

Influence of irrigation frequency on greenhouse gases emission from a paddy soil

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Abstract Water management is known to be a key factor on methane (CH₄), carbon dioxide (CO₂), and nitrous oxide (N₂O) emissions from paddy soils. A field experiment was conducted to study the effect of continuous irrigation (CI) and intermittent irrigation (II) on these emissions. Methane, CO₂, and N₂O emissions from a paddy soil were sampled weekly using a semi-static closed chamber and quantified with the photoacoustic technique from May to November 2011 in Amposta (Ebro Delta, NE Spain). Intermittent irrigation of rice paddies significantly stimulated (N₂O + N₂)–N emission, whereas no substantial N₂O emission was observed when the soil was re-wetted after the dry phase. The cumulative emission of (N₂O + N₂)–N was significantly larger from the II plots (0.73 kg N₂O–N ha⁻¹ season⁻¹, *P* < 0.05) than from the CI plots (–1.40 kg N₂O–N ha⁻¹ season⁻¹). Draining prior to harvesting increased N₂O emissions. Draining and flooding cycles controlled CO₂ emission. The cumulative CO₂ emission from II was 8416.35 kg CO₂ ha⁻¹ season⁻¹, significantly larger than that from CI (6045.26 kg CO₂ ha⁻¹ season⁻¹, *P* < 0.05). Lower CH₄ emission due to water drainage increased CO₂ emissions. The soil acted as a sink of CH₄ for both types of irrigation. Neither N₂O–N nor CH₄ emissions were affected by soil temperature. Global warming potential was the highest in II (4738.39 kg CO₂ ha⁻¹) and the lowest in CI (3463.41 kg CO₂ ha⁻¹). These findings suggest that CI can significantly mitigate

the integrative greenhouse effect caused by CH₄ and N₂O from paddy fields while ensuring the highest rice yield.

Keywords Water management · Intermittent irrigation · Continuous irrigation · Ammonium sulfate

Introduction

Global warming induced by increasing greenhouse gases (GHG) concentration in the atmosphere is a matter of great environmental concern. Methane (CH₄), carbon dioxide (CO₂), and nitrous oxide (N₂O) are important long-living GHG, which have attracted considerable attention during the last decades because of their contribution to global warming. The agroecosystem plays a significant role in the global budget of GHG (Hou et al. 2012). Agriculture is responsible for about 50 % of the global anthropogenic CH₄, and for 60 % of N₂O (IPCC 2007), and can be an important source of trace gases or can act as a major sink. Agricultural CH₄ and N₂O emissions have increased by nearly 17 % from 1990 to 2005 (IPCC 2007), and agricultural N₂O emissions are predicted to increase between 23 and 60 % by 2030 due to increased chemical and manure nitrogen inputs (FAO 2010). Paddy fields are considered to be an important source of anthropogenic CH₄, N₂O (Hou et al. 2012), and CO₂ (Liu et al. 2013). Atmospheric CH₄ from rice fields will further increase with the increasing rice harvested area in the years to come (Cai et al. 2007).

Rice is the staple food of nearly 50 % of the world's population. Rice planting areas account for about 20 % of the world total. In Spain, rice covers about 3 % of the Spanish irrigated area (about 11,0785 ha rice) (<http://www.magrama.gob.es/es/estadistica/temas/estadisticas-agrarias/>

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[agricultura/esyrc/e/](#), accessed 12 January 2015). Hence, it is interesting to study CH₄ and N₂O emissions from Spanish paddy fields.

Water management has been recognized as one of the most important practices that affect CH₄, CO₂, and N₂O emission from paddy fields (Xiong et al. 2007; Liu et al. 2010; Hadi et al. 2010; Hou et al. 2012). In Spain, the most typical water management of paddy fields involves continuous flooding to improve rice growth and increase yields. The influence of water management on CH₄ and N₂O emission from paddy fields under continuous flooding and intermittent irrigation has been well documented in the climatic conditions of China, Japan, and India (Panthak et al. 2003; Zheng et al. 2004; Zheng et al. 2006; Hadi et al. 2010; Hou et al. 2012; Suryavanshi et al. 2013) but not in the Mediterranean climate, especially in the Ebro Delta (Spain). Continuously flooded rice fields have a high potential for CH₄ emission, while N₂O emissions are negligible. However, Nugroho et al. (1994) reported that intermittent drainage did not affect CH₄ emission or, inversely, sometimes resulted in higher CH₄ emission than continuously flooded soil in Indonesia. Nitrous oxide emissions during intermittent irrigation depend greatly on whether or not water logging is present in paddy soils (Zou et al. 2005). Intermittent drainage has been proposed as a water management technique to reduce CH₄ emission from paddy soils; it is also useful for the removal of hazardous organic components in rice rhizosphere and to increase the availability of some nutrients (Hadi et al. 2010). Intermittent irrigation has been shown to mitigate CH₄ emissions compared with continuous flooding (Zou et al. 2009; Liu et al. 2010). Draining a paddy soil prior to harvesting increased N₂O emissions (Hadi et al. 2010).

Intermittent drainage can have a strong effect on soil CO₂ emissions, increasing them considerably (Miyata et al. 2000; Saito et al. 2005). However, the mechanism of CO₂ exchange between rice paddies and the atmosphere is not fully understood (Miyata et al. 2000). For example, using eddy covariance measurements, Miyata et al. (2000) found a significantly larger net CO₂ flux from rice paddy soil to atmosphere when the field was drained compared to when it was flooded. These differences in the CO₂ flux were mainly due to increased CO₂ emissions from the soil surface under drained conditions resulting from the removal of the diffusion barrier caused by floodwater. The existence of floodwater, anaerobic soil, and changes in the micro-meteorological environment influence root activity, photosynthesis, and respiration of the rice plant (Liu et al. 2013).

Soil properties (such as soil moisture, soil oxygen status, soil redox potential, and soil temperature) and microbial activity in continuous irrigation (CI) paddy fields are very different from those in intermittent irrigation (II) rice fields,

and induce changes in CH₄, CO₂, and N₂O emissions. To our knowledge, very few studies have been made on the quantification of CH₄, CO₂, and N₂O emissions from CI and II paddy fields in Spain. In the present study, the CH₄, CO₂, and N₂O–N and (N₂O + N₂)–N emissions from paddy fields with two types of water management (CI and II) were measured during one cropping season (from May to November 2011). The aim of this study was to determine the effect of irrigation frequency (continuous irrigation and intermittent irrigation) on the emission of greenhouse gases (CH₄, CO₂, N₂O–N), N₂, and the global warming potential (GWP) from a paddy soil at the Ebro Delta (NE Spain) to know the environmentally sound irrigation practice.

Materials and methods

Experimental design

The experiment was conducted in 2011 on experimental plots at the Institute for Food and Agricultural Research and Technology (IRTA), in its Amposta station (40°39'19.02"N, 00°47'01.2"W), Catalonia, Spain. This region has a Mediterranean climate with an average annual temperature of 17 °C and a mean annual precipitation of 550 mm. Soil texture in the experimental site is silty clay loam (3.5 % total sand, 61.7 % total silt, and 34.8 % total clay), which is representative of the soil type in this region. The main (0–20 cm depth) properties of this soil are 2.34 % organic matter, 3 mg NO₃[−]–N kg^{−1}, 6 mg NH₄⁺–N kg^{−1}, 44 mg total P–Olsen kg^{−1}, 158 mg total K kg^{−1}, and pH 8.1. The experiment involved two irrigation treatments: continuous irrigation (CI) and intermittent irrigation (II). Each treatment had three replicates established in a randomized block design in six plots of an approximate size of 30 m² (5 × 6 m) each. Irrigation started on 15th March and the field was flooded until 1st June.

In the CI treatment, a water layer of 10 cm was kept until 12th September when the field was drained. Harvesting took place on 20th September. In the II treatment, soil was flooded as in the continuous flooding system (10 cm water layer), but the irrigation was suspended until the water layer disappeared, at that moment it was irrigated again. Ten irrigations were done during the growing season to the II treatment, and each irrigation lasted for approximately 1 day. The final drainage was applied on 12th September and harvesting took place on 16th September. In Spain, rice is permanently flooded for most of the growing season, although short drainage periods may be needed to correct specific crop disorders dependent on herbicide application, etc. Rice (*Oryza sativa* L.) cultivar *Gleva* was

sown directly on site on 9th May in both treatments (II and CI) at a density of 170 kg ha⁻¹. Fertilizers, herbicides, and pesticides were applied in accordance with local conventional practice. To minimize the impact of weeds on yield, Molinate and Bentazona herbicides combined with cultural practices were used. Puddling before sowing and weeding by hand (until the rice plant has become so tall that cannot be weeded without damage) were performed, especially for the so-called wild rice, red rice or “crodo” as it is called in Italy. Two applications of fungicide (Benzotiazol at a dose of 300 g ha⁻¹) were done on 26th July and on 19th August. Nitrogen fertilizer was applied at a rate of 170 kg N ha⁻¹ as ammonium sulfate (21 % N richness). To improve N utilization, N was applied 33 % on the 6th May, 33 % on the 10th June, and 33 % on the 12th July. Phosphorous and K were applied on 6th May to all the plots, at 57 kg P₂O₅ ha⁻¹, and 57 kg K₂O ha⁻¹.

Sampling and measurements

Gas samples were collected using the closed chamber method at an interval of 7 days throughout the period of rice growth (Peng et al. 2011; Hou et al. 2012). The cylindrical (20-cm diameter and 60-cm high) static chamber was made of polyvinyl chloride (PVC) coated with an epoxy resin and was inserted 18 cm into the soil each sampling day on the 6 plots. This cylinder was closed with a vented screwed lid with a three-way key. Air samples from inside the chamber were taken in duplicate immediately after closing the chamber, and 20 and 40 min later. Samples were taken through a Teflon tube connected to the three-way key and into 100-ml plastic syringes adapted with a valve. Before sampling, air within the chamber was mixed by filling and emptying the syringe six times before withdrawing the sample. After taking the gas sample, the syringes were closed by the valve. After 40 min of sampling, the three-way keys were left open until the sampling with acetylene.

The acetylene (C₂H₂) inhibition method (Balderston et al. 1976; Yoshinari and Knowles 1976) was used to inhibit the last step of denitrification (N₂O–N₂). Ten percent (v/v) of the air enclosed in the chamber was replaced by C₂H₂. After C₂H₂ was allowed to diffuse into the soil for 20 min, samples were taken as described in the previous paragraph. After 40 min of sampling, the three-way keys were left open until the following sampling and the chambers were removed from the field and cleaned properly with water.

The syringes were transported to the laboratory and the concentrations of N₂O, CO₂ and CH₄ in the sampled air were analyzed using the photoacoustic technique (Innova 1312 Photoacoustic Multigas Monitor). The N₂O, CO₂, and

CH₄ emission fluxes were determined from the linear increase of gas concentration at each sampling time (0, 20, and 40 min) during the time of chamber closure. The cumulative emission throughout the study period was calculated by integrating the emission curve through time. During N₂O, CO₂, and CH₄ emission monitoring, soil temperature at a depth of 5 cm was determined by means of a thermometer. In this study, water-filled pore space (WFPS) was calculated according to the following equation (Peng et al. 2011):

$$\text{WFPS (\%)} = (\text{Gravimetric water content (\%)}) / (\text{Total soil porosity} \times \text{Soil bulk density} \times 100),$$

where total soil porosity = (1 – soil bulk density)/2.65, with 2.65 g cm⁻³ as the assumed particle density of the soil (Porta et al. 2008).

Nitrogen gas emission was obtained by subtracting N₂O emission without acetylene from N₂O emission with acetylene (Ryden et al. 1979), and then the N₂O–N/(N₂O + N₂)–N ratio was calculated.

Since the chambers were not transparent, it cannot be assumed that the CO₂ flux was the net flux, as photosynthesis was ignored.

Global warming potential (GWP)

Global warming potential (GWP) is an index defined as the cumulative radiative forcing between the present and some chosen later time ‘‘horizon’’ caused by a unit mass of gas emitted now. In GWP estimation, CO₂ is typically taken as the reference gas, and an increase or reduction in emission of CH₄ and N₂O is converted into ‘‘CO₂-equivalents’’ through their GWPs. The GWP for CH₄ (based on a 100-year time horizon) is 25, whereas that for N₂O is 298, when the GWP value for CO₂ is taken as 1 (IPCC 2007). GWP of CH₄, N₂O, and CO₂ emissions was calculated using the following equation (IPCC 2007): GWP = cumulative CO₂ emission + cumulative CH₄ emission × 25 + cumulative N₂O emission × 298.

Statistical analysis

Statistical analyses of data were carried out using the JMP ver. 10 (SAS Institute Inc., Cary, USA). Daily emission fluxes, as well as the estimated seasonal emission data, were checked for normal distribution. A *t* test was used to examine the statistical significance of the parameter estimates. An analysis of variance (ANOVA) *F* test partitioned the total variation of the results. A one-way ANOVA was used to test whether the cumulative emissions depended on the water regime.

Results and discussion

The coefficients of variation (CV%) of N_2O -N and $(\text{N}_2\text{O} + \text{N}_2)$ -N ranged from 199 to 477 %. Such high uncertainties are unfortunately common in this kind of study.

Grant and Pattey (2003) obtained coefficients of variation from 30 to 100 %. Our CV was in the same range as that reported by Flessa et al. (1995) and Thornton et al. (1996) (CV > 150 %), but lower than described by Teira-Esmatges et al. (1998). A review on N_2O studies with a much greater number of replicates than the one reported here was done by Mosier et al. (1996), revealing that it is not the measurement technique that provides most of the uncertainty in N_2O flux values in the literature but rather the diverse combination of physical and biological factors that control gas fluxes.

Nitrous oxide and molecular nitrogen emission

The measured N_2O fluxes differed considerably from day to day, probably due to the changes in soil temperature and water content. The measured emission fluxes ranged from -71 to $384 \text{ g } (\text{N}_2\text{O} + \text{N}_2)\text{-N ha}^{-1} \text{ day}^{-1}$ in CI, and from -113 to $526 \text{ g } (\text{N}_2\text{O} + \text{N}_2)\text{-N ha}^{-1} \text{ day}^{-1}$ in II (Fig. 1), which is broader than many of the ranges reported in the literature (Peng et al. 2011; Hou et al. 2012).

Emission from the soil depends on (Teira-Esmatges et al. 1998) (i) the formation of N_2O during denitrification and nitrification and its diffusion to the headspace, and (ii) the consumption of N_2O through its reduction to N_2 during denitrification and the diffusion rate of N_2O from the headspace into the soil. When N_2O consumption was larger than its emission, the N_2O concentration in the headspace

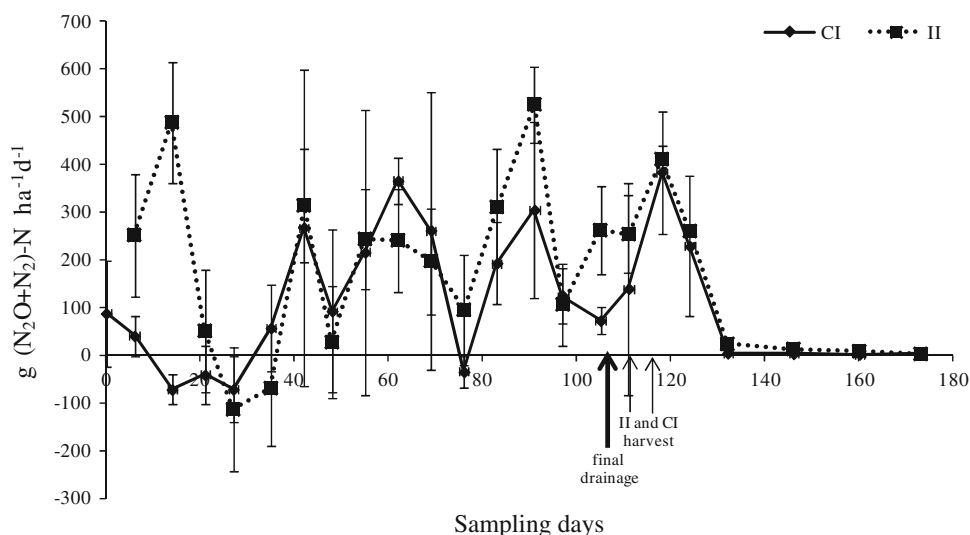
decreased resulting in negative fluxes of N_2O . Negative fluxes indicate that the soil acted as a sink for N_2O .

The evolution of the daily $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ fluxes from the CI is quite parallel to that from the II treatment (Fig. 1). The $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ emission fluxes from the II paddy plots were higher than those from the CI plots for most of the rice-growing season. The average daily flux of $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ in the II fields ($178.2 \text{ g ha}^{-1} \text{ day}^{-1}$) was 1.5 times higher than in the CI fields ($115.18 \text{ g ha}^{-1} \text{ day}^{-1}$). The highest peak of $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ emission flux from the II paddy fields ($526.44 \text{ g ha}^{-1} \text{ day}^{-1}$, 1.72 times that from the CI paddy fields) was observed on measurement day 91 in the physiological maturity period, which coincides with the day when the soil temperature was the highest ($27.0 \text{ }^\circ\text{C}$ in the CI plots and $27.8 \text{ }^\circ\text{C}$ in the II plots). Other four peaks of $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ emission flux from the II fields were observed (Fig. 1). In the CI plots, another three peaks of $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ emission were observed (Fig. 1). The peaks in II are mainly due to mineral fertilizer application (days 14 and 62 of sampling) and to the high soil water content, while peaks in CI are mainly due to the application of mineral fertilizer (day 62 of sampling) and to draining (days 42, 91 and 118 of sampling).

After fertilization, on measurement days 14 and 48, the $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ flux increased along with soil temperature, probably because the concentration of $\text{NH}_4^+\text{-N}$ and of $\text{NO}_3^-\text{-N}$ in soil solution increased. Toshiaki et al. (2007) reported that the development of denitrifying bacteria activity along with the rise of soil temperature might account for the high N_2O emissions.

Under CI, the $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ flux was lower than under II. One possible reason for this may be the dissolution of N_2O in the flooding water. Toshiaki et al. (2007) reported that the N_2O concentration in flood water was as high as between 0.65 and $10.4 \mu\text{g l}^{-1}$. He proposed that the

Fig. 1 Daily flux of $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ from the paddy soils under different water management practices through the sampling season. The vertical arrow on day 112 of measurement indicates the harvest of rice in the II treatment, and that on day 116 indicates the harvest of rice in the CI treatment. II intermittent irrigation, CI continuous irrigation. Vertical lines indicate the standard error of the average ($n = 3$)



$(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ emission can be mitigated considerably by a thin film of flooding water on paddy soils, which is already common practice in the Ebro Delta.

Intermittent irrigation clearly increased the $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ fluxes from the rice paddy. Smith and Patrick (1983) observed that alternate anaerobic and aerobic conditions considerably increased N_2O fluxes relative to continuous anaerobic and aerobic conditions, and that the net N_2O flux increased when the duration of both the anaerobic and aerobic periods was increased from 7 to 14 days.

In this experiment, the time between the disappearance of the floodwater layer and reflooding was usually about 1 week. The results are consistent with those reported by Cai et al. (1997) and Toshiaki et al. (2007) who showed that the soil water content associated with maximum N_2O emissions was normally close to field capacity. At about this soil water content, either nitrifiers or denitrifiers may be N_2O generators. Yagi et al. (1996) showed that water management with very short anaerobic–aerobic cycling induced a very low N_2O emission in a Japanese paddy field.

The results of the present study suggest that II of rice paddies significantly stimulated $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ emission. The cumulative emissions during the sampling period were 25.05 kg $(\text{N}_2\text{O} + \text{N}_2)\text{-N ha}^{-1}$ season⁻¹ in CI and 34.12 kg $(\text{N}_2\text{O} + \text{N}_2)\text{-N ha}^{-1}$ season⁻¹ in II (Table 1). The cumulative $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ emission from the II plots was 1.36 times greater than that from the CI plots. Felber et al. (2012) estimated that the total denitrification loss of nitrogen ($(\text{N}_2\text{O} + \text{N}_2)\text{-N}$) is in the range of 6–26 kg N ha⁻¹ year⁻¹, albeit with uncertainties close to 100 %.

The measured daily $\text{N}_2\text{O-N}$ emission fluxes cover a shorter range of emissions than the $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ emissions though broader than many of the ranges reported in the literature (Aulakh et al. 2001; Ma et al. 2012). They ranged from -119.66 to 116.07 g $\text{N}_2\text{O-N ha}^{-1}$ day⁻¹ in

CI, and from -114 to 79 g $\text{N}_2\text{O-N ha}^{-1}$ day⁻¹ in II (Fig. 2). Ma et al. (2012) and Hadi et al. (2010) also reported negative $\text{N}_2\text{O-N}$ fluxes. In CI, 48 % of the $\text{N}_2\text{O-N}$ flux measurements were negative, and 8.3 % were >100 g of $\text{N}_2\text{O-N ha}^{-1}$ day⁻¹. In II, 27 % of the $\text{N}_2\text{O-N}$ flux measurements were negative and 6.25 % were >50 g of $\text{N}_2\text{O-N ha}^{-1}$ day⁻¹. The drain applied before harvest determined increased $\text{N}_2\text{O-N}$ in both treatments, though this was much clearer in II (Fig. 2).

The cumulative $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ and $\text{N}_2\text{O-N}$ emissions from the studied irrigation types were significantly different. The cumulative $\text{N}_2\text{O-N}$ emissions during the sampling period were 0.73 kg $\text{N}_2\text{O-N ha}^{-1}$ season⁻¹ from II and -1.40 kg $\text{N}_2\text{O-N ha}^{-1}$ season⁻¹ from CI (Table 1). The cumulative $\text{N}_2\text{O-N ha}^{-1}$ emissions with similar management practices found by Hadi et al. (2010) were between -50.3 and -14.8 kg $\text{N}_2\text{O-N ha}^{-1}$ season⁻¹. Aulakh et al. (2001) reported cumulative emissions between 2 and 36.8 kg $(\text{N}_2\text{O} + \text{N}_2)\text{-N ha}^{-1}$ season⁻¹.

Ratio of $\text{N}_2\text{O-N}:(\text{N}_2\text{O} + \text{N}_2)\text{-N}$

The $\text{N}_2\text{O-N}:(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ ratio is an indicator of the extent to which denitrification proceeds to N_2 . In general, this ratio varied a lot indicating that the production of the two gases reacted differently to changes in conditions and that their production is partly independent. This is in agreement with what Ciarlo et al. (2007) reported.

The total N_2O produced in the presence of acetylene may be a good indicator of total denitrification; the production of N_2O in the absence of acetylene may not be solely a product of denitrification but may include nitrification and other aerobic processes as well. In the II plots, WFPS ranged between 73 and 93 % (Fig. 3), suggesting complete reduction of $\text{N}_2\text{O-N}_2$.

The cumulative $\text{N}_2\text{O-N}:(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ ratio was 0.055 for CI and 0.021 for II. The low $\text{N}_2\text{O-N}$

Table 1 Cumulative emission of $\text{N}_2\text{O-N}$, $(\text{N}_2\text{O} + \text{N}_2)\text{-N}$, CO_2 , CH_4 , GWP, rice yield and the statistical significance of the effect of soil temperature, irrigation type, and their interactions ($P < 0.05$; $n = 3$)

Treatment	$\text{N}_2\text{O-N}$ (kg ha ⁻¹ season ⁻¹)	$(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ (kg ha ⁻¹ season ⁻¹)	CO_2 (kg ha ⁻¹ season ⁻¹)	CH_4 (kg ha ⁻¹ season ⁻¹)	GWP (kg CO ₂ ha ⁻¹)	Rice yield (kg ha ⁻¹)
II	0.73 ± 0.08a	34.14 ± 0.35a	8416.35 ± 178.56a	-155.82 ± 1.81b	4738.39	6291.12 ± 682.14b
CI	-1.40 ± 0.1a	25.05 ± 0.67b	6045.26 ± 416.57b	-87.09 ± 6.69a	3463.41	9572.23 ± 156.08a
<i>ANOVA results</i>						
Soil temperature	n.s.	0.0006	0.0001	n.s.		
Irrigation (CI/II)	n.s.	0.0019	0.0150	0.0062		
Soil temp × irrigation	n.s.	n.s.	n.s.	n.s.		

Different letters, per column, indicate significantly different emissions using a *t* test ($P < 0.05$)

II intermittent irrigation, CI continuous irrigation, ns not significant

Fig. 2 Daily flux of N_2O –N from the paddy soils under different water management practices through the sampling season. The vertical arrow on day 112 of measurement indicates the harvest of rice in the II treatment, and that on day 116 indicates the harvest of rice in the CI treatment. II intermittent irrigation, CI continuous irrigation. Vertical lines indicate standard errors of three replications for each treatment

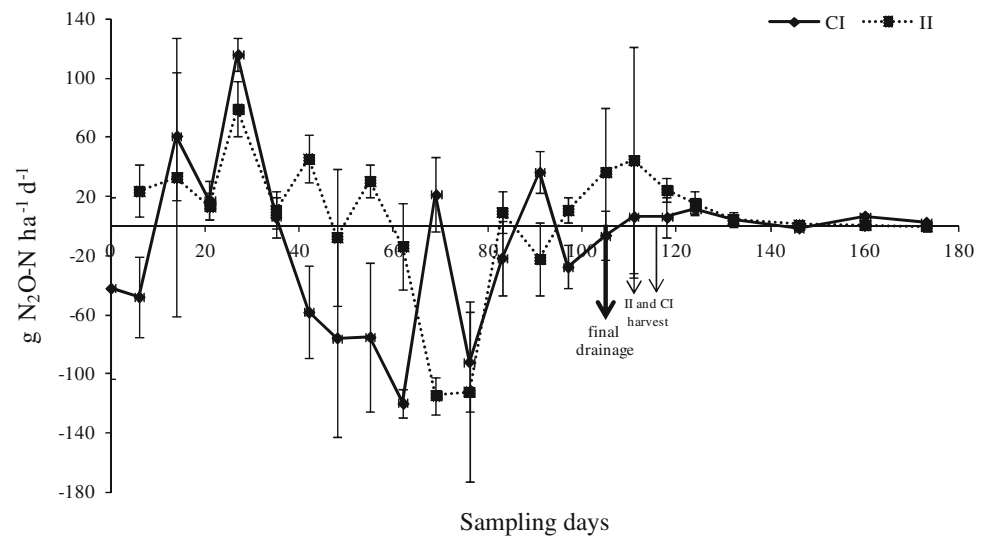
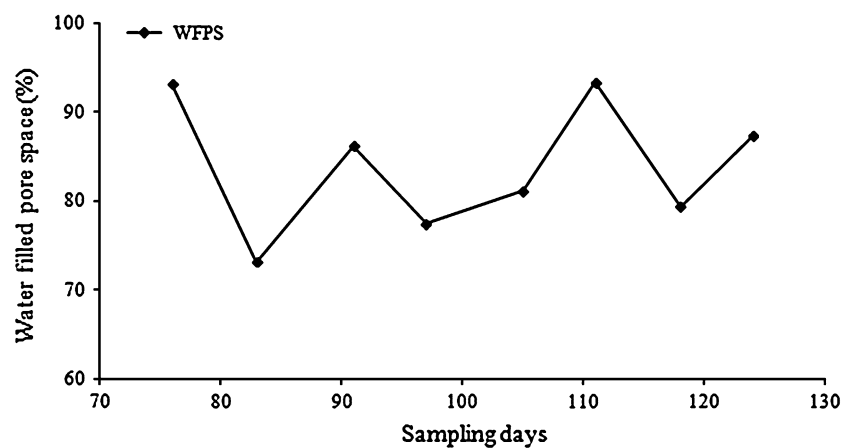


Fig. 3 Average water-filled pore space through the sampling season in the intermittent irrigation treatment (II)



$\text{N}:(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ ratio is probably due to stronger anaerobic conditions created by the presence of a surface water layer, which promoted N_2O reduction to N_2 , as suggested by Xu et al. (2004). In addition, the surface water layer probably limited N_2O upward diffusion, as suggested by Yan et al. (2000), and this probably stimulated N_2O reduction to N_2 . Another possible explanation would be the greater sensitivity of the N_2O reductase than the other denitrifying enzymes to oxygen (Knowles 1982). Ciarlo et al. (2007) found an average $\text{N}_2\text{O}\text{-N}:(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ ratio of 0.051–0.17 when WFPS was between 70 and 120 %. Other researchers have also noted that in continuously wet soil denitrification proceeds rapidly to N_2 and little N_2O is released (Abdirashid et al. 2003).

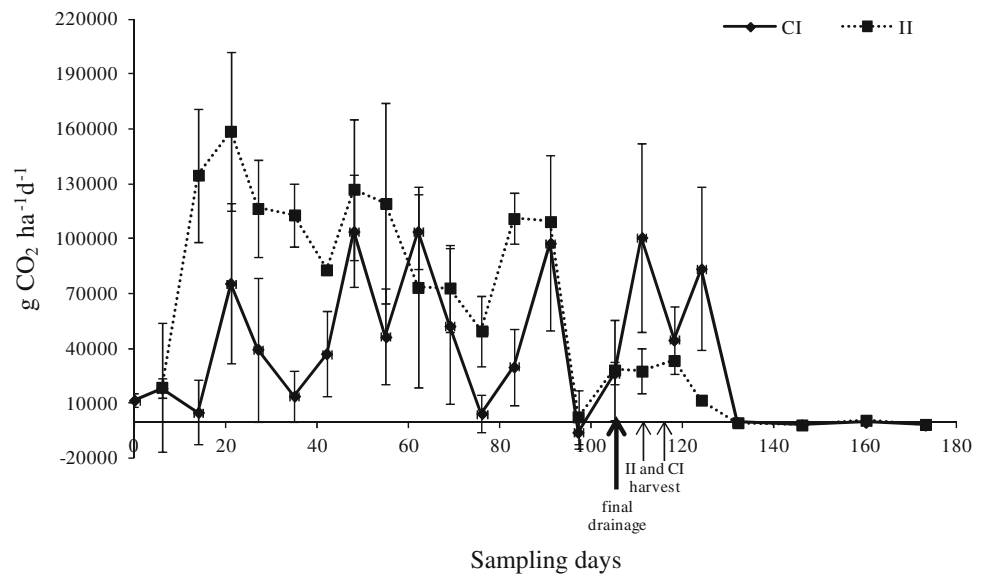
It is known that high soil nitrate content inhibits N_2O reduction to N_2 (Ciarlo et al. 2007). Knowles (1982) explained this phenomenon by stating that nitrate is preferred as an electron acceptor with respect to nitrous

oxide. However, neither N_2O nor the $\text{N}_2\text{O}\text{-N}:(\text{N}_2\text{O} + \text{N}_2)\text{-N}$ ratio was related to soil nitrate contents, probably because of the strong control that moisture exerted on these variables.

Carbon dioxide emission

The daily CO_2 flux is shown in Fig. 4. The measured emission fluxes ranged from -5664.0 to 104140.8 g CO_2 $\text{ha}^{-1} \text{day}^{-1}$ in CI, and from -1574.4 to 158923.2 g CO_2 $\text{ha}^{-1} \text{day}^{-1}$ in II, exhibiting a wide fluctuation during the rice-growing season. The results are consistent with those reported by Li et al. (2004), who found that rice fields may act as a sink for CO_2 , with net CO_2 fluxes of -2191.8 to 6849.3 kg CO_2 $\text{ha}^{-1} \text{day}^{-1}$. Liu et al. (2008) found a range of CO_2 emission fluxes from 6571.2 to 443760.0 mg CO_2 $\text{ha}^{-1} \text{day}^{-1}$. The cumulative CO_2 emissions during the sampling period were 8416.3 kg CO_2 $\text{ha}^{-1} \text{season}^{-1}$ from II and 6045.2 kg CO_2 $\text{ha}^{-1} \text{season}^{-1}$ from CI.

Fig. 4 Daily flux of CO₂ from the paddy soils under different water management practices through the sampling season. The vertical arrow on day 112 of measurement indicates the harvest of rice in the II treatment, and that on day 116 indicates the harvest of rice in the CI treatment. II intermittent irrigation, CI continuous irrigation. Vertical lines indicate standard errors of three replications for each treatment



The CO₂ fluxes during the rice seedling stage ranged from 5241.6 to 18403.2 g CO₂ ha⁻¹ day⁻¹ in CI, and from 17841.3 to 19012.8 g CO₂ ha⁻¹ day⁻¹ in II. The first drainage in II plots took place on 1st June (day 6 of sampling), until which every field had remained flooded. In both treatments, fluxes of CO₂ increased gradually until the mid-seedling stage when two peaks were clearly observed (on day 21). In II plots, the peak appears due to draining and mineral fertilization (ammonium sulfate), while in the CI plots the peak appears due only to the application of ammonium sulfate. In the first part of the tillering stage, two other peaks were observed (Fig. 4), associated also to fertilization (II and CI) and drainage (II).

Fertilization has shown contradictory effects on soil CO₂ flux: enhancement (Fisk and Fahey, 2001; Iqbal et al. 2009; Bhattacharyya et al. 2012), or no effect (Lee et al. 2007). Fertilizer application enhances biomass production and C input. Many experiments have shown a positive effect of N additions on soil organic carbon content, a consequence of the higher biomass generated which, in turn, returns the C in the form of CO₂ to the atmosphere (Iqbal et al. 2009). The results of this experiment are in agreement with those of Xiao et al. (2005) who reported an increased soil CO₂ flux in response to N fertilization from rice paddies in suburban Shanghai, China. The higher CO₂ flux as a response to fertilization can be interpreted in two ways. One interpretation is that the microbial use of C increases when applying nitrogen fertilizer (Fisk and Fahey 2001). Another is that microbial biomass assimilates carbon less efficiently and so respire a greater proportion of C when applying nitrogen fertilizers (Iqbal et al. 2009).

In II plots, the soil CO₂ fluxes increased immediately after flooding and exceeded pre-flooding values by two-thirds. This increase was abrupt (Fig. 4). Replacement of soil air by water must have caused an enriched CO₂ pulse. Subsequently, the CO₂ flux rapidly decreased after the water pulse. In the following days, CO₂ remained at minimum levels (about 2875.2 g CO₂ ha⁻¹ day⁻¹) during flooding. As standing water declined and eventually disappeared, the CO₂ fluxes gradually increased and finally reached maximum levels (about 158923.2 g CO₂ ha⁻¹ day⁻¹). This indicates that draining and flooding cycles play vital roles in controlling CO₂ emissions in paddy soils.

Average soil CO₂ daily flux under flooded conditions was 38651.66 g CO₂ ha⁻¹ day⁻¹ whereas under drained conditions it was 6382.16 g CO₂ ha⁻¹ day⁻¹. It is likely that floodwater decreased topsoil diffusivity and may have decreased soil CO₂ fluxes (Maier et al. 2010). Reduction of oxidizing activity under anoxic conditions may be another reason for low soil CO₂ fluxes during the flooding period (Kogel-Knabner et al. 2010). Miyata et al. (2000), Cai et al. (2003), and Liu et al. (2013) found that the water content of paddy soils had a strong effect not only on CH₄ emissions but also on CO₂ emissions. Lower CH₄ emissions due to water drainage may increase CO₂ emission. However, during the submerged period of paddy rice cultivation, CO₂ production in the soil is severely restricted. This effect can be explained with two basic mechanisms (Liu et al. 2013). Firstly, when flooding biological activity reduction under anoxic conditions slows down rather than completely inhibits CO₂ production. Secondly, when flooding a field for subsequent rice cultivation, water replaces the gaseous phase in the soil pores. Since the CO₂ diffusion rate in water is four orders of magnitude lower

than that in air, part of the produced CO_2 is stored in the soil. Hence, the soil CO_2 fluxes can be dramatically reduced by flooding during rice cultivation (Miyata et al. 2000; Campbell et al. 2001; Saito et al. 2005). Results from the present study provide indirect support for this conclusion, since the soil CO_2 flux rates under flooded conditions were significantly lower than those observed under drained conditions.

The soil CO_2 fluxes from both treatments were generally low during the post-harvest period and reached their maximum negative values at the end of the sampling period (Fig. 4). Low or negative fluxes of CO_2 in the post-harvest period coincide with the period of drainage and a low temperature. This indicates that draining (when included as an integral part of the irrigation treatment) and temperature play vital roles in controlling CO_2 emissions in a paddy soil.

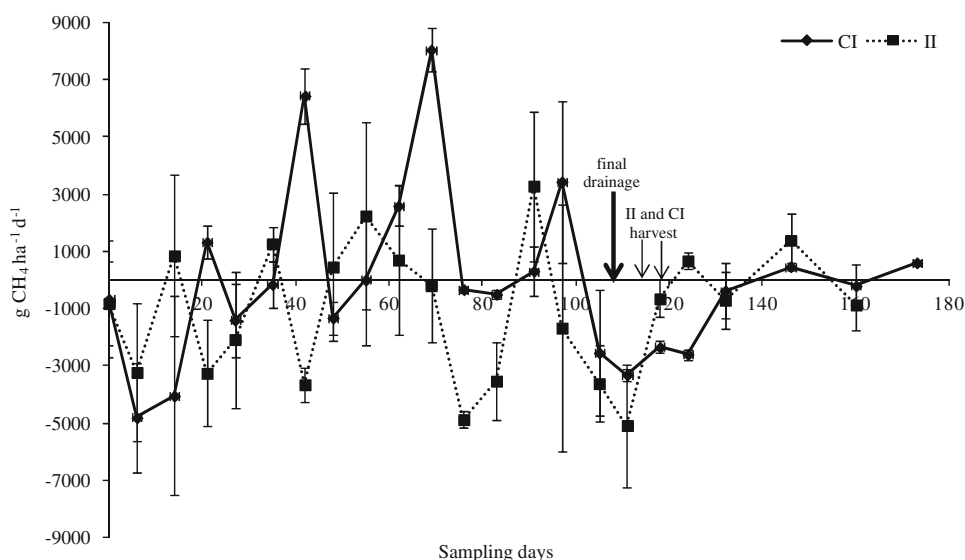
Methane emission

The daily fluxes of CH_4 emission from paddy soils under continuous and intermittent irrigation are shown in Fig. 5. The measured emission fluxes ranged from -4810.93 to $8041.06 \text{ g CH}_4 \text{ ha}^{-1} \text{ day}^{-1}$ in CI, and from -5082.06 to $3285.86 \text{ g CH}_4 \text{ ha}^{-1} \text{ day}^{-1}$ in II, exhibiting a wide seasonal fluctuation during the rice-growing season. The negative fluxes indicate that the soil acts as a net sink. Negative fluxes have also been found by Muhr et al. (2008), Yao et al. (2012), and Feike et al. (2013).

The average daily flux of CH_4 in the II fields was -1063.87 and $-63.05 \text{ g ha}^{-1} \text{ day}^{-1}$ in the CI fields. The first CH_4 efflux peak in continuous flooding ($6432.6 \text{ g CH}_4 \text{ ha}^{-1} \text{ day}^{-1}$) was very high and was observed at the end of the vegetative phase (day 42 of sampling). In the CI, a

second CH_4 efflux peak ($8041.06 \text{ g CH}_4 \text{ ha}^{-1} \text{ day}^{-1}$) during the reproductive phase (day 69 of sampling) was observed. Near flowering, high CH_4 efflux probably occurs due to large root exudation serving as a source of carbon (C) (Wassmann and Aulakh 2000). In addition, at this stage, aerenchymas are fully mature throughout the entire plant, working as continuous channels transporting CH_4 into the atmosphere (Wassmann and Aulakh 2000). The decrease in CH_4 flux during maturation and until harvest is due to the adverse conditions for CH_4 production caused by the end of irrigation and plant senescence, when labile organic C compounds are no longer released by roots (Cai et al. 1997). In the post-harvest period, the daily fluxes of CH_4 emissions were very low. Water management in the field is one of the determinants for CH_4 production in paddy soils, because methanogenesis takes place under strict anaerobic reducing conditions. Methane emission under CI was significantly higher than under II. The results of this study are consistent with the results obtained by Moterle et al. (2013) and Suryavanshi et al. (2013), who observed that there was a pronounced difference in CH_4 production between paddy soils under intermittent irrigation and continuous flooding. The relationship between the period of drainage and the change of CH_4 flux is important to evaluate how long the fields should be drained to mitigate CH_4 emission (Yagi et al. 1996). Draining lasting at least 10 days was needed to reduce CH_4 flux appreciably in the field studied by Yagi et al. (1996). This is probably due to the fact that longer drainage allows oxygen to diffuse into deeper layers of the soil column, in which the redox components are converted from their reduced form to their oxidized form (Fe^{2+} to Fe^{3+} , Mn^{2+} to Mn^{4+} , NH_4^+ to NO_3^-), as shown by Patrick and Jugsujinda (1992). Once the redox components in soil are converted to their

Fig. 5 Daily flux of CH_4 from the paddy soils under different water management practices through the sampling season. The vertical arrow on day 112 of measurement indicates the harvest of rice in the II treatment, and that on day 116 indicates the harvest of rice in the CI treatment. II intermittent irrigation, CI continuous irrigation. Vertical lines indicate standard errors of three replications for each treatment



oxidized form, it takes a certain period to reduce these oxidized components and to decrease soil Eh to a level suitable for CH₄ production after the field is reflooded.

Although some authors such as Dong et al. (2011) argue that the already high N applications to paddy fields will have to increase, because this is the most limiting factor in rice productivity, this is not the case in the Ebro Delta, as fertilizer doses are already well optimized. Low emissions of CH₄ in the two treatments may be due to the use of ammonium sulfate and soil salinity. Linqvist et al. (2012) summarized that sulfate can reduce overall CH₄ emissions by both suppressing methanogenesis as well as contributing to anaerobic CH₄ oxidation. Nitrogen fertilization may directly or indirectly affect the processes involved in the CH₄ budget of rice paddies, i.e., the production, oxidation, and transport of CH₄. However, studies investigating N fertilizer effects on these processes have yielded contradictory results. For example, after fertilization with urea or (NH₄⁺)₂SO₄, lower CH₄ emissions were detected and this was attributed to the direct inhibition of methanogenesis by these fertilizers (Lindau et al. 1990; Cai et al. 1997; Ma et al. 2007). However, higher CH₄ emissions were also observed from paddy fields after applications of ammonium-based non-sulfate fertilizers (e.g., urea, (NH₄⁺)₂HPO₄, which may increase plant growth and carbon supply and thus provide more methanogenic substrates and enhance the efficiency of CH₄ transport to the atmosphere (Singh et al. 1996; Schimel, 2000; Zheng et al. 2006). Dan et al. (2001) and Cai et al. (2007) reported no difference in CH₄ emissions between N fertilized and unfertilized rice paddies. These conflicting findings regarding CH₄ emissions as affected by N fertilizer underscore the need for more research, especially for non-sulfate, non-nitrate N fertilizers, because CH₄ production is generally inhibited by sulfate, as described in previous studies (Schütz et al. 1989; Minami 1995; Scheid et al. 2003), and also because nitrate-based fertilizers are not recommended for use in paddy rice production in order to avoid intensive N₂ loss by denitrification. The above contrasting effects of N fertilizer on CH₄ emissions from paddy fields indicate that soil N availability interacts with other site-specific factors when controlling CH₄ production processes.

The negative cumulative emissions of CH₄ (soil acts as a sink) in both treatments may be due to soil salinity and to a high sulfate (SO₄²⁻) content in irrigation water. In this study, the soil salinity was 4.65 dS m⁻¹ (measured in the soil saturated paste extract). In the study area (Ebro Delta) groundwater is highly salty, sometimes more than sea water. Over extensive areas the groundwater electrical conductivity at 5 m depth varies from 16 to 60 dS m⁻¹ with maximum values over 100 dS m⁻¹ (Casanova 1998). It is possible that salinity caused a reduction in the total

microbial activity, thereby reducing CH₄ production (Patanaik et al. 2000). Biswas et al. (2006) have studied CH₄ emission from the saline rice fields of Sunderban mangroves of the Indian East coast and reported a significant reduction of CH₄ emission from the rice fields reclaimed from mangrove swamp compared to upland rice fields and mangrove forest area. In the present study, the high concentrations of sulfate in the irrigation water (about 150 ppm SO₄²⁻, Casanova 1998) may lead to the suppression of CH₄ emission. In natural systems, Pennock et al. (2010) found that annual CH₄ emissions from a freshwater wetland declined when the concentration of sulfate in the water increased. Segers (1998) summarized that sulfate can reduce overall CH₄ emissions by both suppressing methanogenesis as well as contributing to anaerobic CH₄ oxidation. Three possible mechanisms as to how sulfate (and other electron acceptors) could suppress methanogenesis were proposed. First, the reduction of electron acceptors could reduce substrate concentrations to a value that is too low for methanogenesis. Second, the presence of electron acceptors could result in a redox potential that is too high for methanogenesis. Third, electron acceptors could be toxic for methanogens. In a supra-optimal concentration of sulfate, this element could possibly decrease CH₄ emission, which is presumably due to saturation of the relevant enzyme surfaces, competition for electrons between methanogens and sulfate reducers, and the development of toxicity (Banik et al. 1995). So, the soil salinity together with a high sulfate concentration in the irrigation water could inhibit CH₄ emission.

Global warming potential

The integrative GWP of CO₂, CH₄, and N₂O on a 100-year horizon for the CI treatment was 3463.41 kg CO₂-eq ha⁻¹, which was 26.9 % lower than that for the II paddy fields. The soil acted as a sink for N₂O which resulted in the reduction of the GWP for the CI treatment. In addition, the mean rice yield of the CI paddy fields was 9572.23 kg ha⁻¹, which was higher than that of II paddies by 34.2 % and the difference was significant (*P* > 0.05) (Table 1). These results suggest that CI can significantly mitigate the integrative greenhouse effect caused by CH₄ and N₂O from paddy fields while ensuring the highest rice yield.

Conclusions

From this study, it can be concluded that (1) intermittent irrigation (II) led to significantly higher (N₂O + N₂)-N (1.36 times greater) and CO₂ emissions than continuous irrigation (CI); (2) draining prior to harvesting increased

N_2O – N emissions; (3) draining and flooding cycles controlled CO_2 emissions; (4) lower CH_4 emissions due to water drainage (II) may increase CO_2 and $(\text{N}_2\text{O} + \text{N}_2)$ – N emission, and (5) the soil acted as a CH_4 sink for both types of irrigation.

The integrative GWP of CO_2 , CH_4 and N_2O on a 100-year horizon decreased by 34.2 % in the CI paddy fields compared with the II fields. These results suggest that CI can significantly mitigate the integrative greenhouse effect caused by CH_4 and N_2O from paddy fields while ensuring the highest rice yield.

The present study makes it clear how flooding and drainage affect the exchanges of $(\text{N}_2\text{O} + \text{N}_2)$ – N , CO_2 and CH_4 from rice paddies in the short term. Further measurements throughout rice cultivation to assess the long-term effect of an intermittent drainage practice on the exchange of these gases from rice paddies are needed.

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