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Intermittent irrigation changes production, oxidation, and emission of CH_4 in paddy fields determined with stable carbon isotope technique

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ABSTRACT

Intermittent irrigation is an important option for mitigating CH_4 emissions from paddy fields. In order to better understand its controlling processes in CH_4 emission, CH_4 fluxes, CH_4 production and oxidation potentials in paddy soils, and ^{13}C-isotopic signatures of CH_4 were observed in field and incubation experiments. The relative contribution of acetate to total CH_4 production (f_{ac}) and fraction of CH_4 oxidized (f_{ox}) in the field was also calculated using the isotopic data. At the beginning of the rice season, the theoretical ratio of acetate fermentation: H_2/CO_2 reduction = 2:1 was reached, however, in the late season H_2/CO_2-dependent methanogenesis became dominant. Compared to continuous flooding, intermittent irrigation significantly reduced CH_4 production potential and slightly decreased f_{ac}-value, indicating methanogens, particularly acetate-utilizing methanogens, were inhibited. CH_4 oxidation was very important, especially in paddy fields under intermittent irrigation where 19–83% of the produced CH_4 was oxidized. Intermittent irrigation enhanced CH_4 oxidation potential slightly and raised f_{ox}-value significantly relative to continuous flooding. Intermittent irrigation significantly decreased CH_4 flux creating a more positive $\delta^{13}C$-value of emitted CH_4 by 12–22‰. A significant negative correlation was found between CH_4 fluxes and values of $\delta^{13}CH_4$ suggesting that the less the CH_4 oxidation, the higher the CH_4 emission, and the lower the $\delta^{13}C$-value of emitted CH_4. Collectively, the findings show that intermittent irrigation reduced the seasonal CH_4 production potential by 45% but increased the fraction of CH_4 oxidized by 45–63%, thus decreasing the seasonal CH_4 emission from the paddy fields by 71%, relative to continuous flooding.

1. Introduction

Methane (CH_4), one of the most important greenhouse gases, reached 1774 ppb in 2005 (IPCC, 2007). Irrigated paddy fields are a major anthropogenic source of atmospheric CH_4. The global CH_4 emission from the paddy fields was estimated to be 31–112 Tg CH_4 year^{-1}, accounting for about 5–19% of the total CH_4 emissions (IPCC, 2007). Thus, options for its mitigation were studied intensively by scientists all over the world. A series of useful options for decreasing CH_4 emissions from paddy fields have been confirmed by considerable field data, such as planting winter crops or draining the field dry for winter fallow, adopting intermittent irrigation during the rice season, incorporating organic manure into the field in winter seasons rather than in rice seasons, using compost or chemical fertilizers instead of green manure and other fresh organic manure, and applying CH_4 production inhibitors (Cai et al., 2009). However, both the mechanism of CH_4 emission and its controlling processes are not completely clear.

Intermittent irrigation, an episode of drainage for several days in the middle of the rice season and drying-wetting alternation during the following period, is recognized as an important cultivation practice to mitigate CH_4 emission in rice production (Yan et al., 2005, 2009). Paddy fields under intermittent irrigation emit significantly less CH_4 than paddy fields under continuous flooding during the rice season (Yagi et al., 1996; Xu et al., 2000; Zou et al., 2005; Li et al., 2007, 2011; Zhang et al., 2009a). Moreover, intermittent irrigation has been commonly adopted in China not only for inhibiting ineffective tillers, removing toxic substances, and improving roots activities (Gao et al., 1992), but also for cutting down the cost of field management and saving the water resources. Numerous studies reported CH_4 emissions from rice fields were affected by intermittent irrigation through flux measurements in situ (Sass et al., 1992; Yagi et al., 1996; Cai et al., 2000; Zou et al., 2005; Li et al., 2007, 2011; Zhang et al., 2009a). Further investigations should be conducted on

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its influence on the processes of CH_4 emission related to both CH_4 production and CH_4 oxidation in the paddy fields.

CH_4 production is the terminal step in anaerobic microbial decomposition of organic matter. Before being released to the atmosphere, the CH_4 produced in the field is oxidized to a large extent by methanotrophs in the rhizosphere and at the soil–water interface (Conrad and Rothfuss, 1991; Groot et al., 2003). However, production and oxidation of CH_4 in paddy fields occurs nearly at the same time, and their processes are quite complicated. So, it is difficult to distinguish the effect of CH_4 production from that of CH_4 oxidation on CH_4 emission. Intermittent irrigation possibly inhibits methanogenesis while accelerating methanotrophy, thus decreasing CH_4 emission from rice field. However, it is still not yet clear how intermittent irrigation affects methanogenesis and methanotrophy in the field separately, and consequently the total CH_4 emission. Recently, the stable carbon isotope technique is considered to be a good method for estimating relative contributions of CH_4 production and CH_4 oxidation to the total CH_4 emission (Tyler et al., 1997; Krüger et al., 2002). By determining carbon isotope compositions of CH_4, CO_2 and acetate, and other parameters such as isotopic fractionation factors $\varepsilon_{acetate}$, $\alpha_{CO2/CH4}$, $\varepsilon_{transport}$ and α_{ox}, contributions of the pathways of methanogenesis and the fraction of CH_4 oxidized in rice fields can be calculated separately and reliably (Sugimoto and Wada, 1993; Tyler et al., 1997; Bilek et al., 1999; Conrad et al., 2002; Krüger et al., 2002; Nakagswa et al., 2002; Krüger and Frenzel, 2003; Fey et al., 2004; Conrad and Klose, 2005; Zhang et al., 2011a). Nevertheless, these studies focused mainly on continuously flooded paddy fields, and little on paddy fields under intermittent irrigation or methanogenic pathways, including the fraction of CH_4 oxidized during the rice season.

CH_4 fluxes, CH_4 production and oxidation potentials in paddy soils, and ^{13}C-isotopic signatures were measured through field and incubation experiments during the 2007 rice season in order to investigate the effects of intermittent irrigation on production, oxidation and emission of CH_4 in paddy fields, especially on pathways of methanogenesis, fraction of oxidized CH_4, and $\delta^{13}C$-value of emitted CH_4.

2. Materials and methods

2.1. Experimental site and design

CH_4 flux was monitored in a field located at the Baitu Town, Jurong City, Jiangsu Province, China (31°58′N, 119°18′E) in 2007. The soil of the paddy field is classified as Typic Haplaquepts, and its initial properties are pH (H_2O) 6.9, organic C 11.9 g kg^{-1}, total N 1.5 g kg^{-1}, and $\delta^{13}C$-value of soil carbon −26.8‰.

After the harvest of winter wheat, the stubbles were incorporated to the soil with straw transported to outside of the field. Then, two treatments of water management, i.e. Treatment II (intermittent irrigation) and Treatment CF (continuous flooding) were carried out, each having three replicates, in the paddy field during the rice-growing season. The plots of Treatment II were initially flooded on June 18, drained on July 26 for an 8-day-long midseason aeration, re-flooded for 15 days, and then subjected to drying-wetting alternation (with a cycle of 3-day drying and 5 day-wetting) until October 3, while the plots of Treatment CF were kept flooded from June 18 to October 3. When the crop was almost ready for harvest, all the plots were finally drained on October 4. The rice used in the experiment was "Oryza sativa L. Huajing 3" and was sown into the nursery bed on May 17, transplanted into the plots on June 19, and harvested on October 22. Urea was applied at a rate of 300 kg N ha^{-1}, 50% as basal fertilizer on June 19, 25% as tillering fertilizer on July 11, and 25% as panicle fertilizer on August 12, and Ca $(H_2PO_4)_2$ (450 kg ha^{-1}) and KCL (225 kg ha^{-1}) was also applied together with urea as basal fertilizer.

2.2. Field sampling and measurements

CH_4 flux was monitored using the static chamber technique. The flux chambers (0.5 m long × 0.5 m wide × 1 m high), made of plexiglass, covered six hills of rice plants each in the paddy field. All the chambers were equipped with a fan inside to ensure complete gas mixing. Plastic bases for the chambers were installed before rice transplantation in all the plots, and kept there until rice harvest. Removable wooden boardwalks (2 m long) were set up at the beginning of the rice season to avoid soil disturbances during sampling and measuring. To measure the flux, gas samples were generally collected once every 4–7 days, and four gas samples from each chamber were collected using 18 ml vacuum vials at 15 min intervals between 09:45 and 10:30 in the morning on each sampling day. During the midseason aeration and the following reflooding periods, CH_4 flux was observed at an interval of 1–3 days. CH_4 flux was determined from the slope of linear regression, and expressed in mg CH_4 m^{-2} h^{-1}. To determine carbon isotope composition of the gas, samples were taken at 15–30 day intervals, and only two gas samples were collected using 500 ml bags (aluminium foil compound membrane, Delin gas packing Co., Ltd, Dalian, China) with a small battery-driven pump. The first sample was taken after the chamber was closed for 3–5 min, and the second at the end of the 2 h closure period. $\delta^{13}C$-values of the emitted CH_4 were calculated using the following equation:

$$S = [(B \times b) - (A \times a)]/(B - A) \tag{1}$$

where A and B stands for CH_4 concentration ($\mu l\ L^{-1}$) in the samples at the beginning and at the end, respectively, and a and b for corresponding $\delta^{13}CH_4$-values (‰) of the gas samples.

When CH_4 flux was monitored simultaneously, soil redox potential (Eh) at a depth of 0.1 m was measured, using Pt-tipped electrodes (Hirose Rika Co. Ltd., Japan), which were inserted into the soil of each plot to a depth of 0.1 m in three replicates and kept there throughout the whole observation period, and an oxidation-reduction potential meter with a reference electrode (Toa PRN-41). Depth of the water layer in the field was measured manually with a ruler, and soil temperature at 0.1 m depth was measured with a hand-carried digital thermometer (Yokogawa Meter and Instruments Corporation, Japan).

Soil cores from the top soil layer (0–0.15 m), 3 in each plot, were collected at about 15–30-day intervals (Zhang et al., 2011b). Then, the 3 soil cores were mixed together. Of the mixture, two soil samples, around 50 g (dry weight) each, were promptly taken and transferred into two 250-ml Erlenmeyer flasks separately. Soil samples in the flasks were turned into slurries with N_2-flushed deionized sterile water added to a soil/water ratio of 1:1. During the whole process, N_2 was constantly flushed through the samples to remove O_2 and CH_4, and the flasks containing these samples were then sealed for anaerobic incubation. Other flasks with air headspace were sealed directly for aerobic incubation. All the flasks were sealed with rubber stoppers fitted with silicon septum that allowed sampling of headspace gas. Finally, they were stored under N_2 at 4 °C and transported back to the lab as soon as possible for further analysis. A small portion of the soil mixture was taken for further determination of soil mineral N concentration.

2.3. Laboratory experiment

CH_4 production potential in the soil slurries was determined as described in Zhang et al. (2011b). The flasks of slurry, with air