in combinations.

## 2.5. Statistical Analysis and Uncertainty Analysis

[17] The statistic analyses were conducted by using R-program for Windows XP ([www.r-project.org](http://www.r-project.org)). The simple linear regressions were used to obtain long-term changing rates of driving forces and CH<sub>4</sub> flux caused by global change factors, individually or in combination. A multiple linear regression was used to evaluate the contribution of multiple factors on the inter-annual variations in CH<sub>4</sub> emission.

[18] In our previous studies, we have estimated the uncertainties of CH<sub>4</sub> fluxes over North America [Xu, 2010] and entire China [Tian *et al.*, 2011a] by using a Monte Carlo method. Same method was used in this study to estimate the uncertainties of CH<sub>4</sub> emission from marshland across China. Totally ten parameters (Table S1 in the auxiliary material) were chosen and 300 simulations were carried out.<sup>1</sup>

## 2.6. Model Performance Testing

[19] Model performance testing is an important step before model application; it is used to show the model's ability in simulating variables of interest [Haefner, 2005]. In this study, we conducted the model performance testing from four aspects: 1) site-level validation of CH<sub>4</sub> flux at a daily scale (Figure 1 and Table 3); 2) regional validation of site-level CH<sub>4</sub> flux across China at a monthly scale; 3) national or regional comparison of simulated CH<sub>4</sub> flux with previous estimates by extrapolation approach at an annual scale; and 4) validation of factorial contribution to CH<sub>4</sub> flux from China's marshlands (Tables 4 and 5).

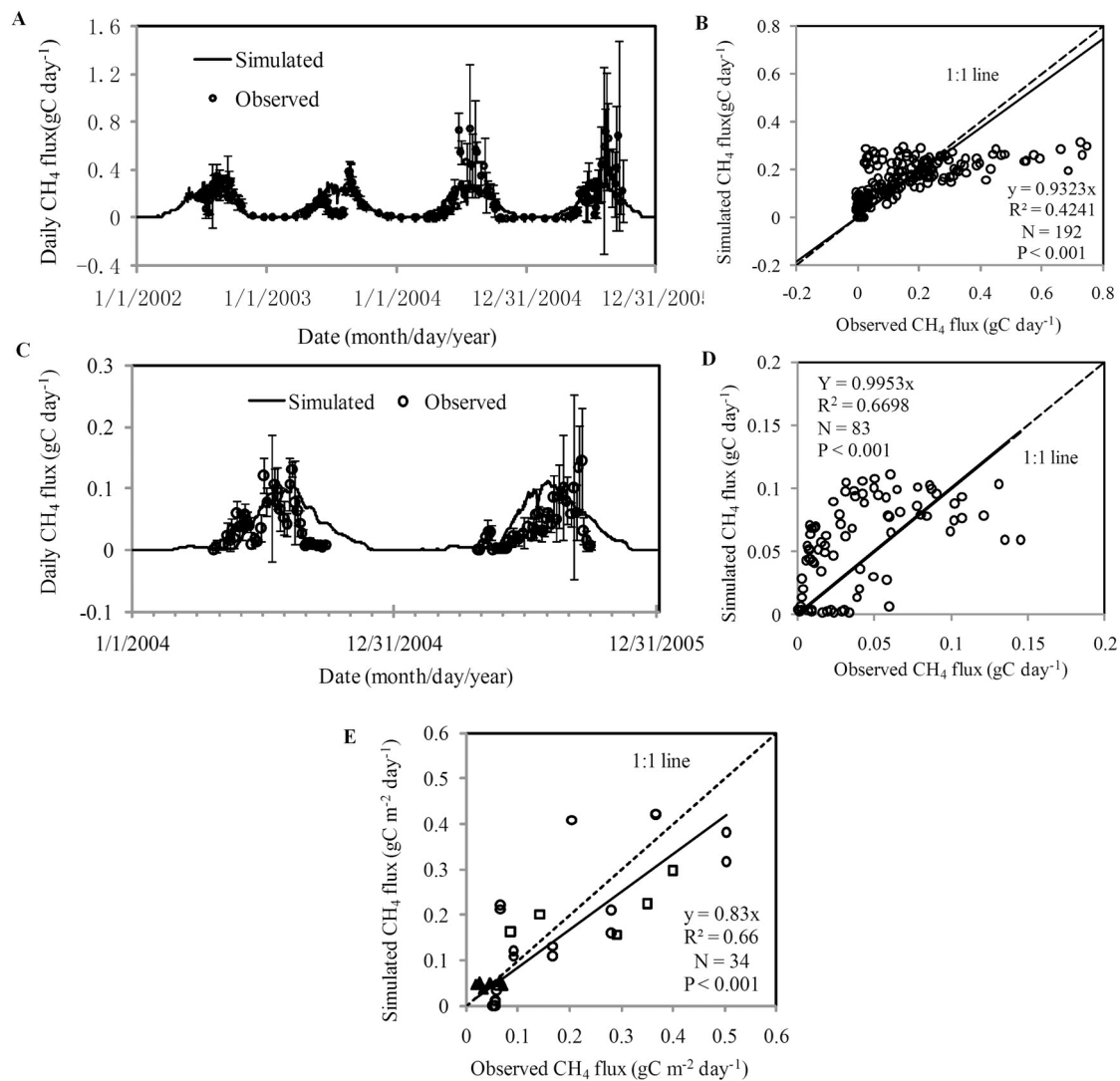
[20] For the site-level validation, we compared our simulated CH<sub>4</sub> fluxes with observations at marshlands in Sanjiang

and Ruergai stations (Figures 1a–1d). The model driving forces were listed in the Table 3. The daily climate data were provided by the China Meteorological Administration (CMA) ([www.cma.org.cn](http://www.cma.org.cn)); we used the climate data from the CMA site that is close to the validation site. The atmospheric CO<sub>2</sub> concentration is from National Oceanic and Atmospheric Administration (NOAA). Nitrogen deposition and O<sub>3</sub> pollution were retrieved from our regional data set, while other soil and geological properties were from Chinese Ecological Research Network (CERN) or published literatures. The validations for two sites were shown in Figure 1. Figures 1a and 1b showed that the simulated and observed CH<sub>4</sub> fluxes are tightly consistent for first two years in the Sanjiang stations, while several observed “hot moments” in CH<sub>4</sub> emission were not captured by the DLEM. The overall comparison showed the good agreement between observation and simulations (slope = 0.9323; P < 0.001) (Figure 1b). Meanwhile, the simulated daily CH<sub>4</sub> fluxes are quite consistent with the observed CH<sub>4</sub> fluxes in the Ruergai marshland (Figures 1c and 1d). The overall comparison showed the consistency between simulations and observations (slope = 0.9953, P < 0.001), while the discrepancies were showed between simulated and observed CH<sub>4</sub> fluxes during growing season (Figure 1c).

[21] We further collected more site-level CH<sub>4</sub> fluxes across China to verify the simulated CH<sub>4</sub> flux at regional scale. Due to the scarcity of observations of CH<sub>4</sub> flux across China, we only got monthly CH<sub>4</sub> fluxes for three more sites [Cui *et al.*, 1998; Hirota *et al.*, 2004; Wang *et al.*, 2009] (Figure 1e). These three sites are representative of the three major areas with dense marshland in China: North China, Northeast China, and Northwest China. The total site-month is 34. The comparison shows the consistencies between observed CH<sub>4</sub> flux and simulated CH<sub>4</sub> flux across China (slope = 0.83; P < 0.001). This consistency indicated that the simulated CH<sub>4</sub> fluxes are reliable at regional scale.

[22] We also conducted a regional or national comparison of our simulated CH<sub>4</sub> flux with estimates from previous studies [Ding *et al.*, 2004b; Ding and Cai, 2007; Jin *et al.*, 1999; Khalil *et al.*, 1993]. The results showed that our simulated regional or national estimates are consistent with previous studies (Table 4). For example, we estimated that the national CH<sub>4</sub> emission from China's marshland was 1.37 Tg C a<sup>-1</sup> (1 Tg = 10<sup>12</sup> g) for 2001–2002, which is consistent with 1.32 (1.28–1.37) Tg C a<sup>-1</sup> estimated by

<sup>1</sup>Auxiliary materials are available in the HTML. doi:10.1029/2010GB003946.



**Figure 1.** Verification of model performance on simulating CH<sub>4</sub> flux over marshland over China. (a) Time series comparison of observed and simulated CH<sub>4</sub> flux in the Sanjiang marshland, (b) scatterplot of simulated and observed CH<sub>4</sub> flux for the Sanjiang marshland, (c) time series comparison of observed and simulated CH<sub>4</sub> flux in the Ruoergai marshland, (d) scatterplot of simulated and observed CH<sub>4</sub> flux for the Ruoergai marshland, and (e) scatterplot showing comparison between simulated and observed CH<sub>4</sub> flux over China (circles represent observations from Wang *et al.* [2009], squares represent observations from Cui *et al.* [1998], and triangles represent observations from Hirota *et al.* [2004]).

Ding *et al.* [2004b]. For the time period of 1995–2004, our estimate ( $1.38 \pm 0.03 \text{ Tg C a}^{-1}$ ) is consistent with that from Ding and Cai [2007] ( $1.32 \text{ Tg C a}^{-1}$ ). And our estimated national CH<sub>4</sub> emission for 1996 was  $1.37 \text{ Tg C a}^{-1}$ , slightly lower than  $1.5 \text{ Tg C a}^{-1}$  estimated by Jin *et al.* [1999], while our estimated CH<sub>4</sub> emission was  $1.52 \text{ Tg C a}^{-1}$  for 1990, which is slightly higher than  $1.28 \text{ Tg C a}^{-1}$  estimated by Khalil *et al.* [1993]. For the regional comparison for the Northeast China, our result was  $0.88 \text{ Tg C a}^{-1}$  in 2001–2002 which is highly consistent with  $0.89 \text{ Tg C a}^{-1}$  in Ding *et al.*'s [2004b] study.

[23] Because our estimations are based on multiple driving forces, we further verified the factorial contribution to CH<sub>4</sub> flux (Table 5). Currently, there was only one field study conducted on nitrogen input impacts on CH<sub>4</sub> emission from

marshland in China [L. H. Zhang *et al.*, 2007]; there was no study reported for the effects of elevated atmospheric CO<sub>2</sub> and O<sub>3</sub> pollution on CH<sub>4</sub> flux from marshland in China. We collected the observed CH<sub>4</sub> flux in response to elevated atmospheric CO<sub>2</sub>, O<sub>3</sub> pollution, and nitrogen input conducted in other countries for the factorial validation in this study. The results were shown in Table 5. Our results showed that the doubled atmospheric CO<sub>2</sub> lead to a 4.15% increase in CH<sub>4</sub> emission from marshland over China, which falls in the low end of previous studies [Cheng *et al.*, 2006; Dacey *et al.*, 1994; Hutchin *et al.*, 1995; Megonigal and Schlesinger, 1997; Saarnio *et al.*, 1998; Saarnio and Silvola, 1999; Saarnio *et al.*, 2003; Vann and Megonigal, 2003]; this is because that the field studies are plot-specific, while our estimate are based on the simulation result over the

**Table 3.** Site-Level Driving Force for Simulation at the Sanjiang and Ruoergai Marshland<sup>a</sup>

Sites	Variables (Unit)	Time Period or Value	Sources	
Sanjiang	Climate data	1980–2008	CMA	
	CO <sub>2</sub> (ppm)	1900–2008	NOAA	
	Nitrogen deposition (gN/m <sup>2</sup> /year)	1900–2005	<i>Lu and Tian</i> [2007]	
	O <sub>3</sub> concentration (AOT40: ppb hr/day)	1900–2005	<i>Ren et al.</i> [2007]	
	Latitude	47.58	CERN	
	Longitude	133.52	CERN	
	Multiple-year mean precipitation (mm)	600	CERN	
	Multiple-year ET (mm)	511	CERN	
	Altitude (m)	56	CERN	
	Slope (°)	0	Spatial data set	
	Aspect (°)	168	Spatial data set	
	Soil pH	6	CERN	
	Clay (%)	31.4	CERN	
	Sand (%)	29.1	CERN	
	Soil organic matter (%)	30	<i>Song et al.</i> , 2009	
	Ruoergai	Climate data	1957–2008	CMA
		CO <sub>2</sub> (ppm)	1900–2008	NOAA
Nitrogen deposition (gN/m <sup>2</sup> /year)		1900–2005	<i>Lu and Tian</i> [2007]	
O <sub>3</sub> concentration (AOT40: ppb hr/day)		1900–2005	<i>Ren et al.</i> [2007]	
Latitude		32.79	CERN	
Longitude		102.53	CERN	
Multiple-year mean precipitation (mm)		732	CERN	
Multiple-year ET (mm)		713	CERN	
Altitude (m)		3470	CERN	
Slope (°)		6	Spatial data set	
Aspect (°)		1	Spatial data set	
Soil pH		6	CERN	
Clay (%)		20	CERN	
Sand (%)		46.9	CERN	
Soil organic matter (%)		40	<i>Wang</i> [2001]	

<sup>a</sup>CMA: China Meteorology Administration; NOAA: National Oceanic and Atmospheric Administration; CERN: Chinese Ecosystem Research Network; Spatial data set represent the regional data set for model simulation.

entire China; the upscaling process might cause large discrepancies. Our estimated nitrogen-induced CH<sub>4</sub> emission was 63.3 mg C g N<sup>-1</sup> m<sup>-2</sup> a<sup>-1</sup>, much higher than 8 ± 4 mg C g N<sup>-1</sup> m<sup>-2</sup> a<sup>-1</sup> summarized on few observations [*Liu and Greaver*, 2009], while is much lower than a field experiment in China which estimates the nitrogen-induced CH<sub>4</sub> emission was 135–676 mg C g N<sup>-1</sup> m<sup>-2</sup> a<sup>-1</sup> [*L. H. Zhang et al.*, 2007]. Zhang et al.'s field experiment was conducted on one species, while our model study is for

entire marshland ecosystem which is usually consisted of many species; the ecosystem adaptation to N input [*Langley and Megonigal*, 2010] and differential nitrogen responses of different species [*Nykänen et al.*, 2002] might explain this difference. The simulated O<sub>3</sub> pollution effect is very small, which is consistent with field experiments [*Mörsky et al.*, 2008; *Niemi et al.*, 2002].

### 3. Results

#### 3.1. Environmental Changes Across Marshland in China

[24] A great amount of marshland has been converted to cropland in China over past half century [*An et al.*, 2007]. The total area of marshland over China has decreased from 179.32 × 10<sup>3</sup> km<sup>2</sup> in 1949 to 94.10 × 10<sup>3</sup> km<sup>2</sup> in 2008, with a significant decreasing rate of 1.65 ± 0.02 × 10<sup>3</sup> km<sup>2</sup> a<sup>-1</sup>. Meanwhile, China's marshlands have experienced substantial changes of environmental conditions [*Fang and Kiang*, 2006; *Fu*, 2008]; for instance, over the study period the atmospheric CO<sub>2</sub> concentration has increased from 310.75 ppm in 1949 to 385.57 ppm in 2008, with a significant increasing rate of 1.29 ± 0.03 ppm a<sup>-1</sup>; nitrogen deposition has increased from 0.64 g N m<sup>-2</sup> a<sup>-1</sup> in 1949 to 1.46 g N m<sup>-2</sup> a<sup>-1</sup> in 2008, with a significant increasing rate of 13.54 ± 0.44 mg N m<sup>-2</sup> a<sup>-2</sup>, and O<sub>3</sub> pollution has showed a significant increasing trend with changing rate of 1.87 ± 0.06 ppb hr d<sup>-1</sup> a<sup>-1</sup> (Table S2). Furthermore, China's marshlands also experienced dramatic changes of climate; for example, the maximum, minimum, and average temperatures shown significant increasing trends at rates of 0.19 ± 0.03°C decade<sup>-1</sup>, 0.34 ± 0.04°C decade<sup>-1</sup>, 0.26 ± 0.03°C decade<sup>-1</sup>, respectively; while the relative humidity and precipitation changed, yet not significant, at rates of -0.07 ± 0.07% decade<sup>-1</sup>, 2.14 ± 3.99 mm decade<sup>-1</sup>, respectively (Table S2).

#### 3.2. Temporal Variations of CH<sub>4</sub> Emission From China's Marshland

[25] A significant decreasing trend of CH<sub>4</sub> flux from marshland from 1949 to 2008 with a strong inter-annual variation was observed (Figure 2). The shaded area in Figure 2 represents the simulated CH<sub>4</sub> flux at 95% confidence level derived from the 300 simulations. The maximum annual CH<sub>4</sub> emission was 2.4 Tg C a<sup>-1</sup>, with 95% confident boundaries of 1.8 ~ 3.4 TgC a<sup>-1</sup> in 1952, while the minimum was 1.3 Tg C a<sup>-1</sup>, with 95% confident boundaries of 1.0 ~ 1.9 TgC a<sup>-1</sup> in 2003 (Figure 2). Over the past 60 years, the significant decreasing rate of CH<sub>4</sub> emission was 18.7 Gg C a<sup>-1</sup> (P < 0.001), with 95% confidence boundaries of 17.6 ~ 19.8 Gg C a<sup>-1</sup>.

**Table 4.** Regional Validation of CH<sub>4</sub> Emission From Marshland Across China

CH <sub>4</sub> Emission	Time Period	This Study (Tg C a <sup>-1</sup> )	Other Studies (Tg C a <sup>-1</sup> )	References
China	2001–2002	1.37	1.32 (1.28~1.37)	<i>Ding et al.</i> [2004b]
China	1995–2004	1.38 ± 0.03	1.32	<i>Ding and Cai</i> [2007]
China	1996	1.37	1.50	<i>Jin et al.</i> [1999]
China	1990	1.52	1.28	<i>Khalil et al.</i> [1993]
Northeast China (Heilongjiang, Jilin, Liaoning, portions of Neimenggu Provinces)	2001–2002	0.88	0.89	<i>Ding et al.</i> [2004b]

**Table 5.** Factorial Validation of CH<sub>4</sub> Emission From Marshland in China

Environmental Factors	This Study	Other Reports	Experiments	References
Elevated CO <sub>2</sub>	4.15% <sup>a</sup>	0–146%	Free Air CO <sub>2</sub> Enrichment	<i>Cheng et al.</i> [2006], <i>Dacey et al.</i> [1994], <i>Hutchin et al.</i> [1995], <i>Megonigal and Schlesinger</i> [1997], <i>Saarnio et al.</i> [1998], <i>Saarnio and Silvola</i> [1999], <i>Silvola et al.</i> [2003], and <i>Vann and Megonigal</i> [2003]
Nitrogen input	63.3 <sup>a</sup> (mg C gN m <sup>-2</sup> a <sup>-1</sup> )	8 ± 4 (mg C gN m <sup>-2</sup> a <sup>-1</sup> ) 135–676 (mg C gN m <sup>-2</sup> a <sup>-1</sup> )	Meta-analysis Gradient field experiment	<i>Liu and Greaver</i> [2009] <i>L. H. Zhang et al.</i> [2007]
O <sub>3</sub> pollution	–1.24% <sup>a</sup>	Negative yet insignificant	Double O <sub>3</sub>	<i>Mörsky et al.</i> [2008] and <i>Niemi et al.</i> [2002]

<sup>a</sup>Calculated based on the time series of simulated CH<sub>4</sub> flux and nitrogen input.

[26] To further show the decadal variation of national CH<sub>4</sub> emission, we summarized the decadal fluxes of CH<sub>4</sub> over the past 60 years (Table 6). The decadal mean of CH<sub>4</sub> flux also showed a decreasing trend, from 2.25 ± 0.08 Tg C a<sup>-1</sup> for the 1950s, to 2.06 ± 0.11 Tg C a<sup>-1</sup> for the 1960s, 1.79 ± 0.09 Tg C a<sup>-1</sup> for the 1970s, 1.59 ± 0.08 Tg C a<sup>-1</sup> for the 1980s, 1.42 ± 0.06 Tg C a<sup>-1</sup> for the 1990s, and 1.38 ± 0.02 Tg C a<sup>-1</sup> for the time period of 2000–2008. Overall, the coefficient variance of the decadal CH<sub>4</sub> flux increased from the 1950s to the 1960s, and then decreased to 2000–2008 (Table 6).

### 3.3. Spatial Variations of CH<sub>4</sub> Emission From China's Marshland

[27] There was a substantial spatial variation of CH<sub>4</sub> emission from China's marshland, with relatively high emission rate in the eastern China while relatively low emission rate in the western China. The CH<sub>4</sub> emission from marshland in the southeastern China was as high as 30 g C m<sup>-2</sup> a<sup>-1</sup>, and the CH<sub>4</sub> emission rate ranged from 10 g C m<sup>-2</sup> a<sup>-1</sup> to 30 g C m<sup>-2</sup> a<sup>-1</sup> for the northeast marshland. The Western China had low CH<sub>4</sub> emission rate of <10 g C m<sup>-2</sup> a<sup>-1</sup>, or even < 2 g C m<sup>-2</sup> a<sup>-1</sup> (Figure 3).

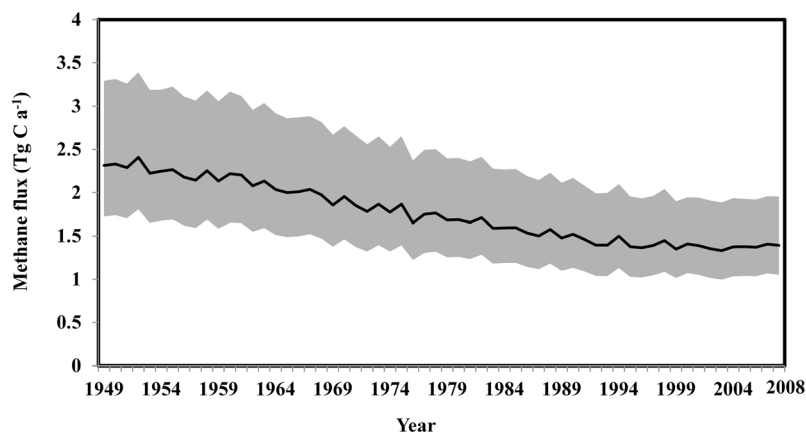
[28] We further summarized the total CH<sub>4</sub> emission along the latitudinal and longitudinal directions (Figure 4). Along the latitudinal direction, there was a peak of CH<sub>4</sub> emission around 29°N, and a high CH<sub>4</sub> emission from 45° N to 50° N. Overall, northern and southern China have high CH<sub>4</sub> emission,

while the central has lower CH<sub>4</sub> release. Along the longitudinal direction, an increasing trend was observed from the West to the East. Between 80°W and 110°W, the CH<sub>4</sub> emission was quite low; however, it was much higher for the regions between 110°W and 135°W.

### 3.4. Factorial Contributions to the CH<sub>4</sub> Emission From China's Marshland During 1949–2008

[29] Figure 5 shows the accumulated CH<sub>4</sub> emissions from China's marshland from 1949 through 2008. Over the study period, all global change factors together cumulatively reduced CH<sub>4</sub> emission by 33.08 Tg C (95% confidence range: 24.5 ~ 47.2 Tg C); of which, marshland loss contributed 37.9 Tg C (95% confidence range: –28.0 ~ 54.1 Tg C); in addition, O<sub>3</sub> pollution cumulatively reduced CH<sub>4</sub> emission by 0.08 Tg C (95% confidence range: 0.06 ~ 1.2 Tg C); while elevated atmospheric CO<sub>2</sub>, nitrogen deposition, climate change, and multiple-factor interaction cumulatively increased CH<sub>4</sub> emission by 0.23 Tg C (95% confidence range: 0.17 ~ 0.33 Tg C), 0.15 Tg C (95% confidence range: 0.11 ~ 0.22 Tg C), 4.4 Tg C (95% confidence range: 3.3 ~ 6.3 Tg C), and 0.09 Tg C (95% confidence range: 0.6 ~ 1.2 Tg C), respectively (Table 7). Of the climate impacts on CH<sub>4</sub> emission from China's marshland ecosystems during 1949–2008, the temperature, precipitation and their interactions contributed 61.2%, 51.6% and –12.8%, respectively.

[30] Over the study period, marshland loss and O<sub>3</sub> pollution continuously decreased CH<sub>4</sub> emission, while elevated



**Figure 2.** Temporal variation of CH<sub>4</sub> emission from China's marshland during 1948–2008 (shaded area shows the upper and lower boundaries of simulated CH<sub>4</sub> flux at 95% significance level).

**Table 6.** Decadal Fluxes of CH<sub>4</sub> From China's Marshland<sup>a</sup>

Time Period	CH <sub>4</sub> Emission (Tg C a <sup>-1</sup> )		
	Mean	SD	CV (%)
1950–1959	2.25	0.08	3.75
1960–1969	2.06	0.11	5.32
1970–1979	1.79	0.09	5.13
1980–1989	1.59	0.08	4.79
1990–1999	1.42	0.06	4.12
2000–2008	1.38	0.02	1.79

<sup>a</sup>SD: standard deviation; CV: coefficient variance.

atmospheric CO<sub>2</sub>, nitrogen deposition, climate change continuously enhanced CH<sub>4</sub> emission, and multiple-factor interaction played a complicated role in determining CH<sub>4</sub> emission; it decreased CH<sub>4</sub> emission during 1949–1958, while enhanced CH<sub>4</sub> emission since 1959 onward (Figure 5). We also analyzed the factorial contribution to the inter-annual variability of CH<sub>4</sub> emission from China's marshland by using a multiple linear regression. The results showed that the climate variation played a predominate role in controlling inter-annual variations in CH<sub>4</sub> emission ( $t = 412.06$ ;  $P < 0.001$ ); meanwhile, marshland loss ( $t = -3.73$ ;  $P < 0.001$ ), elevated atmospheric CO<sub>2</sub> ( $t = 3.60$ ;  $P = 0.001$ ), and nitrogen deposition ( $t = 2.24$ ;  $P = 0.029$ ) also made contributions.

### 3.5. Regional Contributions to CH<sub>4</sub> Emission Induced by Global Change Factors

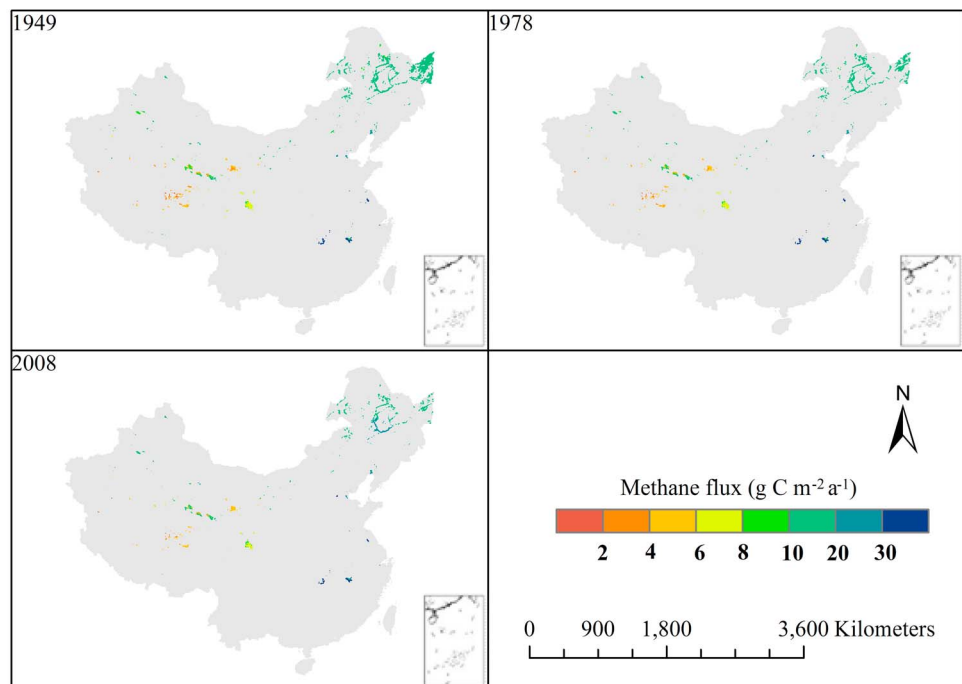
[31] China features a variety of landscapes, soil properties and climate condition, which might contribute to the spatial variations of CH<sub>4</sub> emission from marshland across China. We analyzed the regional contribution to CH<sub>4</sub> emission from China's marshland induced by global change factors, individually or in combination (Figure 6). China's marshland

was divided into five regions including the Northwest, North, Northeast, Southeast, and Southwest (Figure 6). Over the past 60 years, the Northeast China contributed 62% to the national CH<sub>4</sub> emission; the Northwest, Southeast, North and Southwest China contributed 15%, 13%, 7%, and 3%, respectively, to the national CH<sub>4</sub> emission from marshland (Figure 6).

[32] Of the baseline CH<sub>4</sub> emission, the Northeast China contributed 68%, followed by 13% from the Northwest, 11% from Southeast, 6% from North, and 2% from Southwest China (Figure 6). For the interactive effects of multiple global change factors, the Southeast China made the greatest contribution at 72%. The Northeast China also made greatest contribution to land conversion-induced, climate change-induced, and elevated atmospheric CO<sub>2</sub> induced CH<sub>4</sub> emissions. The Southeast China made the biggest contribution to nitrogen deposition-induced, and O<sub>3</sub> pollution-induced CH<sub>4</sub> emission; the North China made minor contribution to the multiple-factor interactive effects on CH<sub>4</sub> emission (Figure 6).

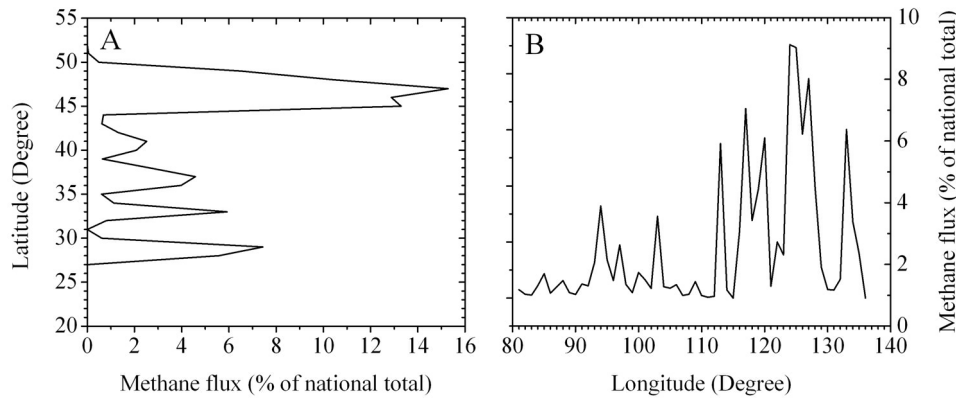
### 3.6. Factorial Contribution to CH<sub>4</sub> Emissions From China's Marshland During 1949–2008 at Regional Scale

[33] Using a set of model simulations (Table 2), we attributed the variations in temporal CH<sub>4</sub> emission to various global change factors. The results showed that factorial contributions to the CH<sub>4</sub> emission from marshland varied among regions. The common phenomena for all five regions was the baseline CH<sub>4</sub> emission made the biggest contribution, while all other factors together reduced CH<sub>4</sub> emission from marshland (Table 8). For the Northeast, marshland loss reduced more than one third of the baseline emission with another 0.03% from O<sub>3</sub> pollution; while climate change, followed by elevated atmospheric CO<sub>2</sub>, multiple-factor interaction, and nitrogen deposition, enhanced



**Figure 3.** Spatial distribution of CH<sub>4</sub> emission from China's marshland.





**Figure 4.** Latitudinal and longitudinal variation of CH<sub>4</sub> emission from marshland over China in the year 2000.

CH<sub>4</sub> emission. For the Southeast, except the negative effects from marshland loss, O<sub>3</sub> pollution contributed  $-0.38\%$  to the CH<sub>4</sub> emission; while elevated atmospheric CO<sub>2</sub> and nitrogen deposition, and climate change enhanced by 0.62%, 0.95%, and 0.83%, respectively, of the CH<sub>4</sub> emission. For the other three regions, marshland loss, elevated atmospheric CO<sub>2</sub> and nitrogen deposition contributed slightly to CH<sub>4</sub> emission.

[34] Comparison of the factorial contributions for the five regions reveals that climate change made bigger contributions to CH<sub>4</sub> emission in northern regions (the Northwest, North, and Northeast China), while made minor contributions to CH<sub>4</sub> emission in the southern regions (the Southwest and Southeast China). Elevated atmospheric CO<sub>2</sub>, nitrogen deposition, and O<sub>3</sub> pollution made large contributions in the Southeast China, while made minor contribution in other regions.

## 4. Discussion

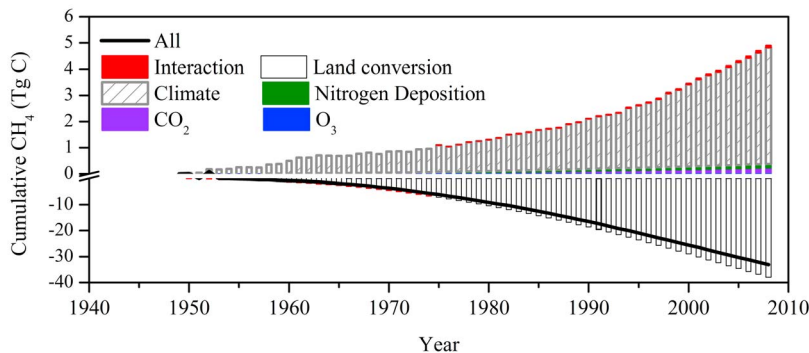
### 4.1. Source Strength of CH<sub>4</sub> Emission From China's Marshland

[35] This study reported that the source strength of CH<sub>4</sub> emission from China's marshland was approximately

$1.38 \pm 0.03$  Tg C for the time period of 1995–2004 at a wetland area of  $94.10 \sim 101.6 \times 10^3$  km<sup>2</sup>. So the areal CH<sub>4</sub> emission was  $13.63 \sim 14.98$  g C m<sup>-2</sup> a<sup>-1</sup>. And a study for the continental United States reported a total CH<sub>4</sub> emission rate of 4.13 Tg a<sup>-1</sup> with a wetland area of  $210 \times 10^3$  km<sup>2</sup> [Potter *et al.*, 2006]; the areal CH<sub>4</sub> emission is 19.64 g C m<sup>-2</sup> a<sup>-1</sup>. So the areal CH<sub>4</sub> emission from marshland in China is lower than those in the United States. The areal CH<sub>4</sub> emission from China's marshland is also lower than the summarized CH<sub>4</sub> emission rates from temperate swamps, temperate marshes, and temperate floodplains [Bartlett and Harriss, 1993]. This is probably due to a great amount of marshland in the northeastern China, which features low temperature and has relatively lower emission rate of CH<sub>4</sub> (Figure 3).

### 4.2. Major Factors Responsible for Temporal Variation of CH<sub>4</sub> Emission Over China's Marshland

[36] We identified marshland loss as the dominate factor contributing to CH<sub>4</sub> emission reduction in China's marshland over the past 60 years. This is consistent with one study which reported the primary role of marshland loss on global warming potential reduction over the northeastern China [Huang *et al.*, 2010]. Compared to other natural ecosystems,



**Figure 5.** Factorial contribution to the national CH<sub>4</sub> emission from China's marshland during 1949–2008. All: the contribution from all factors combined together; Interaction: the contribution from multiple-factor interaction; Land conversion: the contribution from marshland loss; Climate: the contribution from climate change; O<sub>3</sub>: the contribution from O<sub>3</sub> pollution; Nitrogen deposition: the contribution from nitrogen deposition; CO<sub>2</sub>: the contribution from elevated atmospheric CO<sub>2</sub>; noted that the negative contribution of O<sub>3</sub> is very small, see Tables 7 and 8 for exact estimates.

**Table 7.** Factorial Contribution to the National CH<sub>4</sub> Emission From Marshland in China<sup>a</sup>

Time Period	Elevated Atmospheric CO <sub>2</sub>	Nitrogen Deposition	O <sub>3</sub> Pollution	Climate Change	Marshland Loss	Interaction	All Factors	Baseline Emission
1949-1958	4.2 (3.0 ~ 5.9)	3.9 (2.8 ~ 5.6)	-0.1 (-0.09 ~ -0.2)	352.0 (256.1 ~ 501.7)	-849.0 (-617.6 ~ -1210.2)	-4.2 (-3.0 ~ -6.0)	-493.3 (-358.8 ~ -703.1)	23150.8 (17255.3 ~ 32924.2)
1959-1968	15.6 (11.6 ~ 21.7)	15.0 (11.2 ~ 20.9)	-1.4 (-1.0 ~ -1.9)	416.9 (311.4 ~ 580.8)	-2766.8 (-2066.4 ~ -3854.5)	1.4 (1.1 ~ 2.0)	-2319.3 (-1732.2 ~ -3231.0)	23150.8 (17255.3 ~ 32924.2)
1969-1978	29.4 (22.0 ~ 41.3)	25.4 (19.0 ~ 35.7)	-7.8 (-5.9 ~ -11.0)	348.1 (260.3 ~ 488.7)	-5436.3 (-4064.3 ~ -7631.7)	9.4 (7.0 ~ 13.2)	-5031.8 (-3761.9 ~ -7063.8)	23150.8 (17255.3 ~ 32924.2)
1979-1988	46.4 (34.4 ~ 65.8)	32.4 (24.1 ~ 46.0)	-18.4 (-13.7 ~ -26.2)	610.8 (453.6 ~ 867.1)	-7703.6 (-5721.1 ~ -10936.7)	10.6 (7.9 ~ 15.1)	-7021.8 (-5214.8 ~ -9968.9)	23150.8 (17255.3 ~ 32924.2)
1989-1998	58.4 (43.3 ~ 83.3)	35.9 (26.6 ~ 51.2)	-22.1 (-16.4 ~ -31.5)	1081.3 (801.7 ~ 1542.8)	-9994.2 (-7409.8 ~ -14259.9)	21.9 (16.3 ~ 31.3)	-8818.8 (-6538.3 ~ -12592.9)	23150.8 (17255.3 ~ 32924.2)
1999-2008	75.1 (55.0 ~ 109.1)	38.1 (27.9 ~ 55.4)	-34.3 (-25.1 ~ -49.8)	1632.1 (1194.0 ~ 2370.7)	-11151.7 (-8158.0 ~ -16198.2)	46.6 (34.1 ~ 67.7)	-9394.0 (-6872.2 ~ -13645.1)	23150.8 (17255.3 ~ 32924.2)
1949-2008	229.0 (169.5 ~ 326.8)	150.8 (111.6 ~ 215.5)	-84.2 (-62.3 ~ -120.1)	4441.1 (3286.4 ~ 6336.3)	-37901.6 (-28046.9 ~ -54075.3)	85.9 (63.5 ~ 122.5)	-33078.9 (-24478.2 ~ -47194.7)	138904.8 (103531.5 ~ 197544.9)

<sup>a</sup>Gg C/10 years or Gg C/60 years; the range in brackets represent the 95% confidence ranges for each estimate. The sum of factorial contribution may not equal to all combined results because of rounding in calculation.

marshlands embrace higher CH<sub>4</sub> emission rate [Bartlett and Harriss, 1993; Song et al., 2009; Tian et al., 2010a]; so the marshland loss dominated the CH<sub>4</sub> emission reduction. Another site-level study also reported that marshland loss has significantly reduced CH<sub>4</sub> emission in the northeastern China [Jiang et al., 2009].

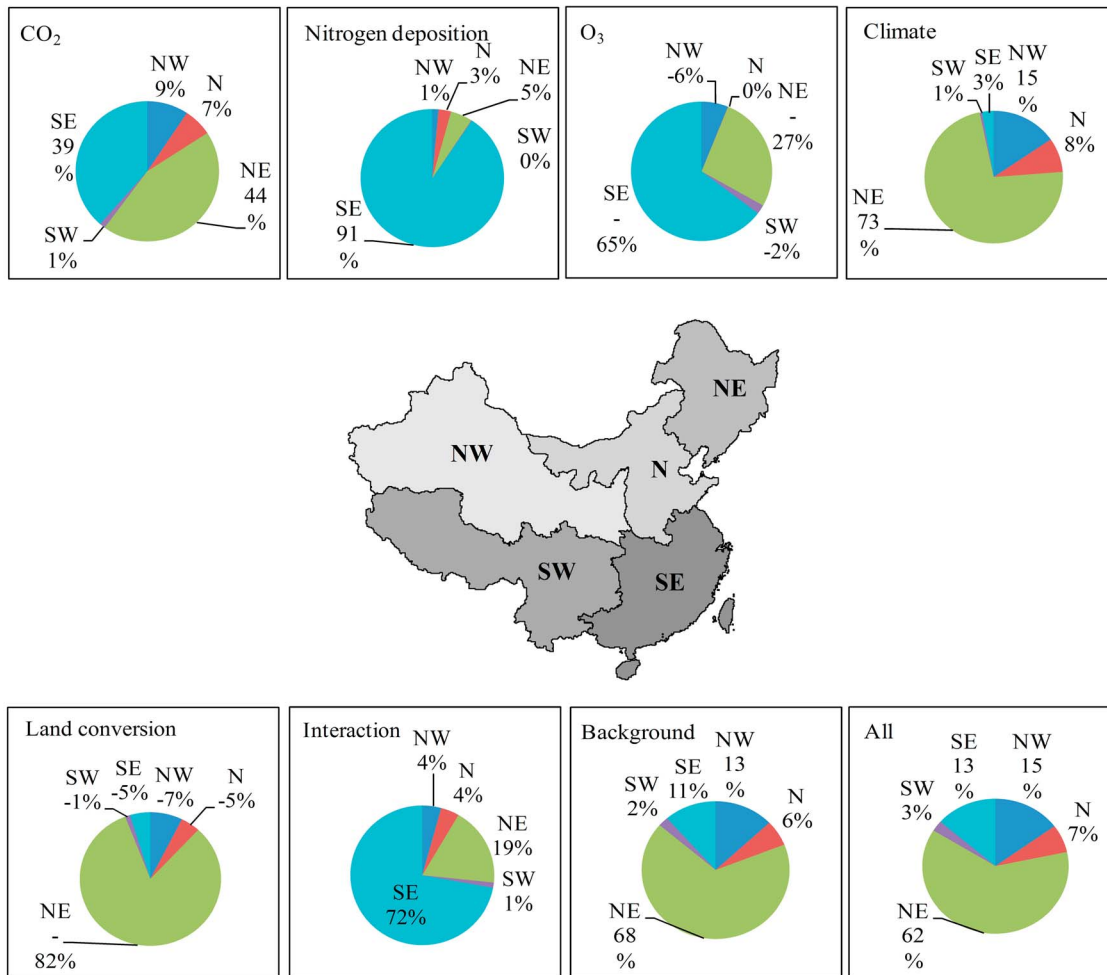
[37] The O<sub>3</sub> pollution decreased CH<sub>4</sub> emission by its inhibition of CH<sub>4</sub> production by reducing carbon assimilation to the system. In the DLEM, the effects of O<sub>3</sub> pollution were simulated as its suppressing effects on photosynthesis [Ren et al., 2007]; the lower carbon assimilation to the system directly reduces the carbon allocation to methane production [Whiting and Chanton, 1993]. The stimulating effects of elevated atmospheric CO<sub>2</sub> and nitrogen deposition on CH<sub>4</sub> emission are due to the positive effects of elevated atmospheric CO<sub>2</sub> and N on photosynthesis which would provide more substrates for CH<sub>4</sub> production [Whiting and Chanton, 1993].

### 4.3. Major Factors Responsible for Spatial Variations of CH<sub>4</sub> Emission Over China's Marshland

[38] The spatial distribution of CH<sub>4</sub> emission along latitudinal and longitudinal directions is consistent with the spatial distribution of marshland over China. The peak at the 29° N is associated with the marshland at downstream of the Changjiang river [Zhao, 1999]. The high CH<sub>4</sub> emission at mid-high latitude is associated with the marshland in the northeastern China [Zhao, 1999; Liu and Ma, 2000; Wang, 2002]. The CH<sub>4</sub> emission along the longitudinal direction is associated with the wetland distribution and spatial pattern of areal CH<sub>4</sub> emission rate. More wetlands are located in northern parts of China; and the areal CH<sub>4</sub> emission rate is higher in the eastern than the western China (Figure 3).

[39] Due to the high spatial heterogeneity of driving forces over China, the simulated CH<sub>4</sub> emission rate from China's marshland showed a substantial spatial variation. A number of studies have reported CH<sub>4</sub> production and oxidation controlled by some factors such as soil pH, soil texture, temperature, soil moisture, soil nutrient content etc [Conrad, 1996; Lai, 2009; Le Mer and Roger, 2001]. All the driving forces in this study are highly varied across the nation, which would lead to spatial variations in CH<sub>4</sub> production and oxidation. The highly spatial heterogeneity of nitrogen deposition, and O<sub>3</sub> pollution might also contributed to spatial variations in CH<sub>4</sub> emission from China's marshland. Given the lower contribution of these global change factors to CH<sub>4</sub> emission (Table 8), their contribution to spatial variations in CH<sub>4</sub> flux might be relatively low.

[40] We have found that the climate change made significant contribution to CH<sub>4</sub> emission in the northern regions, while minor contributions to CH<sub>4</sub> emission in the southern region. This might be due to two reasons. One is the higher changing rates of climate variables in the northern regions; our climate data indicate that temperature (daily maximum, minimum and average temperature) significantly increased over the northern regions; while no significant changing trends were observed for the southern regions. The second reason might be the higher sensitivity of soil organic matter to climate change in the northern regions. A number of studies have confirmed that the soil organic matter in cool



**Figure 6.** Regional contributions to CH<sub>4</sub> emission induced by global change factors, individually and in combination.

region is more sensitive to temperature increase than those in warm regions [Davidson and Janssens, 2006; Denman et al., 2007]. The northern region in China is experiencing temperature-limitation [Zhu et al., 2007], so the soil organic matters in northern regions are more sensitive to climate warming. The high temperature sensitivity of CH<sub>4</sub> emission in northern regions indicates that climate warming might produce more organic carbon for methane production, thus trigger a positive feedback for climate warming over wetlands in the mid-high latitudes. This indicates that the natural wetland in the mid-high latitudes worldwide might be very important for global climate change given the high

density of carbon storage and high temperature sensitivity of the stored carbon which might be released as either CO<sub>2</sub> or CH<sub>4</sub>.

**4.4. Multiple-Factor Interactive Effects on CH<sub>4</sub> Emission From Marshland**

[41] Global change is a complicated issue which involves a number of aspects of environmental change and their interactions. For example, the elevated atmospheric CO<sub>2</sub> [Hutchin et al., 1995] and nitrogen deposition [L. H. Zhang et al., 2007] have been found to enhance CH<sub>4</sub> emission, however, their combined effects on CH<sub>4</sub> is negative (-0.03 Tg C for

**Table 8.** Baseline and Factorial Contribution to Regional CH<sub>4</sub> Emission, Individually and in Combination<sup>a</sup>

Regions	Elevated Atmospheric CO <sub>2</sub>	Nitrogen Deposition	O <sub>3</sub> Pollution	Climate Change	Land Conversion	Interaction	Baseline	Total
Northwest	0.14%	0.01%	-0.03%	4.31%	-17.69%	0.02%	113.24%	100.00%
North	0.21%	0.06%	<0.01%	5.26%	-23.95%	0.05%	118.37%	100.00%
Northeast	0.15%	0.01%	-0.03%	4.92%	-47.37%	0.02%	142.29%	100.00%
Southwest	0.11%	0.01%	-0.07%	0.95%	-16.11%	0.04%	115.06%	100.00%
Southeast	0.62%	0.95%	-0.38%	0.83%	-12.34%	0.43%	109.88%	100.00%

<sup>a</sup>The sum of factorial contribution may not equal to 100% because of rounding in calculation.

60 years), which is consistent with a recent study [Pancotto *et al.*, 2010]. This interactively negative effect of elevated atmospheric CO<sub>2</sub> and nitrogen deposition is caused by nitrogen accumulation in soil and vegetation, named as progressive nitrogen limitation [Luo *et al.*, 2004]. So the influence of multiple-factor interaction on CH<sub>4</sub> emission from marshland is critically important for further understanding of CH<sub>4</sub> emission and mitigation. In this study, the multiple-factor interaction represents the interaction among marshland loss, climate change, elevated atmospheric CO<sub>2</sub>, nitrogen deposition, and O<sub>3</sub> pollution. It varied from negative to positive, depending on the time period, location and factors.

[42] Our comparative study on factorial contributions for five sub-regions reveals that the highest interaction in the southeastern China had relatively uniform contributions from elevated atmospheric CO<sub>2</sub>, nitrogen deposition, climate change, and marshland loss (Table 8). This indicates that the interactions would be strong when all factors are more convergent. However, if one or two factors are predominant, their effects would out-compete others and lead to negligible interactive effects. A comprehensive understanding of interactions among these factors is needed to be acquired through field experiments [Dermody, 2006].

#### 4.5. Uncertainties and the Way Forward

[43] Using a process-based ecosystem model, we estimated the magnitude, spatiotemporal patterns of CH<sub>4</sub> emission from China's marshland, and further attributed the variations to multiple global change factors. There are several limitations in data and model might bring some uncertainties in this study, and need further improvement in future study. First, the wetland distribution data used in this study might need improvements; currently, there are several sources of wetland distribution in China [Liu *et al.*, 2005; Niu *et al.*, 2009]; however, not all data set are publicly accessible; the uncertainties caused by different sources of wetland data might need to be evaluated in future work. Second, the constant geological properties along the study period might cause some uncertainties; for example, the constant pH over the 60-year study period is a source of uncertainty; yet no explicit data for the changes of pH over the study period at large scale are available at current stage; dynamic soil properties in model simulations would be a great improvement in ecosystem modeling studies. Third, the temperature sensitivity of CH<sub>4</sub> flux is a very important parameter for regional estimation of CH<sub>4</sub> flux in the changing climate scenario, which needs further investigation in regional studies. Fourth, it has been found that the aerenchyma is highly related to CH<sub>4</sub> transport capacity in wetland vegetation. Although the DLEM incorporates this mechanism in simulating CH<sub>4</sub> transport [Tian *et al.*, 2010a; Xu *et al.*, 2010], the various species responses would be important and need to be improved in future work.

[44] Last but not least, the factorial contributions to CH<sub>4</sub> emission from marshland in China has been validated by comparing with published results conducted in other countries, rather than local field observations due to the scarcity of field experiment across China's marshlands. Local parameterization and validation of the process-based model with field observations in China will be important for

evaluating the accuracy and precision of regional simulations of CH<sub>4</sub> flux.

## 5. Conclusions

[45] The temporal and spatial patterns of CH<sub>4</sub> emissions from marshland in China during 1949–2008 were examined. The results indicated that over the past 60 years, CH<sub>4</sub> emission from China's marshland continuously decreased; the marshland loss, followed by O<sub>3</sub> pollution, is the major reason for the decline of CH<sub>4</sub> emission from China's marshland. While elevated atmospheric CO<sub>2</sub>, nitrogen input enhanced CH<sub>4</sub> emission from marshland. The climate change generated largest positive effects on CH<sub>4</sub> emission; of which the temperature had larger effects than precipitation. The effect of multiple-factor interaction is complex; it could be positive or negative, depending on specific year. The major contribution to the national decline is from the Northeast China, where experienced dramatic land conversion from marshland to cropland [Liu and Ma, 2000; Wang, 2002]. The strong stimulating effects of CH<sub>4</sub> emission by rising temperature in the midlatitude indicates that the positive feedback between climate system and the soil carbon in the mid-high latitude might be not too far away as expected.

[46] This study provided the first national estimation of multiple-factor-induced CH<sub>4</sub> emission over China's marshland. Even though marshland loss caused negative effects on biodiversity and ecosystem services, it reduced the CH<sub>4</sub> release to the atmosphere. Meanwhile the changing global environment including elevated atmospheric CO<sub>2</sub> and nitrogen input did stimulate CH<sub>4</sub> emission. Given that a large amount of marshland was converted to other usages, and the present marshland is experiencing severe environmental changes, multiple factorial field experiments will be essential for better understanding CH<sub>4</sub> emission from marshland and the underlying mechanisms. While modeling approach needs to be more associated with field studies for better addressing factorial contribution on CH<sub>4</sub> flux at a large scale, the data-model fusion approach will be a major direction for future research on CH<sub>4</sub> flux estimation and mitigation.

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