

## Estimation of nitrous oxide emissions from rice paddy fields using the DNDC model: a case study of South Korea

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### ABSTRACT

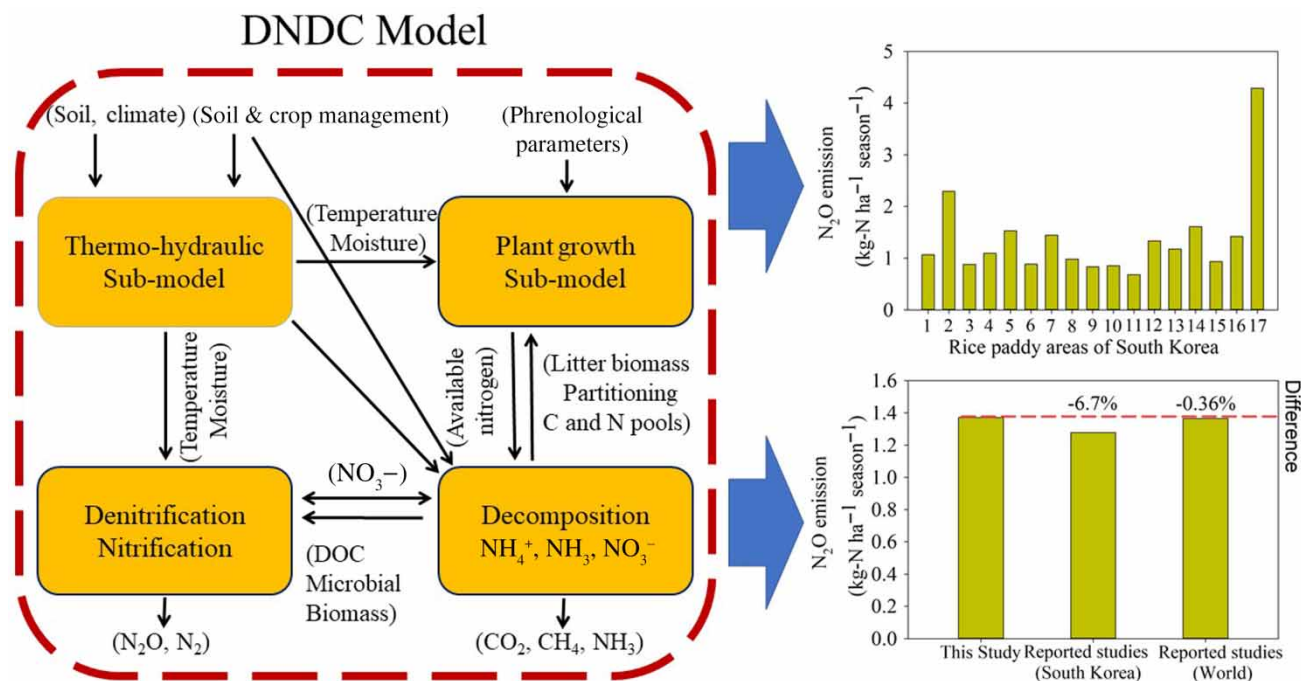
The Denitrification-Decomposition (DNDC)-Rice is a mechanistic model which is widely used for the simulation and estimation of greenhouse gas emissions [nitrous oxide (N<sub>2</sub>O)] from soils under rice cultivation. N<sub>2</sub>O emissions from paddy fields in South Korea are of high importance for their cumulative effect on climate. The objective of this study was to estimate the N<sub>2</sub>O emissions and biogeochemical factors involved in N<sub>2</sub>O emissions such as ammonium (NH<sub>4</sub><sup>+</sup>) and nitrate (NO<sub>3</sub><sup>-</sup>) using the DNDC model in the rice-growing regions of South Korea. N<sub>2</sub>O emission was observed at every application of fertilizer and during end-season drainage at different rice-growing regions in South Korea. Maximum NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> were observed at 0–10 cm depth of soil. NH<sub>4</sub><sup>+</sup> increased at each fertilizer application and no change in NO<sub>3</sub><sup>-</sup> was observed during flooding. NH<sub>4</sub><sup>+</sup> decreased and NO<sub>3</sub><sup>-</sup> increased simultaneously at end-season drainage. Minimum and maximum cumulative N<sub>2</sub>O emissions were observed at Chungcheongbuk-do and Jeju-do regions of South Korea, respectively. The simulated average cumulative N<sub>2</sub>O emission in rice paddies of South Korea was 1.37 kg N<sub>2</sub>O-N ha<sup>-1</sup> season<sup>-1</sup>. This study will help in calculating the total nitrogen emissions from agriculture land of South Korea and the World.

**Key words:** ammonium and nitrate, biogeochemical model, DNDC-Rice model, nitrous oxide, rice paddy

### HIGHLIGHTS

- We successfully simulated N<sub>2</sub>O emission from rice paddy fields of South Korea.
- DNDC model was used to estimate N<sub>2</sub>O emission in South Korea.
- Biogeochemical factors of soil such as NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> concentration were also focused upon.
- Simulated N<sub>2</sub>O emission in this study was strongly correlated as  $R^2 = 0.88-0.90$ .
- Results may help to estimate the nitrogen budget of South Korea as well as the world.

## GRAPHICAL ABSTRACT



## 1. INTRODUCTION

Nitrous oxide (N<sub>2</sub>O) is well known greenhouse gas for its global warming potential of 298 times greater than carbon dioxide (CO<sub>2</sub>) (IPCC 2007). N<sub>2</sub>O is also known for its role in damaging the stratospheric ozone layer (Fan *et al.* 2020). The soil ecosystem accounts for 65% (6.8 Tg N<sub>2</sub>O-N year<sup>-1</sup>) of the total N<sub>2</sub>O emitted to the atmosphere mainly due to nitrogen fertilizer application (IPCC 2007). Nitrogen is the most deficient nutrient in soils and essential for plant growth. Nitrogen fertilizer is extensively used to meet the plant need and food demand at large. However, 1–5% of N fertilizer applied is lost into the atmosphere as N<sub>2</sub>O emissions (Tang *et al.* 2018; Liu *et al.* 2019a). Nitrification and denitrification in the soil are core pathways of N<sub>2</sub>O emissions (Wei *et al.* 2017; Duan *et al.* 2019). Nitrification occurs under aerobic conditions in which primary nitrifiers (*Nitrosomonas* sp.) oxidize ammonium (NH<sub>4</sub><sup>+</sup>) to nitrite (NO<sub>2</sub><sup>-</sup>) and NO<sub>2</sub><sup>-</sup> is further converted to nitrate (NO<sub>3</sub><sup>-</sup>) by secondary nitrifiers (*Nitrobacter* sp.) (Giguere *et al.* 2018; Chen *et al.* 2019b). Oxidation of ammonia leads to nitrification-driven N<sub>2</sub>O emissions (Hu *et al.* 2015). Denitrification occurs under anaerobic conditions in which NO<sub>3</sub><sup>-</sup> is stepwise transformed into N<sub>2</sub>O and N<sub>2</sub> by facultative anaerobic bacteria (Xu *et al.* 2017; Giguere *et al.* 2018; Zhang *et al.* 2018a).

Rice (*Oryza sativa* L.) is cultivated on nearly 155 million ha around the world and feeds approximately 50% of the world's population (Fahad *et al.* 2019; Bashir *et al.* 2020). Paddy soils offer a diverse environment of biochemical cycles due to variation in water management practices (Kimura 2000). Nitrification occurs in the rhizosphere zone of paddy soils where nitrogen fertilizers release NH<sub>3</sub> (Wei *et al.* 2019; Yao *et al.* 2020). The NO<sub>3</sub><sup>-</sup>-N and NO<sub>2</sub><sup>-</sup>-N are stepwise transformed into N<sub>2</sub>O and N<sub>2</sub> through the denitrification process (Ishii *et al.* 2009). A significant amount of N<sub>2</sub>O is released from paddy soils to the atmosphere during the mid-season drainage period (Liu *et al.* 2019b; Liao *et al.* 2020). However, N<sub>2</sub>O could be further transformed into N<sub>2</sub> during the anaerobic state of soils that creates an anaerobic environment in paddy soils (Abbas *et al.* 2019). N<sub>2</sub>O emission during one growing season of rice paddy is estimated to be between 9 and 35 Gg N/year (Cai 2012). Previously, most studies have focused on N<sub>2</sub>O emissions from agricultural soils, wetlands, and grasslands (Brenzinger *et al.* 2018; Zhang *et al.* 2019; Ogle *et al.* 2020; Žurovec *et al.* 2021). However, few studies have reported the N<sub>2</sub>O emission from rice paddy at a national scale.

Recently, process-based models such as denitrification decomposition (DNDC) have been developed to add the feature of estimating N<sub>2</sub>O emissions at a regional scale (Gaillard *et al.* 2018; Tian *et al.* 2018; Yue *et al.* 2019). Additionally, these models are able to accurately predict N<sub>2</sub>O emissions under different agronomic management practices (Dutta *et al.* 2018;

Goglio *et al.* 2018; Jiang *et al.* 2020). The effect of climate change on crop yields can also be assessed using these models (Rafique *et al.* 2014; He *et al.* 2019). These models are frequently used for the identification of estimation factors of N<sub>2</sub>O emissions (Gaillard *et al.* 2016; He *et al.* 2019). Different models such as DayCent, EPIC, CERES-EGC, and SWAT have been developed for these applications (Gaillard *et al.* 2018; Massara *et al.* 2018; Giltrap *et al.* 2020). At first, the DNDC model was explicitly developed for simulation and estimation of N<sub>2</sub>O emissions, but now it has been further developed to include other ecosystems (Cui & Wang 2019; Yin *et al.* 2020; Zhao *et al.* 2020). The DNDC model can be used to simulate CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, NO, and NH<sub>3</sub> emissions (Shen *et al.* 2018; Jiang *et al.* 2020; Pandey *et al.* 2021). DNDC was previously used to estimate N<sub>2</sub>O emissions from agricultural fields (Abdalla *et al.* 2020b; Jiang *et al.* 2021; Pandey *et al.* 2021), animal farms (Brown *et al.* 2001; Shen *et al.* 2019), and CH<sub>4</sub> emissions from paddy fields (Li *et al.* 2002; Wang *et al.* 2021b). This model connects ecological drivers such as soil properties, vegetation type, climate, and anthropogenic activities to soil and environmental variables. The links between these variables simulate transformation processes of organic carbon and nitrogen, through which CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, NO, and NH<sub>3</sub> are estimated (Camarotto *et al.* 2018; Zhang *et al.* 2018b; Li *et al.* 2019). The two operating modes of the DNDC model are site and regional modes. The site mode can simulate trace gas emissions at a particular site and therefore can be compared with field observations. The regional mode can estimate trace gas emissions from the entire region which is based on statistical uncertainty estimates.

Previously, specific site and treatment-oriented studies on N<sub>2</sub>O emission from rice paddy fields have been reported in South Korea (Berger *et al.* 2013b; Kim *et al.* 2014a, 2016a; Pramanik *et al.* 2014). However, the present study is the first to estimate N<sub>2</sub>O emission from rice paddy fields on a national scale in the Republic of Korea (South Korea) using the DNDC model. The objective of this study was to estimate N<sub>2</sub>O emissions from rice-growing regions of South Korea and identify biogeochemical factors such as NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> that are known to affect N<sub>2</sub>O emissions. In this paper we will briefly show the preliminary results of a DNDC model of N<sub>2</sub>O fluxes from 17 different rice paddy fields of South Korea.

## 2. MATERIALS AND METHODS

### 2.1. Region/site description and data collection

The official name of South Korea is the Republic of Korea (ROK), located in eastern Asia and covers the southern part of the Korean peninsula, which is located between the Sea of Japan (East Sea) in east, the Yellow Sea in west, and the Korea Strait, a sea passage between South Korea and Japan in the south. South Korea shares a land border with North Korea in north. The country also shares maritime borders with China and Japan (Macdonald & Clark 2018). The total area of South Korea is 99,678 km<sup>2</sup> the country is about the size of Iceland, or slightly smaller than the United States' State of Pennsylvania (You 2009). South Korea is divided into six provinces namely Chungcheongbuk-do, Chungcheongnam-do, Gyeongsangbuk-do, Gyeongsangnam-do, Jeollabuk-do, and Jeollanam-do, one autonomous province Jeju-do, six metropolitan cities namely Busan, Daegu, Daejeon, Gwangju, Incheon, and Ulsan. Seoul is the capital city of the country. South Korea has a population of 50.8 million inhabitants in 2016. The official language is Korean. Forests cover 64% of the total land area in South Korea (Lee *et al.* 2019). Rice is the most important crop here, accounting for about 90% of the country's total grain production and over 40% of farm income. Barley, soybeans and potatoes are the other major broad acre crops. Fruit, particularly citrus, and vegetables are also widely grown (Neszmelyi 2017).

N<sub>2</sub>O emissions were estimated from 17 rice-growing regions across South Korea, as shown in Table 1. To compile data sets on different management practices of agricultural systems such as (1) soil properties, (2) irrigation, and (3) fertilizer application we referred to reported studies on a rice paddy fields in South Korea as shown in Table 2, and supplementary materials Tables S1, and S2, respectively. Reported agronomic practices varied slightly such as date of sowing, fertilizer application, irrigation, and harvesting were different. For missing data on input parameters, we used average values as prevalent in the region. The date of sowing was 12-06-2019 and the date of harvesting was 18-10-2019. Based on data available in Table 2 we selected soil texture silt loam, average bulk density, soil pH, and soil organic carbon as 1.17 g cm<sup>-3</sup>, 6.2, and 0.0201 (kg C·kg<sup>-1</sup>), respectively. The date of flooding and the date of end-season drainage were selected from Table S1 and was 11-06-2019 and 29-09-2019, respectively. The dates of fertilizer application at different growing stages of the crop such as basal, tillering, and panicle stage were 11-06-2019, 29-06-2019, and 23-07-2019, respectively (Table S2). Climatic characteristics of each region, such as minimum and maximum temperature, precipitation, wind speed, solar radiation, and air humidity, were collected from Korean metrological administration reports 2019 (KMA 2019).

**Table 1** | Cultivated area of rice paddies in different regions of South Korea (KOSIS 2019)

Site order	City/Province	Total arable area (ha)	Rice paddy area (ha)	Rice paddy area (%)
1	Seoul	149	131	87.9
2	Busan	2,503	2,383	95.2
3	Daegu	3,560	2,970	83.4
4	Incheon	11,610	10,419	89.7
5	Gwangju	5,302	4,988	94.1
6	Daejeon	1,259	1,109	88.1
7	Ulsan	4,402	4,102	93.2
8	Sejong	4,226	3,970	93.9
9	Gyeonggi-do	88,586	78,484	88.6
10	Gangwon-do	39,643	29,710	74.9
11	Chungcheongbuk-do	43,846	35,069	80.0
12	Chungcheongnam-do	143,288	134,035	93.5
13	Jeollabuk-do	136,294	118,340	86.8
14	Mr	186,954	161,442	86.4
15	Gyeongsangbuk-do	113,413	99,551	87.8
16	Gyeongsangnam-do	77,640	67,895	87.4
17	Jeju-do	6,562	113	1.7
	Total	869,236	754,713	86.8

**Table 2** | Soil and crop parameters for denitrification and decomposition model

Order	Region	GPS coordinates	Year	Sowing	Harvesting	Soil texture	Soil pH	SOC (kg C/kg)	BD (g/cm <sup>3</sup> )	Reference
1	Chungbuk	NA*	2000	12-Jun	04-Oct	NA*	5.9	0.0379	NA*	Ok <i>et al.</i> (2011)
2	Gimje	36°44'E;127°52'N	2012–13	21-Jun	20-Oct	Silt loam	5.6	0.0323	NA*	Chun <i>et al.</i> (2016)
3	Jinju	36°50'N;128°26'E	2006–7	10-Jun	20-Oct	Clay loam	6.1	0.0384	1.1	Ali <i>et al.</i> (2008)
4	Jinju	36°50'N;128°26'E	2011–12	10-Jun	20-Oct	Silt loam	6.9	0.0085	NA*	Haque <i>et al.</i> (2016)
5	Jinju	35°08'N;128°05'E	2010	08-Jun	21-Oct	Fine silty, mixed	6.6	0.015	NA*	Kim <i>et al.</i> (2013)
6	Jinju	35°08'N;128°05'E	2009–10	15-Jun	15-Oct	Fine silty, mixed	6.6	0.0098	NA*	Kim <i>et al.</i> (2012)
7	Jinju	35°06'N;128°07'E	2007–11	15-Jun	15-Oct	Fine silty, mixed	6.2	0.0144	NA*	Kim <i>et al.</i> (2016b)
8	Jinju	35°06'N;128°07'E	2011–12	15-Jun	15-Oct	Fine silty, mixed	6.32	0.0144	NA*	Kim <i>et al.</i> (2014b)
9	Jinju	36°50'N;128°26'E	2011	11-Jun	21-Oct	Silt loam	6.2	NA*	NA*	Pramanik <i>et al.</i> (2013)
10	Jinju	36°50'N;128°26'E	2014	11-Jun	21-Oct	Silt loam	6.32	0.011	NA*	Pramanik & Kim (2014)
11	Milyang	36°36'N;128°45'E	2008	05-Jun	20-Oct	Silt loam	5.98	0.023	1.23	Lee <i>et al.</i> (2010)
12	Sacheon	35°08'N;128°05'E	2010–11	12-Jun	24-Oct	Silty clay loam	6.6	0.0169	NA*	Gutierrez <i>et al.</i> (2013)

NA\*Data not available.

## 2.2. DNDC-Rice model description

The DNDC-Rice model has three main segments: (1) soil climate, (2) crop growth, and (3) biogeochemistry of soil. The scientific aspects and background information of this model are explained by Fumoto *et al.* (2008). The DNDC model is a process-based computer simulation model based on carbon and nitrogen biogeochemistry that was originally developed to estimate N<sub>2</sub>O emissions in agroecosystems. The model is based on two important components such as physicochemical and

biochemical components. The physicochemical component includes (soil climate, crop growth, and decomposition sub-models) that predicts the redox potential, pH, soil temperature, soil moisture, and substrate concentration profiles driven by ecological drivers such as climate, soil, vegetation, and anthropogenic activities. The biochemical component includes nitrification, denitrification, and fermentation sub-models and predicts the gas emissions from plant–soil systems such as carbon dioxide, nitrous oxide, nitric oxide, dinitrogen, methane, and ammonia (Wang *et al.* 2022).

All the input factors in each component of the DNDC-Rice model are reported in Fumoto *et al.* (2010). The soil climate simulates the functions of moisture, temperature, and  $O_2$  in the soil layers (0–50 cm depth) and it also simulates the greenhouse gases based on physicochemical properties of soil, daily weather, and agronomic practices. The movement of oxygen in undisturbed soil cores of croplands was explained with Buckingham–Burdine–Campbell diffusion model (Osozawa & Kubota 1987). The crop growth in the DNDC-Rice model is simulated with the computer program MACROS modules (Penning de Vries *et al.* 1989). In the crop growth section, nitrogen availability, the atmospheric concentration of  $CO_2$ , respiration of soil, and allocation of C are considered as main components of the photosynthesis process of a plant (Zhang & Niu 2016). The fluxes of carbon from rice paddy roots to soil are assessed as a part of the C balance. In soil biogeochemistry section, different biogeochemical processes are simulated as a part of soil and environmental properties. The organic carbon decomposition is assessed with first-order reaction kinetics, where the controlling factors are soil water content, temperature, proportion of clay content, concentration of  $O_2$ , nitrogen deficiency, and soil tillage. In this section, hydrogen ( $H_2$ ) and dissolved organic carbon (DOC) production is calculated under anaerobic conditions. Both  $H_2$  and DOC are worked as electron donors for the reduction of oxidants such as  $Mn^{4+}$ ,  $Fe^{3+}$ , and  $SO_4^{2-}$  while helping in the production of  $CH_4$ . When paddy soil is drained, both oxidants and  $CH_4$  are oxidized by atmospheric  $O_2$  diffusion into paddy soil and results in  $N_2O$  emissions.

### 2.3. $N_2O$ sampling and validation of the model output

The simulated results of this study were validated using the experimental records of field studies conducted in South Korea as published previously (Berger *et al.* 2013a; Kim *et al.* 2014a). The same procedure of validation of DNDC simulations was adopted by Cai *et al.* (2003) and Fumoto *et al.* (2008). Berger *et al.* (2013a) conducted field experiments and  $N_2O$  flux measurements that were done between 11 May 2010 and 23 October 2010 at the FDFM paddy and between 6 May and 15 September 2011 at all three paddies. To measure  $N_2O$  exchange at the soil–water/atmosphere interface, closed-chamber measurements in conjunction with a photoacoustic infrared gas analyses (Multigas Monitor 1312, INNOVA, Ballerup, Denmark) was used every 2 days at each experimental site. One day before the measurement, eight polyvinylchloride (PVC) cylinders (20 cm long and 19.5 cm wide) were installed 6 cm deep in the soil, so that depending on the water level of the rice paddy they poked out of the paddy water at least 2 cm. At each rice paddy, four of them contained rice plants, the other four were installed on spots without rice plants. For the measurement days, the cylinders were connected to chambers with a tubing connection to the gas analyzer, which determined the  $N_2O$  concentration of the chamber's headspaces after 0, 8, 16, 24, and 36 min. The reproducibility of one single  $N_2O$  concentration measurement was  $\pm 32$  ppb. From a linear increase or decrease of the  $N_2O$  concentration in the chambers' headspaces the  $N_2O$  flux was calculated considering the total chamber volume of the gas analyzing system, including the chamber head-space volume, volume of the two 25 m long Teflon tubes and of the  $CO_2$  and  $H_2O$  gas traps. Cumulative  $N_2O$  emissions were calculated by multiplying the  $N_2O$  emission rates of 2 consecutive measurement days with the corresponding time period. These time-weighted  $N_2O$  flux means were then summed up over the measurement period.

Kim *et al.* (2014a) conducted a field experiment in which rice cultivation experiments were carried out at the National Academy of Agricultural Science Research Farm (37°15'N; 126°59'E; 12 m elevation), Rural Development Administration, Suwon, Korea, in 2008. Methane and  $N_2O$  emission characteristics were investigated during the rice cropping season using the closed-chamber method (Rolston 1986). In each plot, three transparent acryl chambers (width 62 cm, length 62 cm, and height 112 cm) were placed permanently into the soil after transplanting the rice seedlings. There were four holes in the bottom of each chamber to control the amount of floodwater. The chamber was equipped with a circulating fan to ensure complete gas mixing during the period of sampling. Eight rice plants were covered by each chamber. Gas sampling was carried at 11:00–13:00 to determine the average daily  $N_2O$  emission rates during the cropping season. Briefly, gas samples in triplicates were collected once a week using 50-mL air-tight plastic syringes at 0-, 15-, and 30-min intervals after manually closing the chamber. The collected gas samples were transferred into 30-mL air-evacuated glass vials sealed with a butyl rubber septum. Nitrous oxide concentrations were determined using gas chromatograph (Shimadzu, GC-2010) with a stainless-steel column packed with Porapak Q column (Q 80–100 mesh) and equipped with a 63 Nielelectron capture detector.

The temperatures of the column, injector, and detector were adjusted at 70, 80, and 320 °C, respectively. Helium and hydrogen gases were used as the carrier and burning gases, respectively.

#### 2.4. Statistical analysis

The SPSS Statistics 20 software package was used for analysis. The Pearson correlation test was performed to determine the correlation between simulated and observed N<sub>2</sub>O emission.

### 3. RESULTS AND DISCUSSION

#### 3.1. Climatic factors

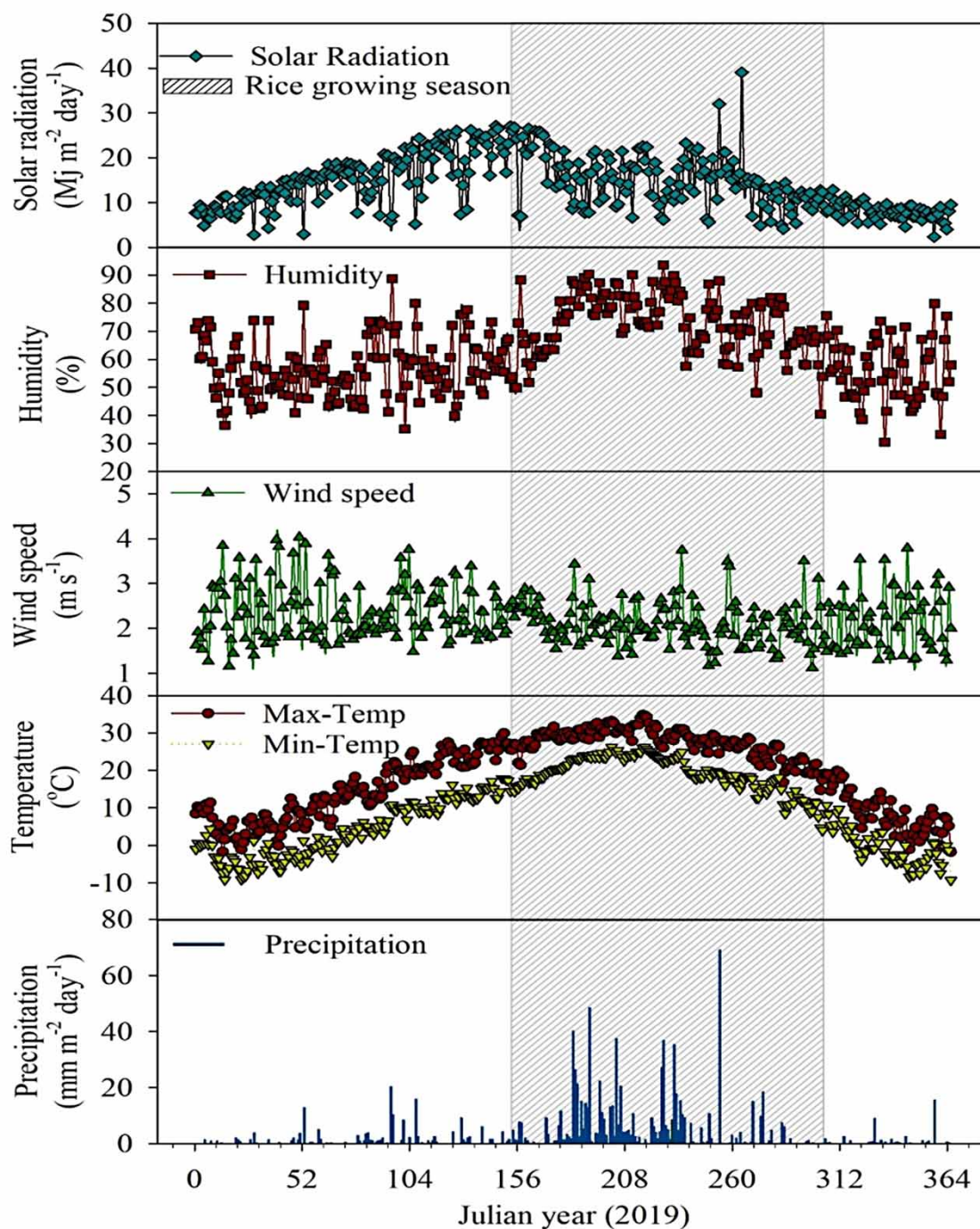
Climatic factors of all regions used in the DNDC model were averaged as shown in Figure 1 Average solar radiation, humidity, wind speed, minimum temperature, maximum temperature, and precipitation during the rice-growing season were 15.5 MJ m<sup>-2</sup> day<sup>-1</sup>, 74.7%, 2.1 m s<sup>-1</sup>, 20.2 °C, 28.2 °C, and 6.1 mm m<sup>-2</sup> day<sup>-1</sup>, respectively. Annual average climatic factors were significantly lower than during the growing season except for wind speed and solar radiation. Annual average solar radiation, humidity, wind speed, minimum temperature, maximum temperature, and precipitation during the rice-growing season were 14.2 MJ m<sup>-2</sup> day<sup>-1</sup>, 63.3%, 2.2 m s<sup>-1</sup>, 9.5 °C, 18.8 °C, and 2.8 mm m<sup>-2</sup> day<sup>-1</sup>, respectively.

#### 3.2. Comparison between observed and simulated N<sub>2</sub>O emissions to validate the results of DNDC model

Since the DNDC model simulates N<sub>2</sub>O emission based on available data of soil climate, crop growth, and biogeochemistry of soil, to validate the simulated results it is therefore very necessary to compare the results with N<sub>2</sub>O emission observed in the field. Observed N<sub>2</sub>O emissions were compared with simulated emissions from rice paddies to evaluate the performance of the DNDC-Rice model (Figure 2). Observed N<sub>2</sub>O emissions in 2010 during rice paddy growing season ranged from -21.5 to 87.4 µg m<sup>-2</sup> h<sup>-1</sup> while the simulated values were in the range of -15.4 to 78.1 µg m<sup>-2</sup> h<sup>-1</sup> (Figure 2(a)). However, the difference between observed and simulated results of N<sub>2</sub>O emission is not significant in this study. The N<sub>2</sub>O emission from paddy fields vary due to soil water content, thermal regime, soil organic matter, and nitrogen fertilizer application (Xing *et al.* 2009; Pandey *et al.* 2014; Xu *et al.* 2020). In this study, the slight difference in soil water content and nitrogen fertilizer application reflected in change in N<sub>2</sub>O emission. In 2008, minimum observed and simulated N<sub>2</sub>O emissions were 0.03 and 2.8, respectively while maximum observed and simulated N<sub>2</sub>O emissions were 54.2 and 50.5, respectively (Figure 2(b)). The difference between simulated seasonal emissions and observed emissions was 5.0% and 6.4% in 2010 and 2008, respectively. A similar pattern between simulated and observed N<sub>2</sub>O emission was reported (Abdalla *et al.* 2010). The simulated N<sub>2</sub>O emission was positively correlated with observed N<sub>2</sub>O emission in 2010 ( $R^2 = 0.96$ ,  $n = 45$ ) (Figure 3(a)). Similarly, the simulated N<sub>2</sub>O emission in 2008 was positively correlated with observed N<sub>2</sub>O emission ( $R^2 = 0.90$ ,  $n = 34$ ) (Figure 3(b)). Thus, the DNDC model successfully simulated N<sub>2</sub>O emissions at sites one and two. Wu & Zhang (2014) Reported ( $R^2 = 0.892$ ,  $n = 28$ ,  $p = 0.01$ ) consistency between observed and simulated N<sub>2</sub>O emissions from paddy fields under water-saving irrigation. Correlation between observed and simulated seasonal N<sub>2</sub>O fluxes from different sites was reported as  $R^2 = 0.93$ ,  $n = 9$ ,  $P < 0.01$  (Babu *et al.* 2006). Another correlation between observed and simulated N<sub>2</sub>O fluxes were reported  $R^2 = 0.75$  (Bhanja *et al.* 2019).

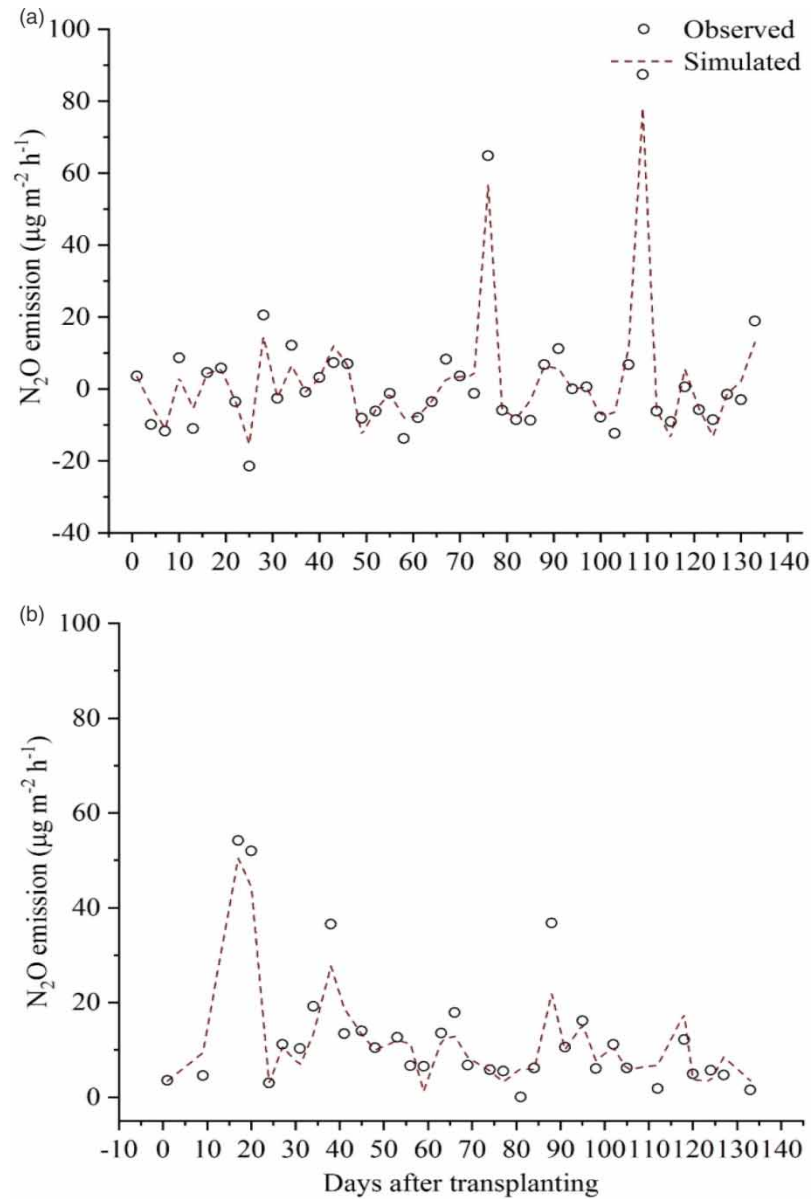
#### 3.3. N<sub>2</sub>O emissions in different regions of South Korea

Rice paddies were cultivated in 17 different regions of South Korea as shown in supplementary material Figure S1. Rice paddies are cultivated on 754,713 hectares that correspond to 87% of the total arable land of South Korea (Table 1). A maximum 161,442 hectares of rice paddies was cultivated in Jeollanam-do and a minimum of 113 hectares in Jeju-do. In each region N<sub>2</sub>O emission was observed only at basal, supplementary one, and supplementary two fertilizer applications and after end-season drainage. N<sub>2</sub>O emission after the dates of basal N fertilizer application was reported (Chen *et al.* 2019a). N<sub>2</sub>O emission was not observed between all three fertilizer applications. It was also not observed between supplementary fertilizer two and end-season drainage. Oxidation and reduction take place in paddy fields. The application of ammonium N containing fertilizers is nitrified in the oxidized layer, at the water-soil interface, forming nitrate, which moves downwards and denitrified in the reduced layer, producing N<sub>2</sub>O. In the presence of a water layer on the soil N<sub>2</sub>O is further reduced and escapes to the atmosphere as N<sub>2</sub>. N<sub>2</sub>O is emitted mainly through the soil surface in the absence of floodwater (Xing *et al.* 2009; Majumdar 2013; Wu *et al.* 2022). Since there was continuous flooding between three fertilizer applications and



**Figure 1** | Average climatic factors of rice paddy regions of South Korea.

between supplementary fertilizer and end-season drainage  $N_2O$  was further reduced and escaped to the atmosphere as  $N_2$ , therefore  $N_2O$  emission was not observed during these periods of the growing season. Minimum and maximum  $N_2O$  emission were observed at basal fertilizer application and after end-season drainage, respectively.  $N_2O$  emission varied significantly due to the change in climatic factors and physicochemical properties of each region. The continuous flooding, periodic drainage, seasonal tillage, and other agricultural activities during the rice-growing season make paddy soils in a state of dry-wet alternation (Liao *et al.* 2020; Zuo *et al.* 2022). Consequently, different soil layers have different

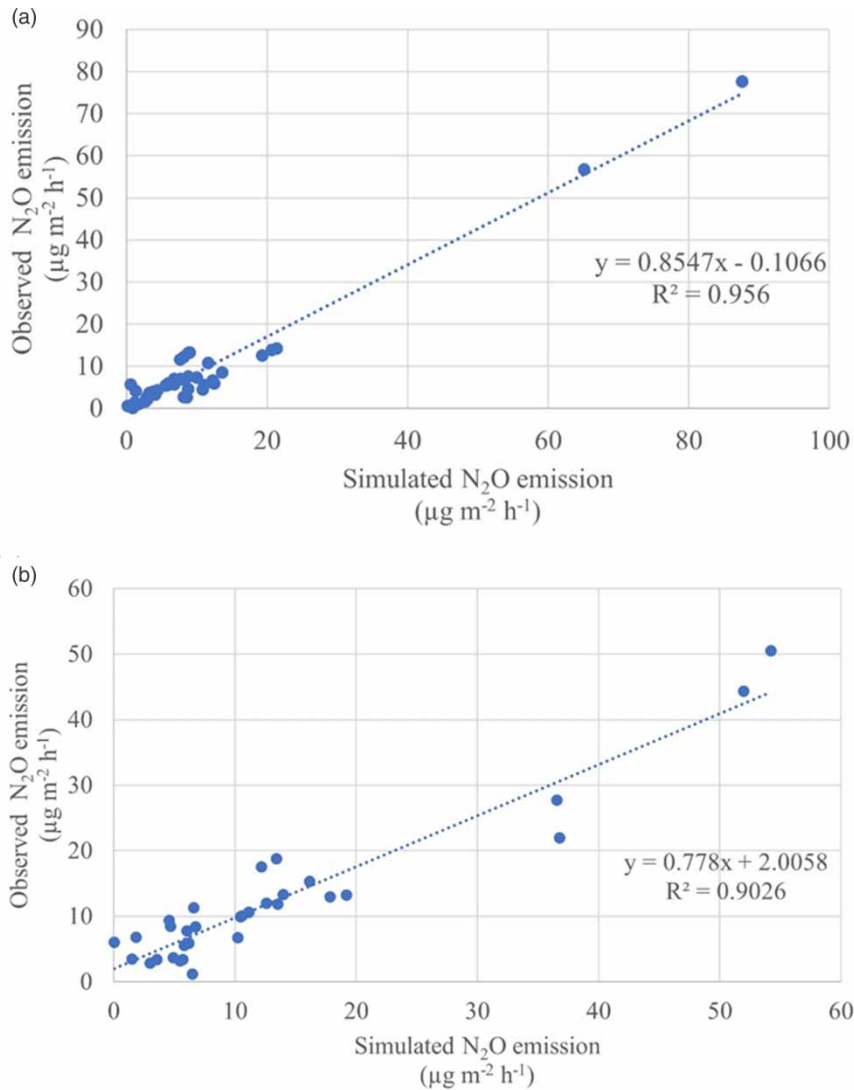


**Figure 2** | (a) Observed and simulated seasonal patterns of N<sub>2</sub>O emission in 2010 (Berger *et al.* 2013a), (b) N<sub>2</sub>O emission in 2008 (Kim *et al.* 2014a). (The observed values were digitized from the above-mentioned publications using Origin software.)

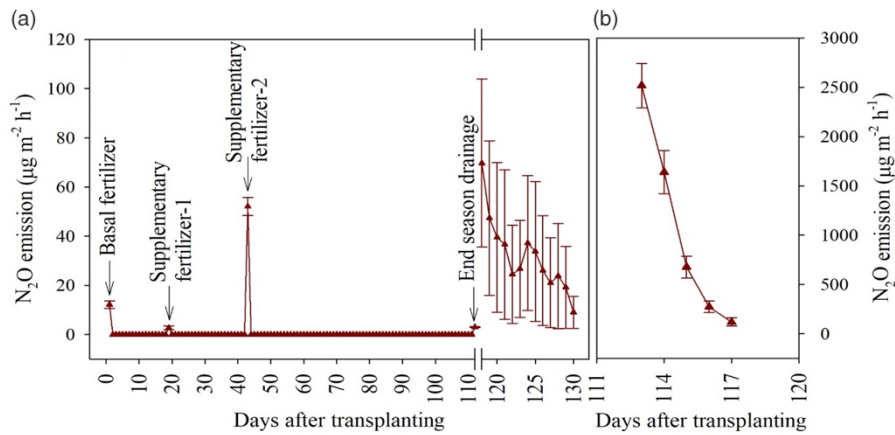
water content and aeration statuses, leading to the changes in chemical composition such as soil redox, pH, state of nitrogen, and other properties in time and space (Wang *et al.* 2021a; Zuo *et al.* 2022). In addition, environmental changes such as the changes in soil moisture, temperature, can affect the composition of soil microbial communities, thus, in turn, affecting the nitrification and denitrification of soil that may further lead to either increases or decreases in N<sub>2</sub>O emission (Wang *et al.* 2017; Qin *et al.* 2020).

N<sub>2</sub>O emission from rice paddy fields of all regions was averaged as shown in Figure 4. Average N<sub>2</sub>O emissions at basal, supplementary-one and supplementary-two fertilizer applications were  $12.2 \pm 1.5$ ,  $2.7 \pm 0.81$ , and  $52.1 \pm 3.7$  µg m<sup>-2</sup> h<sup>-1</sup>, respectively. Maximum N<sub>2</sub>O emission  $2,518.3 \pm 225$  was observed on the first day of end-season drainage Figure 4(b). N<sub>2</sub>O emission decreased gradually after end-season drainage until the last day of N<sub>2</sub>O simulation. Maximum 79% N<sub>2</sub>O emission of the entire season occurred following end-season drainage when soil is dry and creating suitable conditions for nitrification and denitrification (Adviento-Borbe *et al.* 2015; Abdalla *et al.* 2020a).





**Figure 3** | (a) Correlation between observed and simulated N<sub>2</sub>O emission in 2010 (Berger *et al.* 2013a), (b) Correlation between observed and simulated N<sub>2</sub>O emission in 2008 (Kim *et al.* 2014a).



**Figure 4** | (a) Temporal variation in average N<sub>2</sub>O emission in rice paddies in all South Korea. (b) Maximum N<sub>2</sub>O emission readily after end-season drainage.

### 3.4. Concentration of ammonium and nitrate

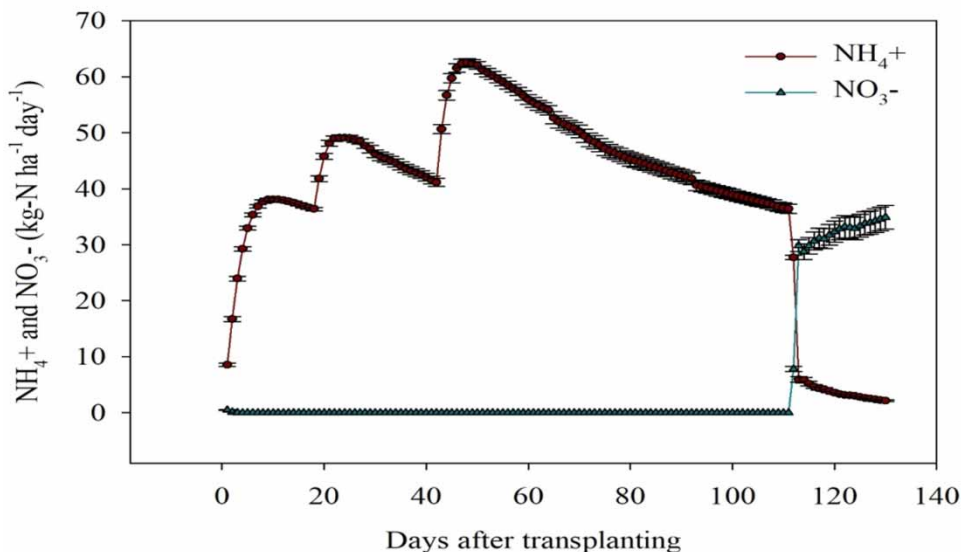
Ammonium ( $\text{NH}_4^+$ ) and nitrate ( $\text{NO}_3^-$ ) were simulated at different soil depths up to 50 cm from all rice paddy growing regions of South Korea (Figure 5, Tables 3 and 4). Maximum response of both  $\text{NH}_4^+$  and  $\text{NO}_3^-$  was only observed at 0–10 cm depth in all rice paddy growing regions of South Korea.  $\text{NH}_4^+$  increased at basal, supplementary one and two fertilizer applications.  $\text{NH}_4^+$  decreased abruptly on the first day of end-season drainage and then decreased gradually. Total simulated  $\text{NH}_4^+$  during the entire season was 4,929.3, 5,497.0, 5,398.6, 5,131.8, 5,225.9, 5,112.2, 5,522.3, 5,094.5, 5,253.6, 4,846.3, 4,897.0, 4,968.8, 5,165.4, 5,277.8, 4,994.1, 5,384.7, and 3,979.9  $\text{kg N ha}^{-1} \text{ season}^{-1}$  recorded at sites 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, and 17, respectively; site names are mentioned in Table 1. Throughout the season,  $\text{NO}_3^-$  was only observed after end-season drainage.  $\text{NO}_3^-$  increased rapidly for the first two days after end-season drainage and then decreased gradually up to the last day of  $\text{NO}_3^-$  simulation. Total simulated  $\text{NO}_3^-$  during the entire season was 580.7, 419.9, 603.2, 637.1, 607.6, 622.5, 654.7, 652.1, 602.9, 666.7, 654.3, 606.5, 662.1, 644.8, 637.7, 644.3, and 70.6  $\text{kg N ha}^{-1} \text{ season}^{-1}$  recorded in 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, and 17, respectively. Nitrous oxide is produced when nitrification and denitrification take place under aerobic and anaerobic conditions in soil, respectively (Katayanagi *et al.* 2012). Nitrification mainly regulates  $\text{N}_2\text{O}$  production in rice paddy soils as a function of  $\text{NH}_4^+$  where it is further oxidized to  $\text{NO}_3^-$  (Pathak *et al.* 2005; Katayanagi *et al.* 2012).

### 3.5. Comparison of cumulative $\text{N}_2\text{O}$ emissions

Cumulative  $\text{N}_2\text{O}$  emissions during the entire season in this study were compared with cumulative  $\text{N}_2\text{O}$  emissions of reported studies worldwide and in South Korea (Figure 6). Reported cumulative  $\text{N}_2\text{O}$  flux in rice paddies worldwide and in South Korea ranged from  $-0.34$  to  $15.27$  and  $0.014$  to  $4.2 \text{ kg-N ha}^{-1} \text{ season}^{-1}$ , respectively as shown in supplementary material Table S3. In this study, simulated cumulative  $\text{N}_2\text{O}$  emissions in all rice paddy growing regions of South Korea were in the range  $0.68$ – $4.29 \text{ kg-N ha}^{-1} \text{ season}^{-1}$ . Cumulative  $\text{N}_2\text{O}$  emissions were underestimated by 4–19% by the DNDC model (Yu *et al.* 2017, 2018).

## 4. CONCLUSION

Using the DNDC model, we simulated  $\text{N}_2\text{O}$  emissions in rice paddies of all rice paddy growing regions of South Korea. Reported studies in South Korea and simulated  $\text{N}_2\text{O}$  emissions in this study were strongly correlated  $R^2 = 0.88$ – $0.90$ . The difference between reported and simulated  $\text{N}_2\text{O}$  emissions was 5–6%.  $\text{N}_2\text{O}$  emissions in all rice paddy growing regions of



**Figure 5** | Average concentration of ammonium and nitrate at 0–10 cm depth of rice paddy soils of South Korea. Error bars show standard error of the mean between all sites.

**Table 3** | The average amount of ammonium ( $\text{kg-N ha}^{-1} \text{ season}^{-1}$ ) in rice paddy soils of different regions of South Korea

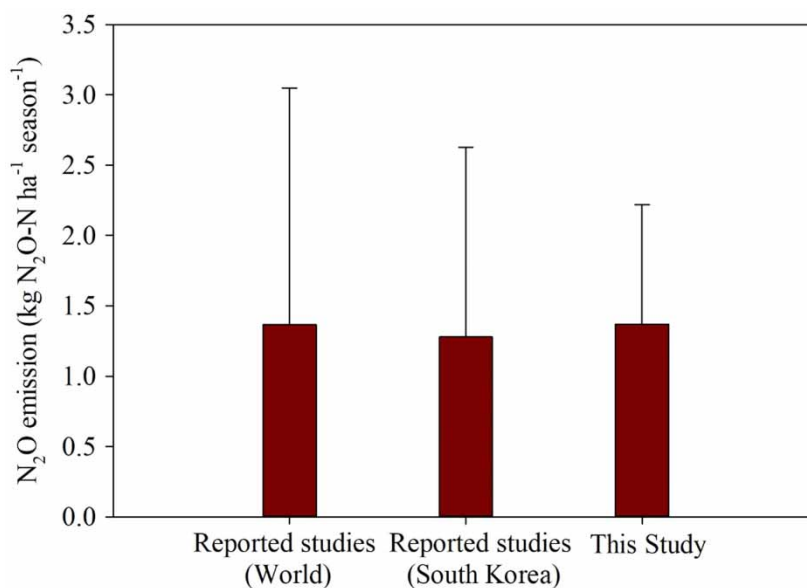
Soil depth (cm)	Mean	1 <sup>a</sup>	2 <sup>a</sup>	3 <sup>a</sup>	4 <sup>a</sup>	5 <sup>a</sup>	6 <sup>a</sup>	7 <sup>a</sup>	8 <sup>a</sup>	9 <sup>a</sup>	10 <sup>a</sup>	11 <sup>a</sup>	12 <sup>a</sup>	13 <sup>a</sup>	14 <sup>a</sup>	15 <sup>a</sup>	16 <sup>a</sup>	17 <sup>a</sup>
0–10	Avg	37.9	42.3	41.5	39.5	40.2	39.3	42.5	39.2	40.4	37.3	37.7	38.2	39.7	40.6	38.4	41.4	30.6
	SE	15.7	18.1	17.1	16.6	17.5	16.6	18.3	16.8	16.3	16.2	16.3	16.8	17.4	17.9	16.5	18.1	13.8
10–20	Avg	3.4	4.9	4.4	3.5	4.2	4.0	4.2	4.0	3.9	1.4	1.6	1.7	1.8	1.6	1.7	1.5	1.8
	SE	0.6	0.2	0.3	0.4	0.2	0.3	0.3	0.3	0.4	0.7	0.8	0.8	0.9	0.8	0.9	0.9	1.1
20–30	Avg	3.7	3.8	2.7	2.2	2.7	2.5	2.7	2.5	3.4	1.0	1.1	1.2	1.3	1.2	1.2	1.4	1.2
	SE	0.4	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.3	0.4	0.5	0.5	0.5	0.5	0.5	0.4	0.5
30–40	Avg	1.7	1.4	1.3	1.0	1.3	1.2	1.2	1.2	1.6	0.5	0.5	0.6	0.6	0.5	0.5	0.7	0.6
	SE	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2	0.2
40–50	Avg	0.8	0.7	0.6	0.5	0.6	0.5	0.6	0.5	0.8	0.2	0.2	0.3	0.3	0.3	0.2	0.3	0.3
	SE	0.10	0.05	0.05	0.04	0.05	0.04	0.05	0.04	0.08	0.06	0.07	0.08	0.08	0.08	0.07	0.06	0.09

<sup>a</sup>Names of different regions as listed in Table 1.

**Table 4** | The average amount of nitrate ( $\text{kg-N ha}^{-1} \text{ season}^{-1}$ ) in rice paddy soils of different regions of South Korea

Soil depth (cm)	Mean	1*	2*	3*	4*	5*	6*	7*	8*	9*	10*	11*	12*	13*	14*	15*	16*	17*
0–10	Avg	4.5	3.2	4.6	4.9	4.7	4.8	5.0	5.0	4.6	5.1	5.0	4.7	5.1	5.0	4.9	5.0	0.5
	SE	11.0	8.0	11.5	12.1	11.5	11.9	12.4	12.4	11.5	12.7	12.5	11.5	12.6	12.3	12.1	12.2	2.0
10–20	Avg	0.1	0.1	0.1	0.1	0.2	0.1	0.1	0.1	0.1	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0.0
	SE	0.2	0.3	0.2	0.2	0.4	0.3	0.2	0.3	0.2	0.5	0.2	0.3	0.2	0.3	0.2	0.1	0.1
20–30	Avg	0.005	0.005	0.002	0.003	0.004	0.004	0.003	0.004	0.003	0.010	0.004	0.004	0.004	0.004	0.003	0.003	0.001
	SE	0.02	0.03	0.02	0.02	0.03	0.02	0.02	0.02	0.02	0.04	0.03	0.03	0.03	0.03	0.03	0.03	0.01
30–40	Avg	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
	SE	0.011	0.010	0.012	0.012	0.012	0.012	0.011	0.012	0.011	0.016	0.016	0.015	0.015	0.015	0.016	0.014	0.007
40–50	Avg	0.000	0.000	0.001	0.001	0.001	0.001	0.001	0.001	0.000	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.000
	SE	0.005	0.005	0.006	0.006	0.006	0.006	0.006	0.006	0.005	0.008	0.008	0.007	0.007	0.007	0.007	0.007	0.004

\*Names of different regions as listed in Table 1.



**Figure 6** | Comparison of cumulative N<sub>2</sub>O emission in rice paddies of this study with reported studies worldwide and in South Korea. Error bars show the standard deviation between N<sub>2</sub>O fluxes from different locations.

South Korea were mainly due to NH<sub>4</sub><sup>+</sup> and NH<sub>3</sub>- during flooding (wet period) and after end-season drainage (dry period), respectively. Minimum and maximum N<sub>2</sub>O emissions were observed at basal fertilizer application and during end-season drainage, respectively. There was no significant difference between the reported average N<sub>2</sub>O emissions in rice paddies worldwide, in South Korea, and that assessed in this study. This comparison of N<sub>2</sub>O emissions reflected the reliability of N<sub>2</sub>O estimation and our simulated results are within the range of reported studies.

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## DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

## CONFLICT OF INTEREST

The authors declare there is no conflict.

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