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A simplified sampling procedure for the estimation of methane emission in rice fields

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Abstract Manual closed chamber methods are widely used for CH_4 measurement from rice paddies. Despite diurnal and seasonal variations in CH_4 emissions, fixed sampling times, usually during the day, are used. Here, we monitored CH_4 emission from rice paddies for one complete rice-growing season. Daytime CH_4 emission increased from 0800 h, and maximal emission was observed at 1200 h. Daily averaged CH_4 flux increased during plant growth or fertilizer application and decreased upon drainage of plants. CH_4 measurement results were linearly interpolated and matched with the daily averaged CH_4 emission calculated from the measured results. The time when daily averaged emission and the interpolated CH_4 curve coincided during the daytime was largely invariant within each of the five distinctive periods. One-hourly sampling during each of these five periods was utilized to estimate the emission during each period, and we found that five one-hourly samples during the season accurately reflected the CH_4 emission calculated based on all 136 hourly samples. This new sampling scheme is simple and more efficient than current sampling practices. Previously reported sampling schemes yielded estimates 9 to 32% higher than the measured CH_4 emission, while our suggested scheme yielded an estimate that was only 5% different from that based on all 136-h samples. The sampling scheme proposed in this study can be used in rice paddy fields in Korea and extended worldwide to countries that use similar farming practices. This sampling scheme will help in producing more accurate global methane budget from rice paddy fields.

Highlights
?• Measured CH_4 were interpolated and matched with the average daily CH_4 emission.
• The time when the two were the same was invariant within each of the five periods.
• One-hourly sampling in each period can be used to estimate the emission.

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Introduction

Global methane (CH_4) emission reached 556 ± 56 Tg year^{-1} in 2011, of which 354 ± 45 Tg year^{-1} was contributed by anthropogenic sources (IPCC 2013). Agriculture accounts for 47% of anthropogenic CH_4 emissions (IPCC 2007). Rice cultivation generates 33 to 40 Tg year^{-1} of methane (IPCC 2013), which is 10 and 20% of the total anthropogenic and agricultural CH_4 emission, respectively (van Groenigen et al. 2013). Seasonal and yearly CH_4 emissions are based on the measurement of CH_4 level using some type of sampling scheme. The micrometeorological eddy covariance method has been used for continuous CH_4 flux measurements and is considered as an alternative technique to avoid chamber-related problems (Hendriks et al.

2007; Zona et al. 2009; Kroon et al. 2010). The eddy covariance method measures net CH_4 fluxes in the atmosphere; these fluxes represent the integrated net fluxes from the landscape upwind from the measurement point. The eddy covariance method does not disturb the soil surface microenvironment; it integrates over larger areas, and it can measure CH_4 fluxes continuously over long periods (Dugas 1993). However, this method also has a wide range of limitations, such as how it is most applicable over flat terrains and in atmospherically stable conditions. Total CH_4 flux can be underestimated due to low turbulence conditions at nighttime (Long et al. 2010).

The automated closed chamber can measure CH_4 fluxes continuously at a much higher frequency such as (once per hour). This system is useful for monitoring the short-term temporal variability of greenhouse gases and collecting the data over long periods of time (Minamikawa et al. 2012). However, these automatic systems are expensive and require a power supply, which are major limitations (Weller et al. 2015). Chamber methods are often criticized because it covers small patches of soil that may disturb soil temperature, moisture, and air under the chamber. In recent chamber arrangements, these effects have been eliminated or were negligible (Denmead 2008).

A manual closed chamber method is more widely used because of its easy portability, operational simplicity, low cost, and low energy consumption (Hoffmann et al. 2015; Weishampel and Kolka 2008). Fewer samples, however, are taken when manual measurement is used. Air samples are collected manually with a syringe from the headspace of closed chamber and then

Table 1 Physiochemical properties of the soil in this research

Property	Initial	Final	% Increased or decreased
EC (dS m^{-1})	0.95	0.92	−3.16
T-N (%)	0.317	0.30	−5.36
CEC (cmol kg^{-1})	11.54	9.94	−13.86
P_2O_5 (mg kg^{-1})	159.34	210.34	32.01
TOC (%)	1.92	3.46	80.21

analyzed using gas chromatography. The CH_4 fluxes are then calculated by measuring the rates of change in the CH_4 concentrations inside the chamber. Manual closed chambers usually provide periodic measurements to estimate the daily and even annual CH_4 fluxes using linear interpolations or regression models (Song et al. 2009; Chen et al. 2011). Manual closed chamber method is a labor-intensive and time-consuming process; therefore, CH_4 cannot be sampled frequently. Several different sampling schemes for manual systems were proposed. Triplicate sampling at 0600, 1200, and 1800 hours was suggested to replace an automated system (Buendia et al. 1998). Once per day CH_4 sampling at 1000 hours resulted in ± 10% error from the results of automated systems (Minamikawa et al. 2012). Yun et al. (2013) reported that the best diurnal interpolation time for CH_4 emission was 1000–1100 hours.

CH_4 sources and sinks are well defined, but there is great uncertainty regarding the magnitude of fluxes and the factors that regulate these fluxes (IPCC 2007; Kirschke et al. 2013; Yvon-Durocher et al. 2014). Methanogens play critical roles in CH_4 production, and

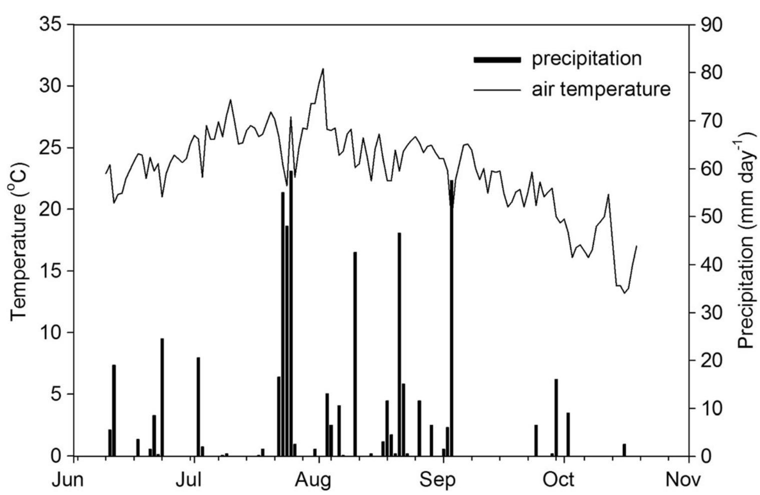

Fig. 1 Daily mean air temperature and precipitation during the rice-growing seasons at the experimental site

the magnitude of CH_4 emission is dependent on several factors, such as water management, temperature, fertilizer, soil pH, and so on (Conrad 2004; Jain et al. 2004; Oyewole 2012; Singh et al. 2003). Continuous flooding of rice paddy fields makes the soil anaerobic. Mid-season drainage supplies oxygen and reduces CH_4 emission (Jain et al. 2004; Anand et al. 2005). An increase in temperature from 15 to 30 °C resulted in a 2- to 2.2-fold increase in CH_4 emission from rice paddy fields (Dakua et al. 2013). Nitrogen fertilizer also strongly affects CH_4 emission. Type, quantity, and method of application changes the amount of CH_4 emitted (Minami 1995; Dong et al. 2011). Variation in soil pH affects methanogenic CH_4 production, with maximal emission found at pH 6.9–7.1 (Jain et al. 2004). Therefore, CH_4 emission changes with the growth of rice plants (Jia et al. 2001; Lee et al. 2010; Dakua et al. 2013).

Existing sampling schemes for measuring CH_4 emission from rice paddies use fixed sampling time throughout the season. However, there are several factors that can increase or decrease CH_4 emission from rice paddies. Under this complex environment, sampling with manual closed chamber at a fixed time throughout a season can lead to incorrect estimation of diurnal and ultimately seasonal CH_4 emission. Therefore, it is important to observe the most influencing factors that cause fluctuation in diurnal flux and find representative sampling time for daily average CH_4 flux. To address this issue, we monitored CH_4 emission from rice paddies for one complete rice-growing season. Our objectives were (1) to investigate the effect of CH_4 sampling time on diurnal/seasonal CH_4 emission estimation and (2) to propose a general sampling scheme for the manual closed chamber method, which provides

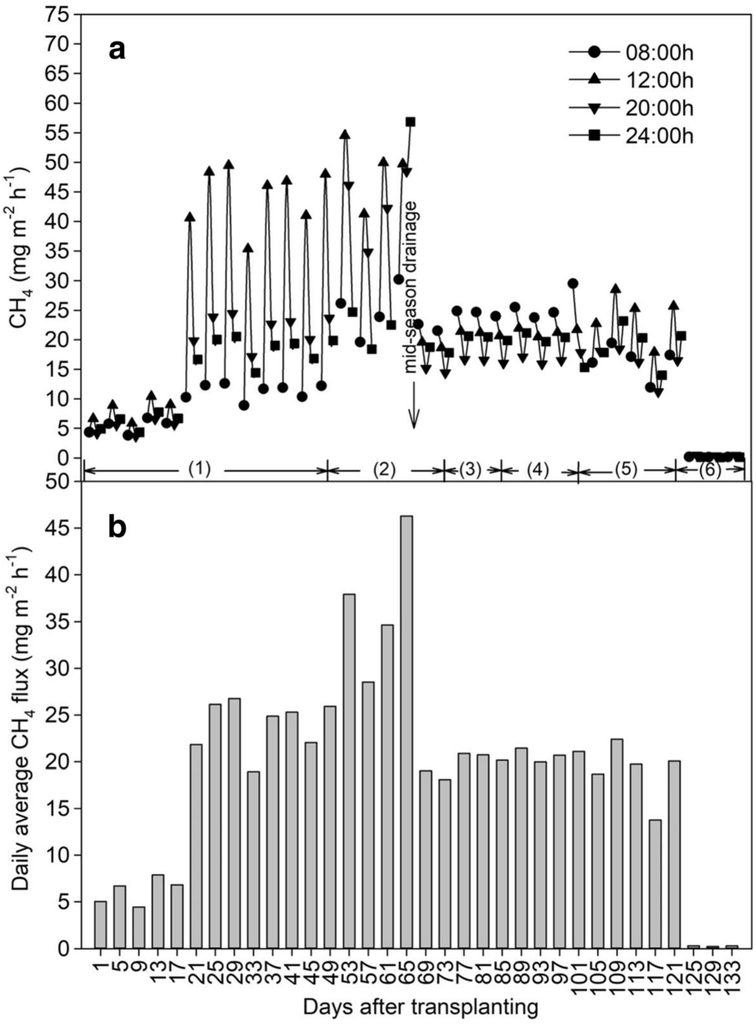

Fig. 2 **a** Measured CH_4 emission from the experimental rice field at different stages ((1) tillering, (2) jointing, (3) booting, (4) heading, (5) milky, (6) ripening) and **b** daily averaged CH_4 emission

a more accurate estimate of CH_4 emission from rice paddy fields.

Material and methods

Experimental design

The experiment was conducted on Hanyang University campus, Seoul, Republic of Korea (37° 33′ 16″ N, 127° 02′ 38″ E). A monsoon climate prevailed in this area with mean annual air temperature and precipitation of 12.5 °C and 1450.5 mm, respectively. Average air temperature during the experimental period was 23.2 °C, and total precipitation was 543.4 mm (Fig. 1). Both pots and chambers in this research were custom-made using 10-mm-thick acryl sheets. The dimensions of each pot were 720 × 720 × 520 mm. Styrofoam of 20 mm thickness was used to cover all sides of each pot to reduce heat exchange. Triplicated pots were filled with 260 kg of 4-mm sieved and air-dried soil. Above the soil surface, 100 mm high free space was left for irrigation. Each pot was thoroughly soaked initially, and the water level was maintained at 5–7 cm above the soil for the entire season except for mid-season drainage and harvest. The soil used in this research was obtained from a plow layer (150–200 mm depth) in a paddy field, located in Jeonsu-ri, Kyunggi-do, Republic of Korea (37° 28′ 55.37″ N, 127° 25′ 27.13″ E).

Soil physiochemical characterization

Soil organic matter was determined using the modified Walkey and Black method and the Tyurin titrimetric method with acid-wet oxidation and dichromate (Lee et al. 2014). Soil P_2O_5 content was determined by the Lancaster method (Heczko and Zaujec 2009). Kjeldahl distillation was used to analyze the total N (RDA 1988). Cation exchange capacity (CEC) was measured using the ammonium acetate extracting method at pH 7.0 (Sumner and Miller 1996). Electrical conductivity (EC) was determined by using a conductivity meter (CM-25R). Soil texture was determined using the soil hydrometer method and was classified by USDA criteria. Soil texture was sandy loam with sand, silt, and clay proportions of 59.8, 38.6, and 1.6%, respectively. Other physicochemical properties of the soil were analyzed before crop sowing and after harvesting (Table 1).

One chamber was used for each pot. A chamber consisted of three parts without a bottom panel: base, middle, and top with sides of 150, 500, and 500 mm, respectively. The inner dimensions of a chamber were 500 × 600 mm. Each chamber was equipped with a battery-operated fan inside for air mixing. A thermometer

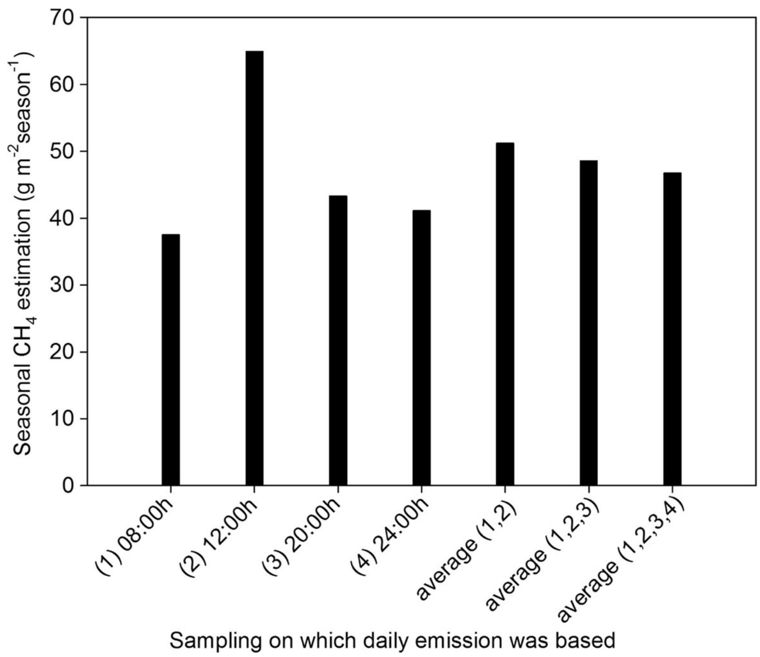

Fig. 3 CH_4 emission in a season estimated using different sampling schemes

and gas sample collection tubes were also installed on the top of each chamber.

Mineral fertilizers were applied at rates of 110 kg N ha^{-1}, 31 kg P_2O_5 ha^{-1}, and 66 kg K_2O ha^{-1}, using urea, fused superphosphate, and potassium chloride. The basal mineral fertilizers applied 1 day before paddy transplanting were 55 kg N ha^{-1}, 31 kg P_2O_5 ha^{-1}, and 66 kg K2O ha^{-1}. Basal fertilizers were mixed manually within the top 100 mm of soil under flooding. Twenty-day-old nursery seedlings (three plants per hill) were transplanted by hand at a spacing of 300 × 150 mm, resulting in eight rice hills per pot. Mid-tillering fertilizer (27.5 kg N ha^{-1}) was broadcasted approximately 3 weeks after rice transplanting, and the panicle fertilizer (27.5 kg N ha^{-1}) was broadcasted 9 weeks after transplanting.

CH_4 sampling, analysis, and flux calculations

CH_4 samplings at 0800, 1200, and 1600 hours were reported for daily average CH_4 estimation from rice paddies in Korea (Gutierrez et al. 2013; Haque et al. 2016; Kim et al. 2016). Since all the three samplings were in daytime, samplings at nighttime should be included. CH_4 sampling was performed four times a day on every fourth day in this research: at approximately 0800, 1200, 2000, and at 2400 hours. To determine the hourly flux at each sampling time, 60 ml samples were collected with an

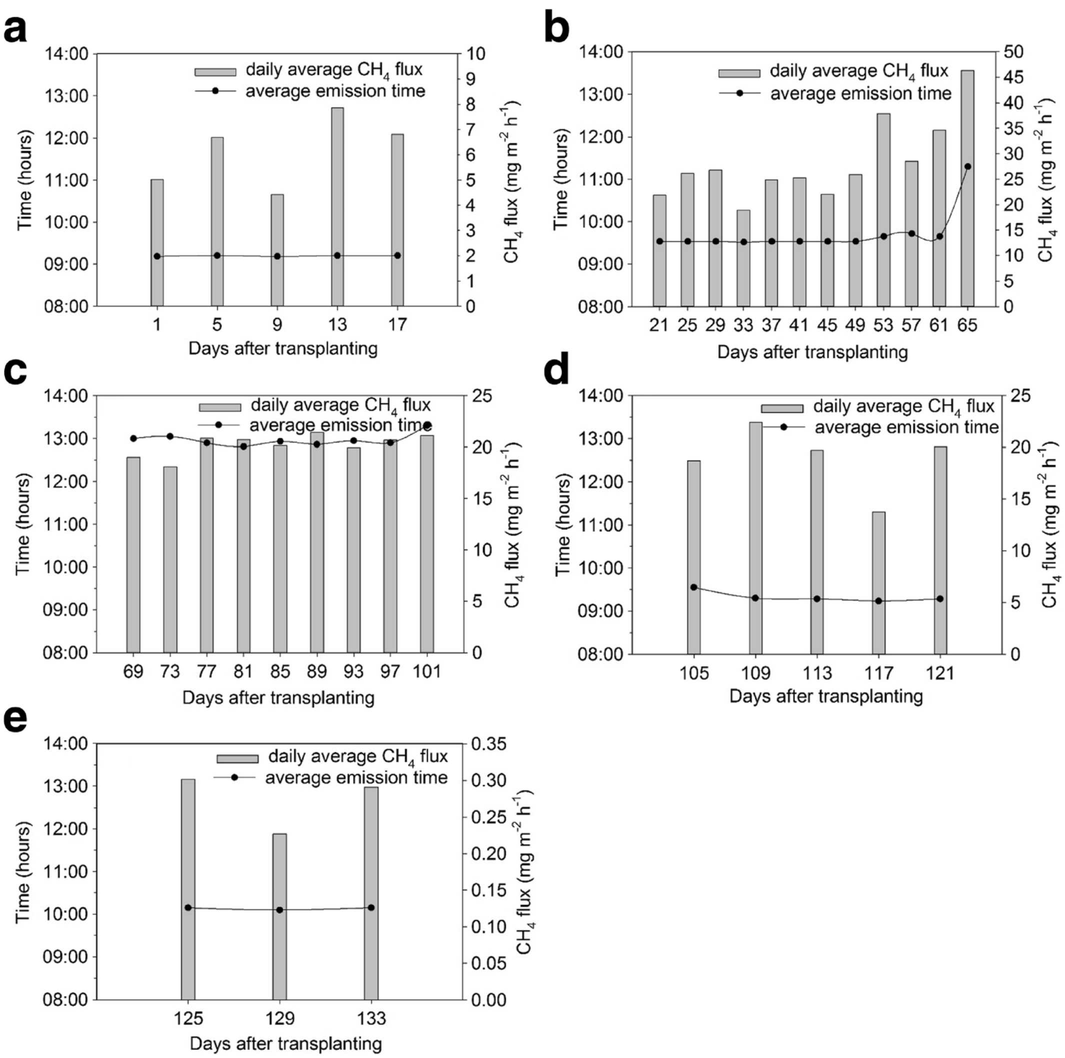

Fig. 4 Average CH_4 emission time and daily averaged CH_4 flux for each of five distinctive period: **a** transplanting to mid-tillering, **b** mid-tillering to jointing, **c** jointing to heading, **d** heading to ripening, and **e** ripening to harvesting

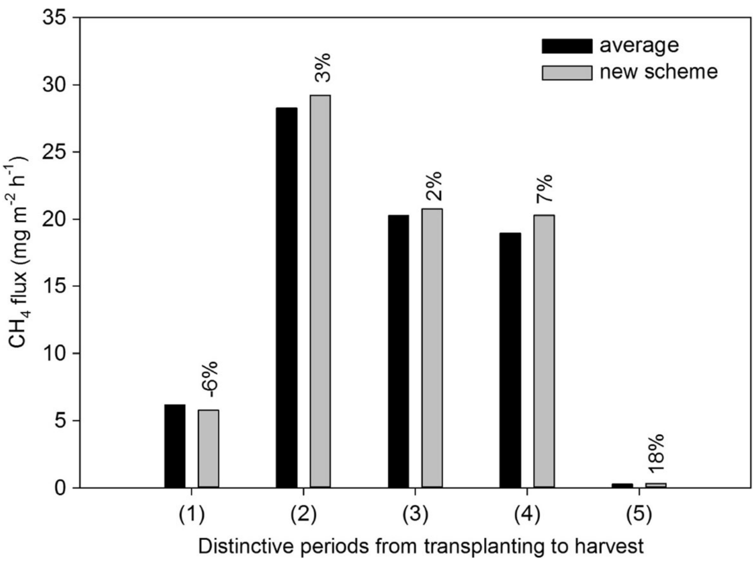

Fig. 5 Comparison of average CH_4 emission from actual measurements and CH_4 emission with the suggested sampling scheme at each of the five distinctive periods: (1) transplanting to mid-tillering, (2) mid-tillering to jointing, (3) jointing to heading, (4) heading to ripening, (5) ripening to harvesting

airtight syringe at 0, 30, and 60 min, respectively, after chamber closure at the hour (Jia et al. 2001; Frei et al. 2007). CH_4 samples were analyzed within 2 h after sampling using a gas chromatograph (YL 6100, Young Lin Instrument Co., Korea) equipped with a flame ionization detector and HP-PLOT Q Agilent column (length, 30 m; inner diameter, 0.5 mm; and film thickness, 40 μm). The temperatures of the column, injector, and detector were 80, 150, and 250 °C, respectively. Helium was used as the carrier gas at a flow rate of 30 ml min^{-1}. Temperatures of ambient air, the air inside the chamber, and the soil were also recorded at the time of each CH_4 sampling.

Hourly CH_4 flux was calculated from the change in gas concentration in the chamber over a 60-min period (Rolston 1986; Cheng et al. 2007):

$$F = \frac{V}{A} \times \frac{dc}{dt} \times \left(\frac{273}{273 + T}\right) \qquad (1)$$

where F is the hourly CH_4 flux (mg m^{-2} h^{-1}), V is the gas volume at standard condition (m^3), A is the area of the chamber base (m^2), $\frac{dc}{dt}$ is the rate of CH_4 concentration change over a 60-min period in the chamber (mg m^{-3} h^{-1}), and T is the air temperature inside the chamber (°C). Daily CH_4 flux was calculated from the measured hourly flux. Zadoks, Feekes, and Haun scales are widely used to define crop-growth stages (Ali 2010). The Zadoks scale defines growth stages as germination, tillering, jointing, booting, heading, milky, drought, and ripening. CH_4 emission was investigated according to the Zadoks scale in this study. Because CH_4 sampling was carried out after transplanting, CH_4 emission from germination to transplanting was not investigated. The drought was considered part of the ripening stage. CH_4 flux for a stage was calculated by taking the averages of the daily fluxes. Total CH_4 emission flux (g m^{-2}) over the entire crop season was calculated as the sum of CH_4 fluxes at each stage.

Results and discussion

Measured and daily averaged CH_4 emission varied over the season (Fig. 2). Emission increased until the jointing

Table 2 CH_4 emission in Korea estimated using different sampling times and frequencies

Reference	Sampling time	Sampling interval (days)	Average CH_4 flux (mg m^{-2} h^{-1})
Kim et al. 2016	0800, 1200, and 1600 hours	6	11.83
Kim et al. 2014b	1100 and 1200 hours	3	12.94
Gutierrez et al. 2013	0800, 1200, and 1600 hours	3	14.98
Haque et al. 2016	0800, 1200, and 1600 hours	6	6.00
Kim et al. 2014a	1100 and 1300 hours	6	5.78

stage, and then decreased. Variation over the course of a day was quite noticeable from the mid-tillering to the milky stage, as shown in Fig. 2a. Relatively smaller daily variation was observed after the mid-season drainage than before. From mid-tillering to jointing stages before mid-season drainage, lower emission was observed at 0800 h, while higher emission was observed at 1200 h. Higher and lower emissions were observed at 0800 hours and at 2000 hours, respectively, after mid-season drainage to the heading stage. Low emission at ripening was probably due to reduced permeability of the root epidermal layer associated with plant aging (Gogoi et al. 2005). CH_4 emission varies with plant-growth stage due to an increase in the size of the plant aerenchyma and roots (Jia et al. 2001; Dakua et al. 2013). Water management in rice paddies has a major effect on CH_4 emission (Zou et al. 2005; Kumar et al. 2016). In response to continuous flooding, CH_4 emission gradually increases after transplanting and reaches a maximal level at the heading stage, and then decreases (Wang et al. 1999; Lu et al. 2000). Irrigation with mid-season drainage is a widely adopted water management practice to improve rice growth and yield (Cai et al. 1997; Lu et al. 2000). Mid-season drainage in our study resulted in a switch from anaerobic to aerobic conditions, which reduced CH_4 emission, as shown in Fig. 2b (Tyagi et al. 2010; Li et al. 2011).

Estimation of CH_4 emission from a field using a manual system is based on sampling time and frequency. Average daily emission was calculated based on the average of four samples in this research: 0800 h, 1200 h, 2000 h, and 2400 h. If sampling was performed differently and daily emissions were estimated based on those samples, the estimated emission could be quite different. Figure 3 shows estimates of seasonal CH_4 emission based on different combinations of actual samplings. The highest estimation was produced with one-time sample at 1200 hours (38% higher than the average of the four time samples). The averages of 0800 and 1200 hours and the 0800, 1200, and 2000 hours samplings resulted in estimates 9.6 and 3.9% higher than the average of the four sampling times, respectively. Different CH_4 sampling schemes can produce different estimates of seasonal CH_4 emission. CH_4 emission at 1200 hours was much higher than at the rest of a day; therefore, estimation based on a one-time sample near 1200 hours can be misleading. It should be noted that quite a few studies have reported CH_4 emission based on one or two time samples taken near 1200 hours.

When CH_4 measurement results were linearly interpolated and matched with the daily averaged CH_4 emission calculated from the measured results, the daily average and measured CH_4 curve coincided twice in

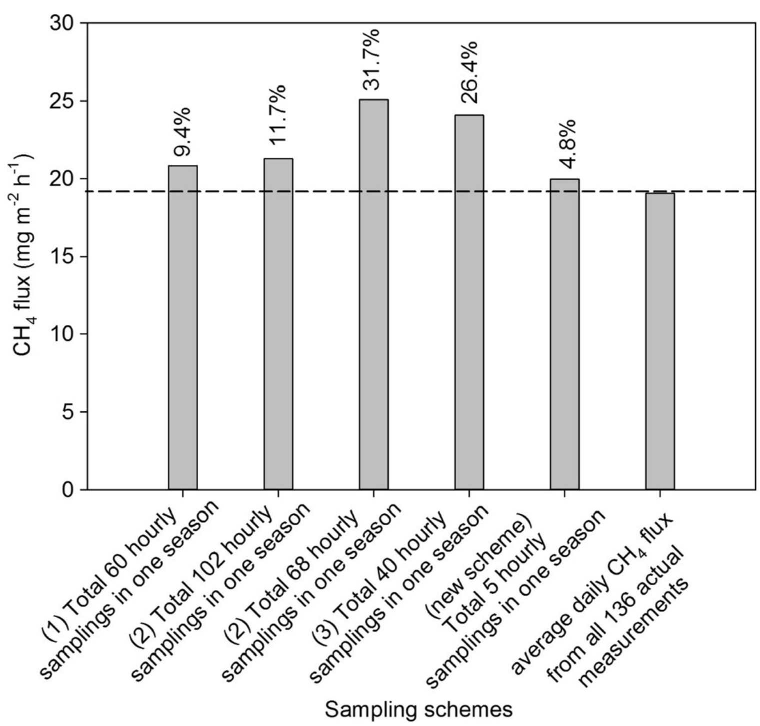

Fig. 6 Change in average CH_4 emission according to different sampling schemes based on actual 24-h interpolated CH_4 flux measurements: (1) 0800, 1200, and 1600 hOURS at 6-day intervals; (2) 0800, 1200, and 1600 hours at 3-day intervals; (3) 1100 and 1200 hours at 3-day intervals; and (4) 1100 and 1300 hours at 6-day intervals

one day: once during the daytime, and once during nighttime. The change in the time when the daily average and interpolated CH_4 curves coincided in the daytime (average emission time) is shown in Fig. 4. The whole season was divided into five periods. In each period, the variation in average emission time was negligible. Application of second and third doses of fertilizer at 17 and 61 days after transplanting increased CH_4 emission and changed the average emission time. Mid-season drainage reduced CH_4 emission significantly.

As average emission time within each of the five periods was rather invariant, we hypothesized that CH_4 estimation based on several daily samplings during each period could be replaced with one-hourly sampling per period (Fig. 4). For instance, CH_4 measurement can be carried out 1 day after transplanting, 3 days after fertilizer application at mid-tillering growth stage, 3 days after mid-season drainage, when heading of rice paddies occurs, and at the ripening stage. In other words, only five hourly samples would have to be taken per season. Average CH_4 emission on each of the 5 days can then be calculated based on hourly CH_4 flux measurement from 0900 to 1000 hours at the first (transplanting to mid-tillering), second (mid-tillering to jointing), and fourth (heading to ripening) stages. Hourly measurement from 1200 to 1300 hours and from 1000 to 1100 hours can be used at the third (jointing to heading) and fifth (ripening to harvesting) stages, respectively. Daily CH_4 estimation using the suggested scheme was compared with average daily CH_4 flux from all actual measurements for each of the five periods. Overall, there was a 4.8% difference between the two schemes for the season (Fig. 5). This new sampling scheme is a simpler and less labor-intensive alternative to continual sampling over a season.

Reported CH_4 flux from Korean rice paddy fields using the same farming practices as used in this research varied from 5.78 to 14.98 mg m^{-2} h^{-1} when different sampling schemes were used, as shown in Table 2. Sampling was carried out two or three times a day once or twice a week. Four different sampling schemes are shown in Table 2. Hourly CH_4 flux measurement at 0800, 1200, 2000, and 2400 hours in this research was linearly extrapolated to 24 h, and the four different sampling schemes were applied to this 24-h interpolated CH_4 flux to compare the effect of different sampling schemes on CH_4 flux estimation. The results were compared with average daily CH_4 flux from all actual measurements and the new sampling scheme proposed in this research (Fig. 6). Sampling schemes (1) to (4) showed 9.4, 11.7, 31.7, and 26.4% difference from the average CH_4 based on actual measurements (Fig. 6). This confirmed that our suggested sampling scheme, which only requires five one-hourly samplings a season, is an excellent alternative to existing sampling schemes.

Conclusions

We measured variation in daily averaged CH_4 flux and average sampling time to develop a simpler and more efficient sampling scheme. Average emission time was rather invariant within each of the five distinctive periods: (1) transplanting to mid-tillering, (2) mid-tillering to jointing, (3) jointing to heading, (4) heading to ripening, and (5) ripening to harvesting. Five hourly samples taken on the first day of each period allowed the average daily emission during each period to be estimated with the average calculated for the season. Existing sampling schemes require more frequent and numerous hourly samplings. The daily averaged CH_4 flux calculated using these sampling schemes was found to be much higher than the daily averaged CH_4 flux found in this research. This higher estimation was due to the use of fixed sampling times throughout the season and sampling at high CH_4 emission hours. Our new sampling scheme requires only five 1-h samples for the entire season. This scheme can be adopted anywhere in Korea or the world that uses similar rice paddy farming practices. The limitation of the suggested scheme is that, in case of different farming practices, diurnal flux measurements should be performed for at least one season to identify distinctive periods and to calculate the average emission for these periods.

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