ORIGINAL RESEARCH

Soil Methane Production, Anaerobic and Aerobic Oxidation in Porewater of Wetland Soils of the Minjiang River Estuarine, China

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Abstract

Wetlands are important sources of methane emission. Anaerobic oxidation, aerobic oxidation and production of methane as well as dissolved methane are important processes of methane metabolism. We studied methane metabolism and the soil influencing factors. Potential soil methane production, anaerobic oxidation and aerobic oxidation rates, and dissolved methane in soil porewater changed seasonally and the annual average was 21.1 ± 5.1 µg g^{-1}d^{-1}, 11.0 ± 3.9 µg g^{-1}d^{-1}, 20.9 ± 5.8 µg g^{-1}d^{-1}, and 62.9 ± 20.6 µmol l^{-1}, respectively. Potential soil methane production and anaerobic and aerobic oxidation were positively correlated among themselves and with soil pH and negatively correlated with soil redox potential (Eh). Potential soil methane production and aerobic and anaerobic oxidation rates were negatively related to pore soil methane concentration. Thus, the more water-saturated the soil (the lower Eh), the higher its capacity to produce methane. The potential soil capacity for methane oxidation was higher both in the same anaerobic circumstances and when the soil was suddenly subjected to aerobic conditions. The results of this study suggested a buffer effect in the methane balance in wetland areas. The environmental circumstances favoring methane production are also favorable for anaerobic methane oxidation.

Keywords Methane production · Methane anaerobic oxidation · Methane aerobic oxidation · Dissolved methane · Minjiang River estuarine wetland

Introduction

Although the total area occupied by wetlands is only 4.6% of the total land area (Costanza et al. 1997), their ecosystem services value accounts for 32% of the total value of the world. The ecosystem services value of coastal wetlands accounts for 17% of the total global value (Costanza et al. 1997). Wetland is sensitive to external stress and is thus the ideal area for global change research (Simas et al. 2001). Methane is one of the important greenhouse gases affecting global climate change. Although wetlands only represent a small fraction of the Earth's land surface, they are the main sources of methane to the atmosphere, representing 20%–39% of global methane emissions (Laanbroek 2010). The relative increase of methane on a scale of 100 years is about 25 times that of carbon dioxide (IPCC 2014). Thus, there is a need to improve the knowledge of methane metabolism in wetland soils.

Methane metabolism is in several phases: production, oxidation, dissolved methane, transport and emission (Buckley et al. 2008). Methane production, oxidation and dissolved methane in water have great impacts on the ultimate reduction of methane emissions in wetlands (Singh 2011). In the twenty-first century, anaerobic methane oxidation has become a core issue (Raghoebarsing et al. 2006; Kniemeyer et al. 2007).

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However, reports about anaerobic methane oxidation in coastal estuarine wetlands are few. Furthermore, the relationships between potential anaerobic soil methane oxidation capacity and potential soil methane production as well as potential aerobic soil methane oxidation capacity are uncertain in wetland areas. Whether relationships exist between the capacity of the soil to produce and oxidize methane in anaerobic conditions and also between production and oxidation in wet conditions in dry-aerobic periods are two important questions that warrant research.

Methane metabolism in wetlands is strongly influenced by environmental factors that vary both spatially and temporally (Datta et al. 2013). The availability of electron acceptors and donors in soils plays a key role in regulating CH_4 production and consumption (Moran et al. 2008; Ettwig et al. 2010; Ro et al. 2011) thereby controlling dissolved methane and the emission of methane. Electron acceptors (e.g., Fe^{3+}, NO_3^-, and sulfate) are reduced during wet periods, but regenerated (oxidized) during dry periods (Neubauer et al. 2007). Soils can also provide carbon substrates for mediating CH_4 production and enhancing plant growth which in turn govern more than 90% of CH_4 transport to microbes (Le Mer and Roger 2001). Other environmental variables, including soil temperature, pH, redox potential (Eh) and salinity, also influence CH_4 metabolism (Song et al. 2008; Wang et al. 2017). Better knowledge and characterization of the metabolism and transport of CH_4 are essential for proper understanding and characterizing of GHG emissions from wetlands. Advancements in the knowledge of soil and soil porewater circumstances can favor the production and oxidation processes in soil media. The final methane contents in soil-atmosphere interface and the emission can give clues to choose between soil and plant community management strategies to diminish methane emissions as much as possible. This information can also provide clues to improve the models and projections of methane production and emissions on regional and global scales.

China has a coastline of 18,000 km with numerous estuaries and bays and diverse coastal wetlands which are important components of China's, as well as the world's wetlands. Minjiang River estuarine wetland is the main natural wetland of southeast coast of China. The wetland of Minjiang River estuary is rich in biological species and abundant in waterbird resources (Liu et al. 2006). Moreover, due to its unique location at the transition between central and southern subtropical climatic zones, the Minjiang River estuary is an important tidal wetland ecosystem (Zheng et al. 2006). Tidal wetlands are rich in animal and plant biodiversity (Zhou et al. 2006), and Minjiang River estuary serves as an excellent site representing the wide coastal wetland areas of this part of China coast-.

We studied: (1) the seasonal variation potential of soil methane production, anaerobic soil methane oxidation, aerobic soil methane oxidation, dissolved methane, methane emission and the relationships among these variables (2) the soil variables that have significant relationships with methane-related variables in Minjiang River estuarine wetland along the year. The results obtained in this study were also aimed to provide a scientific basis for suitable management of wetland avoiding CH_4 emissions as much as possible.

Materials and Methods

Study Area

This study was conducted in the Shanyutan wetland (26°01′46″N; 119°37′31″E, Fig. 1), the largest tidal estuarine wetland (approximately 3120 ha) in the estuary of the Minjiang River.

The climate in this region is relatively warm and wet with a mean annual temperature of 19.6 °C and mean annual precipitation of 1346 mm (Zheng et al. 2006). The soil surface was submerged across the study site beneath 10–120 cm of water for 3–3.5 h during each tidal inundation. Soil surfaces of the entire wetland were exposed at low tides during 24 h, and the weight percentage of water in the soil and soil redox potential were 116.39% and 12.57 mV respectively. Soil remains flooded at some depths. The average salinity of the tidal water between May and December 2007 was $4.2 \pm 2.5\%o$. *C. malaccensis* is one of the two dominant species of plants in this estuarine wetland. *C. malaccensis* is a native plant typically found in the upper (mid to high) portions of mudflats that grow between April and October, the highest population height is about 1.5 m, and the density is about 1000 m^{-2}. Below-ground rhizomes are creeping growth in the topsoil layers.

Experimental Design

Seasonal variation samples were collected from April 2012 to March 2013 from Shanyutan wetland in Minjiang River estuary. We established a plot of 900 m^2 in Shanyutan wetland and then collected the *C. malaccensis* wetland soil randomly after selecting three quadrats (100 m^2) within the big plot. Soil samples of 0–20 cm were collected with a small core sampler (length and diameter, 0.3 and 0.1 m). The sampling was conducted every month during the one-year period. Thus, a total of 36 soil samples (one wetland type × one soil layer × twelve months' × three replicate plots) were collected.

Measurements of Potential Methane Production

The potential soil methane production rate was determined for each sampled soil by placing 30 g of the fresh soil sample in a 120 ml incubation bottle and injecting 30 ml of distilled water (water: soil ratio was thus 1:1) (Wang et al. 2010; Bergman et al. 2000). The incubation bottles were filled with oxygen-free nitrogen through a small hole in the bottle stopper to
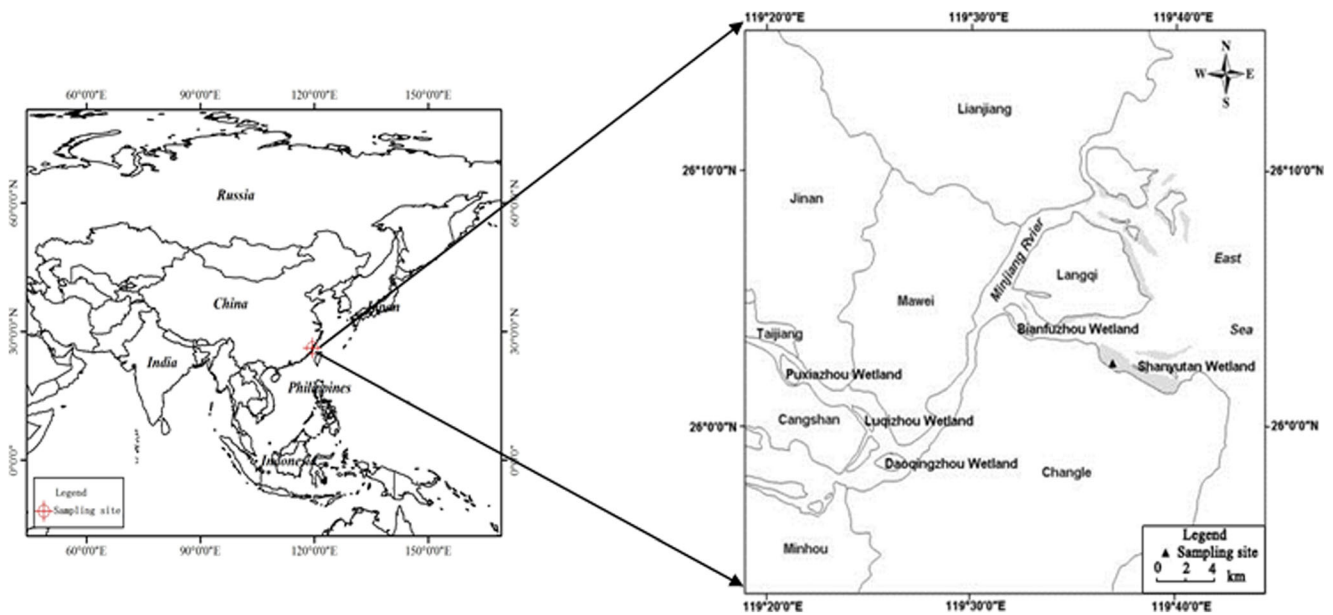
Fig. 1 Study area and sampling site (▲) in southeastern China

eliminate the possibility of methane consumption caused by carrying oxygen during the sampling process and slowly equilibrated with the atmospheric pressure for 24 h to consume the residual oxygen in incubation bottles. This precaution ensures that the soil sample is in a strictly anaerobic environment (Smemo and Yavitt 2007; Wrede et al. 2012). Three replicates were set and placed in an anaerobic incubator (YQX-II, Shanghai Yuejin Medical Equipment Factory) in a dark place using the average soil temperature in situ. Then the gas samples were taken at 0, 24, 48, 72, and 96 h, and the sample incubation bottles were gently swirled for 1–2 min before gas sampling. Each extraction was 2 ml and supplemented with the corresponding volume of oxygen-free nitrogen. Methane concentration was determined by a GC-2010 gas chromatograph (Shimadzu Scientific Instruments, Kyoto, Japan). The potential methane production rate was calculated by the methane concentration increment during the incubation time in the incubation bottles.

Measurements of Potential Methane Anaerobic Oxidation

In each collected soil sample, potential soil methane anaerobic oxidation rate was determined by placing 30 g of the fresh soil sample in a 120 ml incubation bottles and injecting 30 ml of 40 mmol l^{-1} of methane production inhibitor (BES, bromoethanesulfonate) solution (Müller et al. 1993; Hoehler et al. 1994). The water: soil ratio was 1:1 (Bergman et al. 2000). The incubation bottles were filled with oxygen-free nitrogen through a small hole in the bottle stopper to eliminate the possibility of methane consumption caused by carrying oxygen during the sampling process and slowly equilibrated

with the atmospheric pressure for 24 h to consume the residual oxygen in the incubation bottles. This precaution ensured that the soil sample was in a strictly anaerobic environment (Smemo and Yavitt 2007; Wrede et al. 2012). Then, pure Methane standard gas was injected into each incubation bottles until the concentration of methane in the incubation bottle was 10,000 µmol mol^{-1}. Three replicates were set and placed in an anaerobic incubator (YQX-II, Shanghai Yuejin Medical Equipment Factory) in a dark place using the average soil temperature in situ. The gas samples were taken at 0, 24, 48, 72, and 96 h, and the sampled incubation bottles were gently swirled for 1–2 min before gas sampling. Each extraction was 2 ml and supplemented with the corresponding volume of oxygen-free nitrogen. Methane concentration was determined by a GC-2010 gas chromatograph (Shimadzu Scientific Instruments, Kyoto, Japan). Potential methane anaerobic oxidation rate was calculated by the methane concentration decrement as the incubation time in the incubation bottles.

Measurements of Potential Methane Aerobic Oxidation

In each soil sample, potential soil methane aerobic oxidation rate was determined, following Krüger et al. (2002) and Supparattanapan et al. (2009), by placing 30 g of the fresh soil sample in a 120 ml incubation bottle and injecting 30 ml of distilled water into it. The water: soil ratio was 1:1 (Wang et al. 2010; Bergman et al. 2000). Pure methane standard gas was then injected into each incubation bottles so that the concentration of methane in the incubation bottles was 10,000 µmol mol^{-1}. Three replicates were set and incubated in a dark place using the average soil temperature in situ. The gas samples

were taken at 0, 24, 48, 72, and 96 h, and the incubation bottles were gently swirled for 1–2 min before gas sampling. Each extraction was 2 ml and supplemented with the corresponding volume of oxygen-nitrogen. Methane concentration was determined by a GC-2010 gas chromatograph (Shimadzu Scientific Instruments, Kyoto, Japan). Potential soil methane anaerobic oxidation rate was calculated by the methane concentration decrement as the incubation time in the incubation bottles.

Measurement (In Situ) of Porewater Dissolved CH_4 Concentration

Porewater was sampled in situ once each month. Three specially designed stainless steel tubes (2.0 cm inner diameter) were installed to a depth of 30 cm in each plot. Porewater samples were collected immediately after the measurements of CH_4 emission using 50-ml syringes to inject it into pre-evacuated vials (20 ml) stored in a cooling box in the field. After transporting them to the laboratory, the samples in the vials were stored at −20 °C until the analysis of CH_4 concentration. Before analysis, the vials were first thawed at room temperature and were then vigorously shaken for 5 min to equilibrate the CH_4 concentrations between the porewater and the headspace. The gas samples were taken from the headspace of the vials and analyzed for CH_4 concentration with the above gas chromatograph (Ding et al. 2003).

Determination of Methane Concentrations

Methane concentrations in the headspace air samples were determined by gas chromatography (Shimadzu GC-2010, Kyoto, Japan) using a stainless steel Porapak Q column (2 m long, 4 mm outer diameter, 80/100 mesh). A flame ionization detector (FID) was used for the determination of the methane concentrations. The operating temperatures of the column, injector and detector for the determination of methane concentrations were adjusted to 70, 200 and 200 °C. The gas chromatograph was calibrated before and after each set of measurements using 1.01, 7.99 and 50.5 μL methane L^{-1} in He (CRM/RM Information Center of China) as primary standards.

Calculation of Potential Methane Production, Anaerobic and Aerobic Oxidation, and Porewater Dissolved CH_4 Concentration

Potential methane production, anaerobic oxidation, and aerobic oxidation rates were estimated by (Wassmann et al. 1998):

$$P = \frac{dc}{dt} \cdot \frac{V_H}{W_S} \cdot \frac{MW}{MV} \cdot \frac{T_{st}}{T_{st} + T}$$

where P is the potential rate of anaerobic oxidation, aerobic oxidation and production of methane (μg^{-1} g^{-1} d^{-1}), dc/dt is

the recorded change in the mixing ratio of C (methane) in the headspace over time (mmol mol^{-1} d^{-1}), V_H is the volume of the headspace (L), Ws is the dry weight of the soil (g), MW is the molecular weight of methane (g), MV is the molecular volume (L), T is the temperature (K) and T_{st} is the standard temperature (°K).

The concentration of CH_4 dissolved in the porewater was calculated following (Ding et al. 2003):

$$C = \frac{Ch \cdot Vh}{22.4 \cdot Vp}$$

where Ch is the CH_4 concentration (μl l^{-1}) in the air sample from the vials, Vh is the volume of air in the bottle (ml), and Vp is the volume of the porewater in the bottle (ml).

Measurements of Soil Properties

Total soil porewater (collected by centrifugation at 5000 r min^{-1}) dissolved organic-C (DOC) concentration was measured using a TOC-V CPH total carbon analyzer (Shimadzu Scientific Instruments, Kyoto, Japan). Porewater (collected by centrifugation at 5000 r min^{-1}) NO_3^-, SO_4^{2-} and Cl^- concentrations were determined by ICS2100 ion chromatography (American Dionex Production, Sunnyvale, USA). Soil temperature, Eh and pH were measured with an Eh/pH/temperature meter (IQ Scientific Instruments, Carlsbad, USA), and salinity was measured using a 2265FS EC Meter (Spectrum Technologies Inc., Paxinos, USA). Total Fe content was determined by digesting fresh soil samples with 1 mol HCl L^{-1}. Ferrous ions were extracted using 1,10-phenanthroline and measured spectrometrically (Wang et al. 2012). Ferric concentration was calculated by subtracting the ferrous concentration from the total Fe concentration.

Statistical Analyses

The significance of the differences in potential methane production, anaerobic oxidation, aerobic oxidation and dissolved methane, soil variables and other properties among the seasonal variation were assessed by One-Way ANOVA. We analyzed the relationships of the potentials of soil methane production, anaerobic soil methane oxidation, aerobic soil methane oxidation and dissolved methane among themselves and with soil DOC, soil temperature, Eh, pH, salinity, soil NO_3^-, SO_4^{2-} and Cl^- concentrations, and plant biomass. Plot and time of sampling (month) were introduced into the models as random factors. If a variable was non-normally distributed, we transformed it to normalize its distribution. In concrete, soil nitrate and soil ferric concentrations were log-transformed to reach their normal distribution. We used the "*nlme*" (Pinheiro et al. 2016) and "*lme4*" (Bates et al. 2015) R packages with the "*lme*" and "*lmer*" functions to conduct the

mixed linear models. We chose the best model for each dependent variable using Akaike information criteria. We used the MuMIn (Barton 2012) R package in the mixed models to estimate the percentage of variance explained by the model. We presented the total variance explained by the model including the fixed and random factors (R^2c) and also the variability explained by only the fixed factor (R^2m) in significant relationships.

We used Principal component analyses (PCA) to assess the multiple correlations among total potential methane production, anaerobic oxidation, aerobic oxidation and dissolved methane and environmental factor and the analyzed soil variables and their relative importance in the separation of soil samples from different seasons. The PCA was performed using Statistica 6.0 (StatSoft, Inc. Tule, Oklahoma, USA).

Results

Potential Soil Methane Production, Anaerobic Methane Oxidation, Aerobic Methane Oxidation and Dissolved Methane during the one-Year Study Period

Potential soil methane production rates changed seasonally in the Shanyutan wetland of Minjiang River estuary (Figs. 2, 3), with a maximum value of 57.4 ± 7.7 µg g^{-1} d^{-1} in January 2013 and a minimum value of 4.85 ± 1.1 µg g^{-1} d^{-1} in August 2012. The annual average value was 21.1 ± 5.1 µg g^{-1} d^{-1}. In general, potential soil methane production rate was significantly higher in winter than that of the summer (Fig. 3, $P < 0.05$). However, there were not significantly different across other seasons (Fig. 3, $P > 0.05$).

Potential anaerobic soil methane oxidation rates changed seasonally in the Shanyutan wetland of Minjiang River estuary (Figs. 2, 3), with a maximum value of 41.8 ± 13.4 µg g^{-1} d^{-1} in January 2013 and a minimum value of 3.46 ± 0.97 µg g^{-1} d^{-1} in August 2012. The annual average value was 11.0 ± 3.9 µg g^{-1} d^{-1}. In general, the potential anaerobic soil methane oxidation production rate was significantly higher in winter than in spring and autumn (Fig. 3, $P < 0.05$). However, there was no significant difference across other seasons (Fig. 3, $P > 0.05$).

Potential aerobic soil methane oxidation rates changed seasonally in the Shanyutan wetland of Minjiang River estuary (Figs. 2, 3), with a maximum value of 70.2 ± 24.5 µg g^{-1} d^{-1} in January 2013 and a minimum value of 6.55 ± 1.42 µg g^{-1} d^{-1} in May 2012. The annual average value was 20.9 ± 5.8 µg g^{-1} d^{-1}. In general, potential aerobic soil methane oxidation production rate was not significantly different across the seasons (Fig. 3, $P > 0.05$).

Dissolved methane in soil porewater changed seasonally in the Shanyutan wetland of Minjiang River estuary (Figs. 2, 3), with a maximum value of 261 ± 39 µmol l^{-1} in August 2012 and a minimum value of 7.52 ± 0.37 µmol l^{-1} in January 2012. The annual average was 62.9 ± 20.6 µmol l^{-1}. In general, dissolved methane in soil porewater was significantly higher in summer than other seasons (Fig. 3, $P < 0.05$). However, there was no significant difference across other seasons (Fig. 3, $P > 0.05$).

The Relationship among Potential Soil Methane Production, Anaerobic Methane Oxidation, Aerobic Methane Oxidation and Dissolved Methane during the one-Year Study Period

The linear mixed models showed that the potential anaerobic soil methane oxidation, potential soil methane production and potential aerobic soil methane oxidation rates were positively related to each other (Table 1). The statistical models of the corresponding three relationships (including plots and time as random factors) had very high total significance ($R^2c = 0.99$, $P < 0.0001$) (Table 1). Methane concentrations in soil porewater were negatively correlated with potential soil anaerobic oxidation rates ($R^2m = 0.15$, $R^2c = 0.91$), potential soil methane production rates ($R^2m = 0.24$, $R^2c = 0.99$) and potential aerobic soil methane oxidation ($R^2m = 0.13$, $R^2c = 0.94$) (Table 1).

Seasonality in Environment Variables

Soil temperature changed seasonally in the Shanyutan wetland of Minjiang River estuary (Fig. 4), with a maximum value of 31.5 ± 0.1 °C in September 2012 and a minimum value of 12.4 ± 0.6 °C in February 2013. The annual average was 21.4 ± 2.0 °C.
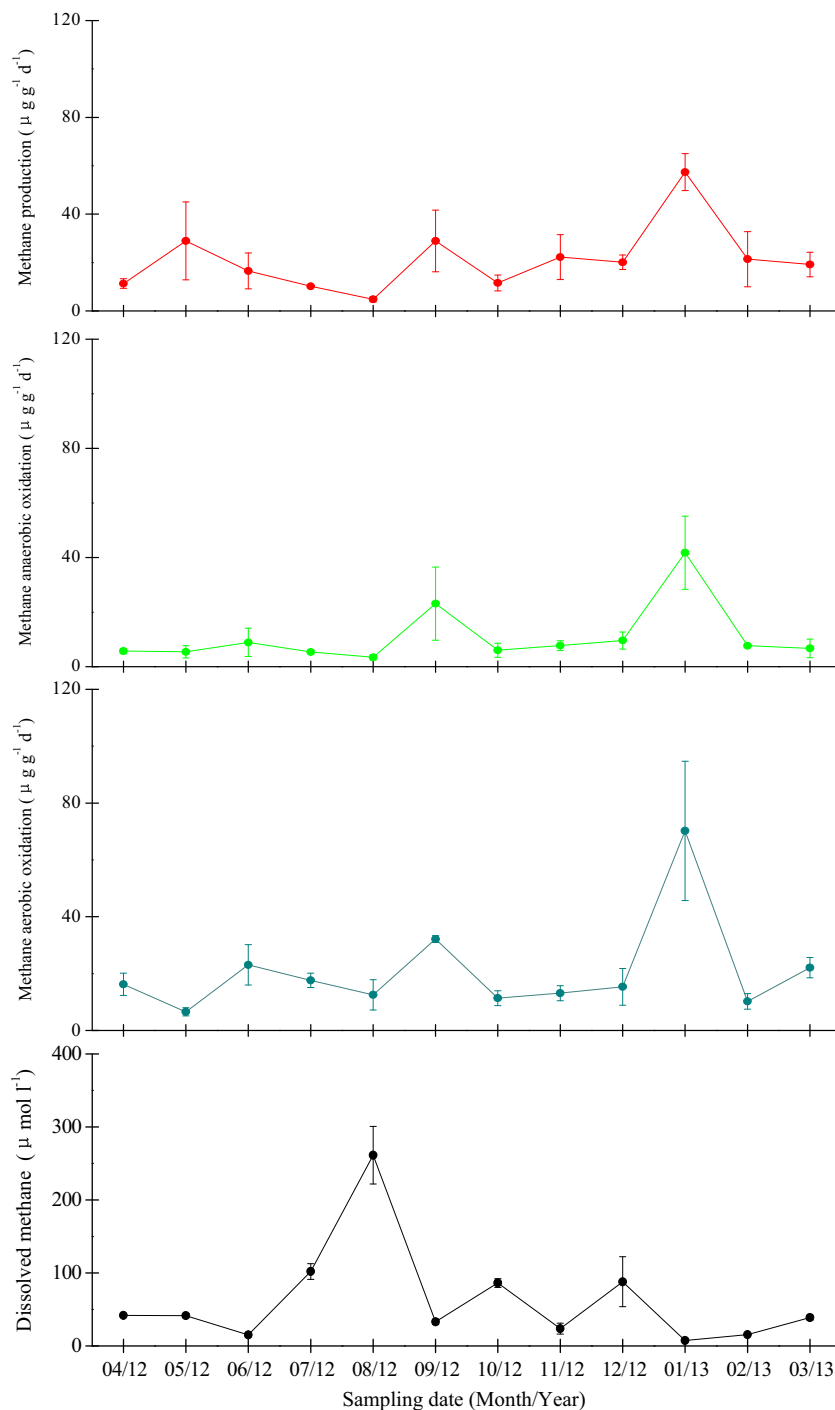
Soil ferric concentration changed seasonally in the Shanyutan wetland of Minjiang River estuary (Fig. 4), with a maximum value of 87.0 ± 9.9 mg g^{-1} in June 2012 and a minimum value of 12.2 ± 2.3 mg g^{-1} in September 2012. The annual average was 37.8 ± 6.9 mg g^{-1}.

Soil pH changed seasonally in the Shanyutan wetland of Minjiang River estuary (Fig. 4), with a maximum value of 7.62 ± 0.01 in January 2013 and a minimum value of 6.30 ± 0.02 in November 2012. The annual average was 6.72 ± 0.11.

Soil Eh changed seasonally in the Shanyutan wetland of Minjiang River estuary (Fig. 4), with a maximum value of 41.1 ± 0.7 mV in November 2012. January 2013 had the lowest value, -36.5 ± 0.7 mV. The annual average was 16.3 ± 6.5 mV.

Soil salinity changed seasonally in the Shanyutan wetland of Minjiang River estuary (Fig. 4), with a maximum value of 4.21 ± 0.71 mS cm^{-1} in April 2012. February 2012 had the lowest value, 1.91 ± 0.33 mS cm^{-1}. The annual average was 3.08 ± 0.21 mS cm^{-1}.

Fig. 2 Monthly variation of
potential soil methane production
rate, potential anaerobic soil
methane oxidation, potential
aerobic soil methane oxidation
rate, and dissolved methane
concentration in soil porewater

Dissolved sulfate in soil porewater concentration changed seasonally in the Shanyutan wetland of Minjiang River estuary (Fig. 4), with a maximum value of 362 ± 36 mg l^{-1} in December 2012 and a minimum value of 128 ± 12 mg l^{-1} in February 2013. The annual average was 227 ± 25 mg l^{-1}.

Dissolved nitrate in soil porewater concentration changed seasonally in the Shanyutan wetland of Minjiang River estuary (Fig. 4), with a maximum value of 2.46 ± 0.44 mg l^{-1} in

December 2012. In October 2012, the lowest value, 0.106 ± 0.039 mg l^{-1}, was recorded. The annual average was 0.828 ± 0.212 mg l^{-1}.

The concentration of dissolved chloridion in soil porewater changed seasonally in the Shanyutan wetland of Minjiang River estuary (Fig. 4), with a maximum value of 4595 ± 279 mg l^{-1} in November 2012. In August 2012, the lowest value, 1412 ± 92 mg l^{-1}, was recorded; the annual average was 2821 ± 283 mg l^{-1}.

Fig. 3 Seasonal values of potential soil methane production rate (**A**), potential anaerobic soil methane oxidation rate (**A**), potential aerobic soil methane oxidation rate (**A**), and dissolved methane concentration in soil porewater (**B**). Different letters indicate significant difference among seasons
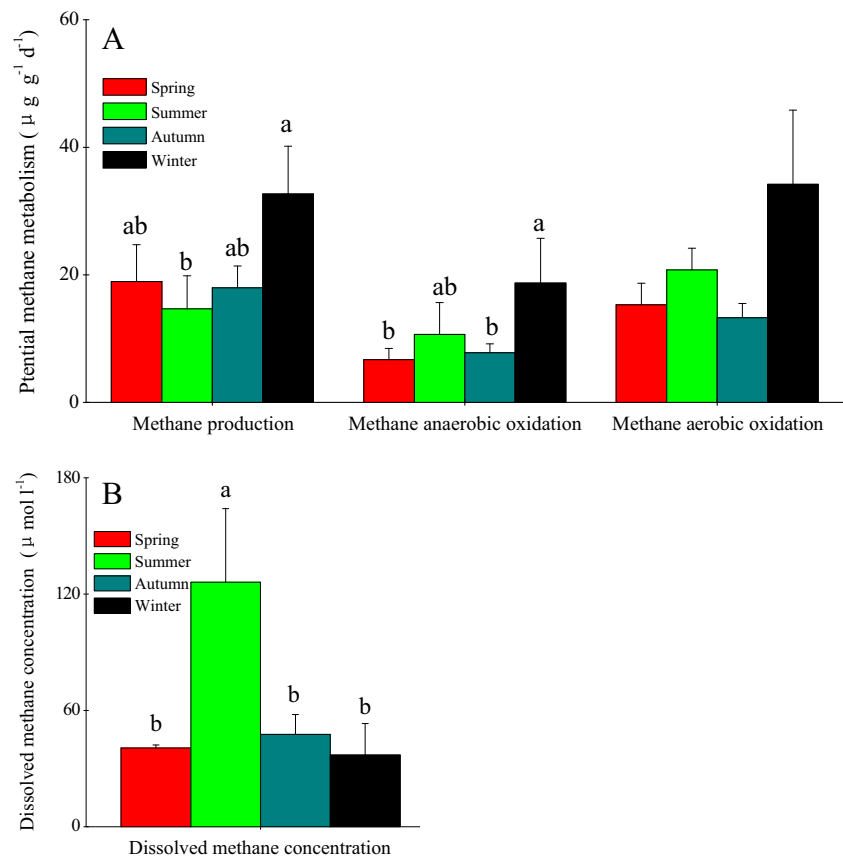
Plant biomass changed seasonally in the Shanyutan wetland of Minjiang River estuary (Fig. 4), with a maximum value of 2313 ± 657 g^{-2} in December 2012. April 2012 had the lowest value, 759 ± 320 g^{-2}. The annual average was 1462 ± 198 g^{-2}.

Air temperature changed seasonally in the Shanyutan wetland of Minjiang River estuary (Fig. 4), with a maximum value of 35.5 ± 0.0 °C in July 2012. February 2013 had the lowest value, 10.2 ± 0.1 °C, and the annual average was 23.0 ± 2.4 °C.

Potential Soil Methane Production, Anaerobic Methane Oxidation, Aerobic Oxidation Rates, Dissolved Methane, and their Relationships with Soil Properties

The linear mixed models showed that potential soil methane production rates were positively related to soil pH (R^2m = 0.15, R^2c = 0.90) and negatively related to soil Eh (R^2m = 0.15, R^2c = 0.90) and temperature (R^2m = 0.13, R^2c = 0.90) (Table 1). Potential anaerobic soil methane oxidation rates were positively related to soil pH (R^2m = 0.14, R^2c = 0.99) and negatively with soil Eh (R^2m = 0.15, R^2c = 0.99) (Table 1). Finally, methane concentration in soil porewater was positively related to soil temperature (R^2m = 0.32, R^2c = 0.99) (Table 1).

The PCA analysis was completely consistent with previous commented univariate analyses. Soil samples collected in summer were located across the PC1 axis coinciding with higher porewater methane concentrations, higher soil and air temperature and soil Eh and lower soil pH and potential soil methane production and oxidation rates, both anaerobic and aerobic (Fig. 5). The contrary patterns were related to soil samples collected in winter that were placed on the other side of the PC1 axis (Fig. 5).

Discussion

Seasonal Variation of Potential Methane Production, Anaerobic Oxidation, Aerobic Oxidation and Dissolved Methane

Potential soil methane production rate was higher in winter than in summer. In winter, there are lots of plant litter input. The soil carbon concentration which is the most important substrate for methane production thus increases (Yagi and Minami 1990), thereby promoting soil methane production (Van der Gon and Neue 1995). Moreover, the optimum temperature of methane production is about 20 °C (Wagner and Pfeiffer 1997). In our study, the average temperature was 14.4 and 29.2 °C in winter and summer, respectively, thus more

Table 1 Significant observed relationships of the potentials of soil methane production, soil methane anaerobic oxidation and soil methane aerobic oxidation among them and with soil properties. Plot and time of sampling (month) were introduced in the models as random factors

model <− lme(Variable~fixed factor, data = dades, random = ~1|plot/time,method = "REML")

Variable	Fixed factor	Fixed factor statistics	Model statistics (R^2m = fixed factor, R^2c = fixed + random factor)
Potential soil anaerobic CH_4 oxidation	Potential soil CH_4 production	Estimates = 0.578 F = 43.7 $P < 0001$	R^2m = 0.56 R^2c = 0.99
Potential soil aerobic CH_4 oxidation	Potential soil CH_4 production	Estimates = 0.707 F = 21.2 $P < 0001$	R^2m = 0.38 R^2c = 0.99
Potential soil aerobic CH_4 oxidation	Potential soil anaerobic CH_4 oxidation	Estimates = 1.14 F = 49.0 $P < 0001$	R^2m = 0.58 R^2c = 0.99
CH_4 pore-water soil concentration	Potential soil CH_4 production	Estimates = −0.029 F = 11.8 $P = 0.0016$	R^2m = 0.24 R^2c = 0.91
CH_4 pore-water soil concentration	Potential soil anaerobic CH_4 oxidation	Estimates = −0.42 F = 6.31 $P = 0.017$	R^2m = 0.15 R^2c = 0.91
CH_4 pore-water soil concentration	Potential soil aerobic CH_4 oxidation	Estimates = −0.46 F = 5.09 $P = 0.031$	R^2m = 0.13 R^2c = 0.94
Potential soil anaerobic CH_4 oxidation	Soil pH	Estimates = 6.17 F = 5.47 $P = 0.026$	R^2m = 0.14 R^2c = 0.99
Potential soil anaerobic CH_4 oxidation	Soil Eh	Estimates = −0.016 F = 5.89 $P = 0.021$	R^2m = 0.15 R^2c = 0.99
Potential soil CH_4 production	Soil pH	Estimates = 5.80 F = 6.55 $P = 0.015$	R^2m = 0.15 R^2c = 0.90
Potential soil CH_4 production	Soil Temperature	Estimates = −0.045 F = 5.87 $P = 0.021$	R^2m = 0.13 R^2c = 0.90
Potential soil CH_4 production	Soil Eh	Estimates = −0.015 F = 6.76 $P = 0.014$	R^2m = 0.15 R^2c = 0.90
CH_4 pore-water soil concentration	Soil Temperature	Estimates = 0.082 F = 16.4 $P < 0001$	R^2m = 0.32 R^2c = 0.99

closely to the optimum temperature for soil methane production in winter than in summer. Moreover, in summer, the plant growth was higher than in other seasons, and more O_2 was released into the soil, generating soil redox conditions which were not proper for methane production. Furthermore, in Fujian province, the acid rain was high, and summer was the main rainy season, so the soil pH decreased in summer thereby inhibiting methane production. In contrast, winter was the dry season with relatively higher pH, favoring methane production. In our study, the linear mixed models showed that potential soil methane production rates were positively related to soil pH and negatively related to soil Eh and temperature.

Potential anaerobic soil methane oxidation rates were also higher in winter than in spring and autumn, also than in summer, although not significantly different. As commented on Fujian province, acid rain was high, and summer was the main rainy season, so in summer the soil pH decreases thereby inhibiting the microbial activity of anaerobic methane oxidation. In contrast, winter was the dry season with relatively higher pH which favoured the growth of microbes involved in anaerobic methane oxidation. In our study, the linear mixed models showed that potential anaerobic soil methane oxidation rates were positively related to soil pH, supporting these comments. Moreover, Nauhaus et al. (2002) showed that the optimum temperature for anaerobic methane oxidation is 4–16 °C. In our
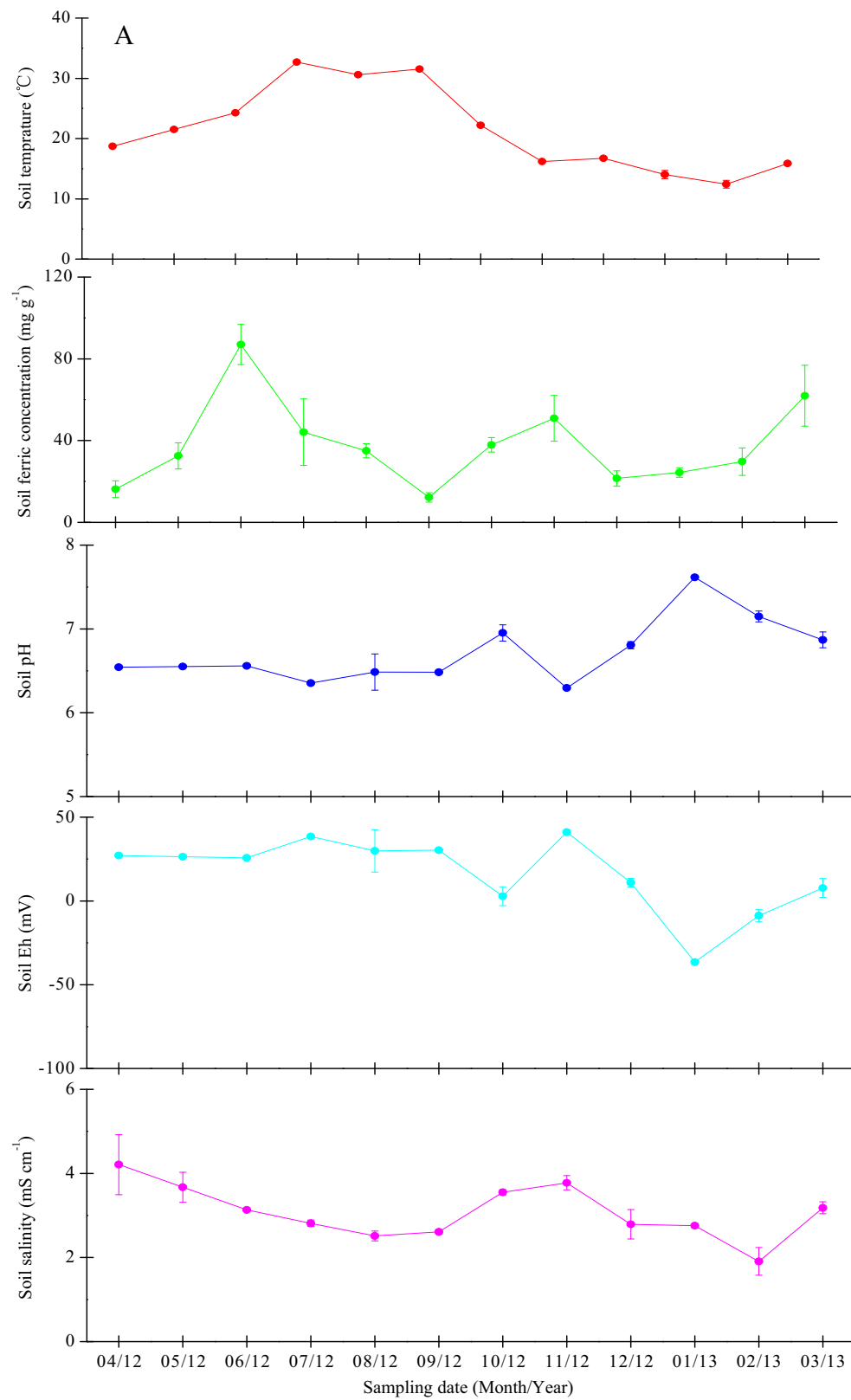
Fig. 4 Monthly variation of soil properties (**A**), porewater properties (**B**), plant biomass (**C**) and air temperature (**D**)
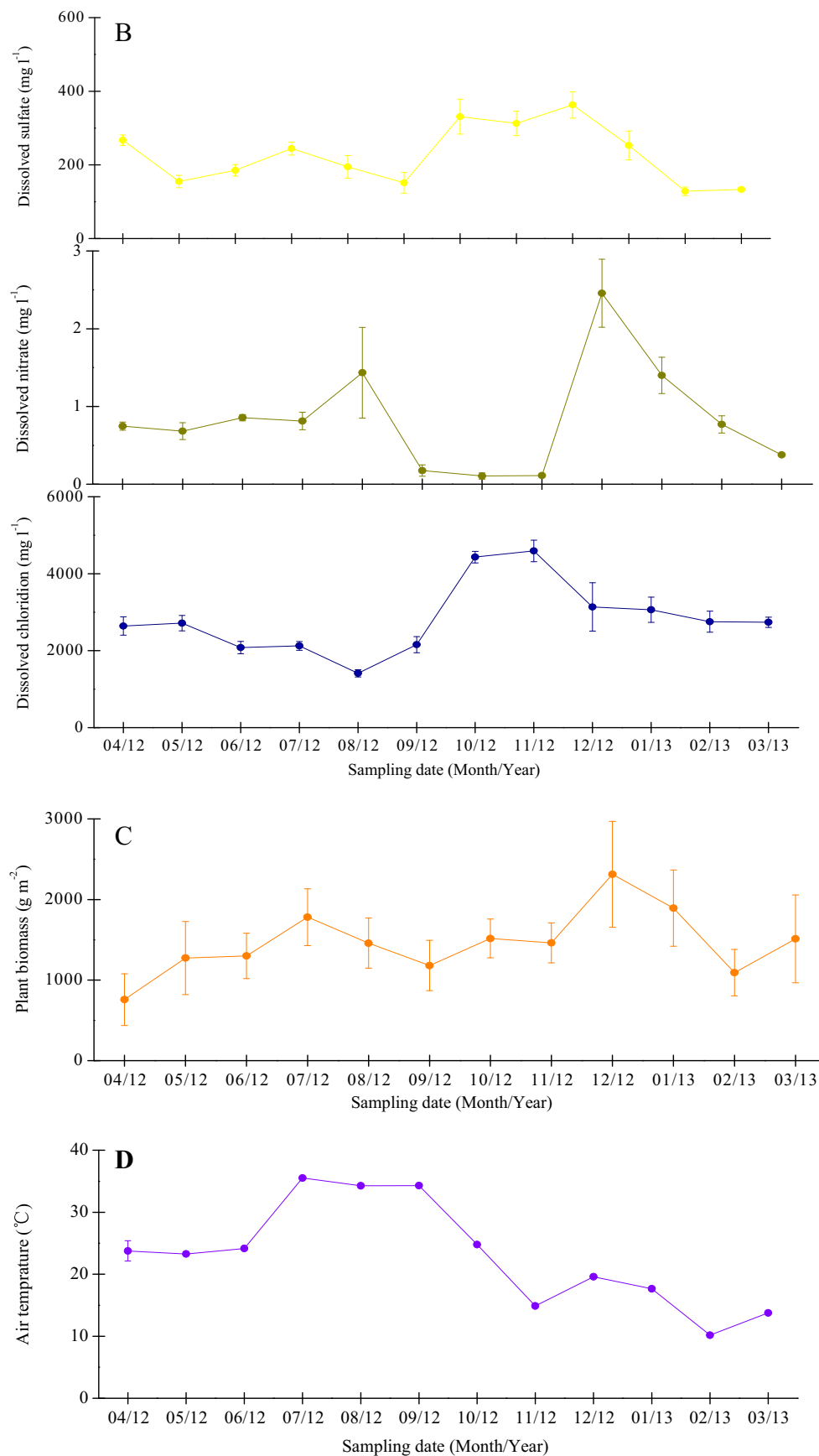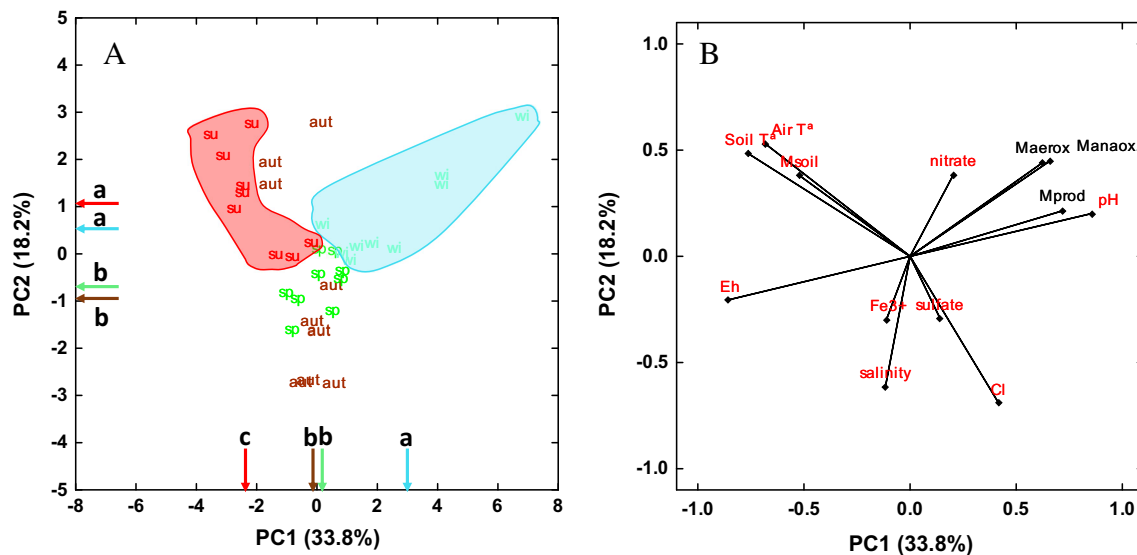
Fig. 4 (continued)

Fig. 5 Principal component analyses (PCA) to observe the multiple correlations among potential anaerobic soil methane oxidation, potential soil methane production, potential aerobic soil methane oxidation and the environmental factors and the analyzed soil variables. We represented the position of different cases (soil samples) (**A**) and loads of the commented variables (**B**) in the layout generated by the two first PCA axes (explaining together a 52.0% of the total variance). Ait T^a = Air temperature, Cl = concentration of Cl- in soil, Eh = soil potential redox, Fe^{3+} = Soil Fe^{3+} concentration, Maerox = Potential soil methane aerobic oxidation, Manaox = Potential soil methane anaerobic oxidation, Mprod = Potential soil methane production, Msoil = concentration of methane in soil porewater, pH = soil pH, salinity = soil salinity, Soil T^a = soil temperature, sulfate = soil sulfate concentration, aut = autumn, su = summer, sp. = spring, wi = winter

study, the average seasonal temperatures were 18.7, 29.2, 23.3 and 14.4 °C for spring, summer, autumn and winter respectively. Winter thus proved to be the most suitable season for anaerobic methane oxidation.

Potential aerobic soil methane oxidation rate was not significantly different among the seasons. However, there was a trend of higher values in winter than in the other seasons. The reason would be the same as for anaerobic methane oxidation: winter is the dry season with relative higher soil pH. In our study, the linear mixed models showed that potential aerobic soil methane oxidation rates were also positively related to soil pH. Moreover, Dasselaar et al. (1998) found that temperature promotion of the aerobic methane oxidation was higher when the temperature was 4~12 °C than when it was 12~18 °C. In our study, the temperatures closer to 12 °C were those of winter.

Dissolved methane in soil porewater was instead higher in summer than in the other seasons, which had no significant differences among them. The dissolved methane in soil porewater resulted from many factors, such as methane production, oxidation and transportation, etc. The lower summer anaerobic and aerobic methane oxidation were likely the most determining factors of these higher values of dissolved methane in summer.

The soils with highest soil pH and lowest Eh were those with the highest potentials of methane production and anaerobic oxidation. However, the most interesting result was that of soil samples with the highest soil pH and lowest Eh and soil samples with the highest potentials of aerobic

methane oxidation. Consistent with these results, Kettunen et al. (1999) also observed that the maximum potential capacity of aerobic methane oxidation was higher in soils below than above table level. Similar results have also been observed in boreal pine fen areas (Saarnio et al. 1997).

These results thus suggested a buffer effect in the methane balance in wetland areas. Environmental and soil conditions favoring methane production are also more favorable for anaerobic methane oxidation during the same circumstances and also in drier periods, for aerobic methane oxidation. In fact, alternation between wet-dry periods related to wetland source-sink of methane has been widely observed (Juutinen et al. 2003; Knorr et al. 2008; Brown et al. 2014; Goodrich et al. 2015). The fact that same soil conditions are favorable for methane production and oxidation was observed in both flooded (anaerobic) and dry (aerobic) periods. This observation warrants future research to corroborate this possibly general pattern.

Relationship among Potential Methane Production, Anaerobic Oxidation, Aerobic Oxidation and Dissolved Methane

Potential anaerobic methane oxidation and potential methane production showed a very significant positive correlation. This pattern has been observed previously in tropical and boreal wet soils and peatlands where these two variables correlated significantly, in agreement with our results (Smemo and Yavitt 2011; Blazewicz et al. 2012). The relationship between

anaerobic methane oxidation and methane production was mainly related to the functional microbial association. Anaerobic methanotroph (ANME) *archaea,* which can also participate in the anaerobic methane oxidation, is the main microorganism involved in methane production (Alperin and Hoehler 2009; Lloyd et al. 2011). Methane-producing *archaea* can oxidize methane as observed in pure culture experiments (Moran et al. 2005; Joye and Samarkin 2009; Roberts and Aharon 1994). Also, there was a significant positive correlation between anaerobic and aerobic methane oxidation in this study. Recent studies have demonstrated that aerobic and anaerobic methane oxidation bacteria can coexist in the same places, suggesting that the proportion of different species can depend on the availability of oxygen and methane and also that diverse microbial activity is important to sustain methanotrophic activity (Siniscalchi et al. 2017). Eller et al. (2005) observed the co-occurrence of aerobic and anaerobic methane process in same soil samples and water columns. Moreover, potential aerobic methane oxidation and production processes showed a very significant positive correlation as expected with methane being the substrate of methane oxidation (Nesbit and Breitenbeck 1992). However, negative relationships between methane concentrations in soil porewater and the studied potential methane production rates and also potential soil methane oxidation, both in anaerobic and aerobic conditions, were also observed. These results suggest that methane production is not the most important factor controlling dissolved methane in soil porewater. However, porewater-dissolved methane was directly limited by anaerobic and aerobic methane oxidation in Minjiang estuarine wetland. These results were not in agreement with the fact that methane storage is the key factor in the oxidation of methane in coastal sulphate-rich marine sediments (Nauhaus et al. 2002; Treude et al. 2005; Orcutt et al. 2005). This possible explanation was consistent with the linear mixed models showing the inverse relationships between methane present in porewater and the potential soil capacity for methane production and oxidation.

Conclusions

1. Potential methane production, anaerobic oxidation and aerobic oxidation were all shown to be higher in winter than other seasons. However, the dissolved methane in soil porewater was higher in summer than other seasons.
2. The concentration of soil pH and Eh are the studied factors with stronger relationships with potential soil methane production and anaerobic and aerobic oxidation rates. Thus, this showed strong relationships among the different soil metabolic methane processes and the basic potential chemical activities of soils.
3. The positive correlation between methane production, anaerobic methane oxidation and aerobic methane oxidation suggested that at least some of the soil conditions and overall set of microorganisms communities that favor methane production also favor its oxidation.
4. The negative relationships between methane concentrations in soil porewater and the potential soil of methane production and oxidation in anaerobic and aerobic conditions suggest that the higher the soil potential to produce methane, the higher the potential soil capacity to oxidize methane in aerobic and anaerobic conditions.

Table 1. Significant relationships observed among the potentials of soil methane production, anaerobic soil methane oxidation and aerobic soil methane oxidation and with soil properties. Plot and time of sampling (month) were introduced in the models as random factors.

Compliance with Ethical Standards

Conflicts of Interest   The authors declare no conflicts of interest.

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