



# Methane and nitrous oxide emissions from paddy fields with no fertilizer use under continuous irrigation with treated municipal wastewater

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## Abstract

Treated wastewater (TWW) irrigation has been recommended as an environmentally friendly agricultural practice and has been applied in many countries for decades. The effects of wastewater irrigation on rice yield and quality, as well as on the environment, with particular focus on greenhouse gas emissions from paddy fields with municipal wastewater irrigation, have gained substantial attention. In this study, bench-scale experiments were conducted in two cultivation seasons where seedlings of Bekoaoba, a large-grain high-yield rice variety, were transplanted and irrigated with TWW without fertilization. A control experiment was performed to simulate the cultivation conditions of normal paddy fields. The study aimed to quantify the effects of TWW irrigation on rice yield and quality, in addition to CH<sub>4</sub> and N<sub>2</sub>O emissions. The highest rice yield (10.4 t ha<sup>-1</sup>) and protein content in brown rice (13.8%) was achieved when the soil was repeatedly subjected to bottom-to-top TWW irrigation without any synthetic fertilizer. Bottom-to-top TWW irrigation decreased CH<sub>4</sub> emissions by up to 95.6% when compared with tap water irrigation, whereas bottom-to-top and top-to-top TWW irrigation increased N<sub>2</sub>O emissions by 5 and 15 times, respectively. Bottom-to-top irrigation of TWW could be considered a promising solution for reducing greenhouse gas emissions as TWW irrigation resulted in a lower combined global warming potential than tap water irrigation. Further, bottom-to-top irrigation of TWW produced less CH<sub>4</sub> and N<sub>2</sub>O than top-to-top irrigation.

**Keywords** Treated wastewater irrigation · Bottom-to-top irrigation · High yield and quality rice · Greenhouse gas · Soil redox potential

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## Introduction

Climate change is a serious environmental problem caused by increased greenhouse gas (GHG) emissions. Thus, reducing GHG emissions entering the atmosphere is urgently required to mitigate the adverse impacts of climate change (Wang et al. 2017). Methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) are two important GHGs that have contributed to an estimated 0.6–0.7 °C increase in the Earth's surface temperature during the last century (S. Yang et al. 2012). Globally, agriculture accounted for approximately 47% and 58% of anthropogenic CH<sub>4</sub> and N<sub>2</sub>O emissions in 2005, respectively, representing a 17% rise since 1990 (IPCC 2007). Atmospheric concentrations of CH<sub>4</sub> and N<sub>2</sub>O have increased rapidly from pre-industrial levels (722 ppb and 270 ppb, respectively) to the present levels of 1830 ppb and 324 ppb, respectively (IPCC 2013a).

Paddy fields are recognized as a significant source of atmospheric GHGs, including CH<sub>4</sub> and N<sub>2</sub>O (Meijide et al.

2016). Rice is a leading cereal crop that provides approximately 20% of the daily calorie requirements for more than 3.5 billion people through 150 million hectares of global rice land, which supply 550–600 million tons of rough rice each year (Pham and Watanabe, 2017). Rice is highly sensitive to the water regime, and paddy rice is estimated to consume approximately 50% of global irrigation water. However, irrigation water for rice paddies does not typically require the same quality as drinking water (Pham and Watanabe, 2017). As a result, municipal wastewater has been considered as an alternative source of irrigation water and has been applied and studied globally due to its high concentrations of nutrients (N and P) and organic matter, which are essential for plant growth (Hanjra et al. 2012). Although the use of wastewater for rice production would benefit farmers by enhancing the rice yield and reducing the need for fertilizers, it also has the potential to cause adverse effects to human health and the environment. Thu (2001), Yoon et al. (2001), Nyomora (2015), and Pham et al. (2017) reported that greater rice yields were achieved through wastewater irrigation than freshwater irrigation. Mastro et al. (2009) found that sewage effluent irrigation enhanced soil texture and increased the soil carbon content. Furthermore, some studies have reported that sewage effluent irrigation increases soil concentrations of heavy metals such as Pb, Cd, Cu, and Zn (Q. W. Yang et al. 2006; Rogan et al. 2009; Chung et al. 2011).

The emission fluxes of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  from paddy fields can be affected by numerous factors, including water regime, organic matter, and nitrogen sources.  $\text{CH}_4$  is generated under anaerobic conditions by methanogens, and then 60–90% is oxidized by methanotrophs under the aerobic conditions of the rhizosphere to form  $\text{CO}_2$  (Wang et al. 2017). Contrary to  $\text{CH}_4$  emissions,  $\text{N}_2\text{O}$  emissions from flooded paddy fields are mostly negligible because the anaerobic conditions of paddy soil transfer the end product of denitrification processes toward  $\text{N}_2$  (Weller et al. 2014). Domestic sewage effluent is rich in organic matter and nitrogen, which are important sources of  $\text{CH}_4$  and  $\text{N}_2\text{O}$ . Thus, sewage irrigation for rice cultivation may promote the emission of these gases from paddy fields. For example, Zou et al. (2009) observed that increased emissions of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  from paddy fields accompanied an increased rice yield in their study using domestic wastewater irrigation with the same amount of N-fertilizer input. However, Xu et al. (2017) found that treated sewage irrigation incorporated with straw returns significantly decreased  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions when compared with tap water irrigation.

In this study, we hypothesize that treated municipal wastewater (TWW) irrigation increases  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions from rice paddies, compared with freshwater irrigation. To investigate this, bench-scale experiments were conducted in Tsuruoka, Japan, under both tap water and TWW irrigation. The primary aim is to quantify the effect of TWW irrigation on rice yield and quality, in addition to  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions.

## Materials and methods

### Experimental design and cultivation management

The experiments were conducted using the same bench-scale apparatus used in previous studies (Muramatsu et al., 2015; Pham et al., 2019; Pham et al., 2017; Watanabe et al., 2016). The apparatus consists of a TWW feeding system and a bench to simulate a paddy field (Fig. 1). The feeding system comprised a 500-L influent tank, a peristaltic pump, and a 100-L effluent tank for each treatment. The bench-scale paddy field consisted of a 30 cm × 60 cm plastic box with a depth of approximately 35 cm. A horizontal drainpipe was fixed at the bottom of the field to supply water upward, and to drain all the water out from the field when necessary. The drainpipe was covered with a thin layer of gravel to avoid clogging, and then further covered by a 15-cm thick layer of soil. An overflow pipe was fixed at a height of 5 cm from the soil surface. Four treatments without replicates were employed under different conditions (Table 1). In 2016, bottom-to-top irrigation was applied, whereby TWW at a flow rate of 4.5 L/day continuously entered the field from the bottom, passed through the soil layer, and flowed out to the effluent tank via the overflow pipe (Treatments no. 1 and no. 2). Treatment no. 3 used top-to-top irrigation, whereby TWW was continuously supplied to the surface of the field at the same flow rate as treatments no. 1 and no. 2. Treatment no. 4 was designed as the control by adding tap water to make up the water loss due to evapotranspiration with supplementation of N-P-K fertilizers. In the 2017 season, the flow rate of treatment no. 2 was changed to 2.0 L/day, whereas the other treatments maintained the same conditions as the 2016 season.

The water used for the experiments was taken from the effluent of the municipal wastewater treatment plant (WWTP) in Tsuruoka, Yamagata, Japan, which employs a conventional technology with a standard activated sludge process and chlorine disinfection. The soils for treatments no. 1 and no. 3 in the 2016 season and treatment no. 4 in both seasons were sampled from the surface layer (0 to 20 cm) of a paddy field in the farm of Yamagata University (Tsuruoka, Yamagata, Japan) (New), whereas treatment no. 2 in the 2016 season reused the soil from the 2015 season (Pham et al. 2017) and the other treatments in the 2017 season repeatedly used the soil from the 2016 season (Pham et al. 2019).

In the 2016 season, 30-day-old rice seedlings of Bekoaoba, a large-grain high-yield variety, were transplanted on May 20, at a rate of five plants per hill and four hills per container. A water depth of 5 cm was maintained during the growing season from transplanting to harvesting on September 28, except for the period from July 4 to 11. This period is known as the midsummer drainage (MSD) and refers to the time when the water supply was stopped, and the paddy soil was kept in a dried condition in order to enhance rice root growth by serving

**Table 1** Experimental conditions

Cultivation condition	Treatment no. 1		Treatment no. 2		Treatment no. 3		Treatment no. 4 (control)
	2016	2017	2016	2017	2016	2017	2016 and 2017
Soil	New	Old	Old	Old	New	Old	New
Water	TWW	TWW	TWW	TWW	TWW	TWW	Tap water
Flow rate (L day <sup>-1</sup> )	4.5	4.5	4.5	2.0	4.5	4.5	Depends on evapotranspiration
Flow direction	B-to-T	B-to-T	B-to-T	B-to-T	T-to-T	T-to-T	No flow
Water supply	Cont.	Cont.	Cont.	Cont.	Cont.	Cont.	As required
Chemical fertilizer	No	No	No	No	No	No	N, P, K (for basal) and N-K (before flowering)

*B-to-T* bottom-to-top irrigation, *T-to-T* top-to-top irrigation, *Cont.* continuous irrigation

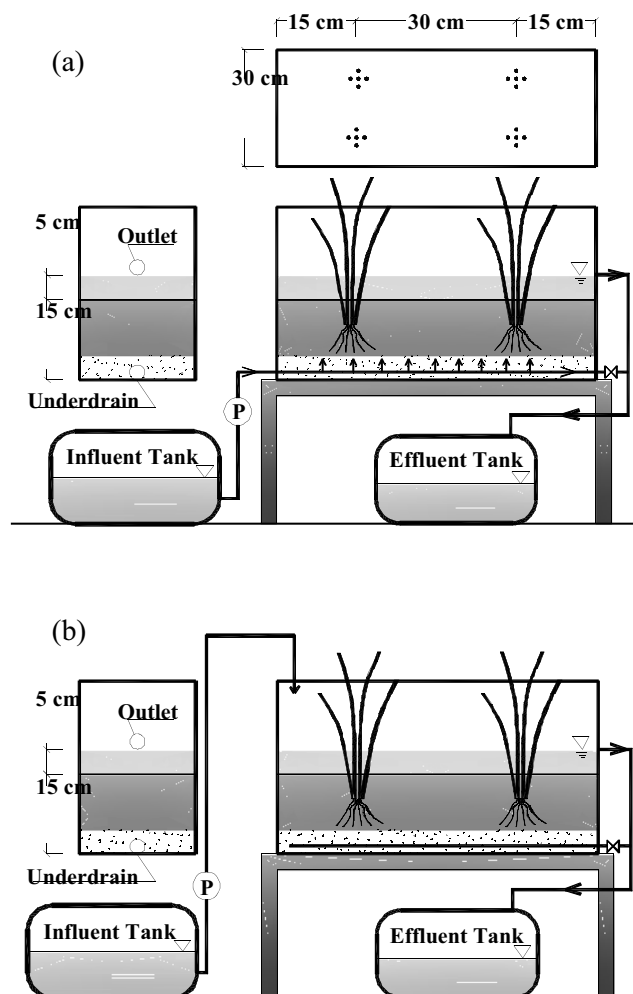
oxygen to the rice root zone. The 2017 cultivation season was conducted in the same way as 2016, with transplanting, MSD, and harvesting occurring on May 17, July 10–16, and October 01, respectively.

In treatment no. 4, basal N-P-K fertilizer was applied at a rate of 160 kg/ha before transplantation, and N-K fertilizer was applied as top-dressing at a rate of 100 kg/ha on July 11

and 16, in 2016 and 2017, respectively. This was almost twice the amount of fertilizer used for rice as staple food, following the local farmer's practice. No fertilization was applied for the other treatments in either season.

### CH<sub>4</sub> and N<sub>2</sub>O measurement

In the 2016 season, only CH<sub>4</sub> was sampled using manual static chambers. In the 2017 season, samples of both CH<sub>4</sub> and N<sub>2</sub>O were collected using the same chambers as the 2016 season. The chambers were made of acrylic with a 20-cm × 22-cm footprint and two heights: 60 cm for the early rice growing stage and 115 cm for the middle and later rice-growing stages. Each chamber covered two hills of rice and was installed with an air-circulating fan to ensure complete gas mixing during the sampling period. Chambers were placed on the frame of the apparatus in each plot prior to gas sampling. Sampling was conducted four times at intervals of 20 min, once a week in the morning (10:30 to 12:00), using a 60-ml syringe through a silicon tube embedded at the flank of the chamber. The samples were then immediately transferred to 10-ml glass vials for measurement with a gas chromatography instrument (Hitachi GC-163) with a flame ionization detector (FID) for CH<sub>4</sub> and a Shimadzu GC-14B gas chromatography instrument with an electron capture detector (ECD) for N<sub>2</sub>O. The fluxes of gas emissions were calculated according to the equation proposed by Minamikawa et al. (2015).



**Fig. 1** Simulated paddy fields with different directions of continuous irrigation: **a** bottom-to-top irrigation and **b** top-to-top irrigation

### GWPs of CH<sub>4</sub> and N<sub>2</sub>O

Global warming potential (GWP) is an index that attempts to integrate the overall climate impacts of a specific action. In the GWP calculation, CO<sub>2</sub> is typically used as the reference gas. The GWPs were calculated over a specific time interval by multiplying the seasonal total CH<sub>4</sub> and N<sub>2</sub>O emissions by their respective radiative forcing potentials. In this study, we calculated the GWPs for 20-year and 100-year periods. The radiative forcing potentials for CH<sub>4</sub> and N<sub>2</sub>O were 84 and 264 for

the 20-year period, and 28 and 265 for the 100 year period, respectively, for a CO<sub>2</sub> value of 1 (IPCC 2013).

**Other data measurement**

During the growing season, various rice plant growth parameters were measured, including plant height, tiller number, and leaf chlorophyll concentration detected using a soil plant analysis development (SPAD) meter (Markwell et al. 1995). The rice yield and yield components (number of ears per square meter, number of kernels per ear, grain weight, matured kernels, and rice weight of grain per hectare) and dry mass were measured after harvesting. Nitrogen contents in the rice grain were analyzed using an automatic high-sensitivity NC analyzer (SUMIGRAPH NC-220F, SCAS, Japan).

The properties of irrigation water were measured once a week for basic parameters, including total nitrogen (TN), N components (NH<sub>4</sub>, NO<sub>3</sub>), total organic carbon (TOC), total phosphorus (TP), dissolved oxygen (DO), pH, electrical conductivity (EC), oxidation-reduction potential (ORP), and heavy metal contents, according to the methods described by Pham et al. (2017). Soil properties before and after the experiments were measured using the standard methods of Homer and Pratt, 1962 for TC, TN, TP, and heavy metals. A platinum Eh electrode (EP-201, Fujiwara, 24 cm) was installed permanently at 10 cm soil depth during the growing season. The soil

redox potential (Eh) was measured during gas sampling using an Eh meter (PRN-41, Fujiwara, DKK-TOA Corporation).

The overall balance of nitrogen in the experiment was calculated by multiplying the dry weight of soil, whole rice plant, or volume of irrigation water by its concentrations in these samples. The difference from N input (soil, irrigation water, and fertilizer) to its output (soil and rice plant) can be explained by the release of N<sub>2</sub>O and N<sub>2</sub> into the air.

**Statistical analysis**

The treatments were not replicated owing to resource limitations. Instead, four plants of rice seedlings were transplanted into each treatment and were considered as replicates during data analysis (Pham et al. 2019). An analysis of variance (ANOVA) was conducted at 5% probability, and the mean differences were detected using a least significant difference (LSD) test. The statistical package SPSS 24.0 was used for the data analysis.

**Results**

**Irrigation water quality and soil properties**

The monthly chemical characteristics of irrigation water in 2017 are illustrated in Table 2. The pH of the irrigation TWW varied

**Table 2** Monthly average quality of irrigation water in 2017

Parameters	Units	Tap water	TWW					Average	Permissible limits <sup>a</sup>	Data in 2016 <sup>b</sup>
			Month							
			May	Jun.	Jul.	Aug.	Sep.			
pH	–	NA	7.3	7.3	7.6	7.2	7	7.3	NA	7.1 (7.0–7.3)
ORP	mV	NA	197	211	219	234	279	228	NA	236 (296–178)
EC	mS m <sup>-1</sup>	NA	71.7	61.7	67.4	67.7	64.2	66.5	NA	63.6 (56.7–72.5)
DO	mg L <sup>-1</sup>	NA	4.8	2.9	3.2	2.9	3	3.4	NA	3.7 (1.9–5.9)
TN	mg L <sup>-1</sup>	<0.1	40	31.9	28.5	33.5	27.8	32.3	NA	27.6 (23.0–40.0)
NH <sub>4</sub> -N	mg L <sup>-1</sup>	NA	27.5	17.0	18.9	20.4	15.6	18.9	NA	20.4 (4.2–35.6)
NO <sub>3</sub> -N	mg L <sup>-1</sup>	NA	2.8	4.8	4.6	9.7	11.0	11.2	NA	9.54 (1.0–26.8)
TOC	mg L <sup>-1</sup>	<0.6	6.3	6.3	5.8	5.5	5.3	5.8	NA	6.6 (4.8–9.3)
TP	mg L <sup>-1</sup>	NA	0.27	0.3	0.44	0.21	0.18	0.28	NA	0.17 (0.15–0.23)
Cu	µg L <sup>-1</sup>	NA	8.5	7.7	7.8	7.4	6.5	7.6	200	11.2 (8.4–15.4)
Cr	µg L <sup>-1</sup>	NA	<0.3	<0.3	<0.3	<0.3	<0.3	<0.3	100	0.64 (0.60–0.80)
Zn	µg L <sup>-1</sup>	NA	25.4	14.1	13.5	12.1	18.5	16.7	2000	47.5 (40.2–58.0)
Cd	µg L <sup>-1</sup>	NA	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	10	NA
Pb	µg L <sup>-1</sup>	NA	3.2	1.2	4.1	0.9	1.6	2.2	5000	0.68 (0.6–0.8)
As	µg L <sup>-1</sup>	NA	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	100	NA

NA not available

<sup>a</sup> Adapted from (FAO 1985)

<sup>b</sup> Average (Min–Max) (Pham et al., 2019)

**Table 3** Volume and nitrogen contents in effluents in 2017

Treatment	Volume (L)	TN (mg L <sup>-1</sup> )				
		May	Jun.	Jul.	Aug.	Sep.
No. 1	385	4.6	8.3	9.3	2.3	1.4
No. 2	155	5.9	0.7	5.9	1.2	0.9
No. 3	390	23.6	23.8	14.6	13.1	20.4

from 7.0 to 7.6. The ORP varied from 197 to 279 mV. The average TN ranged from 27.8 to 40.0 mg L<sup>-1</sup>, and TP fluctuated between 0.18 and 0.44 mg L<sup>-1</sup>. NH<sub>4</sub>-N was the dominant component of nitrogen in the TWW throughout the period of experiment, as the result of nitrification control in the wastewater treatment process for phosphorus removal. The concentrations of trace metals in TWW complied with the regulation on water quality for agriculture (FAO 1985). The similar characteristics of irrigation water were observed in 2016 (Pham et al. 2019). The average TN and TOC in tap water were lower than 0.1 and 0.6 mg L<sup>-1</sup>, respectively (Table 3).

Table 4 shows the basic elements in the soil prior to the experiments. The TN and TP in the soils varied from 1.3 to 1.7 g kg<sup>-1</sup> and from 0.5 to 1.2 g kg<sup>-1</sup>, respectively. The concentrations of heavy metals in the soils were comparable, except for Cu. The contents of Cu in the old soils (used for treatment no. 2 in 2016 and treatments no. 1, no. 2, and no. 3 in 2017) were significantly higher than in new soils (used for treatments no. 1, no. 3, and no. 4 in 2016 and treatment no. 4 in 2017). This was attributed to the oxidation of copper cables used for the experiment in 2015 (Pham et al. 2019).

### Grain yield, grain quality, and whole plant dry biomass

The yield and protein content of rice grains, in addition to the dry mass of the complete rice plant, is provided in Table 5.

Generally, for the treatments irrigated with TWW, the 2016 season exhibited higher values of rice yield, rice grain protein, and rice plant dry mass than the 2017 season. In contrast, the rice yield and dry mass of the control were higher in the 2017 season compared with the 2016 season, indicating that reductions in the rice yield, rice protein content, and plant dry mass in the treatments using TWW were not due to climatic differences. Instead, the P remaining in the soil from the 2015 season must have been used effectively during the 2016 season (Table 4).

### Soil redox potential during cultivation

Soil Eh of the four treatments during the growing periods of 2016 and 2017 are illustrated in Fig. 2. The soil redox potentials ranged from -210 to +225 mV in the 2016 season, and from -160 to +500 mV in the 2017 season. Data for the first 3 weeks of the 2016 season are absent due to late preparation for the measurement. In general, soil Eh decreased gradually after the beginning of the season until MSD, when the fields were drained. After MSD, when the soil was re-flooded again, the soil redox potential continued to decrease until the end of the season. In the 2016 season, the soil redox potential values in most treatments were less than 0 mV.

### Seasonal variation of CH<sub>4</sub> emission

Seasonal variations of CH<sub>4</sub> emission fluxes from the paddy fields in 2016 and 2017 are illustrated in Fig. 3. In 2015, CH<sub>4</sub> emissions were not observed, which is likely due to the inhibitory effect of high copper levels in the soil (Pham et al., 2017). After transplantation, the paddy soil was maintained in a flooded state with a 5-cm water depth, and the CH<sub>4</sub> emissions gradually increased with the development of soil-reductive conditions and plant growth until week 7, immediately prior to MSD. After MSD with no measurement of

**Table 4** Basic elements in the soils before the experiments

Parameters	Units	2016		2017	
		Treatments no. 1, no. 3, no. 4	Treatments no. 2	Treatments no. 1, no. 2, no. 3	Treatment no. 4
TN	g kg <sup>-1</sup>	1.3	1.4	1.7	1.6
TP	g kg <sup>-1</sup>	0.7	1.2	0.5	0.7
TC	g kg <sup>-1</sup>	21.5	17.1	21.5	28.3
Cu	mg kg <sup>-1</sup>	17.7	97.1	59.6	16.7
Cr	mg kg <sup>-1</sup>	20.5	28.5	34.3	38.5
Zn	mg kg <sup>-1</sup>	98.0	119.2	88.5	82.4
Cd	mg kg <sup>-1</sup>	0.06	0.07	0.12	0.14
Pb	mg kg <sup>-1</sup>	12.8	16.1	16.6	18.4
As	mg kg <sup>-1</sup>	1.5	3.3	2.0	2.6

**Table 5** Crude protein, grain yield, and biomass

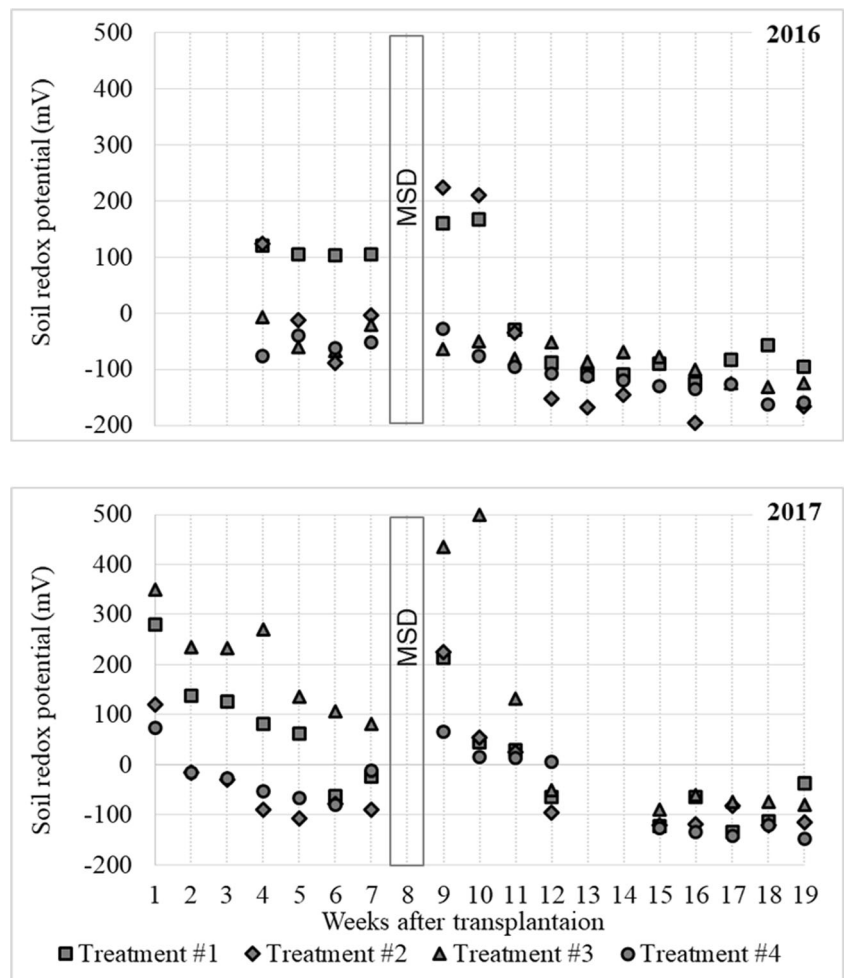
Treatment	Rice yield (t/ha)		Protein content (%)		Dry biomass (t/ha)	
	2016*	2017	2016*	2017	2016*	2017
Treatment no. 1	14.1a	10.4a	13.5ab	12.8ab	15.5a	13.2a
Treatment no. 2	12.3a	9.1a	14.6a	12.3bc	15.0a	11.1a
Treatment no. 3	10.3a	9.4a	13.2ab	10.3c	11.8a	10.4a
Treatment no. 4	9.0a	10.1a	10.5c	10.4c	10.4b	11.7a

\*Pham et al., 2019

GHG, the paddy fields were re-flooded, and CH<sub>4</sub> emission rates increased gradually again, reaching a peak during the heading stage (week 12), before finally dropping to the same level as the initial stage during the late ripening stage (weeks 18 and 19). The emission fluxes of CH<sub>4</sub> from all treatments varied from 0.03 to 0.52 mg m<sup>-2</sup> h<sup>-1</sup> in 2016 and from 0.01 to 7.69 mg m<sup>-2</sup> h<sup>-1</sup> in 2017, which were much lower than those reported from normal paddy fields (S. Yang et al. 2012; Win et al. 2013; Liu et al. 2015; Riya et al. 2015). Interestingly, the 2017 season control revealed

that CH<sub>4</sub> emissions began to increase sharply from week 12 until a peak flux of 7.69 mg m<sup>-2</sup> h<sup>-1</sup> in week 17. This flux was much greater than in the other growing stages and treatments throughout the experiment. Seasonal CH<sub>4</sub> average emission fluxes in treatments no. 1, no. 2, no. 3, and no. 4 were 0.12, 0.14, 0.15, and 0.16 mg m<sup>-2</sup> h<sup>-1</sup> in 2016 and 0.12, 0.09, 0.12, and 2.06 mg m<sup>-2</sup> h<sup>-1</sup> in 2017, respectively (Table 6). There was no significant difference in emission fluxes between treatments, except for treatment no. 4 in 2017.

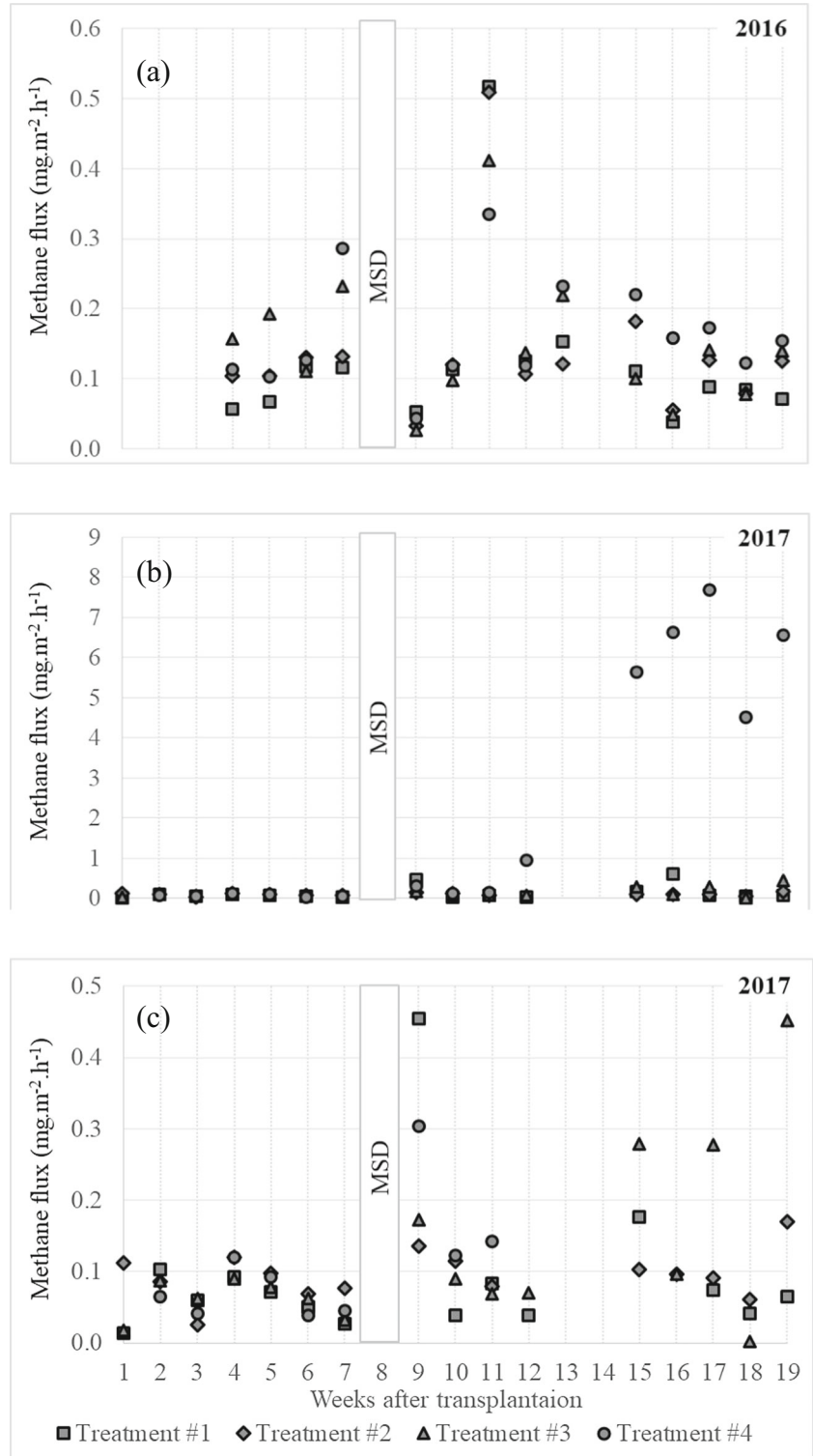
**Fig. 2** Variations in the soil redox potential in paddy soils (mV)



**Table 6** CH<sub>4</sub> average emission fluxes (mg m<sup>-2</sup> h<sup>-1</sup>)

Treatment	Treatment no. 1	Treatment no. 2	Treatment no. 3	Treatment no. 4
2016	0.12	0.14	0.15	0.16
2017	0.12	0.09	0.12	2.06

**Fig. 3** Seasonal variations in the methane emission flux (mg m<sup>-2</sup> h<sup>-1</sup>) from paddy fields: **a** in the 2016 season, **b** in the 2017 season with full range of Y-axis, and **c** in the 2017 season with short range of Y-axis to show the differences between treatments clearly



**Table 7** N<sub>2</sub>O average emission fluxes ( $\mu\text{g m}^{-2} \text{h}^{-1}$ ) in the 2017 season

Treatment	Treatment no. 1	Treatment no. 2	Treatment no. 3	Treatment no. 4
2017	180	58	545	40

### Seasonal variation of N<sub>2</sub>O emission

Figure 4 shows the seasonal variations in nitrous oxide emission fluxes from the paddy fields in 2017. Generally, TWW irrigation on the surface of the field led to significantly increased N<sub>2</sub>O emissions compared with those irrigated from the bottom of the field or using tap water with synthetic fertilizers. For the first week following rice transplanting, emission fluxes from all treatments were similarly low. Subsequently, in the greening and tillering stages, emission fluxes for TWW irrigation from the bottom at a lower flow rate (treatment no. 2) and in the control treatment remained low. Conversely, emissions increased slightly in subsurface TWW irrigation at a higher flow rate before reaching a peak in week 4 and increased sharply in surface TWW irrigation before reaching a peak in week 5 after transplanting. The highest emission fluxes of treatments no. 1, no. 2, no. 3, and no. 4 were 713, 271, 1578, and 261  $\mu\text{g m}^{-2} \text{h}^{-1}$ , respectively. After MSD in week 8, N<sub>2</sub>O emission fluxes from all treatments were still high, but then decreased gradually until harvesting. In the control, from week 16 at the start of the ripening stage, no emissions were observed. The average seasonal N<sub>2</sub>O emissions over the entire rice season of surface TWW irrigation were significantly higher than those of bottom-to-top irrigation and the control (Table 7). In subsurface irrigation, treatment no. 1 with a higher flow rate produced a larger amount of N<sub>2</sub>O than treatment no. 2, although the difference was not statistically significant.

### Combined GWP of CH<sub>4</sub> and N<sub>2</sub>O emissions

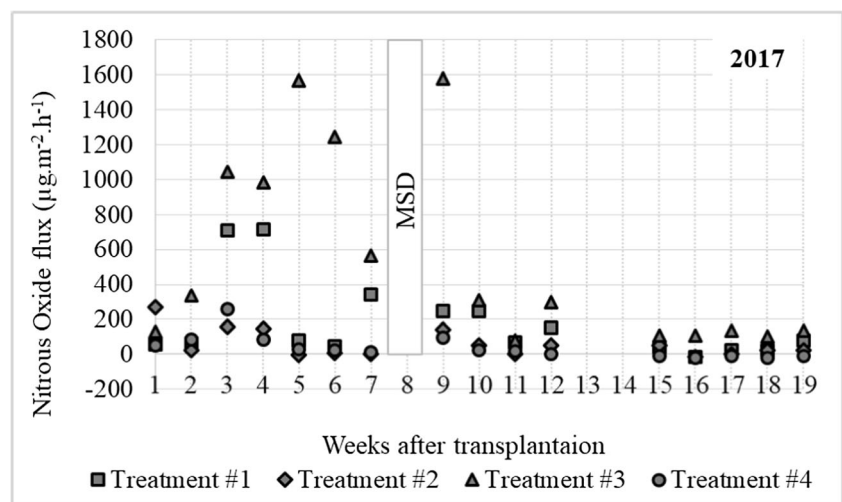
Compared with tap water irrigation or top-to-top TWW irrigation, TWW irrigation from the bottom of the field significantly decreased the net GWP for 20-year or 100-year periods (Table 8). In comparison with synthetic fertilizer application, top-to-top TWW irrigation decreased the net GWP slightly by 7.0% over the 20-year period but significantly increased the net GWP by 57.6% over the 100-year period. For the treatments with bottom-to-top irrigation, a lower irrigation flow rate contributed to a lower GWP (IPCC 2013b).

### Discussion

#### Effect of sub-irrigation of treated municipal wastewater on rice yield and quality

The rice yield and quality data for the 2016 season was discussed in detail in previous research (refer to Pham et al., 2019); therefore, this section only discusses data from the 2017 season. Rice yield in the control treatment in 2017 was higher than that obtained in 2016, indicating better climatic conditions for rice development in the 2017 season. Therefore, the decrease of rice yield observed for other treatments in this season could not be attributed to the climatic conditions. Compared with top-to-top irrigation, bottom-to-top irrigation at the same flow rate resulted in a 10.2% higher rice yield and a 24.3% higher protein content in brown rice. A

**Fig. 4** Seasonal variations in nitrous oxide emission fluxes ( $\mu\text{g m}^{-2} \text{h}^{-1}$ ) from paddy fields in the 2017 season





**Table 8** Net GWPs of CH<sub>4</sub> and N<sub>2</sub>O emissions over the growing season (kg CO<sub>2</sub>-eq ha<sup>-1</sup>) in 2017

Treatment	Treatment no. 1	Treatment no. 2	Treatment no. 3	Treatment no. 4
20-year period	1863	757	4995	5367
100-year period	1639	608	4807	2036

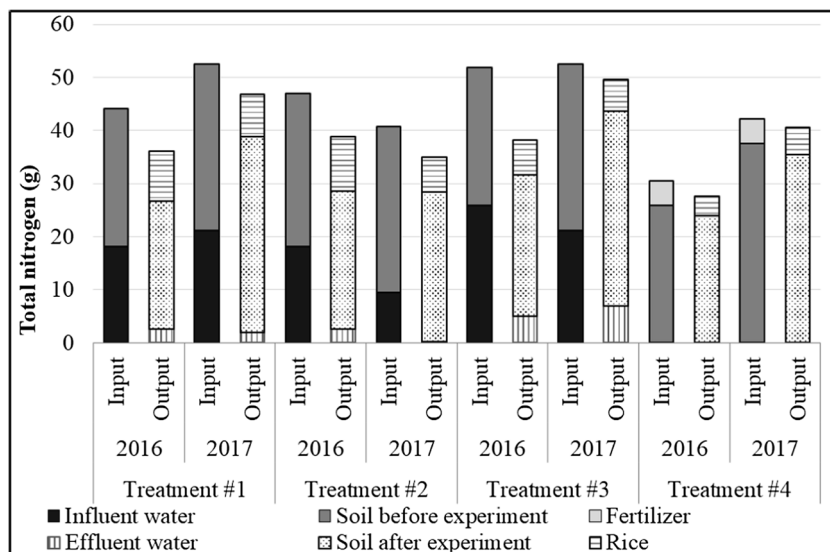
comparison between treatments no. 1 and no. 2, which applied the same irrigation flow direction without fertilization, indicated that a higher flow rate increased the rice yield and rice protein content by 14.3% and 8.5%, respectively. In treatment no. 1, the rice yield and quality decreased in the 2017 season; however, these decreases were not statistically significant, implying that upward TWW irrigation at a high flow rate can produce high rice yield and quality without any fertilizer, even when the soil is used repeatedly. However, long-term experiments should be conducted to verify the rice yield and quality when the paddy soil is irrigated with TWW from the bottom of the field with no fertilization.

### Effect of TWW irrigation on GHG emissions

Zou et al. (2009) reported that, in comparison with river water irrigation, sewage irrigation significantly increases CH<sub>4</sub> emissions from paddy fields. Several explanations were provided for this result. Firstly, sewage is rich in organic matter, which can promote CH<sub>4</sub> production (Zou et al. 2005). Secondly, wastewater irrigation may change the condition of soil physiochemical properties and bacterial communities that encourage CH<sub>4</sub> production. However, the results of the present study are not consistent with their findings. TWW irrigation led to remarkably decreased CH<sub>4</sub> emissions, in comparison with tap water irrigation. This could be attributed to the inhibitory effect of nitrogen in TWW on CH<sub>4</sub> formation. TWW was rich in nitrogen, and denitrification is generally believed to occur

before methanogenesis. Moreover, denitrification intermediates such as NO<sub>2</sub><sup>-</sup> and NO may inhibit methanogenic microorganisms, resulting in reduced CH<sub>4</sub> formation (Chen and Lin 1992). During the phase of reduction of NO<sub>3</sub><sup>-</sup>, NO<sub>2</sub><sup>-</sup>, and N<sub>2</sub>O, the partial pressure of H<sub>2</sub> may decrease, reducing the concentrations required to support CH<sub>4</sub> production (Bao et al. 2016). As shown in Table 2, the dominant form of nitrogen in the TWW used for irrigation was ammonium. If TWW dominated by nitrate is used, further reduction of CH<sub>4</sub> emission is expected as the result of enhanced denitrification.

In normal paddy fields, the peak flux of CH<sub>4</sub> emissions typically occurs during the early stage of rice development (Zou et al. 2009; S. Yang et al. 2012). In contrast, in this study, the peak CH<sub>4</sub> emissions occurred during the ripening stage of the rice plant (Fig. 5). This shift was likely caused by the above inhibitory effect of nitrogen on CH<sub>4</sub> production; a lower concentration of nitrogen in soil could promote CH<sub>4</sub> production in the final stage of rice growth. Furthermore, this rise may have been due to an increase in the available C from root exudates in the reproductive stage. A rice plant during flowering to ripening stages offers more exudates for the methanogenic microbial community (Singh and Dubey 2012). Furthermore, the increase in CH<sub>4</sub> emission rates at the flowering stage is associated with the improved CH<sub>4</sub> transport capacity of the rice plants due to well-developed aerenchyma tissues (Gaihre et al. 2014). Methane emission from the control treatment in 2017 was greater than in 2016, which is likely

**Fig. 5** Overall mass balance of nitrogen in the experiment (g)

a result of the greater concentration of C in the soil in 2017 (426 and 605 g/g in 2016 and 2017, respectively).

Conversely, TWW irrigation significantly increased N<sub>2</sub>O emissions when compared with tap water irrigation, especially when top-to-top irrigation or bottom-to-top irrigation at a higher flow rate was applied. This could be because high nitrogen contained in the TWW may have enhanced nitrification and denitrification processes in the soil and consequently increased N<sub>2</sub>O emissions. The larger amount of nitrogen was released to the air in treatments no. 1, no. 2, and no. 3 than the control (Fig. 2), although it contained both N<sub>2</sub>O and N<sub>2</sub>. Alternatively, carbon is a key factor controlling soil nitrification and denitrification processes; thus, organic matter in the TWW acted as carbon sources to enhance nitrification and denitrification processes in the soil, resulting in increased N<sub>2</sub>O emissions (Ndour et al. 2008). The TOC concentration in TWW was not so high (Table 2) but, under continuous irrigation, it must have supplied a considerable amount of carbon into the soil.

Hou et al. (2000) reported that N<sub>2</sub>O emissions are strongly influenced by the soil redox potential and that less N<sub>2</sub>O is emitted under low redox potential conditions, probably due to the reduction of N<sub>2</sub>O to N<sub>2</sub>. In the 2017 season, both a higher soil redox potential (Fig. 2) and notably greater N<sub>2</sub>O emissions were recorded in treatment no. 3, especially before MSD, in comparison with the other treatments. Moreover, in the top-to-top irrigation (treatment no. 3), N<sub>2</sub>O can be produced in the nitrification process in the surface water regardless of soil redox potential at the deep soil layer (Kampschreur et al. 2009). The ammonium-dominant TWW used for irrigation (Table 2) should have contributed to N<sub>2</sub>O emission in this pathway. For the same reason, nitrate-dominant TWW seems to reduce N<sub>2</sub>O emission in the top-to-top irrigation, while the emission may increase due to the enhanced denitrification in the bottom-to-top irrigation. Although it is known that rice plants can uptake NH<sub>4</sub><sup>+</sup> more easily than NO<sub>3</sub><sup>-</sup> (Fried et al. 1965), the dependency of GHG emission from paddy fields on the nitrogen form in the TWW should be investigated in further studies.

## Conclusions

Bench-scale experiments were conducted under tap water and TWW irrigation conditions in order to quantify the effect of TWW irrigation on rice yield and CH<sub>4</sub> and N<sub>2</sub>O emissions. The main findings are as follows:

- A high rice yield and protein content was achieved over two cultivation seasons by bottom-to-top TWW irrigation at a high flow rate without any fertilizers.
- TWW irrigation decreased CH<sub>4</sub> emissions, but increased N<sub>2</sub>O emissions, resulting in a lower combined GWP than tap water irrigation.

- Bottom-to-top TWW irrigation produced less CH<sub>4</sub> and N<sub>2</sub>O than top-to-top TWW irrigation.

**Authors' contributions** Conceiving experiment: Dong Pham Duy, Toru Watanabe.

Conducting experiment: Dong Pham Duy, Agnes Hoki Briliana Suhono, Nobuo Kaku, Shuhei Masuda, Fumiaki Takakai.

Analyzing the data: Dong Pham Duy.

Preparing the manuscript draft: Dong Pham Duy.

Revising the manuscript: all the authors

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**Data availability** Available.

## Compliance with ethical standards

**Competing interests** The authors declare that they have no competing interests.

**Ethics approval, consent to participate and publish** This manuscript is the original work of authors, which has not been previously published elsewhere.

The manuscript reflects the authors' own research and analysis in a truthful and complete manner.

The manuscript properly credits the meaningful contributions of co-authors.

The results are appropriately placed in the context of prior and existing research.

All sources used are properly disclosed (correct citation).

All the authors have been personally and actively involved in substantial work leading to the paper, and will take public responsibility for its content.

**Abbreviations** *EC*, Electrical conductivity; *GHG*, Greenhouse gas; *MSD*, Midsummer drainage; *ORP*, Oxidation-reduction potential; *SPAD*, Soil plant analysis development; *TN*, Total nitrogen; *TWW*, Treated wastewater; *WWTP*, Wastewater treatment plant

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