Evaluation of N Environmental risks on Andosols from an intensive dairy farming watershed using DNDC

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Abstract

Manure nitrogen (N) in the livestock sector has become a key driver of environmental change. The denitrification–decomposition (DNDC) model was used to evaluate N pollution strengths on Andosols with intensive dairy manure application in Upper Naka River Watershed, Japan. The calibrated model was capable of predicting Andosol N flows because the simulated soil mineral N content, soil nitrogen oxide (N₂O) fluxes, denitrification rate, and crop N uptake matched the patterns and magnitudes of the field observations from a wide range of soil textures, as well as manure management and cropping systems. The simulations showed that current intensive manure application systems caused low crop N use efficiency and a large amount of NO₃⁻-N leaching and N₂O emission. The crop N use efficiency was 27%–42% and 37%–55% of input N for uplands and paddy rice, respectively. The uplands showed much more serious N environmental pollution risks with N leaching 123–362 kg N ha⁻¹yr⁻¹ and N₂O emissions 6.53–11.8 kg N ha⁻¹yr⁻¹ than that in the lowland paddy rice with N leaching 17.4–103 kg N ha⁻¹yr⁻¹ and N₂O emissions 0.59–2.77 kg N ha⁻¹yr⁻¹. Foragerice/barley crop systems have high N removal capability due to the greater crop N uptake which reached to 304 kg N ha⁻¹yr⁻¹. High precipitation stimulated more NO₃⁻-N leaching. Sandy soil also showed higher N leaching and was not

appropriate for paddy rice. Slurry application stimulated more N_2O emission than compost manure. To reduce the current high N pollution and without a decrease in crop harvest, the critical N application rate was recommended to be approximately 380, 470, 640, and 390 $\text{kg N ha}^{-1} \text{ yr}^{-1}$ for loam sand planted with maize/grass, loam soil with maize/grass, forage rice/barley, and rice/fallow with winter manure application, respectively.

Keywords: intensive dairy farming, DNDC, N environmental pollution, Andosol, upland, paddy rice

1. Introduction

Livestock sector is a major environmental threat. It produced 14.5% of anthropogenic greenhouse-gas emissions and over 37% of total human-induced nitrogen(N) pollution (Eisler et al., 2014; FAO 2006). Pelletier and Tyedmers (2010) estimated that the developing of livestock sector will soon reach or exceed the sustainability thresholds in climate change and reactive nitrogen mobilization at a global scale. Clearly, the livestock sector serves a pivotal function in anthropogenic environmental change, especially owing to manure production (Bouwman et al., 2013). From 1950 to 2000, manure N increased by approximately twice and reached to 92 Tg. yr^{-1} which was higher than the chemical fertilizer N 83 Tg. yr^{-1} in 2000 (Bouwman et al., 2013). In order to feed more people in 2050, manure N would increase to 140 Tg. yr^{-1} (Bouwman et al., 2013). Hence, manure N management would be one of the major environmental concerning in future.

Intensive livestock farming is gaining popularity and shifting from traditional, extensive, and decentralized family farms (Bouwman et al., 2013; Ilea, 2009). In Japan, livestock density for each farmer has increased. In 2006, the densities were up to 10, 16, 86, and 543 times that of 1970 for cows, beef cattle, pigs and chickens, respectively (Japanese Ministry of Agriculture, Forestry and Fisheries, 2012). For the intensive livestock system, excessive application of livestock waste was very common because of the limited land available for each farm (Ilea, 2009). Deng et al. (2012) reported that the manure N rate in an intensive dairy farming watershed ranged from 500 $\text{kg N ha}^{-1} \text{ yr}^{-1}$ to 700 $\text{kg N ha}^{-1} \text{ yr}^{-1}$. Highly intensive livestock system was generally contributed to signification environmental degradation (Peyraud et al. 2014). High nitrate concentration in ground water has been reported in livestock regions (Somura et al., 2008). Vergé et al. (2007) reported that intensive livestock has

increased greenhouse gas emission at twice the rate of traditional farming systems, and at more than six fold the rate of production based on grazing. An assessment of manure N pollution from intensive livestock farming is urgently needed to formulate efficient environmental solutions.

To assess N-pollutant levels in intensive manure systems and to suggest mitigation solutions, reliable assessment tools that consider the complex spatial and temporal variability resulting from different soils, vegetation, managements, and climates are needed (Follador et al., 2011; Leip et al., 2008). Process-based biogeochemical models are important tools because of their capability to scale-up the spatial and temporal dynamics of agro-ecosystems in response to management methods (Follador et al., 2011; Giltrap et al., 2010; Kemanian et al., 2011). A number of process-based models have been used to study the impact of agricultural management, such as DNDC (DeNitrification-DeComposition) (Li, 1992), Century (Motavalli et al., 1994) and SWAT (Arnold and Allen, 1996). Among those biogeochemical models, DNDC is suitable for simulating N transformation processes under both aerobic and anaerobic conditions (Li et al., 2006), which has also been intensively validated for agroecosystems worldwide. Beheydt et al. (2007) validated DNDC for 22 long-term N₂O emissions from soil measurements over a variety of crop systems in a range of soil types. Follador et al. (2011) used DNDC to investigate the effects of agriculture measures on N₂O emissions, N surplus and N leaching from European cornfields. Pathak et al. (2006) simulated the N balance in rice-wheat systems of Indo-Gangetic plains. The DNDC model therefore allows simulation of N fluxes for both aerobic and anaerobic agriculture ecosystems with different conditions.

Andosol is a highly productive agricultural soil and distributes worldwide with around 0.8% of the total soil area on earth (Shoji et al., 1993). It is characterized by high organic matter content, high stability, strong phosphate fixation, high porosity, low bulk density, high water retention, and good permeability (Armas-Espinel et al., 2003; Shoji et al., 1993). The existence of amorphous short-range compounds combined with high organic matter content confers a variable charge onto Andosol (Shoji et al., 1993). These characteristics drive a distinct N cycling on Andosol. A lower denitrification capacity than that of non-Andosol has been reported for Andosol

(Deng et al., 2012). Till now the DNDC model has yet been applied to assess N flows on Andosol agriculture systems. Therefore, it has been a challenge to predict N flows on Andosol using DNDC.

The objectives of this study are 1) to calibrate and validate the DNDC model for estimate Andosol N flows in intensive manure crop systems, 2) to assess N environmental pollution risks from different forage crop systems under heavy manure management, and 3) to evaluate potential N pollution mitigation solutions in current intensive livestock agricultural practices.

2. Materials and Methods

2.1 Model description

DNDC is a process-oriented biogeochemical model simulating C and N cycling in various ecosystems. The model consists of four sub-models: soil thermal-hydraulic, crop growth, decomposition, and denitrification (Li, et al., 1992). The model requires daily meteorological data of maximum and minimum air temperatures, precipitation and solar radiation, management data (e.g. the dates and types of planting and harvesting, tillage and fertilization), as well as soil and vegetation properties. After building these initialization parameters, the model calculates daily rates of ecosystem processes, including plant N uptake, litter production, mineralization, nitrification, denitrification, and soil water transformations (Butterbach–Bahl et al., 2009; Fumoto et al., 2010; Li et al., 2006, 1994). In this research, DNDC 9.5 has been used. This is the latest version which integrated manure-DNDC, improved crop growth simulations and hydrological features (Gilhespy et al., 2014). For the manure input parameters, the model need input the type, amount and C/N of manure as well as the application time. Then the model links the environmental factors (e.g., temperature, moisture, pH, Eh) to simulate nitrification and denitrification processes.

2.2 Study location and data collection

The study site was located in Upper Naka River watershed, one of the major dairy farming regions in Japan. The watershed has a typical monsoon climate with a mean annual rainfall of 1493 mm and a mean annual air

temperature of 11.7°C over the last 30 years. According to the world reference base for soil resources, the main soil is Andosol (FAO, 2006). Upland maize and paddy rice systems were the main crops in the watershed. For model calibration, site-specific soil data including: soil texture, bulk density, porosity, pH, organic C and mineral N as well as agricultural management information such as: dates planting/harvest, tillage, fertilization, and flooding events; and fertilization rates are required as model inputs. Data were collected from eight fields (each 700–1500 m² in size) within the Upper Naka River watershed at five locations to represent differences in soil textures, land use types, and manure application rates (Table 1). The selected fields had loam, sandy loam, silt loam and loam sand soil textures. The sandy loam soils had a maize/grass rotation system only, whereas the other soil types also had upland maize (maize/grass or maize/barley rotations) and paddy rice (rice/fallow or forage rice/barley rotation) crop systems. Manure application rates were 400–800 kg N ha⁻¹ yr⁻¹ for uplands and 150–480 kg N ha⁻¹ yr⁻¹ for paddy rice. Chemical N was applied in a few fields with 20–100 kg N ha⁻¹ yr⁻¹ for maize and rice summer crops before planting in some fields. Manure was applied once a year for a summer rice/fallow system; otherwise, manure was applied twice a year, i.e., mixed with surface soil before each crop was planted. Table 1 summarizes the crop and soil management practices for each field, designated as G/M1, G/M2, G/M3, G/M4, and M5 for upland fields and R1, R4, and R5 for paddy rice fields. G, M, and R indicate grass, maize, and rice crops, respectively.

Field data including soil N₂O fluxes, soil temperature, water content, soil NO₃⁻-N and ammonium (NH₄⁺-N) (top 10 cm depth) concentration, crop yield, and N and C content of crop components, were measured from January 2008 to April 2010. From January 2008 to April 2009, the gas sampling and soil measurements did in once for two months. The soil denitrification rate was also measured from January 2008 to April 2009. Detailed sample information could be found in Deng et al. (2012). Kamewada (2007) reported that most of the denitrification enzyme activity(DEA) was from the top soil. Therefore, here the denitrification rate from top 30cm soil was assumed that accounted to 75% of total denitrification rate. Then the total denitrification rate was calculated from the values of top 30cm soil layer. From May 2009 to April 2010, the field intensive sampling was once every two days on May 15 to Jun 30 and October 1 to November 30 and then once or twice monthly

for the other periods. Climate data (2007–2010) were obtained from the Kuroiso (36.98N, 140.02E) and Otawara (36.84N, 140.04E) (Kuroiso and Otawara Meteorological Data Systems, Japan Meteorological Agency) weather stations. The Kuroiso weather data were used for fields G/M1, G/M2, G/M3, G/M4, R1, and R4, whereas the Otawara data were for M5 and R5. The distances between the fields and weather stations were within 10 km.

Table 1. Information of soil properties (0–20 cm) and fertilizer management practices

Site	G/M1	R1	G/M2	G/M3	G/M4	R4	M5	R5
Coordinates(°)	37.02N 139.98E	37.02N 139.98E	139.98E 37.00N 140.00E	37.00N 36.96N 139.91E	36.96N 36.94N 140.00E	36.94N 36.94N 140.00E	36.83N 140.00E	36.83N 140.00E
Soil texture	Loam	Loam	Sandy loam	Loam	Loam Sand	Loam Sand	Silt loam	Silt loam
Clay (%)	18	18	7	20	1	1	6	6
SOC (%) (Mean ± SD)	12.2±0.36	6.64±0.70	10.4±0.74	11.8±0.91	4.62±0.08	3.83±0.17	5.23±0.15	9.61±0.99
C/N of SOM	13	14	14	14	11	11	14	11
pH	6.95	6.56	6.27	6.15	5.69	6.1	6.16	6.15
Bulk density(g cm ³)(Mean ± SD)	0.74±0.04	0.74±0.04	0.68±0.01	0.57±0.02	1.34±0.09	1.34±0.09	0.87±0.03	0.87±0.03
Summer crop	Maize	Rice(H ^a)	Maize	Maize	Maize	Rice(H ^a)	Maize	Rice
Sowing/transplanting date	5/27	5/10	5/26	5/22	6/4	5/10	6/27	5/16
Harvest date	9/25	10/12	10/2	9/28	10/1	10/13	10/29	10/22
Tillage date	5/20	4/10	5/24	5/21	6/3	4/10	6/20	5/10
Chemical fertilizer: Type, Rate (kg N ha ⁻¹),Dates	-	Urea,50,5/9	Urea,100,6/1	-	-	-	-	Urea,20,5/10
Manure: Type, Rate (kg N ha ⁻¹), Dates	CDM ^b , 400,5/16	-	CDM ^b , 300 ,5/16	Slurry, 350,5/17	Slurry, 200,6/1	-	CDM ^b , 250,6/17	Slurry, 230,8/2
Flooding starting date	-	4/15	-	-	-	4/15	-	5/15
Flooding ending date	-	9/10	-	-	-	9/10	-	9/10
Winter crop	IRG ^c	-	IRG ^c	IRG ^c	OCG ^d	-	Barley (H ^a)	Barley(H ^a)
Sowing date	10/10	-	10/10	10/18	10/30	-	11/10	11/10
Harvest date	5/10	-	5/15	5/10	5/24	-	6/10	5/9
Tillage date	10/9	-	10/9	10/18	10/29	-	11/5	11/5
Manure: Type, Rate (kg N ha ⁻¹),Dates	CDM,400,9/29	CDM,150,11/24	CDM,300,10/4	Slurry,350,9/29	Slurry,200,10/13	Slurry,200,12/29	CDM,250,11/4	CDM,250,11/4
C/N of manure	23	21	28	24	24	24	12	12

^a H indicates the crop for human food and others crops used for cow forage. ^b CDM denotes composted dry manure. ^c IRG indicate Italian ryegrass. ^d OCG indicates orchard grass.

2.3 Calibration and Validation

Model calibration was performed on the basis of the observed data from eight fields in January 2008 to April 2009. We calibrated the model by focusing on the following points: 1) the model prediction capability of soil climate, such as temperature and moisture, that suggested the simulation of soil thermo and hydrology transforms; 2) the simulation of crop growth; 3) the simulation of the soil NH_4^+ -N, and NO_3^- -N content dynamic patterns and denitrification rate, which represents soil N transformation prediction; and 4) the soil gas N_2O flux assessments, which indicate the simulation of both nitrification and denitrification processes, as well as the soil gas diffusion capability.

Calibration was performed along the following steps: First, the simulated soil microclimate was verified against the measured soil temperatures and moisture contents. Second, the parameters of crop biomass compartments, namely, grain, stem, leaf, and root, were verified for maximum grain biomass carbon (C) production, biomass allocation, and biomass C/N ratio in each compartment on the basis of the field observations (Table 2). Third, given that Andosols have high soil organic carbon (SOC) contents and a major component comprising a recalcitrant organic C pool (Shindo and Honma, 2001), the humus in this study was assumed to represent 5% of the total SOC to achieve good crop biomass production simulation on these soils (Takata et al 2011). Finally, the soil microbial activity index ranging from 0 to 1 in DNDC, which is used to express the soil toxic material impact on soil microbial activity, was set to 0.1 for Andosols to account for the lower ratios of microbial biomass C/soil organic C than that in non-Andosol soils (Inubushi et al., 2005). This parameter was adjusted further for each manure management practice with consideration of the differences in daily N_2O fluxes and denitrification rate of soils among management systems. A soil microbial activity index of 0.1 or 0.15 was used for compost or chemical fertilizer mixed with surface soil and slurry applications on Andosols, respectively.

To validate the model, soil temperature, moisture, soil NH_4^+ -N and NO_3^- -N content, N_2O fluxes, and crop N uptake from May 2009 to April 2010 were used to determine the model's performance level.

Table 2. The input parameters for crop growth.

	Maize			Grass			Rice			Barley		
	Observed	Default	Modified	Observed	Default	Modified	Observed(H/F) ^a	Default	Modified(H/F) ^a	Observed	Default	Modified
Grain max. biomass(kg C ha ⁻¹)	3512	1920	3500	43	48	45	3277/ 4463	2560	3500/ 4500	1146	1040	1200
Biomass fraction (%)												
Grain	0.51	0.39	0.44	0.01	0.01	0.01	0.41	0.41	0.41	0.3	0.3	0.3
Shoot	0.39	0.49	0.46	0.72	0.45	0.72	0.45/ 0.49	0.54	0.45/ 0.49	0.51	0.47	0.51
Root	0.1	0.12	0.1	0.27	0.54	0.27	0.14/ 0.1	0.05	0.14/ 0.1	0.19	0.23	0.19
C:N ratio												
Grain	37	35	35	33	33	33	35	27	33	21	20	20
Shoot	48	50	50	28	33	28	52	45	50	48	50	50
Root	50	50	50	NA ^b	50	50	36	55	36	NA ^b	50	50
TDD ^{\$}	2505	2550	2500	1100	2500	1000	2507	1500	2500	959	1000	1000

^a H and F denote the crop used for human food and cow forage, respectively. ^b NA and \$TDD represent the data not available and accumulative thermal degree days of crop growth season.

2.4 N cycle environmental risks in current intensive livestock-crop systems

Long-term simulation allows exploring the effects of climatic variability and agriculture management on crop production and soil N dynamics. Thus, long-term simulations of current intensive manure application systems were tested over a 30-year series on the basis of historical weather data (1981–2010) from the aforementioned weather stations. During these 30 years, the annual precipitation and temperature was 888–2051 mm and 9.5 °C–13.1 °C, respectively. Based on simulation results, the main factors influencing N cycling could be determined such as soil texture and land uses. Under those factors, the effect of manure N application rates on crop yields and loss N was analyzed to determine the suitable N application rate.

Regarding the evaluation N environmental risks, since NH_3 volatilization from Andosol was limited and that accounted for only 0.2% of the applied (Hayashi et al. 2011), the NO_3^- -N leaching and N_2O emission would be the important N environmental strengths for Andosol and those two N flows were analyzed in this study. Due to no field monitoring of NO_3^- -N leaching and long-term observations in calibration and validation processes, the NO_3^- -N leaching and N_2O emission data from the long-term simulation were further compared with literature data. For literature review, here we inputted keywords as "Andosol, maize, wheat, barley, Ryegrass or paddy rice" and combined with "nitrogen leaching" or " N_2O emission" to search the articles from the "web of science".

2.5 Data analyses

Model performances in calibration and validation were evaluated using the components of mean square deviation (MSD) (Gauch et al., 2003): nonzero intercept (SB), non-unity slope (NU), and lack of correlation (LC). The following equations were used:

$$\text{MSD} = \frac{\sum (X_i - Y_i)^2}{N} \quad (1)$$

$$\text{SB} = (\bar{X} - \bar{Y})^2 \quad (2)$$

$$\text{NU} = (1 - b)^2 \times (\sum x_i^2 / N) \quad (b = \sum x_i y_i / \sum x_i^2) \quad (3)$$

$$\text{LC} = (1 - r^2) \times (\sum y_i^2 / N) \quad (r^2 = (\sum x_i y_i)^2 / \sum x_i^2 \sum y_i^2) \quad (4)$$

where the sample size is denoted by N , Y_i representing the i^{th} measured value, and X_i corresponds to the i^{th} model predicted value. SB results from the difference of two means; and NU and LC relate transparently to the regression parameters b and r^2 , respectively. The three components SB, NU, and LC, which add up to MSD, have distinct and clear meanings representing the deviation between predicted and measured values derived from translation, rotation, and scatter, respectively. The slope b and r^2 also used to confirm the performance of modeling. The comparison between model simulations and literature observations was tested by ANOVA on ranks in Dunn's test using Sigmastat 12.0.

3. Results and analysis

3.1 Model calibration

Soil moisture contents (WFPS) and soil temperature showed good agreement between model simulation and field observations of upland and paddy rice in January 2008 to April 2009 ($p < 0.01$; Table 3). For soil NO_3^- -N and NH_4^+ -N concentrations and N_2O fluxes, the general patterns were still predicted correctly by the model even without significant correlation between simulated and observed values in both uplands and paddy rice systems (Fig. 1 and Table 3). For denitrification rates, the model was simulated reasonably well in uplands, but exhibited weak agreement in paddy fields (Fig. 1 and Table 3). The simulated values trended to underestimate denitrification rate during flooding period in paddy rice, especially in R5 after slurry application in summer season. In case of R1 and R4, most of fertilizer applied in winter season and the available N would be very limited for denitrifiers in rice growing season. The denitrification rate could be very low in flooding time. The low soil mineral N content have been simulated in R1 and R4 (Fig. 1). On the other hand, the observed denitrification rates also showed big variation. So, the simulation from DNDC in R1 and R4 would be reasonable. But for field R5, after the slurry applied in August, even the denitrification rate from DNDC trend to increased, but it was still very low. Even though, denitrification rate in most of fields has been simulated well. For crop growth simulation, the calibrated model showed good agreement between simulation and observed

values across maize, rice, forage rice, grass, and barley (Fig. 2 and Table 3). Over all, calibrated model was able to predict N transformations in current crop systems.

Fig.1 The calibration of the concentrations of soil $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ (0-10 cm), N_2O fluxes and denitrification rate over all fields. Error bars indicated the standard error of average values. The grey dotted areas and arrows indicated the timing of flooding and fertilization, respectively. G, M and R suggested grass, maize and rice, respectively.

Table 3. Model performance statistics from different crop systems for predicting daily soil physical-chemical properties(0-10cm), soil N₂O emissions, denitrification rate and crop N uptake.

	Upland fields								Paddy rice fields							
	b(n) ^a	R ²	MSD	SB (%)	NU (%)	LC (%)	Observed mean(range)	Modeled mean(range)	b(n) ^a	R ²	MSD	SB (%)	NU (%)	LC (%)	Observed mean(range)	Modeled mean(range)
Calibration																
WFPS (%)	0.87(40)	0.83 ^{**}	43.2	3	0	97	36.9(10.9-60.7)	35.5(7-56)	1.02(27)	0.95 ^{**}	3.82	54	12	33	54.9(19.2-100)	53.6(13-100)
Soil temperature (°C)	0.91(40)	0.94 ^{**}	3.93	84	11	5	10.1(0.58-28.1)	11.8(1.5-26.3)	0.81(27)	0.90 ^{**}	3.67	31	49	20	11.5(-0.5-26.4)	12.6(1.6-26.3)
NO ₃ ⁻ -N content (kg N ha ⁻¹)	-0.01(40)	0.003	198.3	39	1	60	9.28(0.56-72.2)	0.50(0-4.96)	0.26(27)	0.053	70.8	26	32	42	7.68(0.68-26.7)	3.36(0-27.7)
NH ₄ ⁺ -N content(kg N ha ⁻¹)	-0.69(40)	0.008	1159	0	99	1	2.92(0.42-12.4)	4.40(0.03-131)	-0.15(27)	0.006	866	7	78	14	9.51(0.41-56.4)	17.6(0-80.7)
Daily N ₂ O flux(kg N ha ⁻¹ d ⁻¹)	1.32(40)	0.034	0.001	30	60	10	0.005(0-0.03)	0.015(0-0.25)	0.12(24)	0.044	0.001	16	18	66	0.008(0-0.03)	0.003(0-0.019)
Denitrification rate (kg N ha ⁻¹ d ⁻¹)	0.90(35)	0.44 ^{**}	0.005	2	6	93	0.04(0.00-0.57)	0.05(0.00-0.83)	0.001(24)	0.01	13.5	8	0	92	1.04(0-17.9)	0.005(0-0.04)
Crop N uptake	1.02(10)	0.96 ^{**}	5.3	9	27	64	115(48-213)	116(45-196)	0.82(4)	0.92 [*]	87.6	16	76	9	149(72.4-219)	146(91-214)
Validation																
WFPS (%)	0.70(102)	0.33 ^{**}	111	9	21	70	27.7(6.83-61.5)	30.9(2.14-55.3)	1.07(57)	0.92 ^{**}	34.3	50	24	27	63.4(8.83-100)	59.3(1.22-100)
Soil temperature (°C)	0.68(104)	0.71 ^{**}	3.72	4	43	54	17.0(1.83-29.1)	16.7(4.51-24.8)	0.70(52)	0.72 ^{**}	4.87	2	43	55	15.0(2.23-26.0)	15.3(3.41-24.9)
NO ₃ ⁻ -N content (kg N ha ⁻¹)	0.79 (47)	0.18 ^{**}	175	0	19	81	14.2(0.91-68.0)	14.9(0.01-131)	0.36(24)	0.06	111	3	47	50	7.14(0.77-35.3)	5.33(0.01-47.2)
NH ₄ ⁺ -N content(kg N ha ⁻¹)	0.33(49)	0.33 ^{**}	51.9	0	25	75	2.66(0.02-63.1)	2.98(0.02-25.0)	-1.75(24)	0.00	2814	5	95	0	0.68(0.02-2.53)	12.0(0-73.4)
Daily N ₂ O flux(kg N ha ⁻¹ d ⁻¹)	0.55(104)	0.33 ^{**}	0.002	5	28	68	0.033(0-0.39)	0.023(0-0.31)	-0.70(55)	0.01	0.003	1	98	1	0.004(0-0.04)	0.009(0-0.22)
Cumulative N ₂ O fluxes	0.90(10)	0.95 ^{**}	0.05	41	44	15	1.24(0.17-5.86)	1.09(0.07-5.46)	2.88(6)	0.27	3.47	5	77	18	0.15(0.03-0.39)	0.57(0-1.74)
Crop N uptake	1.01(10)	0.95 ^{**}	9.60	1	2	97	97.5(18.8-199)	97.2(20.1-186)	0.95(4)	0.99 ^{**}	8.85	2	96	2	118(18.8-189)	117 (26.9-192)

^a b indicates the slope of the least-squared regression of measured vs. modeled values. MSD indicates mean square deviation. SB, NU and LC mean nonzero intercept, non-unity slope, and lack of correlation, respectively. WFPS means soil water filled pore space. ** suggested the significant linear correlation between simulated values and observed values at the level p<0.01.

Fig.2 The calibration and validation crop nitrogen uptake for all crops. The grey marks indicated the results of calibration and the opened marks were used for validation. Error bars indicated the standard error of average values.

3.2 Model validation

In upland fields G/M1, G/M2, G/M3, G/M4, and M5, soil moisture contents(WFPS), temperatures, and soil inorganic N content were simulated reasonably well during May 15, 2009 to May 14, 2010 (Fig. 3). The simulated values showed significant linear correlation with the observed data ($p < 0.01$) (Table 3). The measured WFPS, soil temperature, NO_3^- -N content, and NH_4^+ -N content averaged 27.7%, 17.0 °C, 14.2 kg N ha⁻¹ d⁻¹, and 2.66 kg N ha⁻¹ d⁻¹ with ranges of 6.83%–61.5%, 1.83 °C–29.1 °C, 0.91–68.0 kg N ha⁻¹ d⁻¹, and 0.02–63.1 kg N ha⁻¹ d⁻¹, respectively. Some jumps of soil mineral N had been simulated in G/M3 and G/M4, mainly caused by the slurry application which have a very high inorganic N content. For other fields, the fertilizer was composed manure and no such big jump observed. This result indicated that DNDC were able to predict N dynamics with different manure. For daily soil N₂O fluxes, the simulated daily N₂O fluxes generally followed the observed seasonal patterns and ranged from 0–0.39 kg N ha⁻¹ d⁻¹ and 0–0.31 kg N ha⁻¹ d⁻¹ for observed and modeled values, respectively (Fig. 3 and Table 3). The DNDC model was able to simulate the variation in daily N₂O emissions in upland fields. Both the modeled and observed values showed that the winter cropping season had higher N₂O emission after fertilization than the summer, and the model explained 95% of the variation in cumulative N₂O fluxes (Fig. 4 and Table 3). In the summer season, the observed emissions ranged from 0.17–0.64 kg N ha⁻¹, whereas the model simulated values ranged from 0.07–0.56 kg N ha⁻¹. In the winter season, the N₂O emissions were 0.26–5.86 kg N ha⁻¹ and 0.46–5.46 kg N ha⁻¹ for observed and modeled values, respectively.

In paddy rice fields R1, R4, and R5, the observed values were in the 8.83%–100% range for WFPS and 2.23 °C–26.0 °C for soil temperature, and the measured patterns were predicted by the model (Fig. 3 and Table 3). The model explained 92% and 72% of the variation in soil water content and soil temperature, respectively (Table 3). However, the soil N transformation in paddy rice from the model simulation showed weak agreement with the field observation. The high non-unity slopes (NU) indicated that the values derived from model overestimated the high-level values of soil NO_3^- -N and NH_4^+ -N content and N₂O fluxes, but underestimated the low-level values. The high LC suggested that the simulation had big variations in both smaller and bigger magnitude values. However, the general patterns predicted well across all fields. The cumulative N₂O emission

from October to November in 2009 winter ranged from 0.39–1.74 kg N ha⁻¹ from the DNDC simulation, but was only 0.22–0.39 kg N ha⁻¹ from the field observed values (Fig. 4). In summer, the N₂O fluxes from both experimental and model simulation were nearly negligible. The lower observed cumulative N₂O fluxes during winter in R4 and R5 fields can be attributed to the limited sampling and some missed N₂O flux peaks (Fig. 3). In the case of R4, the model slightly underestimated N₂O emissions after winter fertilization (Fig. 3).

Overall, the model simulated differences in N₂O emissions in both upland and paddy rice crop systems reasonably well. The daily fluxes from upland fields were approximately 0.01–0.1 kg N ha⁻¹ d⁻¹, higher than those from paddy rice fields (mostly less than 0.01 kg N ha⁻¹ d⁻¹). Manure application stimulated N₂O fluxes. The summer fertilizer season (May–June) showed lower N₂O emissions than the winter fertilizer time (October–November), which may be attributed to the relatively high summer crop nutritional requirement.

The validation of crop N uptake was simulated correctly for maize, rice, forage rice, grass, and barley with rotations (Fig. 2 and Table 3). The modeled crop N-uptakes in all crops were within the measured error ranges. Over 95% of the variation was explained by DNDC. The results suggested that the model can simulate crop growth correctly.

Fig.3 The validation of daily WFPS (0-10 cm), soil temperature (0-10 cm), and the concentrations of soil $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ (0-10 cm) and N_2O fluxes over all fields. Error bars indicated the standard error of average values. Arrows indicate the timing of fertilization. WFPS means soil water filled pore space. G, M and R suggested grass, maize and rice, respectively.

Fig.4 The validation of simulated and observed cumulative N₂O emissions in different seasons for all fields. Error bars indicated the standard error of average values. G, M and R suggested grass, maize and rice, respectively.

3.3 Environmental risk of N cycle in current crop systems

3.3.1 The effect of long-term using current manure management on N flows

In uplands

For upland systems, crop N uptake averaged from 186 ± 25 to 239 ± 30 kg N ha⁻¹yr⁻¹ throughout the 30-year simulation period (Fig. 5). The lowest value was found at the G/M4 field, which had loam sand soil with approximately 1% clay content. No difference was observed in crop N uptake in the other fields. Under the current manure management practices, the crop N use efficiency varied from 27% in G/M1 to 42% in M5 field.

Modeled NO₃⁻-N leaching in each field ranged from 123 kg N ha⁻¹yr⁻¹ to 362 kg N ha⁻¹yr⁻¹, which accounted for 27%–41% of applied N (Fig.5). The lowest value was in G/M4, and the NO₃⁻-N leaching rates in other uplands were over 35% of applied fertilizer N. For all upland crops, NO₃⁻-N leaching was only 13–75 kg N ha⁻¹yr⁻¹ in the first year and then increased over the next 20 years (Fig. 5). Throughout the 30-year simulation period, NO₃⁻-N leaching in upland systems was positively correlated with annual precipitation ranging from 888–2051 mm ($p < 0.05$, data not shown). The highest value of 767 kg N ha⁻¹yr⁻¹ was in the field G/M1 with a compost rate of 800 kg N ha⁻¹yr⁻¹ when precipitation was 1560 mm.

Nitrous oxide emissions showed an annual average of 6.53–11.8 kg N ha⁻¹yr⁻¹, which was equivalent to 0.86%–1.59% of applied N (Fig. 5). The fields G/M3 and G/M4 under slurry application exhibited higher N₂O emissions (1.53%–1.59% of input N) than the fields under compost application (0.86%–1.19%). Similar to the pattern observed for NO₃⁻-N leaching, the N₂O fluxes were increased with the increased annual precipitation ($p < 0.05$, data not shown). The simulated highest N₂O 29 kg N ha⁻¹yr⁻¹ was in the field G/M3 with slurry application of 700 kg N ha⁻¹yr⁻¹ when the precipitation was 1415 and 1440 mm.

In paddy crop systems

The modeled crop N uptake for paddy rice systems varied from 86 kg N ha⁻¹yr⁻¹ to 304 kg N ha⁻¹yr⁻¹, whereas N use efficiency was 37%, 52%, and 55% for the R4, R1, and R5 fields, respectively (Fig. 5). The highest crop N uptake was in R5 with forage rice and barley rotation. Over the 30-year simulation period, crop N uptake was relatively constant in R1 and R5, but decreased in R4 (Fig. 5).

The N leaching and N₂O emission was 17.4–103 kg N ha⁻¹yr⁻¹ and 0.59–2.77 kg N ha⁻¹yr⁻¹, respectively (Fig. 5). In the rice systems, R4 (loam sandy) had the highest N losses with 44% and 1.22% of applied N due to NO₃⁻-N leaching and N₂O emission, respectively. R1 and R5 had N losses of 7.5% and 17% of N input as NO₃⁻-N leaching and 0.26% and 0.40% as N₂O emission, respectively. Compared with upland, both NO₃⁻-N leaching and N₂O emission in paddy fields showed smaller variations over simulated time (Fig. 5).

Fig.5 The long term simulations of crop N uptake, NO_3^- -N leaching and N_2O emission under current intensive manure application crop systems over 30 years using the calibrated model. G, M and R denoted grass, maize and rice, respectively.

3.3.2 the comparisons of NO_3^- -N leaching and N_2O emissions from simulation and literatures

Based on the results, NO_3^- -N leaching and N_2O emission would be the significant N risks in the current intensive manure systems, particularly for uplands and loam sand paddy rice after 20 years. After 20 years, the simulated NO_3^- -N leaching from uplands ranged from 9% to 54% with a median of 34% and an average 35% of total input N, which is the same order of magnitude as the percentage of total input N reported in the literature having a median of 24% and an average 33% of input N (Fig. 6). Leaching N was positively related with nitrogen application rate (Fig.7). For one-season rice crop systems, the modeled value from loam soil, 2.2%–10.4% of input N with a median 7.8% of input N, was generally in the same range as that in the literature, 0.58%–36% of input N and with a median 2.6% of input N rate (Fig. 6). By contrast, the loam sand soil showed a higher value than the literature observation and modeled loam soil. The value ranged from 16.4%–63.8% of input N with a median of 37.0%. In case of forage rice, the DNDC simulated value ranged from 4.8%–26.5% with a median 14.8% of input N, which showed a similar magnitude as that of literature observations of 0.5%–17.0% with an average 7.3% of input N. Comparing the different crop systems, the paddy rice crops showed lower NO_3^- -N leaching rate for both modeled and literature observed values, except in loam sand soil. Forage rice was inclined toward higher NO_3^- -N leaching than one-season rice crop system.

For N_2O emissions, no difference was observed between modeled values and literature observations over uplands, one-season rice, and forage rice crop systems (Fig. 6). The uplands showed greater N_2O emission than paddy rice. The N_2O emission from uplands was modeled at 0.54%–3.83% with an average of 1.61% of input N, whereas that of literature was 0.13%–11.1% with an average of 1.78% of input N. In one-season rice crop systems, the simulated loam sand soil had higher N_2O emission of 0.17%–2.02% with an average of 0.90% of input N than the loam soil with an average of 0.39% of input N. The literature observation was from 0.31%–1.97% with an average of 0.94% of input N. For forage rice, the N_2O emission ranged from 0.11%–1.01% with an average of 0.48% of input N and 0.03%–3.16% with an average of 1.37% of input N for DNDC simulation and literature observations, respectively. The N_2O emissions also increased with the increasing of N application rate (Fig. 7).

Fig.6 Comparison of leaching NO_3^- -N and N_2O -N in different crop systems between DNDC simulation and literatures field observations. The error bars indicated the median, 10th, 25th, 75th and 90th percentiles of the values.

Fig.7 The influence of N fertilizer rate on (a) leaching NO₃⁻-N of uplands and (b) N₂O emission from paddy rice fields

3.4 The critical N loading in current crop systems

To explore the critical N application rate for the typical crop system in the study region, the effect of different N application rates on crop N uptake and loss N was determined on loam soil and loam sand soils for uplands grass/maize and silt loam soil for forage rice/barley under compost application, as well as loam soil for paddy rice/fallow with slurry and urea application, respectively, on the basis of previous results (Fig. 8). In the range of compost application rates of 100–700 kg N ha⁻¹yr⁻¹, the simulation showed that crop N uptake and loss N increased in a negative and positive polynomial manner, respectively, for both loam sand and loam soils with maize and grass rotations. The loam sand soil showed a lower crop N uptake and higher loss N than loam soil. The crop N uptake increased as the increasing of crop yield and reached 50% of applied N till the compost rate increased to 383 and 471 kg N ha⁻¹yr⁻¹ for loam sand and loam soil, respectively. Otherwise, the crop N use efficiency will lower than 50% of input N and loss N will higher than 50% of input N. So, the critical N point was defined as the crop N uptake reached to 50% of applied N where the high crop yield could be expected. In the case of the paddy rice systems, the crop N uptake and loss N in forage rice/barley and rice/fallow systems showed a similar trend as that of upland grass/maize cropping systems (Fig. 8). The critical N application rates for crop N uptake to reach 50% of applied N were 638 kg N ha⁻¹yr⁻¹ in slurry for forage rice/barley and 392 kg N ha⁻¹yr⁻¹ in slurry or urea applied in winter season for rice/fallow system. For the rice/fallow system, a higher crop N uptake and lower loss N was achieved with the use of urea rather than slurry if the application rate was lower than the critical N rate.

Given the current nitrogen application rate in the intensive region, the manure N rates of over 600 kg N ha⁻¹yr⁻¹ in uplands were much higher than the critical N rate, and loss N was higher than 50% of input N (Fig. 8). However, in the case of the paddy rice fields, the loss N from both forage rice and one-season rice systems showed lower N pollution risk, and the fluxes were less than 50% of input N (Fig. 8).

Fig.8 Effects of fertilizer N rate on crop N and loss N under different crop systems. The loss N was calculated from the difference between input N and crop N. The shadow area indicates the current N application rate in different crop systems.

4. Discussion

4.1 Model performance on Andosols

Across all fields, calibration and validation showed a good fitness for both upland and paddy rice crop systems with different soil textures, as well as different manure management strategies. First, the comparison of simulated and observed data for daily WFPS, soil temperature, soil mineral N, denitrification rate, and N₂O fluxes indicated that the model generally captured the timing and magnitude of properties in most fields. The calibrated model better predicted the simulations of uplands than that of paddy rice. In the paddy rice systems, the calibrated model underestimated the denitrification rate during the flooding season. For rice field R4, which is located in lowland areas, soil moisture content and N₂O emissions were also underestimated during unflooding period, and an overestimated soil mineral N was consequently simulated. DNDC is a one-dimensional model that failed to simulate soil water accumulation in lowland areas, which could dilute soil nutrient contents and decrease soil aerobic conditions (Li et al., 2006). This condition may be the main reason for the weak agreement.

For the cumulative N₂O emission of R4 and R5 fields, the model overestimated the values for the winter period from October 1 to November 30 in 2009. This discrepancy was attributed to the limited sampling and absence of N₂O peaks. Despite the weak agreements found in the simulation, the general patterns in soil mineral N content and N₂O fluxes matched well over all rice fields. For example, the model showed high N₂O peaks after the drainage stopped in three rice fields, and similar N₂O fluxes in paddy rice were observed in other field studies (Nishimura et al., 2011; Toyota et al., 2008).

Overall, the model was capable of predicting the cumulative N₂O emissions and crop N uptake over a wide range of soil textures (loam, sandy loam, loam sand, silt loam), manure managements (slurry and compost with an application rate in the range 200–800 kg N ha⁻¹yr⁻¹), and cropping systems (maize/grass, maize/barley, forage rice/barley, and paddy rice/fallow). Therefore, the calibrated DNDC model can reveal the effects of fertilization management on crops and the physical and chemical processes of soil in Andosols. Moreover, the model can

predict soil N fluxes on Andosols under different manure management practices in both upland and paddy rice cropping systems.

4.2 Long-term impacts of intensive manure application

After successful calibration and validation, the long-term simulation showed stable crop N uptake in most fields, indicating that the current manure application rate could be sufficient for the crop nutrient requirements. Duan et al. (2014) reported the wheat/corn system in Zhenzhou and Qiyang of China showed a stable wheat yield over 15 years of observation under sufficient fertilizer application rates. By contrast, the crop yield will decrease when an insufficient amount of fertilizer nutrients was applied, such as in the case of rice field R4. The simulated crop N uptake was influenced by crop system and soil texture. In upland systems, crop N uptake was in the range of 186–239 kg N ha⁻¹yr⁻¹, which was similar with the crop N uptake of 195–285 kg N ha⁻¹yr⁻¹ from a corn–ryegrass system under five different water management systems over a two-year observation period (Zhou et al., 1997). Forage rice variety showed the highest crop N uptake of 304 kg N ha⁻¹yr⁻¹ in paddy forage rice/barley R5. Zhou et al. (2008) reported that N uptake by a type of forage rice reached up to 377 kg N ha⁻¹. Sandy soil (G/M4 and R4) showed a lower crop N uptake than the other soils, possibly because of the water and nutrient deficiencies in sand soil (Ismail and Ozawa 2007). Generally, the paddy rice fields showed a high nitrogen using efficiency ranging 37%–58% and with an average of 50%, while that of uplands was 27%–42%, with an average of 34%.

The long-term simulated NO₃⁻-N leaching and N₂O emission rates were within the range reported in the literature reviews. The NO₃⁻-N leaching and N₂O emission were the significant N environmental pollution sources in the current intensive manure application systems and could thus result in high and stable pollution strength for upland crop systems after 20 years. In the case of NH₃ volatilization, Andosols have low pH and showed only 0.2% NH₃ volatilization of the applied N (Hayashi et al., 2011). Thus, NH₃ volatilization could be ignored if considering the high fluxes of NO₃⁻-N leaching and N₂O emission in our crop systems. In both DNDC simulation and literature observations, higher manure N input resulted in higher leaching rate and N₂O emission.

Over the 30-year simulation, NO_3^- -N leaching and N_2O emission tended to increase with an increase in annual precipitation, particularly for upland systems. The N leaching rate in DNDC depends on the soil NO_3^- -N concentration and hydrology flow (Li et al., 2006). High precipitation would increase soil water discharge flow, such that N leaching rate would consequently increase. For N_2O emission, the high precipitation could stimulate more soil N_2O flux by increasing soil denitrification capacity (Mori and Hojito, 2012). The higher nitrate leaching and N_2O emission during higher precipitation years has also been found in a considerable amount of research (Long and Sun, 2012; Mori and Hojito, 2012). Therefore, a high N pollution disaster could occur under current N management practices during a year with high precipitation.

Greater NO_3^- -N leaching and N_2O emission have been simulated in uplands than in paddy rice. Similarly, the literature review also showed the same trend for NO_3^- -N leaching and N_2O emission in both crop systems. NO_3^- -N leaching ranged from 5.2 kg N ha⁻¹yr⁻¹ to 496 kg N ha⁻¹yr⁻¹ for the uplands, for 85 field observations under 67–1000 kg N ha⁻¹yr⁻¹ input N. However, in the paddy rice fields, only 1.7–47.5 kg N ha⁻¹yr⁻¹ NO_3^- -N leaching with 81–600 kg N ha⁻¹yr⁻¹ fertilizer from 29 observations. From the medians (Figs. 6 and 7), the simulated values trended higher than the literature observations. These high leaching rates from the simulation could be attributed to the existence of groundwater at a depth of 1 m in the study region (Somura et al., 2008; Yamanaka et al., 2003), the relatively high porosity of Andosols, and the high N input with the limited crop N uptake. In the case of N_2O emission, observations in uplands were in the range of 0.07–23.3 kg N_2O -N ha⁻¹yr⁻¹ for Japan (Akiyama et al., 2006) and those for paddy fields were 0.1–17.9 kg N_2O -N ha⁻¹yr⁻¹ from 43 field observations under 90–600 kg N ha⁻¹yr⁻¹ fertilization. In paddy rice, the N_2O emission mainly occurred in the un-flooding period (Minamikawa et al. 2010; Nishimura et al., 2011; Zhao et al., 2011). Compared with the composed manure, the slurry application tends to stimulate more N_2O emission, such as in the case of fields G/M3 and G/M4 with approximately 1.55% of input N, as well as in R4 and R5 with 1.22% and 0.4 % of input N, respectively. Riya et al. (2012) also observed a high N_2O up to 17.9 kg N_2O -N ha⁻¹yr⁻¹ only in forage rice season with cattle slurry of 567 kg N ha⁻¹yr⁻¹. This high N_2O emission could be attributed to the higher anaerobic condition and more available N provided by slurry application and the stimulated soil denitrification process

(Deng et al., 2012; Singla et al., 2014). Compared with that in the literature, the N_2O emission from this simulation tended to have lower fluxes (Fig. 7). This lower emission may be explained by the higher porosity and lower denitrification capability in Andosol (Deng et al., 2012). Given the soil texture, greater N leaching was found in sandy soil (Asada et al., 2013a; Thomsen et al., 2014). For paddy rice R4 of loam sand soil, the leaching N reached 44% of input N.

Generally, the predictions demonstrated that the current intensive manure application agriculture systems have low crop N uptake rate and that a large amount of N (e.g. N_2O emission, NO_3^- -N leaching) would be lost to the environment. The uplands showed more NO_3^- -N leaching and N_2O emission than paddy rice fields. The forage rice/barley crop systems have a high nitrogen removal capability because of greater crop N uptake. In the current intensive manure systems, 36% and 23% of input N was lost as NO_3^- -N leaching from uplands and paddy rice fields, respectively, with a corresponding N_2O emission of 1.24% and 0.63% of input N. Precipitation was one of the main factors regulating soil N transportation. NO_3^- -N leaching and N_2O emission would be significant environmental pollution sources during the heavy rain period. The model showed that the NO_3^- -N leaching and N_2O emission would continue increasing for approximately 20 years and would then stabilize. Long and Sun (2012) also reported that the nitrate leaching was continue increasing for over eight years under pig manure application. Therefore, the NO_3^- -N leaching would become a significant N pollution strength under intensive manure application with a couple of years later.

4.3 Model adequacy for nitrogen environmental risk evaluation

Due to the high nitrogen pollution strength from the current intensive livestock region, an urgent need has emerged to implement efficient agriculture and environmental policies to mitigate the current environmental problems among livestock farmers. In this study, the calibrated DNDC was employed to explore the effects of N cycling on intensive manure application and the critical N loading in current intensive livestock region. The simulated results clearly showed the amount of fertilizer to be applied in different soils and how much environment pollution risk would be lost under current fertilizer situations. Thus, farmers can easily determine

the potential fertilizer rate needed to obtain the highest crop yield and how much crop yield would be obtained under the use of different fertilizers (chemical or organic).

Integrating all the analyses, the N flow in the current study region was influenced by crop systems, soil texture, and fertilizer type. The uplands showed lower N use efficiency and serious leaching N and N₂O emissions than paddy rice crop systems. The critical N rates were 383, 471, 638, and 392 kg N ha⁻¹ yr⁻¹ for loam sand with maize/grass, loam soil with maize/grass, forage rice/barley, and rice/fallow with winter manure application, respectively. The sand soil showed high leaching N and was thus unsuitable for planting with paddy rice. Furthermore, slurry would stimulate more N₂O emission than compost manure.

5. Conclusions

This study is the first to calibrate and validate DNDC by using soil mineral N content, denitrification rate, N₂O fluxes, and crop N uptake across a range of soil textures and crop systems for evaluating N environmental risk. Results showed that the calibrated DNDC model captured the general trend regarding the chemical and physical properties of soils, denitrification rate, and the N₂O fluxes and crop N uptake from both upland and paddy rice crop systems with reasonable accuracy. The good agreement in different crop systems across different soil textures indicated that the calibrated DNDC model can be used satisfactorily to predict N flows on Andosols in both upland and paddy rice systems. Considering all of results from simulations, the current crop N use efficiency is 27%–42% and 37–58% for upland and paddy rice systems, respectively. A significant risk of NO₃⁻-N leaching and N₂O emission, especially in upland crop systems, can emerge in the study region. The level of NO₃⁻-N leaching and N₂O emission would increase when the input N rate and precipitation increases. Sandy soil showed a lower crop N uptake and higher N loss rate than the other soil textures, which underlined the importance of soil texture for N flow. Compared with compost manure, slurry application stimulated soil N₂O emission, indicating the necessity to shift from slurry to compost application to reduce N₂O flux. Over all, the calibrated DNDC model provides insight into potential policy impacts by integrating the complex spatial and temporal variations as a cost-effective manner. It successfully addressed

the N flows with different soils, climates, and manure managements on Andosols. This model represents a useful tool for evaluating N flows on Andosol in livestock manure systems.

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