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Methane dynamics in an estuarine brackish *Cyperus malaccensis* marsh: Production and porewater concentration in soils, and net emissions to the atmosphere over five years

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ABSTRACT

Wetlands can potentially affect global climate change through their role in modulating the atmospheric concentrations of methane (CH_4). Their overall CH_4 emissions, however, remain the greatest uncertainty in the global CH_4 budget. One reason for this is the paucity of long-term field measurements to characterize the variability of CH_4 emissions from different types of wetlands. In this study, we quantified CH_4 emissions from a brackish, oligohaline *Cyperus malaccensis* marsh ecosystem in the Min River Estuary in southeast China over five years. Our results showed substantial temporal variability of CH_4 emissions from this brackish marsh, with hourly fluxes ranging from 0.7 ± 0.6 to 5.1 ± 3.7 mg m^{-2} h^{-1} (mean ± 1 SD) during the study period. The inter-annual variability of CH_4 emissions was significantly correlated with changes in soil temperature, precipitation and salinity, which highlighted the importance of long-term observations in understanding wetland CH_4 dynamics. Distinct seasonal patterns in soil CH_4 production rates and porewater CH_4 concentrations also were observed, and were both positively correlated with CH_4 emissions. The seasonal variations of CH_4 emissions and production were highly correlated with salinity and porewater sulfate levels. The mean annual CH_4 efflux from our site over the five-year period was 23.8 ± 18.1 g CH_4 m^{-2} yr^{-1}, indicating that subtropical brackish tidal marsh ecosystems could release a large amount of CH_4 into the atmosphere. Our findings further highlight the need to obtain high-frequency and continuous field measurements over the long term at multiple spatial scales to improve our current estimates of wetland CH_4 emissions.

1. Introduction

The increasing worldwide concern over global climate change and its effects on environmental and human well-beings calls for a better understanding of the magnitude of global greenhouse gas emissions (Tong et al., 2010). Methane (CH_4) is a potent greenhouse gas with a global warming potential 34 times higher than that of CO_2 per mass unit over a 100-year time scale, and contributes to approximately 20% of the global radiative forcing (IPCC, 2013). Global atmospheric CH_4 levels have increased by threefold since 1750, reaching 1845 ± 2 ppb in 2015 (World Meteorological Organization, 2016). Quantifying the potential source strength of various ecosystems has become one of the top priorities for improving the future predictions of CH_4 emissions.

Wetlands are estimated to contribute 20–39% of the global CH_4 emissions (Laanbroek, 2010), with natural wetlands being the single largest source of CH_4. Over the past few decades, considerable efforts were made to quantify CH_4 emissions from different natural wetlands around the world (e.g. Bubier and Moore, 1994; Kutzbach et al., 2004; Hendriks et al., 2010; Tong et al., 2012). However, the majority of these field campaigns were carried out over a relatively short period of not

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more than two years, which provided little knowledge of the inter-annual variability of CH_4 emissions from most types of wetlands other than a few exceptions in northern wetlands, e.g. Song et al. (2009), Jackowicz-Korczyński et al. (2010), and Moore et al. (2011). Long-term observations over multiple seasons and years are critical for determining accurate ecosystem CH_4 budgets (Song et al., 2009). In addition, the availability of long-term data set will improve ecosystem modelling by providing inputs for model calibration and validation, as well as insights on the key factors regulating wetland CH_4 emissions into the atmosphere (Tian et al., 2008; Song et al., 2009).

Coastal wetlands, located at the interface between the terrestrial and marine environments, are biogeochemically important ecosystems that span widely from the arctic to the tropical zones (Chmura et al., 2003; Wang et al., 2016). Previous studies have shown that the sediments in coastal wetlands are generally small atmospheric sources (Bartlett and Harriss, 1993; Poffenbarger et al., 2011; Livesley and Andrusiak, 2012; Koebsch et al., 2013), or even weak sinks of CH_4 (Sun et al., 2013). The low CH_4 source strength of coastal wetlands is mainly because of the relatively high sulfate concentrations in marine waters, which favour the activities of sulfate-reducing bacteria while at the same time hamper the metabolism of methanogens through intense competition for substrates (Poffenbarger et al., 2011; Callaway et al., 2012; Vizza et al., 2017). However, some short-term field studies provide evidence that large CH_4 emissions from wetlands can occur even when sulfate reduction is a dominant process (Lee et al., 2008; Marín-Muñiz et al., 2015; Holm Jr. et al., 2016). The high uncertainty associated with the magnitude and control of CH_4 emissions from coastal wetlands could partly be related to the inherently dynamic environment which introduces a large temporal variability of CH_4 fluxes that is not adequately accounted for by s infrequent field measurements.

In this study, monthly CH_4 flux measurements were made in a subtropical tidal *Cyperus malaccensis* (shichito matgrass) marsh in the Min River Estuary in southeast China over five years between 2007–2009, and 2013–2014. We hypothesized that there would be significant seasonal and inter-annual variability in CH_4 emissions, which implies that flux estimates would be sensitive to the sampling frequency and study duration. We also investigated the temporal correlations between several environmental variables and soil CH_4 production rate, porewater CH_4 concentration, and net CH_4 emissions.

2. Materials and methods

2.1. Site description

This study was carried out in the Shanyutan wetland (26°00′36″–26°03′42″ N, 119°34′12″–119°40′40″ E), the largest tidal wetland area (ca. 3120 ha) in the Min River Estuary, southeast China (Fig. 1). The Shanyutan wetland is influenced by a subtropical monsoonal climate, with a mean annual temperature of 19.6 °C and an annual precipitation of 1350 mm (Tong et al., 2010). The dominant vegetation species in the Shanyutan wetland included the native *Cyperus malaccensis* and *Phragmites australis*, as well as the invasive *Spartina alterniflora* (smooth cordgrass). The average height of *C. malaccensis* at the site was about 1.4 m. The study site was characterized by semi-diurnal tides, such that the soil surface was submerged for approximately 7 h over a 24 h cycle, and at other times, the soil surface was exposed to air (Tong et al., 2010). The average salinity of the tidal water was 4.2 ± 2.5‰ (Tong et al., 2010).

2.2. Gas sampling and CH_4 flux estimation

Net CH_4 emissions were measured in the intertidal zone in the mid-western part of the Shanyutan wetland (26°01′46″ N, 119°37′31″ E), which was dominated by *C. malaccensis*, a widespread plant species at the site. Triplicate 1 m × 1 m plots, with a distance of < 5 m between plots, were established for regular measurement of CH_4 emissions in the *C. malaccensis* stand. CH_4 flux measurements were carried out monthly from early January to early December in 2007–2009 and 2013–2014. A wooden boardwalk was built to facilitate access to the study plots and minimize potential plot disturbance caused by field measurements. The wooden boardwalk and the study plots were damaged during a major typhoon event in 2010, thus we built a new boardwalk and established new plots adjacent to the damaged ones (< 15 m apart) in 2012. During 2013–2014, we continued with gas flux measurements at the new plots.

CH_4 flux measurements were made using static closed chambers and gas chromatography techniques (Hirota et al., 2004; Song et al., 2009; Moore et al., 2011; Marín-Muñiz et al., 2015) with gas samples collected during the neap tides in the morning. The static chamber consisted of two parts: a 30 cm tall stainless steel bottom collar (length and width of 50 × 50 cm in 2007–2009, and 35 × 35 cm in 2013–2014) and a polyvinyl chloride top chamber (length, width and height of 50 × 50 × 170 cm in 2007–2009, and 35 × 35 × 140 cm in 2013–2014). The bottom collar was inserted into the marsh soils, leaving only 2 cm above the soil surface, approximately 10 days prior to the first flux measurement, and was then left in place for the duration of the study. A fan was installed inside the chamber to mix the headspace air during gas sampling. During each flux measurement, headspace air samples were drawn into air sampling bags (Dalian Delin Gas Packing Co., Ltd., China) at 10-minute intervals over a total duration of 30 min in each sampling plot. The total number of gas samples collected per year was 144 (12 months × 4 time intervals × 3 sites). CH_4 concentrations in the gas samples were determined using a gas chromatograph (GC-2010, Shimadzu, Kyoto, Japan) equipped with a flame ionization detector (FID). The rate of CH_4 emission (mg m^{-2} h^{-1}) was calculated based on the slope of the linear regression between CH_4 concentration in the chamber headspace and time. The annual (cumulative) CH_4 emissions (*AE*, g CH_4 m^{-2}) (Song et al., 2009; Moore et al., 2011; Xiang et al., 2015) were calculated using Eq. (1):

$$AE = \sum MF_i \times D_i \times 24 \quad (1)$$

where MF_i is the CH_4 flux at the *i*th month of the year (mg CH_4 m^{-2} h^{-1}), and D_i is the number of days in the *i*th month of the year.

2.3. Measurement of soil CH_4 production rate

Soil CH_4 production in coastal wetlands has distinct spatio-temporal heterogeneity that could be related to variations in thermal conditions and other abiotic factors (e.g. soil moisture, soil substrate, etc.) (Segers, 1998; Vizza et al., 2017). To assess the variability of soil CH_4 production rates across different depths in our marsh, triplicate sediment cores were randomly collected down to a depth of 100 cm in January (winter), March (spring), July (summer), and October (autumn) of 2012. Intact soil cores were collected using a steel sediment sampler (i.d. = 5 cm), sub-divided into ten sections at 10 cm intervals in the field, and then kept on ice in coolers and transported to the laboratory within 6 h. The rate of soil CH_4 production was measured following the method of Wachinger et al. (2000). The chambers (5 cm inner diameter, 12 cm height) used for the anoxic incubation of soil cores were made of polyoxymethylene, which was gas-impermeable and inert to CH_4. Before the start of incubation, the chambers were flushed with N_2 gas for 15 min to create an anaerobic condition (Wassmann et al., 1998). The cores were then incubated at in situ temperatures, i.e. 10.2, 17.5, 27.5, and 21.5 °C for winter, spring, summer, and autumn, respectively, for a duration of 15 days. We collected 5 mL gas samples from the chamber using a syringe at three day intervals ($n = 5$) over the course of the incubation, with N_2 gas being added after each gas sampling to re-establish the ambient atmospheric pressure. The CH_4 concentrations in gas samples were analysed immediately by gas chromatograph. The CH_4 production rates (μg CH_4 g^{-1} (dry weight) day^{-1}) were calculated based on the rate of change in chamber headspace CH_4 concentrations