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Variations of Stable Carbon Isotopes of CH₄ Emission from Three Typical Rice Fields in China

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ABSTRACT

Little is known about the stable carbon isotopes of methane (CH₄) emitted ($\delta^{13}\text{CH}_{4\text{emitted}}$) from permanently flooded rice fields and double rice-cropping fields. The CH₄ emission and corresponding $\delta^{13}\text{CH}_{4\text{emitted}}$ under various field managements (mulching, water regime, tillage, and nitrogen (N) fertilization) were simultaneously measured in three typical Chinese rice fields, a permanently flooded rice field in Ziyang City, Sichuan Province, Southwest China, a double-rice cropping field in Yingtan City, Jiangxi Province, Southeast China, and a rice-wheat rotation field in Jurong City, Jiangsu Province, East China, from 2010 to 2012. Results showed different seasonal variations of $\delta^{13}\text{CH}_{4\text{emitted}}$ among the three fields during the rice-growing season. The values of $\delta^{13}\text{CH}_{4\text{emitted}}$ were negatively correlated with corresponding CH₄ emissions in seasonal variation and mean, indicating the importance of CH₄ production, oxidation, and transport associated with isotopic fractionation effects to the $\delta^{13}\text{CH}_{4\text{emitted}}$. Seasonal variations of $\delta^{13}\text{CH}_{4\text{emitted}}$ were slightly impacted by mulching cultivation, tillage, and N application, but highly controlled by drainage. Meanwhile, tillage, N application, and especially mulching cultivation had important effects on seasonal mean CH₄ emissions and corresponding $\delta^{13}\text{CH}_{4\text{emitted}}$, with low emissions accompanied by high values of $\delta^{13}\text{CH}_{4\text{emitted}}$. Seasonal mean values of $\delta^{13}\text{CH}_{4\text{emitted}}$ from the three fields were similar, mostly ranging from -60% to -50% , which are well in agreement with previously published data. These demonstrated that seasonal variations of $\delta^{13}\text{CH}_{4\text{emitted}}$ mainly depended on the changes in CH₄ emission from rice fields and further indicated the important effects of methanogenic pathways, CH₄ oxidation, and CH₄ transport associated with isotope fractionation effects influenced by field managements on $\delta^{13}\text{CH}_{4\text{emitted}}$.

Key Words: CH₄ oxidation, CH₄ transport, isotope fractionation, methanogenic pathways, mulching, N application, tillage

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INTRODUCTION

The globally averaged mole fraction of atmospheric methane (CH₄) has recently reached a new high of 1833 ± 1 ppb (WMO, 2015). Rice-based ecosystem is a significant source of atmosphere CH₄, which contributes to 11% of the total anthropogenic emissions (Ciais, 2013). China is the largest rice producer, and CH₄ emission from Chinese rice fields is estimated to be 6.4 Tg year^{-1} (Zhang *et al.*, 2014), accounting for about 18% of the global CH₄ budget from rice fields (Ciais, 2013). Great efforts have been made in the past decades to estimate CH₄ emissions from rice fields at a national scale though uncertainties still exist due to variations in rice cultivation, field management, and climate (Cai *et al.*, 2009 and references therein).

Rice-cropping systems in China include single and double rice-cropping systems. The single rice-cropping

systems, mainly consisting of summer rice-winter wheat (rice-wheat) and summer rice-winter fallow (rice-fallow) rotations, cover about 60% of Chinese total rice cultivated areas (EBCAY, 2014). The rice-wheat rotation system is mostly adopted in the Yangtze River Delta Region in China, where the total nitrogen (N) application is as high as 250–300 kg N ha⁻¹ per rice season, and such a high N application rate has been found to decrease CH₄ production and emission (Zhang *et al.*, 2010). As a special kind of rice-fallow rotation system traditionally flooded year-round, permanently flooded rice fields contribute to 45% of total CH₄ emission just with 12% of cultivated area in China (Cai, 1999). Recently, significant decreases in CH₄ emission have been found in this kind of field using plastic film mulching cultivation (Zhang *et al.*, 2016). In contrast, approximately 40% of double rice-cropping fields produce nearly 35% of the total rice grain yields (EBCAY,

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2014) and contribute to about 50% of the total CH₄ emission from rice fields in China (Zhang *et al.*, 2011). Luckily, soil tillage in winter fallow season is considered to be an effective option for mitigating CH₄ emission (Zhang *et al.*, 2016). Observations on CH₄ emission from typical rice fields in China are considerable (Cai *et al.*, 2000; Khalil *et al.*, 2008a, b). However, measurements of corresponding stable carbon isotopes ($\delta^{13}\text{C}$) of CH₄ emission ($\delta^{13}\text{CH}_4$) in these fields, especially in permanently flooded rice fields and double rice-cropping fields, and their responses to various field managements such as N fertilization, cultivation mode, and tillage are scarcely documented.

The stable carbon isotopic composition of CH₄ in agricultural ecosystems can improve our understanding of the relative source strengths of atmospheric CH₄. It also can provide constraints for the global CH₄ budget as microbe-produced CH₄ has a significantly different isotopic signal than CH₄ from other sources. For example, Chanton *et al.* (2000) concluded that $\delta^{13}\text{C}$ values of the CH₄ from combustion and biomass burning were between -9% and -30% , whereas those from landfills, rice fields, and wetlands were between -50% and -70% . On the other hand, the relative contributions of acetate and dihydrogen (H₂)/carbon dioxide (CO₂) to total methanogenesis as well as the processes of CH₄ oxidation and transport can be differentiated by measuring the stable carbon isotopes from different compartments of rice fields.

Based on a field experiment, apparent differences in $\delta^{13}\text{C}$ values of the carbon substrates and the produced, oxidized, and emitted CH₄ could be observed during the CH₄ emission (Zhang *et al.*, 2013). It is reported that $\delta^{13}\text{C}$ of CH₄ emitted ($\delta^{13}\text{CH}_{4\text{emitted}}$) is regulated by the integrated effects of CH₄ production, oxidation, and transport of rice fields (Tyler *et al.*, 1994; Bergamaschi, 1997; Marik *et al.*, 2002). A better understanding of the processes responsible for determining isotopic fractionation during the production, oxidation, and transport of CH₄ will provide better constraints on our estimates of the sources and sinks in the global CH₄ budget.

In this study, therefore, we simultaneously measured CH₄ fluxes and corresponding $\delta^{13}\text{CH}_{4\text{emitted}}$ from typical Chinese rice fields for two years. The objectives were: 1) to report the measurements of $\delta^{13}\text{CH}_{4\text{emitted}}$ from the typical rice fields in China; 2) to reveal the seasonal variations of $\delta^{13}\text{CH}_{4\text{emitted}}$; and 3) to analyze the effects of different field managements on $\delta^{13}\text{CH}_{4\text{emitted}}$. This is an important attempt to increase carbon isotopic data set in order to reasonably

estimate CH₄ emission from Chinese rice fields and indirectly evaluate the importance of CH₄ production, oxidation, and transport to CH₄ emission.

MATERIALS AND METHODS

Experiment description

Three typical Chinese rice fields with different climatic conditions, field managements, and soil properties were chosen in this study: a permanently flooded rice field, a double-rice cropping field, and a rice-wheat rotation field (Table I). The permanently flooded rice field is located in Ziyang City, Sichuan Province, Southwest China, and the soil of this field is derived from purple shale. Two treatments (traditional and mulching cultivation) were established: rice grown on permanently flooded soil as the traditional cultivation treatment and rice grown on non-flooded (without water layer) ridges that were mulched with transparent plastic film (0.004 mm in thickness) with the ditches being waterlogged except for the aeration period as the mulching cultivation treatment. Nitrogen fertilizer was split into two applications in traditional cultivation, but applied on the ridges as basal fertilizer in mulching cultivation. The plastic film was mulched on the ridge after N fertilization.

The double-rice cropping field is located in Yingtan City, Jiangxi Province, Southeast China, and the soil of the field is classified as a Typical Haplaquept (Soil Survey Staff, 1975). Two treatments (tillage and no-tillage) were performed: rice stubbles (about 30 cm long) incorporated into the soil immediately after late-rice harvest as the tillage treatment and rice stubbles not incorporated into the soil until just prior to rice transplanting in the following early-rice season as the no-tillage treatment. After early-rice harvest, rice straw and residues were all transported away for late-rice transplanting. Water management in both seasons was intermittent irrigation, *i.e.*, continuous flooding in the beginning, followed by a 7–10 d midseason aeration, then dry/wet alternation, and final drainage out before harvest.

The rice-wheat rotation field is located in Jurong City, Jiangsu Province, East China, and the soil is classified as a Typic Haplaquept (Soil Survey Staff, 1975). Two treatments (N300 and N0) were designed: 300 kg N ha⁻¹ fertilizer applied as the N300 treatment and 0 kg N ha⁻¹ as the N0 treatment. Water management was intermittent irrigation in 2010 and continuous flooding in 2011. Before rice transplanting, wheat

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