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Agriculture, Ecosystems and Environment





# Methane emission from rice fields as affected by land use change

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#### ARTICLE INFO

Article history: Received 29 August 2010 Received in revised form 27 October 2010 Accepted 1 November 2010 Available online 3 December 2010

Keywords: Rice paddy Land conversion Methane Electron donor Electron acceptor Methanogens

# ABSTRACT

The purpose of this study was to evaluate how former upland cultivation history affects CH<sub>4</sub> emission from rice paddies. We measured CH<sub>4</sub> flux, methanogen population and *in situ* Fe(III) reduction in the rice paddies following three different lengths of time since upland crop (Soybean) cultivation. Results showed that CH<sub>4</sub> emissions from long-term rice paddy (19 year's continuous cultivation) were significantly higher than recently converted ones. Temporal dynamics of methanogens on rice roots also varied among the plots, and showed a good correlation with CH<sub>4</sub> emission rates. Cumulative Fe(III) reduction acted as the dominant electron acceptor in all plots, accounting for 68–94% of the total electron consumption. Fe(II) concentration was highest in the 19-year plot and lowest in the 1-year plots, indicating lower electron availability in recently converted paddies necessary for Fe reduction and CH<sub>4</sub> production. Anoxic laboratory soil incubation also suggested poor availability of electron donors in the recent paddies. Collectively, our results demonstrate that the conversion of upland to paddy rice cultivation significantly affected CH<sub>4</sub> emission through changing availability of electron donors, redox status of soil Fe and activity of methanogens, which ultimately caused low CH<sub>4</sub> emissions.

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### 1. Introduction

Human activities over the past several centuries have been identified as a leading cause of the increased atmospheric concentration of major greenhouses gases (GHGs): carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O) and halocarbons. Methane is the second most important greenhouse gas after CO<sub>2</sub>, having a radiative forcing capacity of 25 times higher than  $CO_2$  (per mass basis, 100-year time-horizon) if indirect effects are taken into account (Forester et al., 2007; Hansen et al., 2005; Shindell et al., 2009) and roughly responsible for 18% of the total greenhouse effect (Forester et al., 2007). The global atmospheric concentration of CH<sub>4</sub> has increased from its pre-industrial level of about 715 ppb to 1750 ppb in the early 1990s and thereafter remained almost constant (Dlugokencky et al., 1992; IPCC, 2007). However, the plateau might have been transitory and atmospheric CH<sub>4</sub> level may increase again in the near future (Bousquet et al., 2006). In fact, the global mean atmospheric CH<sub>4</sub> concentration has shown signs of renewed growth from the beginning of 2007 (Rigby et al., 2008; Dlugokencky et al., 2009). Together with other greenhouse gases, the increased concentration of  $CH_4$  has been predicted to cause an average increase of the global mean surface temperature in the range of 2–4.5 °C on a 100 years time scale (IPCC, 2007).

Methane often known as "marsh gas" is produced in strictly anaerobic environment such as wetlands, sediments, paddy fields, sewage and landfills. Of these sources, paddy fields have long been identified as major contributor to anthropogenic  $CH_4$  emissions. At present, estimates of the total global rice paddy emissions range from 30 to 50 Mt yr<sup>-1</sup> or 8–17% of the total anthropogenic  $CH_4$  flux to the atmosphere (Neue and Sass, 1998; Sass et al., 2002). This wide range of variability in the estimates may partly be driven by the uncertainty in the  $CH_4$  flux due to continuous change in land use or management practices (IPCC, 2001).

In the past, efforts have been made to quantify changes in  $CH_4$  emission in response to land-use change primarily in natural or semi-natural ecosystems such as peatlands reverting to forest (Fowler et al., 1995; Malijanen et al., 2001; Ball et al., 2002) and upland grasslands becoming forest (Oleg Menyailo et al., 2008). These land-use changes typically result in net consumption of  $CH_4$ . However, little is known about the effect of land-use change on  $CH_4$  emissions in agricultural ecosystems such as the conversion from upland crops to rice paddy cultivation (Kumagai and Konno,

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<sup>0167-8809/\$ -</sup> see front matter © 2010 Elsevier B.V. All rights reserved. doi:10.1016/j.agee.2010.11.003

1998; Nishimura et al., 2004). Recently, Nishimura et al. (2004) have estimated the soil carbon budget from a single cropping paddy, single cropping upland rice field and a double cropping soybeanwheat rotation after converting rice paddy fields to upland crop cultivation. Their study demonstrated that CH<sub>4</sub> fluxes from upland crops were significantly lower than those from rice paddy fields. Kumagai and Konno (1998) reported that CH<sub>4</sub> emissions from a restored paddy after 8 years of upland farming was only 44% of the total emission from a continuous rice paddy. Although these studies considered the effects of land conversion on CH<sub>4</sub> emissions, the mechanisms behind the observed changes remain largely unclear.

Among the many soil properties involved in CH<sub>4</sub> emission, microbially reducible Fe(III) content was found to be one of the most important components (Watanabe and Kimura, 1999; Huang et al., 2009). The reduction of Fe(III) has a quantitative relationship with CH<sub>4</sub> production because of its natural abundance in soil as the dominant electron acceptor that competes for the common electron donors (mostly H<sub>2</sub> or acetate) with CH<sub>4</sub> production during the sequential reduction processes in anoxic paddy soil (Takai, 1961; Frenzel et al., 1999). Competition with Fe reduction strongly inhibits CH<sub>4</sub> production because of differences in energy yield, as predicted by thermodynamic theory (Takai and Kimura, 1966). A change in land use might have marked effects on CH<sub>4</sub> production due to a change in the soils reducing conditions, which are largely controlled by the redox status of Fe. Therefore, a simultaneous monitoring of soil Fe reduction would be highly informative for investigating the response of CH<sub>4</sub> emission to land use change

The objective of our study was to evaluate how the conversion of upland soils into paddy rice cultivation affects  $CH_4$  emission. We hypothesized that change in  $CH_4$  emissions were associated with (i) soil properties (e.g. oxidation–reduction status), (ii) microbial population (e.g. methanogenic archaea) or (iii) substrate availability for microbial decomposition. To this end, we measured  $CH_4$  flux, concomitant Fe(III) reduction, changes in the methanogenic archaeal populations in paddy plots and  $CH_4$  production potential following different lengths of time since upland crop cultivation.

#### 2. Materials and methods

# 2.1. Description of experimental fields and cultivation history

This experiment was conducted from May to September 2008 at the National Agricultural Research Center for Tohoku Region, Morioka, Japan (39°45′N, 141°08′E). A total of four plots were used, each comprising an area of 120 m<sup>2</sup>. One plot has been under rice cultivation for the last 19 consecutive years (i.e.19 years as a paddy), since conversion from upland soybean (*Glycine max* L.). The 2-yearold paddy plot was converted from soybean to rice paddy just 1 year ago. Rice cultivation has just begun in the other two plots (1-year paddy) which were cultivated with soybean for the last 6 years. No organic matter (including rice straw) has been added to these plots.

The soil is a volcanic ash soil (Andisol) and classified as a sandy loam with sand content of 45.2%. We used high yielding *Japonica*-type rice cultivar, *Akitakomachi*, which is common in this region. On May 29th, seedlings were transplanted to all four fields. Each hill contains three rice plants. Nitrogen was applied as control-release urea (Kumiai-42-Hifuku-Nyouso-LP70, Zen-Noh, Tokyo, Japan) at 60 kg-N ha<sup>-1</sup> and ammonium sulfate at 20 kg-N ha<sup>-1</sup> before transplanting. Phosphorus (P) was supplied as fused magnesium phosphate at a rate of 130 kg-P ha<sup>-1</sup> and 125 kg-K ha<sup>-1</sup> was applied as potassium chloride and potassium silicate before flooding. All fields were continuously flooded during the cropping period.

#### 2.2. Measurement of $CH_4$ gas flux

Methane emissions were measured using the closed chamber method with a rectangular, transparent, closed top, acrylic chamber (110 cm high with a basal area of  $30 \text{ cm} \times 35 \text{ cm}$  and 0.5 cmthick) during the rice cultivation period. In each plot, chambers were placed on clamps attached to metallic bars installed vertically at the time of transplanting. The clamps and bottom of the chamber were under the level of paddy water to ensure a gas tight seal. In each plot, the flux measurements were conducted in duplicates. The CH<sub>4</sub> fluxes were measured on a weekly to bi-weekly basis from the day after transplanting to before harvesting. Two hills (i.e. six plants) were covered by each chamber. During the sampling period of 30 min, a gas sample of about 25-ml was drawn with a 35-ml plastic syringe at 0, 15 and 30 min after the chamber was emplaced and injected into 20-ml vacuum bottles with a rubber stopper and screw cap. The bottles were taken back to the laboratory, where the amount of CH<sub>4</sub> was measured by a gas chromatograph (Shimadzu GC-14B, Kyoto, Japan) equipped with a flame ionization detector (FID). The operating temperatures for GC were adjusted to 80, 150, and 150 °C for the column, injector and detector, respectively. Methane fluxes were calculated from the slope of the linear increase of gas concentration inside the chamber for the given time interval and expressed as per unit surface area.

# 2.3. Methane production potential

Methane production potential was measured using an anaerobic laboratory incubation method. Before rice transplanting, triplicate soil samples were collected from each plot, air dried and passed through 1 mm sieve to exclude the visible rice residues and weeds. Approximately 10 g of air dried soil was incubated in 100ml serum bottles at 1:2 soil:water (soil to water, w/w) ratio. The head space was replaced with pure nitrogen gas and the samples were incubated at 30 °C. All samples were run in triplicate for a period of 42 days. Methane production potential from the different paddy plots was estimated as the total amount of  $CH_4$  produced in the incubation bottle over 42 days of incubation (Cheng et al., 2000).

# 2.4. Measurement of Fe(II)

The time course of Fe(III) reduction was monitored by measuring the in situ Fe(II) concentration as a cumulative product at 42, 56, 63, 70 and 92 days after transplanting (DAT) to cover the different plant growth stages. We collected the top 5 cm of soil using a cut-tip plastic syringe. Soil samples were collected from between the hills. Immediately after sampling, the plastic syringes were capped with a butyl rubber stopper to prevent redox reactions occurring prior to analysis. Soil Fe(II) was extracted and quantified following the method of Takai et al. (1958). About 2-3 g of wet soil was transferred to a 30 ml centrifuge tube and then 25 ml of pH 3.0 sodium acetate buffer solution was added. The sample was extracted by shaking for 5 min at room temperature, centrifuging at 2000 rpm for 5 min and filtered through a  $0.4 \,\mu$ m filter paper (Advantec, Tokyo Roshi Kaisha Ltd., Japan). About 0.25-0.5 ml of supernatant was transferred into a 15 ml centrifuge tube (Iwaki Glass Corporation, Tokyo, Japan), mixed with 0.4 ml hydroxylamine hydrochloride and 0.4 ml of 0.2% o-phenanthroline. The supernatant was analyzed for Fe<sup>2+</sup> calorimetrically with a UV/VIS spectrophotometer at 510 nm (Shimadzu UV mini-1240, Kyoto, Japan). The moisture content of the remaining wet soil sample was determined gravimetrically, and the Fe(II) content was calculated on a dry-soil basis.

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