Effects of coastal marsh conversion to shrimp aquaculture ponds on CH₄ and N₂O emissions

P. Yang, David Bastviken, D. Y. F. Lai, B. S. Jin, X. J. Mou, C. Tong and Y. C. Yao

The self-archived postprint version of this journal article is available at Linköping University Institutional Repository (DiVA):

<http://urn.kb.se/resolve?urn=urn:nbn:se:liu:diva-143354>

N.B.: When citing this work, cite the original publication.

Yang, P., Bastviken, D., Lai, D. Y. F., Jin, B. S., Mou, X. J., Tong, C., Yao, Y. C., (2017), Effects of coastal marsh conversion to shrimp aquaculture ponds on CH₄ and N₂O emissions, *Estuarine, Coastal and Shelf Science*, 199, 125-131. <https://doi.org/10.1016/j.ecss.2017.09.023>

Original publication available at:

<https://doi.org/10.1016/j.ecss.2017.09.023>

Copyright: Elsevier

<http://www.elsevier.com/>

1 GRAPHICAL ABSTRACT

Effects of coastal marsh conversion to shrimp aquaculture ponds on CH₄ and N₂O emissions

P. Yang^{a,b}, D. Bastviken^c, D.Y.F. Lai^d, B.S. Jin^a, X.J. Mou^e, C. Tong^{a,b,f,*}, Y.C. Yao^e

^a *School of Geographical Sciences, Fujian Normal University, Fuzhou 350007, China*

^b *Key Laboratory of Humid Subtropical Eco-geographical Process of Ministry of Education, Fujian Normal University, Fuzhou 350007, China*

^c *Department of Thematic Studies-Environmental Change, Linköping University, Linköping 58183, Sweden*

^d *Department of Geography and Resource Management, and Centre for Environmental Policy and Resource Management, The Chinese University of Hong Kong, Shatin, New Territories, Hong Kong SAR, China*

^e *Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences, Changchun 130102, China*

^f *Research Centre of Wetlands in Subtropical Region, Fujian Normal University, Fuzhou 350007, China*

Corresponding author: Chuan Tong

Telephone: 086-0591-87445659,

Fax: 086-0591-83465397

Email: yangping528@sina.cn (P. Yang) and tongch@fjnu.edu.cn (C. Tong)

ABSTRACT

In this study, we compared the CH₄ and N₂O fluxes from a tidal brackish *Cyperus malaccensis* marsh ecosystem and nearby shrimp ponds, converted from *C. malaccensis* marsh in the last 3 – 4 years, in the Min River estuary of southeast China over the aquaculture period of the year. Significant differences in CH₄ and N₂O fluxes were observed in space (between brackish marsh and shrimp ponds) and in time (between sampling occasions that were distributed over the aquaculture period). CH₄ fluxes from the shrimp ponds were on an average 10-fold higher than from the brackish marsh. N₂O emissions, on the other hand, were lower from the shrimp pond (25% of the emissions from the brackish marsh). Accessory data indicates that these patterns were primarily linked to water level variability and temperature (all fluxes), sediment porewater sulphate concentrations (CH₄ flux) and total nitrogen concentrations (N₂O flux). Our research demonstrates that the coastal marsh ecosystem converted to aquaculture ponds considerably alter emissions of CH₄ and N₂O and provides input to the global discussion on how to account for emissions from various types of flooded land in greenhouse gas inventories.

Keywords: Land-use conversion; Methane; Nitrous oxide; Tidal marsh; Shrimp pond; Anthropogenic influence

1. Introduction

Methane (CH_4) and nitrous oxide (N_2O) are key greenhouse gases. The globally averaged CH_4 and N_2O concentrations reached 1845 ± 2 ppb and 328.0 ± 0.1 ppb in 2015, respectively, and their concentrations have increased by 150% and 20% since pre-industrial times ([World Meteorological Organization, 2016](#)). The levels of CH_4 and N_2O now contribute on the order of 16 and 6%, respectively, to the overall greenhouse gas radiative forcing, and the atmospheric concentrations are increasing rapidly ([World Meteorological Organization, 2016](#)). There are still large gaps in the attribution of various sources of CH_4 and N_2O . Therefore, quantifying the potential source strength of individual ecosystems and of land use change are critical for improved emission assessments ([Purvaja and Ramesh, 2001](#); [Song and Liu, 2016](#)).

Land-use change can lead to great changes in hydrology, nutrient cycling characteristics, sediment physicochemical properties, and overall ecosystem function ([Dick and Osunkoya, 2000](#)), which could substantially influence greenhouse emissions. For example, [Verchot et al. \(2000\)](#) and [Wick et al. \(2005\)](#) found that conversion of tropical forests in Amazonia to extensively managed pastures and secondary forests could lead to reduced N_2O emissions and decreased CH_4 uptake. [Inubushi et al. \(2003\)](#) suggested that converting a secondary forest peatland to paddy field increased the annual emissions of CH_4 to the atmosphere, while transforming the secondary forest to upland decreased the emissions. Studies by [Roulet \(2000\)](#), [Jiang et al. \(2009\)](#) and [Huang et al. \(2010\)](#) have concluded that conversion of wetlands to agricultural lands with cropping activities can potentially lead to increases in N_2O emissions and decreases in CH_4 emissions. Furthermore, [Hu et al. \(2016\)](#) and [Liu et al. \(2016\)](#)

found that N_2O and CH_4 emissions decreased following conversion of rice paddies to inland crab – fish aquaculture.

Coastal wetlands, the interface between terrestrial and ocean ecosystems, are sites of intense biological production. Land-use changes have accelerating impacts on coastal wetlands (Liu and Mo, 2016). Recent studies indicates that land reclamation for farmland and other land use have already caused degradation or loss of about 50% of the world's coastal wetlands (Barbier et al., 2011). However, it is presently unclear how natural marsh conversion to other land-use (e.g., aquaculture ponds) affects greenhouse gases (GHGs) emissions.

The Asia-Pacific region is one of the most active regions of wetland degradation due to the increasing population and rapid economic development. It has been estimated that approximately 33% of the available wetland area have been drained or converted to areas for intensive food production in Asia-Pacific region (Huang et al., 2010). The conversion of natural coastal wetlands into aquaculture ponds is very common in the region (Zuo et al., 2013). Prawn is one of the most popular mariculture products in the Asia-Pacific region, and shrimp ponds are one of the main aquaculture pond types. The change from temporarily water logged coastal wetlands to a continuously flooded pond, can strongly affect the emissions of CH_4 and N_2O . Given the extent of this landscape change, it is important to quantify the effects of salt marsh ecosystem conversion to shrimp ponds on CH_4 and N_2O emissions. Unfortunately, there is little information available on the effects of this conversion on trace gases emissions. Therefore, in the present study, we measured the fluxes of CH_4 and N_2O from a brackish marsh ecosystem, and from nearby shrimp ponds which were recently converted from the brackish marsh ecosystem. Our primary aim was to assess the effects of

coastal marsh conversion to aquaculture pond on CH₄ and N₂O emissions. A secondary goal was to provide data that can improve GHG emission inventories and contribute to the ongoing evaluation by the IPCC Task Force on National Greenhouse Gas Inventories (TFI), on how to account for emissions from various types of flooded land - a context in which the urgent need of data on how aquaculture affect GHG emissions has been noted.

2. Materials and methods

2.1. Site description

This study was carried out in the Shanyutan wetland of the Min River estuary, southeast China (26°00'36"–26°03'42" N, 119°34'12"–119°40'40" E; [Fig. 1](#)). This area is located in the southern subtropical monsoonal climate zone, with a mean annual temperature of 19.6 °C and precipitation of 1350 mm ([Tong et al., 2010](#)). In the high inter-tidal zone of the central-western portion of the Shanyutan wetland, one of the main macrophyte marsh communities is dominated by *Cyperus malaccensis* growing approximately 1.0 m tall. At the study site, tides were typical semidiurnal where the marsh soil surface is submerged for approximately 7 h of a 24 h cycle, while at other times the soil surface is exposed to air. There is normally between 10 and 150 cm of water above the soil surface during the tidal inundation period.

Conversion of the *C. malaccensis* marsh ecosystem was performed in the Shanyutan wetland of the Min River estuary in recent years, and almost all of the converted lands were used as shrimp ponds. To assess the effects of coastal marsh conversion to aquaculture ponds on CH₄ and N₂O fluxes, a typical *C. malaccensis* marsh stand was selected, and within the stand three replicate chambers separated by about 5 m were deployed for the gas flux

measurements. In the nearby shrimp ponds zone which was within ~100 m distance to the *C. malaccensis* marsh stand, three shrimp ponds separated by about 20 m were randomly selected. Three replicate floating chambers at distance intervals of 15 m were deployed in each pond for the gas sampling. The shrimp ponds were constructed in 2012, and all marsh vegetation were removed and shallow pond basins with steep sides and homogenous depth were created. The area of each shrimp pond was about 7500 m², and the mean water depth over the study period was 1.3 m. The water in the aquaculture ponds is a mix of salt water pumped from coastal water and fresh water drawn locally. The average salinity of the shrimp pond water (June to October) was $1.48 \pm 0.01\%$, and the average breeding density of the shrimp pond was about 45 shrimps m⁻². The aquaculture period usually starts in June and ends in November.

2.2. Gas flux sampling and measurement

CH₄ and N₂O fluxes in the *C. malaccensis* marsh ecosystem and shrimp pond ecosystem were determined during sampling campaigns using enclosed static chambers ([Tong et al., 2013](#); [Olsson et al., 2015](#)) and floating static chambers ([Bastviken et al., 2010](#); [Yang et al., 2015](#)). Considering the three different culture phases of an early, middle, and late stage for the shrimp ponds and the possibility of sampling in the brackish marsh, the gas flux measurements in the shrimp ponds and marsh ecosystem were carried out in the middle-June, August and October in 2015. During each flux measurement, the headspace gas samples were collected by using 60 ml plastic syringes, equipped with three-way stopcocks, at 15 min intervals over a 45 min period after chamber enclosure (total of four samples) ([Tong et al., 2012](#)), the first sample was collected at 0 min. Samples were injected into pre-evacuated

airtight gas sampling bags (Delin, Dalian, China), transported to the laboratory and analyzed within 24 h using gas chromatography (GC-2010, Shimadzu, Kyoto, Japan) equipped with FID and ECD. The fluxes of CH₄ and N₂O in the chamber were calculated from the slope of the regression between CH₄ and N₂O concentration and time, and expressed as mg m⁻² h⁻¹ and μg m⁻² h⁻¹, respectively. The details of gas flux sampling and measurement process are provided in the [Supporting Information](#). In addition, the floating static chambers are used to measure the total gas fluxes from the ponds, and the diffusive fluxes were calculated using wind speed, finally the difference being the ebullition fluxes. More detailed information can be referenced a previous studies of [Chuang et al. \(2017\)](#).

2.3. Ancillary environmental measurements

During each sampling campaign, surface sediment temperature, pH and electrical conductivity (EC) and salinity at a depth of 10 cm were determined *in situ*. On each sampling date, six sediment core samples per site were taken from the topsoil (0 – 10 cm) using a 5 cm diameter steel cylinder. These sediment samples were used to analyze the physicochemical parameters of sediment. The detailed measurement and analysis process is summarized in the [Supporting Information](#).

2.4. Statistical analyses

The results are presented as means of the replications, with standard error (S.E.). The detailed statistical analysis process is summarized in the [Supporting Information](#). Global warming potentials (GWP) of 34 for CH₄ and 298 for N₂O over 100 years ([IPCC, 2013](#)) were used to convert CH₄ and N₂O emissions to CO₂-eq to compare their greenhouse impacts ([Sun et al., 2013](#)).

3. Results

3.1. Effects of marsh conversion to shrimp ponds on CH₄ emissions

There were large variations in CH₄ emission both among ecosystems and observation periods (Fig. 2A). CH₄ emission fluxes from the brackish marsh ecosystem during the non-waterlogged periods ranged from 4 to 15 mg CH₄ m⁻² h⁻¹, with the lower flux during August and higher flux during October. Furthermore, our previous studies (Tong et al., 2010; Tong et al., 2013) found that the brackish marsh dominated by *C. malaccensis* and *P. australis* together emitted CH₄ in a range from 2 to 13 mg CH₄ m⁻² h⁻¹ at three tidal stages (before flood, during the flooding and ebbing process, and after ebb) from April to October. At the shrimp ponds, the CH₄ emission fluxes was similar to fluxes from the marsh in June, but much higher in both August and October, ranging from 6 to 200 mg CH₄ m⁻² h⁻¹ (Fig. 2A). The mean CH₄ flux from shrimp ponds amounted to 123 ± 48 mg CH₄ m⁻² h⁻¹, which was significantly higher than that of 11 ± 2 mg CH₄ m⁻² h⁻¹ in brackish marsh ($F_{df=1} = 16.707$, $p = 0.015$) (Fig. 2A). Taking the above analyses together, our results indicate that CH₄ emissions increased during the culture period following brackish marsh ecosystem conversion to shrimp ponds. In addition, the ebullition emission from the shrimp ponds were calculated according to the method of Chuang et al. (2017) and followed the order: August (189 mg CH₄ m⁻² h⁻¹) > October (65 mg CH₄ m⁻² h⁻¹) > June (5 mg CH₄ m⁻² h⁻¹) (L.S. Tan, unpublished data, 2015).

3.2. Effects of marsh conversion to shrimp ponds on N₂O emissions

The N₂O emission dynamics of shrimp ponds were clearly different from the brackish marsh ecosystem (Fig. 2B). At the brackish marsh, the N₂O emissions during the non-waterlogged periods showed an increasing trend over the study period, and ranged from

36 to 80 $\mu\text{g N}_2\text{O m}^{-2} \text{ h}^{-1}$. Furthermore, our previous studies (Tong et al., 2013) found that N_2O emissions in the brackish marsh dominated by *C. malaccensis* and *P. australis* (relative coverage was 50%) ranged from 44 to 116 $\mu\text{g N}_2\text{O m}^{-2} \text{ h}^{-1}$ during the tidal inundation period. In comparison, the shrimp ponds showed much less temporal variation and generally lower N_2O emissions (ranged from 11 to 26 $\mu\text{g N}_2\text{O m}^{-2} \text{ h}^{-1}$). The mean N_2O flux from shrimp ponds amounted $17 \pm 3 \mu\text{g N}_2\text{O m}^{-2} \text{ h}^{-1}$, which was lower than that of $58 \pm 15 \mu\text{g N}_2\text{O m}^{-2} \text{ h}^{-1}$ in brackish marsh ($F_{df=1} = 7.350$, $p = 0.035$) (Fig. 2B). Taking the above analyses together, our preliminary results indicated that conversion decreased N_2O emissions during the culture period with the conversion of the brackish marsh ecosystem to shrimp ponds.

3.3. Ancillary environmental measurements

The brackish marsh had higher levels of total carbon (TC) and total nitrogen (TN) and slightly lower pH and lower bulk sediment density (Table 1). Sediment temperatures ranged between 23.97 and 31.37 °C with the August sampling being warmest (Fig. 3). In the June and October sampling, temperatures were similar among the systems while the shrimp pond sediments were a few degrees warmer during the August measurements. Sedimentary pore water SO_4^{2-} levels were on an average > 6-fold higher in the brackish marsh (411 mg L^{-1}) than in the shrimp pond (61 mg L^{-1}) (Table 1). However, such large differences were only apparent during measurements in August and October, while levels were similar among the systems at the June sampling (Fig. 3). The mean porewater ammonium and nitrate in the brackish marsh was $29.27 \pm 2.15 \text{ mg L}^{-1}$ and $0.40 \pm 0.06 \text{ mg L}^{-1}$, respectively, which was significantly higher than that in the shrimp pond of $13.01 \pm 1.64 \text{ mg L}^{-1}$ and $0.29 \pm 0.04 \text{ mg L}^{-1}$ ($p < 0.05$, Table 1). In the brackish marsh both ammonium and nitrate levels seemed to

slightly increase between the measurements (Fig. 3).

4. Discussion

4.1. Effects of marsh conversion to shrimp ponds on trace greenhouse gases emissions

4.1.1. Higher CH₄ emissions from the aquaculture ponds than from original marsh wetland ecosystems

CH₄ emission fluxes in shrimp ponds were clearly higher than in *C. malaccensis* brackish marsh during the mid and later parts of the aquaculture season (Fig. 2A), which suggest that the conversion of brackish marsh to shrimp ponds could increase CH₄ emissions during the culture period. One partial explanation for this is the change to continuous inundation following marsh wetland conversion to shrimp ponds. In the present study, the brackish marsh soil experienced tidal flooding for approximately 7 h of a 24 h cycle, and at other times the soil surface was exposed to air. However, the shrimp ponds soil was continuously flooded during the aquaculture period, with the mean depth of 1.3 m. The continuous water cover could effectively reduce oxygen penetration into soils/sediments favouring anaerobic decomposition and CH₄ production by methanogens and while lowering the potential for CH₄ oxidation (Kettunen et al., 1999; Yang et al., 2013). The brackish marsh on the other hand, the regular exposure of sediments to air which would favour CH₄ oxidation and disfavor CH₄ production relative to the shrimp ponds.

Another effect of the brackish marsh conversion to aquaculture ponds that is likely to influence CH₄ emissions is the change of SO₄²⁻ concentrations (Table 1). Multiple studies have indicated that sulfate (SO₄²⁻) reducing bacteria (SRB) compete for H₂/CO₂ and acetate (CH₃COO⁻) with methanogens, and have a stronger affinity to these substrates; hence the

coexistence of SRB and SO_4^{2-} is known to inhibit the production and emission of CH_4 (e.g. [van der Gon et al., 2001](#); [Sun et al., 2013](#)). In addition, SO_4^{2-} -driven CH_4 oxidation can be important in coastal marine sediments. CH_4 fluxes from the brackish marsh and shrimp ponds both showed significant negative correlations with porewater SO_4^{2-} concentration during the observation period ($r^2 = 0.69$, $p = 0.006$; [Fig. 2A](#) and [Fig. 3B](#)). During the June sampling, when SO_4^{2-} levels were similar across the systems, CH_4 fluxes were also similar, while the later differences in SO_4^{2-} both between systems and between measurement times within the systems reflected patterns in CH_4 emissions. These results suggest that reduced SO_4^{2-} concentration was an important factor that together with the permanent flooding caused the elevated fluxes from the shrimp ponds. We also observed large changes in sediment bulk density and grain composition along the land use conversion ([Table 1](#)). In the present study, the mean sediment bulk density in shrimp ponds was about 103% higher than in the brackish marsh. Moreover, the percentage of clay and silt were significantly higher than that of the brackish marsh during the observation period ($p < 0.05$). Furthermore, a significant positive relationship ($r^2 = 0.43$, $p = 0.003$, $n = 18$) between sediment bulk density and $\log_{10}$ CH_4 flux were found during the observation period. The high bulk density, clay and silt in the shrimp ponds contributed to more stable anaerobic conditions for methanogenesis ([Wagner et al., 1999](#); [Veldkamp et al., 2008](#)), but the importance of this potential explanation relative to the more established effects of SO_4^{2-} and flooding is unclear at present.

There was also a temporal similarity between relative levels of NH_4^+ , sediment temperature, and pH versus CH_4 flux in the shrimp pond ([Fig. 2A](#) and [Fig. 3](#), $p < 0.05$). The production of both NH_4^+ and CH_4 would be favoured by high rates of anaerobic

decomposition. Notably highest levels of NH_4^+ and CH_4 flux in the aquaculture pond occurred in August when the sediment temperature was highest providing an explanation to the temperature correlation. Methanogenesis, and also methane fluxes have recently been shown to be exponentially related with temperature (Yvon-Durocher et al., 2014; Natchimuthu et al., 2015). Previous studies indicate that the methanogens are pH sensitive and grow best at pH 7.7 in the coastal wetland (Chang and Yang, 2003). The average sediment pH during the August was 7.3 which might have facilitated the optimum growth of methanogens during the culture period, which in turn may have facilitated higher CH_4 production. Hydrogenotrophic methanogenesis consumes CO_2 , thereby increasing pH, which may contribute to the positive correlation between flux and pH. Hence, elevated pH may not have been a direct driver for methanogenesis but rather a result of processes favourable for methanogenesis. Altogether, SO_4^{2-} levels and temperature are the most likely factors directly influencing the CH_4 production over time in the shrimp pond, while it is more unclear if NH_4^+ and pH are indirectly or directly related with CH_4 production.

While permanent flooding, the reduced SO_4^{2-} concentrations, and temperature all were likely contributors to high CH_4 production in the shrimp ponds, efficient and rapid flux pathways by which CH_4 can escape methane oxidation are needed for high fluxes. In the open water shrimp pond system, no plant mediated fluxes were possible in contrast to the brackish marsh. However, the shallow water, with complete mixing could rapidly transport CH_4 released from sediments to the atmosphere. In addition, the high shrimp density leads to an extraordinary capacity for bioturbation triggering ebullition (bubble flux from the sediment). The much higher fluxes during later shrimp growth stages, in combination with the

observations that high CH₄ fluxes were captured with low variability among replicate chambers and the three studied ponds during 45-minute flux measurements, indicate that ebullition induced by shrimp activity was likely the main transport pathway when the highest fluxes were measured. For developing predictive models of shrimp pond emissions we therefore suggest that water level fluctuations, SO₄²⁻ concentrations, temperature and shrimp growth stage and density be considered.

4.1.2. N₂O Emissions were lower in aquaculture ponds compared to the original coastal marsh ecosystems

In contrast with CH₄ emission fluxes, we found higher N₂O emissions during the culture period in the brackish marsh ecosystem compared to the shrimp ponds (Fig. 2B). The results are mainly attributable to the intermittent flooding and the interactions of these hydrologic patterns and other biotic/abiotic factors. Especially, the increased availability of dissolved oxygen during regular flushing events (e.g., tidal cycle) is likely very important. The regular tidal induced changes between aerobic and anaerobic conditions in surface sediments could have disturbed nitrogen cycling processes and resulted in N₂O production. For example, denitrification could be disrupted upon shifts from anaerobic to aerobic conditions, stimulating aerobic bacterial (i.e., nitrobacteria) activity (Regina et al., 1996; Dinsmore et al., 2009) and N₂O production rate (Yang et al., 2013). The release of bioavailable nutrients from plant residues can provide a large source of available nitrogen for microbial production of N₂O (Dinsmore et al., 2009, Yang et al., 2013). The continuous flooding in the shrimp pond reduced oxygen penetration into the soil/sediment, restricted the diffusion of N₂O, and favoured N₂O reduction to N₂ by denitrifiers (Rusch and Rennenberg, 1998).

N₂O fluxes from both the brackish marsh and shrimp ponds showed significant positive correlation with NO₃⁻-N ($r^2 = 0.37$, $p = 0.048$) and NH₄⁺-N (shrimp ponds) ($r^2 = 0.72$, $p = 0.004$) concentration during the observation period, respectively. These results suggest that the difference in available nitrogen substrates influenced N₂O emissions together with effects of the water cover discussed above from two environments. Similar results were presented by [Yang et al. \(2013\)](#) who found that N₂O emissions at low water table (0–11 and 11.3–28 cm below the soil surface) were 60–110% greater than at high water table position (0–14 cm above the soil surface), which were caused by the changes in nitrogen substrates (NH₄⁺-N) at different water table levels.

N₂O fluxes in the shrimp pond were also positively correlated with temperature ($r^2 = 0.64$, $p = 0.010$; based on differences between measurement occasions of sediment temperature). Significant positive relationships between temperature and N₂O emissions have been reported in many studies ([Beaulieu et al., 2010](#); [Clough et al., 2011](#)).

Previous studies have suggested that high levels of salinity and SO₄²⁻ can inhibit nitrification, which would result in lower N₂O production and emission ([Joye and Hollibaugh, 1995](#); [Wang et al., 2009](#)). However, this study showed the opposite trend ($p < 0.01$; [Fig. 3](#)). Similar results were observed by [Marton et al. \(2012\)](#) and by [Reddy and Crohn \(2014\)](#) who reported N₂O emissions were increased with increasing salinity level. A proposed explanation for these latter patterns, supported by our study, is that the osmotic stress at the higher salinity levels may hamper the denitrifiers responsible for reducing N₂O to N₂, or alternatively salt could interfere with N₂O reductase activity or its de novo synthesis under aerobic conditions ([Reddy and Crohn, 2014](#)). Similarly, [Marton et al. \(2012\)](#) found that N₂O reductase enzymes

were inhibited at high salinity levels thereby resulting in greater N₂O emissions under aerobic conditions.

4.2. Perspective, limitations and further outlook on GHGs from coastal marsh conversion to aquaculture ponds

Because our study focused on evaluating the effects of coastal wetland conversion to aquaculture ponds on CH₄ and N₂O emissions during the aquaculture period of the year, CH₄ and N₂O flux measurements were taken only during the local aquaculture period (from May to October) rather than over a whole annual cycle. Therefore, the results of this study are preliminary and need to be validated further, preferably with long-term field measurements of all three major greenhouse gases (CH₄, N₂O, CO₂) in multiple systems taking various factors into consideration, such as regional climate, marsh ecosystem and aquaculture pond type, specific aquaculture management practice and reclamation history.

Aquaculture is increasing with an average rate of 8.7% per year and has over the past 40 years been the fastest-growing food-production sector ([Herbeck et al., 2013](#)). With rapid aquaculture development, coastal wetlands have suffered great losses and degradation from wetland conversion over past decades in many regions of the world. It was reported that roughly 16% of the world's mangrove area have been lost between 1980 and 2005 when aquaculture experienced the greatest increase ([FAO, 2007](#)), and aquaculture ponds construction are regarded as one of the major causes for this decline ([Páez-Osuna, 2001](#); [Alongi 2002](#); [Herbeck et al., 2013](#)). In the China, roughly 16% of the natural coastal wetlands have been lost between the 1970s and 2007 ([Zuo et al., 2013](#)). Since 1970, the area of aquaculture ponds converted from coastal wetlands increased by 1,260 km², or by 1,700% in

relative terms (Zuo et al., 2013). The subtropical region (include Guangdong, Guangxi, Fujian, Zhe Jiang, Shanghai and Jiangsu province) where approximately 49% of the China's aquaculture production takes place, is one of the most active regions of aquaculture in China (Fig. 1) (Yao et al., 2016). Given this extensive and rapid land use change, it is important to improve the GHG emission inventory in China's coastal zone by quantifying the effects of coastal marsh ecosystem conversion to aquaculture ponds on GHG fluxes. If assuming that the results of this study are representative for the whole relevant area and time periods the average change in CH₄ and N₂O flux would be 1.387 Tg yr⁻¹ and 0.0019 Tg yr⁻¹ for the total areas converted so far (1260 km²; only the aquaculture part of the year considered in this extrapolation). Expressed as global warming potentials (CO₂ equivalent units) the net effect in terms of CH₄ and N₂O flux changes would be 34.73 Tg CO₂-eq yr⁻¹ over a 100-year timeframe. This represents about 8% of the net emissions from terrestrial natural ecosystems of China (e.g., natural wetland, forestlands, grasslands) (Cai et al., 2012). Obviously more data is needed to increase the precision in this estimate, but our calculation example for China clearly show that the conversion to aquaculture ponds can be very important for large scale GHG balances.

5. Conclusion

This study provided an early insight into differences in CH₄ and N₂O fluxes caused by conversion of coastal brackish marshes to shrimp ponds. The results showed significantly increased CH₄ emissions, but decreased N₂O emissions. However, this study results from a limited area across a limited time period. In the future work, there would need to be more points in time and more study sites, especially, covering different temperatures and wetland

types.

Acknowledgements

This research was financially supported by the National Science Foundation of China (No. 41671088 and 41371127), the Program for Innovative Research Team of Fujian Normal University (IRTL1205) and National Basic Scientific and Technological Work (2013FY111805). Contribution by David Bastviken was supported by the Swedish Research Council VR and Linköping University. We thank anonymous reviewers and Prof. Dr. Tim Jennerjahn for their valuable comments and suggestions.

References

- Alongi, D.M., 2002. Present state and future of the world's mangrove forests. *Environ. Conserv.* 29(03), 331-349. <http://dx.doi.org/10.1017/S0376892902000231>.
- Barbier, E.B., Hacker, S.D., Kennedy, C., Koch, E.W., Stier, A.C., Silliman, B.R., 2011. The value of estuarine and coastal ecosystem services. *Ecol. Monogr.* 81, 169-193. <http://dx.doi.org/10.1890/10-1510.1>.
- Bastviken, D., Santoro, A.L., Marotta, H., Pinho, L.Q., Calheiros, D.F., Crill, P., Enrich-Prast, A., 2010. Methane emissions from Pantanal, South America, during the low water season: toward more comprehensive sampling. *Environ. Sci. Technol.* 44(14), 5450-5455. <http://dx.doi.org/10.1021/es1005048>.
- Beaulieu, J.J., Shuster, W.D., Rebholz, J.A., 2010. Nitrous oxide emissions from a large, impounded river: The Ohio River. *Environ. Sci. Technol.* 44(19), 7527-7533. <http://dx.doi.org/10.1021/es1016735>.
- Cai, Z.C., 2012. Greenhouse gas budget for terrestrial ecosystems in China. *Sci. China Earth Sci.* 55(2), 173-182. <http://dx.doi.org/10.1007/s11430-011-4309-8>
- Chang, T.C., Yang, S.S., 2003. Methane emissions from wetlands in Taiwan. *Atmos. Environ.* 37, 4551-4558. [http://dx.doi.org/10.1016/S1352-2310\(03\)00588-0](http://dx.doi.org/10.1016/S1352-2310(03)00588-0).
- Chuang, P.-C., Young, M.B., Dale, A.W., Miller, L.G., Herrera-Silveira, J.A., Paytan, A., 2017. Methane fluxes from tropical coastal lagoons surrounded by mangroves, Yucatán, Mexico. *J. Geophys. Res. Biogeosci.* 122, <http://dx.10.1002/2017JG003761>.
- Clough, T.J., Buckthought, L.E., Casciotti, K.L., Kelliher, F.M., Jones, P.K., 2011. Nitrous oxide dynamics in a braided river system, New Zealand. *J. Environ. Qual.* 40, 1532-1541. <http://dx.doi.org/10.2134/jeq2010.0527>.
- Dick, T.M., Osunkoya, O.O., 2000. Influence of tidal restriction floodgates on decomposition of mangrove litter. *Aquat. Bot.* 68, 273-280. [http://dx.doi.org/10.1016/S0304-3770\(00\)00119-4](http://dx.doi.org/10.1016/S0304-3770(00)00119-4).

- Dinsmore, K.J., Skiba, U.M., Billett, M.F., Rees, R.M., 2009. Effect of water table on greenhouse gas emissions from peatland mesocosms. *Plant Soil*. 318, 229-242. <http://dx.doi.org/10.1007/s11104-008-9832-9>.
- Food and Agricultural Organization (FAO), 2007. *The world's mangroves 1980-2005*. FAO Forestry paper 153. Rome: Forest Resources Division, FAO, 77 pp.
- Herbeck, L.S., Unger, D., Wu, Y., Jennerjahn, T.C., 2013. Effluent, nutrient and organic matter export from shrimp and fish ponds causing eutrophication in coastal and back-reef waters of NE Hainan, tropical China. *Cont. Shelf Res.* 57, 92-104. <http://dx.doi.org/10.1016/j.csr.2012.05.006>.
- Hu, Z.Q., Wu, S., Ji, C., Zou, J.W., Zhou, Q.S., Liu, S.W., 2016. A comparison of methane emissions following rice paddies conversion to crab-fish farming wetlands in southeast China. *Environ. Sci. Pollut. Res.* 23(2), 1505-1515. <http://dx.doi.org/10.1007/s11356-015-5383-9>.
- Huang, Y., Sun, W.J., Zhang, W., Yu, Y.Q., Su, Y.H., Song, C.C., 2010. Marshland conversion to cropland in northeast China from 1950 to 2000 reduced the greenhouse effect. *Global Change Biol.* 16, 680-695. <http://dx.doi.org/10.1111/j.1365-2486.2009.01976.x>.
- Inubushi, K., Furukawa, Y., Hadi, A., Purnomo, E., Tsuruta, H., 2003. Seasonal changes of CO₂, CH₄ and N₂O fluxes in relation to land-use change in tropical peatlands located in coastal area of South Kalimantan. *Chemosphere* 52(3), 603-608. [http://dx.doi.org/10.1016/S0045-6535\(03\)00242-X](http://dx.doi.org/10.1016/S0045-6535(03)00242-X).
- IPCC (2013) In: Stocker, T.F., Qin, D., Plattner, G.K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P.M., (Eds.), *Climate Change 2013: the Physical Science Basis*. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.
- Jiang, C.S., Wang, Y.S., Hao, Q.J., Song, C.C., 2009. Effect of land-use change on CH₄ and N₂O emissions from freshwater marsh in Northeast China. *Atmos. Environ.* 43, 3305-3309. <http://dx.doi.org/10.1016/j.atmosenv.2009.04.020>.
- Joye, S.B., Hollibaugh, J.T., 1995. *Influence of sulfide inhibition of nitrification on nitrogen regeneration in sediments*. *Science* 270, 623-625.
- Kettunen, A., Kaitala, V., Lethinen, A., Lohila, A., Alm, J., Silvola, J., Martikainen, P.J., 1999. Methane production and oxidation potentials in relation to water table fluctuations in two boreal mires. *Soil Biol. Biochem.* 31(12), 1741-1749. [http://dx.doi.org/10.1016/S0038-0717\(99\)00093-0](http://dx.doi.org/10.1016/S0038-0717(99)00093-0).
- Liu, Q., Mo, X., 2016. Interactions between surface water and groundwater: key processes in ecological restoration of degraded coastal wetlands caused by reclamation. *Wetlands* 36 (Suppl 1), S95-S102. <http://dx.doi.org/10.1007/s13157-014-0582-6>.
- Liu, S.W., Hu, Z.Q., Wu, S., Li, S.Q., Li, Z.F., Zou, J.W., 2016. Methane and nitrous oxide emissions reduced following

- conversion of rice paddies to inland crab–fish aquaculture in southeast China. *Environ. Sci. Technol.* 50 (2), 633-642.
<http://dx.doi.org/10.1021/acs.est.5b04343>.
- Marton, J.M., Herbert, E.R., Craft, C.B., 2012. Effects of salinity on denitrification and greenhouse gas production from laboratory-incubated tidal forest soils. *Wetlands* 32 (2), 347-357. <http://dx.doi.org/10.1007/s13157-012-0270-3>.
- Natchimuthu, S., Sundgren, I., Gålfalk, M., Klemetsson, L., Crill, P., Danielsson, Å., Bastviken, D., 2015. Spatio-temporal variability of lake CH₄ fluxes and its influence on annual whole lake emission estimates. *Limnol. Oceanogr.* 61(S1), S13-S26. <http://dx.doi.org/10.1002/lno.10222>.
- Olsson, L., Ye, S., Yu X., Wei, M., Krauss, K.W., Brix, H., 2015. Factors influencing CO₂ and CH₄ emissions from coastal wetlands in the Liaohe Delta, Northeast China. *Biogeosciences* 12, 4965-4977. <http://dx.doi.org/10.5194/bg-12-4965-2015>
- Páez-Osuna, F., 2001. The environmental impact of shrimp aquaculture: a global perspective. *Environ. Pollut.* 112, 229-231.
[http://dx.doi.org/10.1016/S0269-7491\(00\)00111-1](http://dx.doi.org/10.1016/S0269-7491(00)00111-1).
- Purvaja, R., Ramesh, R., 2001. Natural and anthropogenic methane emission from coastal wetlands of south India. *Environ. Manage.* 27, 547-557. <http://dx.doi.org/10.1007/s002670010169>.
- Reddy, N., Crohn, D.M., 2014. Effects of soil salinity and carbon availability from organic amendments on nitrous oxide emissions. *Geoderma* 235-236, 363-371. <http://dx.doi.org/10.1016/j.geoderma.2014.07.022>.
- Regina, K., Nykänen, H., Silvola, J., Martikainen, P.J., 1996. Fluxes of nitrous oxide from boreal peatlands as affected by peatland type, water table level and nitrification capacity. *Biogeochemistry* 35(3), 401-418.
<http://dx.doi.org/doi:10.1007/BF02183033>.
- Roulet, N.T., 2000. *Peatlands, carbon storage, greenhouse gases, and the Kyoto Protocol: prospects and significance for Canada*. *Wetlands* 20(4), 605-615.
- Rusch, H., Rennenberg, H., 1998. Black alder (*Alnus Glutinosa* (L.) Gaertn.) trees mediate methane and nitrous oxide emission from the soil to the atmosphere. *Plant Soil.* 201(1), 1-7. <http://dx.doi.org/10.1023/A:1004331521059>.
- Song, H.L., Liu, X.T., 2016. Anthropogenic effects on fluxes of ecosystem respiration and methane in the Yellow River Estuary, China. *Wetlands* 36 (Suppl 1), S113-S123. <http://dx.doi.org/10.1007/s13157-014-0587-1>.
- Sun, Z.G., Wang, L.L., Tian, H.Q., Jiang, H.H., Mou, X.J., Sun, W.L., 2013. Fluxes of nitrous oxide and methane in different coastal *Suaeda salsa* marshes of the Yellow River estuary, China. *Chemosphere* 90(2), 856-865.
<http://dx.doi.org/10.1016/j.chemosphere.2012.10.004>.
- Tong, C., Huang, J.F., Hu, Z.Q., Jin, Y.F., 2013. Diurnal variations of carbon dioxide, methane, and nitrous oxide vertical fluxes in a subtropical estuarine marsh on neap and spring tide days. *Estuar. Coast.* 36(3), 633-642.
<http://dx.doi.org/10.1007/s12237-013-9596-1>.

- Tong, C., Wang, W., Huang, J., Gauci, V., Zhang, L., Zeng, C., 2012. Invasive alien plants increase CH₄ emissions from a subtropical tidal estuarine wetland. *Biogeochemistry* 111(1-3), 677-693. <http://dx.doi.org/10.1007/s10533-012-9712-5>.
- Tong, C., Wang, W.Q., Zeng, C.S., Marrs, R., 2010. Methane (CH₄) emissions from a tidal marsh in the Min River estuary, south-east China. *J. Environ. Health. Part A* 45, 506-516. <http://dx.doi.org/10.1080/10934520903542261>.
- van der Gon, H.A.D., van Bodegom, P.M., Wassmann, R., Lantin, R.S., Metra-Corton, T.M., 2001. Sulfate-containing amendments to reduce methane emissions from rice fields: mechanisms, effectiveness and costs. *Mitigation Adaptation Strategies* 6, 71-89. <http://dx.doi.org/10.1023/A:1011380916490>.
- Veldkamp, E., Purbopuspito, J., Corre, M.D., Brumme, R., Murdiyarso, D., 2008. Land use change effects on trace gas fluxes in the forest margins of Central Sulawesi, Indonesia. *J. Geophys. Res. Biogeosci.* 113(G2), G02003, <http://dx.doi.org/10.1029/2007JG000522>.
- Verchot, L.V., Davidson, E.A., Cattânio, J.H., Ackerman, I.L., 2000. Land-use change and biogeochemical controls of methane fluxes in soils of eastern Amazonia. *Ecosystems* 3, 41-56. <http://dx.doi.org/10.1007/s100210000009>.
- Wagner, D., Pfeiffer, E.-M., Bock, E., 1999. Methane production in aerated marshland and model soils: effects of microflora and soil texture. *Soil Biol. Biochem.* 31(7), 999-1006. [http://dx.doi.org/10.1016/S0038-0717\(99\)00011-5](http://dx.doi.org/10.1016/S0038-0717(99)00011-5).
- Wang, D.Q., Chen, Z.L., Sun, W.W., Hu, B.B., Xu, S.Y., 2009. Methane and nitrous oxide concentration and emission flux of Yangtze delta plain river net. *Sci. China Ser. B-Chem.* 52(5), 652-661. <http://dx.doi.org/10.1007/s11426-009-0024-0>.
- Wick, B., Veldkamp, E., De Mello, W.Z., Keller, M., Crill P., 2005. [Nitrous oxide fluxes and nitrogen cycling along a pasture chronosequence in Central Amazonia, Brazil](#). *Biogeosciences* 2(3), 175-187.
- World Meteorological Organization, 2016. WMO Greenhouse Gas Bulletin No.12 (October 2016). https://library.wmo.int/opac/doc_num.php?explnum_id=3084. pdf.
- Yang, J.S., Liu, J.S., Hu, X.J., Li, X.X., Wang, Y., Li, H.Y., 2013. Effect of water table level on CO₂, CH₄ and N₂O emissions in a freshwater marsh of Northeast China. *Soil Biol. Biochem.* 61, 52-60. <http://dx.doi.org/10.1016/j.soilbio.2013.02.009>.
- Yang, P., He, Q.H., Huang, J.F., Tong, C., 2015. Fluxes of greenhouse gases at two different aquaculture ponds in the coastal zone of southeastern China. *Atmos. Environ.* 115, 269-277. <http://dx.doi.org/10.1016/j.atmosenv.2015.05.067>.
- Yao, Y.C., Ren, C.Y., Wang, Z.M., Wang, C., Deng, P.Y., 2016. Monitoring of salt ponds and aquaculture ponds in the coastal zone of China in 1985 and 2010. *Wetl. Sci.* 14(6), 874-882 (in Chinese).
- Yvon-Durocher, G., Allen, A.P., Bastviken, D., Conrad, R., Gudas, C., St-Pierre, A., Thanh-Duc, N., del Giorgio, P.A., 2014. Methane fluxes show consistent temperature dependence across microbial to ecosystem scales. *Nature* 507, 488-491. <http://dx.doi.org/10.1038/nature13164>.
- Zuo, P., Li, Y., Liu, C.A., Zhao, S.H., Guan, D.M., 2013. Coastal wetlands of China: changes from the 1970s to 2007 based

on a new wetland classification system. *Estuar. Coast.* 36(2), 390-400. <http://dx.doi.org/10.1007/s12237-012-9575-y>.

Fig. 1. (A) Distribution of aquaculture ponds along the coastline of China; (B) Location of study site in the central-western portion of the Min River estuary in Fujian province; (C) A photo showing the brackish *C. malacensis* marsh; (D) A photo showing the adjacent shrimp pond.

Fig. 2. A comparison of mean (A) CH₄ and (B) N₂O fluxes between the brackish marshes and shrimp ponds. Bars represent mean $\pm$ SE ($n = 3$), and the different lowercase letters above the bars indicate significant differences at the $p < 0.05$ level among sampling sites.

Fig. 3. Mean (A) sediment temperature, and porewater concentrations of (B) SO_4^{2-} , (C) $\text{NH}_4^+\text{-N}$, and (D) $\text{NO}_3^-\text{-N}$ at a depth of 10 cm in June, August, and October in the brackish marsh and shrimp ponds. Bars represent mean $\pm$ SE ($n = 3$), and the different lowercase letters above the bars indicate significant differences at the $p < 0.05$ level among sampling sites.

Table 1

Environmental parameters of the sediments and water in the brackish marsh and shrimp ponds over the sampling campaign. Values are means ($\pm$ S.E.) of samples ($n = 3$ for TC, TN and sediment grain composition; $n = 9$ for other environmental parameters) collected from the *C. malaccensis* marsh and shrimp ponds over all sampling periods. The salinity in the brackish marsh was measured in the tidal water (Tong et al., 2012), NH_4^+ -N, NO_3^- -N and SO_4^{2-} concentrations were measured in sediment pore water, and other environmental variables were measured in dried sediments collected at a depth of 10 cm below the sediment-water interface.

Type	Sediment TC g kg ⁻¹	Sediment TN g kg ⁻¹	Porewater ion concentration (mg L ⁻¹)			Water salinity ‰	pH	Bulk density g cm ⁻³	Sediment grain composition (%)		
			NH ₄ ⁺ -N	NO ₃ ⁻ -N	SO ₄ ²⁻				Clay	Silt	Sand
Brackish marsh	21.69 $\pm$ 0.99 ^b	2.40 $\pm$ 0.06 ^b	29.27 $\pm$ 2.15 ^b	0.40 $\pm$ 0.06 ^b	411.32 $\pm$ 11.38 ^b	4.20 $\pm$ 2.50 ^b	6.37 $\pm$ 0.16 ^a	0.80 $\pm$ 0.01 ^a	9.80 $\pm$ 2.29 ^a	62.45 $\pm$ 1.52 ^b	27.76 $\pm$ 3.76 ^a
Shrimp pond	16.19 $\pm$ 0.63 ^a	1.86 $\pm$ 0.09 ^a	13.01 $\pm$ 1.64 ^a	0.29 $\pm$ 0.04 ^a	60.81 $\pm$ 13.56 ^a	1.48 $\pm$ 0.01 ^a	6.98 $\pm$ 0.12 ^b	1.62 $\pm$ 0.16 ^b	16.11 $\pm$ 0.91 ^b	67.35 $\pm$ 0.42 ^a	16.54 $\pm$ 1.12 ^b

Statistically significant differences in environmental parameters between the brackish marsh and shrimp ponds are indicated by different lower-case letters within each column ($p < 0.05$, independent sample T-test).

Supporting Information

Title: Effects of coastal marsh conversion to shrimp aquaculture ponds on CH₄ and N₂O emissions

P. Yang^{a,b}, D. Bastviken^c, D.Y.F. Lai^d, B.S. Jin^a, X.J. Mou^e, C. Tong^{a,b,f,*}, Y.C. Yao^e

^a *School of Geographical Sciences, Fujian Normal University, Fuzhou 350007, China*

^b *Key Laboratory of Humid Subtropical Eco-geographical Process of Ministry of Education, Fujian Normal University, Fuzhou 350007, China*

^c *Department of Thematic Studies-Environmental Change, Linköping University, Linköping 58183, Sweden*

^d *Department of Geography and Resource Management, and Centre for Environmental Policy and Resource Management, The Chinese University of Hong Kong, Shatin, New Territories, Hong Kong SAR, China*

^e *Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences, Changchun 130102, China*

^f *Research Centre of Wetlands in Subtropical Region, Fujian Normal University, Fuzhou 350007, China*

Corresponding author: Chuan Tong

Telephone: 086-0591-87445659,

Fax: 086-0591-83465397

Email: yangping528@sina.cn (P. Yang) and tongch@fjnu.edu.cn (C. Tong)

22 **Supporting Information Summary**

23 **No. of pages: 8**

24 **Page S3–S6:** Materials and Methods

25 **Page S7–S8:** References

2. Materials and methods

2.2. Gas flux sampling and measurement

CH₄ and N₂O fluxes in the *C. malaccensis* marsh ecosystem were determined during sampling campaigns using enclosed static chambers (Tong et al., 2013; Olsson et al., 2015). The enclosed static chambers were made of two parts: a stainless steel bottom collar (35 × 35 cm; 30 cm in depth) and a chamber made of Plexiglas® (35 × 35 cm; 100 cm in height). The bottom collar was inserted into the marsh sediment to a depth of 30 cm two weeks before flux measurement. To minimize possible disturbance of the measurement sites during sampling, a wooden boardwalk was built to access the sampling sites. The CH₄ and N₂O fluxes from shrimp ponds were measured using floating static chambers similar to those described by Bastviken et al. (2010) and Yang et al. (2015). The floating chambers consisted of two parts: a floating bottom collar and an open-bottomed chamber made of opaque Plexiglas® (inner diameter 35 cm × height 30 cm). We drove a small boat to the centre of the ponds to collect the gas samples.

Considering the three different culture phases of an early, middle, and late stage for the shrimp ponds and the possibility of sampling in the brackish marsh, the gas flux measurements in the shrimp ponds and marsh ecosystem were carried out in the middle-June, August and October in 2015. The gas samples from the tidal marsh ecosystem were collected only at the before flood stages because of safety concerns. The gas sampling from shrimp ponds and marsh ecosystem usually occurred at between 09:00 and 11:00 A.M. at local time because previous studies (Yang et al.,

2012; Tong et al., 2013) and the preliminary experiments in this sites (data not shown) all support that the flux at between 09:00 and 11:00 A.M. is almost equal to the daily mean flux. During each flux measurement, the headspace gas samples were collected by using 60 ml plastic syringes, equipped with three-way stopcocks, at 15 min intervals over a 45 min period after chamber enclosure (total of four samples) (Tong et al., 2012), the first sample was collected at 0 min. Samples were injected into pre-evacuated airtight gas sampling bags (Delin, Dalian, China), transported to the laboratory and analyzed within 24 h using gas chromatography (GC-2010, Shimadzu, Kyoto, Japan) equipped with FID and ECD. The gas chromatography configurations for analyzing the mixing ratios of CH₄ and N₂O were according to the method of Tong et al. (2012). The gas flux (F , mol m⁻² h⁻¹) was calculated according to the following equation (1):

$$F = \frac{dn}{dt} \times \frac{1}{A} \quad (1)$$

where dn/dt is the slope of amount of substance for CH₄ and N₂O over sampling period (mol h⁻¹); A is the chamber area (m²). The amount of CH₄ and N₂O in the chamber at different times was calculated from the following equation (2):

$$n = ppm \times 10^{-6} \times \frac{P_{tot} \times V}{R \times T} \quad (2)$$

where ppm is parts per million (usually comes from GC measurement); P_{tot} is total air pressure (usually around 1 atm = 1013.15 hPa); V is the chamber volume (L); R is common gas constant (0.082056 L atm K⁻¹ mol⁻¹); T is the absolute temperature during sampling (K).

In present study, the unit of CH₄ flux was reported as mg m⁻² h⁻¹ ($= F \times M \times 10^3$),

and N_2O flux was showed as $\mu\text{g m}^{-2} \text{h}^{-1}$ ($= F \times M \times 10^6$). M is molar mass of CH_4 and N_2O (mg mol^{-1}), CH_4 : 16, N_2O : 44.

2.3. Ancillary environmental measurements

During each sampling campaign, surface sediment temperature, pH and electrical conductivity (EC) and salinity at a depth of 10 cm were determined *in situ* by IQ150 instrument (IQ Scientific Instruments, USA) and 2265FS EC Meter (Spectrum Technologies Inc., USA) (Tong et al., 2012). Surface water salinity at a depth of 10 cm was determined *in situ* by using a salinity meter (Eutech Instruments-Salt6, USA). On each sampling date, six sediment core samples per site were taken from the topsoil (0 – 10 cm) using a 5 cm diameter steel cylinder. Three sediment samples were dried to constant weight at 60 °C for determination of bulk density (Olsson et al., 2015). Three additional sediment samples were freeze-dried and homogenized for analyzing total carbon (TC), total nitrogen (TN), $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$ and sediment grain composition (Sun et al., 2013). Total carbon (TC) and total nitrogen (TN) were analyzed on oven-dried subsamples and were ground to pass a 2 mm sieve, on a CHN Elemental Analyzer (Elementar Vario MAX CN, Germany). Sediment grain composition was analyzed by using Malvern Laser Particle Size Analyzer (Mastersizer 2000, UK). Sediment porewater $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ were extracted followed the description of by De Vittor et al. (2012) and Zhang et al. (2013), then were measured by sequence flow analyzer (San⁺⁺ SKALAR, Netherlands). The sediment porewater SO_4^{2-} at depth of 10 cm collection and measurement followed the description of Tong et al. (2015).

2.4. Statistical analyses

Before statistical analysis, the greenhouse gases (CH₄ and N₂O) fluxes and environmental variables were log₁₀ transformed resulting in normal distribution of data. We tested the statistical differences in log transformed greenhouse gases fluxes and environmental variables (except TC, TN and sediment grain composition%) between salt marshes and adjacent shrimp ponds during the study period by repeated measures analysis of variance (RMANOVA). Independent-Sample T Test was used to examine the differences in TC, TN and grain composition of the sediments between the brackish marsh and adjacent shrimp ponds. Correlations between environmental variables and GHG fluxes were tested by Pearson correlation analysis. All statistical analyses and graphics were conducted with SPSS version 17.0 (SPSS, Inc., USA) and OriginPro 7.5 (OriginLab Corporation, USA), respectively, and were considered significant at the 0.05 significance level. All data are reported as mean $\pm$ standard error (SE).

References

- Bastviken, D., Santoro, A.L., Marotta, H., Pinho, L.Q., Calheiros, D.F., Crill, P., Enrich-Prast, A., 2010. Methane emissions from Pantanal, South America, during the low water season: toward more comprehensive sampling. *Environ. Sci. Technol.* 44(14), 5450-5455. <http://dx.doi.org/10.1021/es1005048>.
- De Vittor, C., Faganeli, J., Emili, A., Covelli, S., Predonzani, S., Acquavita, A., 2012. Benthic fluxes of oxygen, carbon and nutrients in the Marano and Grado Lagoon (northern Adriatic Sea, Italy). *Estuar. Coast. Shelf S.* 113, 57-70.
- Olsson, L., Ye, S., Yu X., Wei, M., Krauss, K.W., Brix, H., 2015. Factors influencing CO₂ and CH₄ emissions from coastal wetlands in the Liaohe Delta, Northeast China. *Biogeosciences* 12, 4965-4977. <http://dx.doi.org/10.5194/bg-12-4965-2015>
- Sturm, K., Yuan, Z., Gibbes, B., Werner, U., Grinham, A., 2014. Methane and nitrous oxide sources and emissions in a subtropical freshwater reservoir, South East Queensland, Australia. *Biogeosciences* 11, 5245-5258. <http://dx.doi.org/10.5194/bg-11-5245-2014>.
- Sun, Z.G., Wang, L.L., Tian, H.Q., Jiang, H.H., Mou, X.J., Sun, W.L., 2013. Fluxes of nitrous oxide and methane in different coastal *Suaeda salsa* marshes of the Yellow River estuary, China. *Chemosphere* 90(2), 856-865. <http://dx.doi.org/10.1016/j.chemosphere.2012.10.004>.
- Tong, C., Huang, J.F., Hu, Z.Q., Jin, Y.F., 2013. Diurnal variations of carbon dioxide, methane, and nitrous oxide vertical fluxes in a subtropical estuarine marsh on neap and spring tide days. *Estuar. Coast.* 36(3), 633-642. <http://dx.doi.org/10.1007/s12237-013-9596-1>.
- Tong, C., She, C.X., Yang, P., Jin, Y.F., Huang, J.F., 2015. Weak correlation between methane production and abundance of methanogens across three brackish marsh zones in the Min River Estuary, China. *Estuar. Coast.* 38(6), 1872-1884. <http://dx.doi.org/10.1007/s12237-014-9930-2>.
- Tong, C., Wang, W., Huang, J., Gauci, V., Zhang, L., Zeng, C., 2012. Invasive alien plants increase CH₄ emissions from a subtropical tidal estuarine wetland. *Biogeochemistry* 111(1-3), 677-693. <http://dx.doi.org/10.1007/s10533-012-9712-5>.
- Van den Pol-van Dasselaar, A., Van Beusichem, M.L., Oenema, O., 1999. Methane emissions from wet grasslands on peat soil in a nature preserve. *Biogeochemistry* 44, 205-220. <http://dx.doi.org/10.1007/BF00992979>.
- Yang, P., Tong, C., He, Q.H., Huang, J.F., 2012. Diurnal variations of greenhouse gas fluxes at the water-air interface of aquaculture ponds in the Min River estuary. *Environ. Sci.* 33(12), 4194-4204. (in Chinese with English Abstract)

136 Yang, P., He, Q.H., Huang, J.F., Tong, C., 2015. Fluxes of greenhouse gases at two different aquaculture ponds in
 137 the coastal zone of southeastern China. Atmos. Environ. 115, 269-277.
 138 <http://dx.doi.org/10.1016/j.atmosenv.2015.05.067>.
 139 Zhang, L., Wang, L., Yin, K.D., Lü, Y., Zhang, D.R., Yang, Y.Q., Huang, X.P., 2013. Pore water nutrient
 140 characteristics and the fluxes across the sediment in the Pearl River estuary and adjacent waters, China. Estuar.
 141 Coast. Shelf S. 133, 182-192.