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Emissions of nitrous oxide (N_2O) from soil surfaces and their historical changes in East Asia: a model-based assessment

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Abstract

This study assessed historical changes in emissions of nitrous oxide (N_2O), a potent greenhouse gas and stratospheric ozone-depleting substance, from the soils of East Asia to the atmosphere. A process-based terrestrial ecosystem model (VISIT) was used to simulate the nitrogen cycle and associated N_2O emissions as a function of climate, land use, atmospheric deposition, and agricultural inputs from 1901 to 2016. The mean regional N_2O emission rate in the 2000s was estimated to be 2.03 Tg N_2O year^{-1} (1.29 Tg N year^{-1}; approximately one-third from natural ecosystems and two-thirds from croplands), more than triple the rate in 1901. A sensitivity analysis suggested that the increase of N_2O emissions was primarily attributable to the increase of agricultural inputs from fertilizer and manure. The simulated N_2O emissions showed a clear seasonal cycle and interannual variability, primarily in response to meteorological conditions and nitrogen inputs. The spatial pattern of the simulated N_2O emissions revealed hot spots in agricultural areas of China, South Korea, and Japan. The average N_2O emission factor (emission per unit nitrogen input) was estimated to be 1.38%, a value comparable to previous estimates. These biogeochemical modeling results will facilitate identifying ways to mitigate global warming and manage agricultural practices in this region.

Keywords: Global warming, Land use change, Nitrogen cycle, Regional budget, Terrestrial ecosystem

Introduction

Nitrous oxide (N_2O) is one of the long-lived anthropogenic greenhouse gases, which are attracting particular attention in the context of mitigating global warming. In addition, N_2O is recognized as the most important ozone-depleting substance in the stratosphere (Ravishankara et al. 2009). Despite the relatively small contribution N_2O has made to historical climatic warming (about 10% of the effect of carbon dioxide [CO_2] on historical radiative forcing; Intergovernmental Panel on Climate Change [IPCC] 2013), N_2O is among the most remarkable of the greenhouse gases in several respects. First, N_2O emissions are intimately linked with the biogeochemical cycle of nitrogen (N), which has been greatly impacted by anthropogenic effects that include fertilizer use and industrial activities that lead to atmospheric deposition of reactive nitrogen (e.g., Davidson 2009; Zaehle et al. 2011). Evaluation of the magnitude of N_2O emissions requires a model of the N cycle, and the data needed for calibration and validation of such a model differ from those used to estimate CO_2 and methane (CH_4) emissions. Second, N_2O emissions from soil, one of the major sources, are quite heterogeneous spatially and vary greatly temporally (e.g., Nishina et al. 2012; Bellingrath-Kimura et al. 2015). In soils, instantaneous N_2O production occurs within microzones (i.e., hot spots) of high-microbial activity and bursts of N_2O emissions often occur after rainfall and thawing of frozen soil (e.g., Kim et al. 2012; Muhr et al. 2008). Such high variability is intriguing from the standpoint of biogeochemical mechanisms but confounds quantification of emission rates with a small number of instruments.

Several studies have attempted to evaluate broad-scale (e.g., national, regional, and global) N_2O budgets. Galloway et al. (2004) have synthesized an overview of the global N cycle from available data and have shown that global continental N_2O emissions in 1860 and in the early 1990s were about 8 and 11 Tg N year^{-1}, respectively. Del Grosso et al.

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(2009) used the DAYCENT model to estimate that global annual N_2O emissions from corn, wheat, and soybean croplands in the 1990s totaled 1.15 Tg N year^{-1}. Syakila and Kroeze (2010) analyzed the global N_2O budget from 1500 to 2006 and estimated the global agricultural and natural soil emissions in 2000 to be 4.9 and 6–7 Tg N year^{-1}, respectively. Xu-Ri et al. (2012) used the DyN-LPJ model to simulate the global terrestrial N cycle and estimated total N_2O emissions in the twentieth century of 8.2–9.5 Tg N year^{-1}. Hashimoto (2012) has used an empirical model to estimate mean global soil N_2O emissions of 4.4 Tg N year^{-1}. Recently, more mechanistic models have been developed and used to evaluate global N_2O emissions from soils, the largest source to the atmosphere (Tian et al. 2018). However, large uncertainties remain in the present estimates of the broad-scale greenhouse gas budget because relevant data and understanding of biogeochemical processes are incomplete.

The global N_2O budget is very sensitive to human activities in Asia, especially East Asia. Rapid growth of the human population in Asia and associated economic expansion have led to a considerable increase of fertilizer use and industrial emissions that have caused serious environmental problems such as air pollution and water body eutrophication. This region is under the influence of the Asian monsoon climate system, and there is a clear gradient in precipitation from wet coastal areas to dry inland areas. A credible evaluation of the regional N_2O budget of East Asia requires taking account of such region-specific conditions. Shindo et al. (2003), for example, used a model developed for this region to conduct a study focused on N fluxes in the food production–supply system and related environmental impacts. Several country-specific studies have also been conducted. Yan et al. (2003) have used IPCC guidelines to estimate nitrogenous gas emissions of Asian countries. For China, Tian et al. (2011) used the Dynamic Land Ecosystem Model and estimated N_2O emissions from terrestrial ecosystems of 0.6 Tg N year^{-1}. For Japanese forests, Hashimoto et al. (2011) used a statistical model and estimated N_2O emissions of 0.00194 Tg N year^{-1}. Because of the paucity of regional studies that used up-to-date forcing data, it is still a challenging task to estimate N_2O emissions from East Asia.

In this study, we used a process-based terrestrial biogeochemical model of the East Asia region and the historical record of various driving factors (climate, land use, N inputs, and atmospheric CO_2 concentrations) to estimate the history of N_2O emissions from natural and agricultural soils. Riverine export of N and emissions from inland waters and coastal areas are interesting aspects of the N cycle, but they were excluded from the scope of this study. We analyzed the spatial patterns and temporal variability of the simulated N_2O emissions. Sensitivity simulations were conducted to relate the contributions of the driving factors to changes of historical emissions. We then examined the contributions of N_2O emissions from East Asia to the regional and global budgets. Finally, we assess the possibility of mitigating the impact of those emissions for climate management in the future, and we delineate future research needs and opportunities for improving model reliability.

Methods/Experimental

Description of biogeochemical model

We used a process-based terrestrial ecosystem model, Vegetation Integrative SImulator for Trace gases (VISIT; Inatomi et al. 2010; Ito and Inatomi 2012a). The model consists of component schemes that simulate the energy budget, hydrology, carbon (C) cycle, and N cycle in various types of terrestrial ecosystems, and it can be applied at global scales by selecting appropriate spatial grid systems. The model was developed for terrestrial biogeochemical and ecophysiological studies; the associated water and C cycle schemes have been described and examined in previous papers (Ito and Oikawa 2002; Ito and Inatomi 2012b). To consider the effects of land use, the model simulates natural vegetation and croplands separately at each grid. The hydrological scheme is represented by two (near-surface and below) soil water and snow pools, and it simulates evapotranspiration from the soil surface and vegetation canopy surface, infiltration, and runoff discharge. The carbon cycle scheme consists of three (leaf, stem, and root) plant and two (litter and humus) soil carbon pools and simulates carbon flows, including photosynthetic assimilation, respiratory emission, allocation to C pools, litterfall, and microbial soil decomposition. Each material flow or process is simulated in a mechanistic manner, i.e., on the basis of environmental regulation and biological responsiveness. The model has been validated by comparisons with measurements of atmosphere–ecosystem CO_2 exchange (Ito 2008; Inatomi et al. 2010) and satellite-based estimates of photosynthetic parameters (Ito et al. 2017).

The N cycle scheme consists of two (photosynthetic and non-photosynthetic) plant, two (litter and humus) soil-organic and two (ammonium [NH_4^+] and nitrate [NO_3^-]) soil-inorganic pools, as well as a microbial pool (see Inatomi et al. 2010 for the schematic diagram). The N cycle scheme simulates major N flows such as mineralization, nitrification, denitrification, NH_3 volatilization, nitrate leaching, microbial immobilization, and plant assimilation. To facilitate broad-scale application, the model takes account of these processes with an intermediate complexity; i.e., the simulations use forcing and characteristic data available at regional scales to account for specific processes. The principal processes that emit N_2O, nitrification and denitrification, are simulated separately by using the generalized parameterization of

Parton et al. (1996) developed on the basis of laboratory and field data. The nitrification-associated N_2O emission rate (FN_2O_{nit}) is calculated as follows:

$$FN_2O_{nit} = fW_{nit} \cdot fT_{nit} \cdot fpH_{nit}(K_{nit} + MX_{nit} \cdot fNH_4) \quad (1)$$

where fW_{nit}, fT_{nit}, fpH_{nit}, and fNH_4 are the coefficients that quantify the influence of soil water, temperature, pH, and NH_4^+ availability on the nitrification rate; K_{nit} is the N turnover coefficient; and MX_{nit} is the maximum rate of N_2O emission associated with nitrification when there is excess soil NH_4^+. The denitrification-associated N_2O emission rate (FN_2O_{den}) is calculated as follows:

$$FN_2O_{den} = \min(fNO_3, fCO_2) \cdot fW_{den} / (1 + R_{N2/N2O}) \quad (2)$$

where fNO_3 and fCO_2 represent the effects of soil NO_3^- and soil respiration (a proxy of C availability), fW_{den} is a metric of the effect of soil water on denitrification; min () denotes the smallest value of the term in the parenthesis; and $R_{N2/N2O}$ is the N_2 to N_2O emission ratio from denitrification-associated emissions and is formulated as another function of soil NO_3^-, soil respiration, and soil moisture content. The effect terms, fNH_4 and fNO_3, and the coefficients, fW_{nit}, fT_{nit}, fCO_2, fW_{den}, and $R_{N2/N2O}$, are provided as empirical functions (Parton et al. 1996) and are calculated at each grid for every time step. The fpH_{nit} coefficient is calculated at each grid using soil data and is assumed to be constant through time. K_{nit} and MX_{nit} are constant parameters. Soil moisture content simulated by the hydrology scheme and soil respiration by the carbon cycle scheme is used to estimate N_2O emissions.

The N cycle in the model thus responds directly to climate conditions such as temperature and precipitation. Atmospheric CO_2, which affects the ecosystem C cycle by its fertilization effect (e.g., Norby et al. 2010), can influence the N cycle by altering plant-root N uptake and soil respiration. The driving forces of the terrestrial N cycle include biological N fixation (estimated using the parameterization of Cleveland et al. 1999), atmospheric deposition of NH_4^+ and NO_3^-, and fertilizer inputs to croplands. In this study, the external drivers for the ecosystem (i.e., atmospheric deposition and fertilizer input) were obtained from appropriate datasets (explained later). Land use changes between natural (unmanaged) and agricultural (managed) ecosystems also affect regional N budgets by altering cropland area and are considered by the model (Kato et al. 2013).

Simulations and analyses

The continental-scale simulations were conducted at a spatial resolution of $0.5° \times 0.5°$ in latitude and longitude, and then results for East Asian countries (alphabetically, China, Chinese Taipei, Japan, Mongolia, and North and South Korea) were extracted by using common-use national boundary data. The total land area was about 11.6×10^6 km^2, and the total population in 2010 was about 1.58 billion. After a spin-up period of at least 300 years to obtain stable carbon and N budgets under a stationary climate condition, the simulation was started in January 1901 and ended in December 2015. Atmospheric CO_2 concentrations for the historical period were derived from atmospheric observations (e.g., Keeling and Whorf 2009). Monthly temperature, precipitation, humidity, and cloud cover during the simulation period were obtained from the CRU TS3.25 dataset (Harris et al. 2014) for each grid. Soil properties such as bulk density, clay/sand composition, and field capacity were derived from the International Satellite Land Surface Climatology Project dataset (Hall et al. 2006). A map of soil pH was derived from IGBP-DIS (2000). Transitional land use change was derived from the global land use dataset compiled by Hurtt et al. (2011) for climate studies. Croplands were classified on the basis of global crop distribution maps (Monfreda et al. 2008) into the following three categories: rice, generic C_3 crops (except rice, e.g., wheat), and generic C_4 crops (e.g., maize). To facilitate regional implementation of the model, agricultural management practices were classified in a generic way. For example, cultivation of legume species with symbiotic N fixers was not explicitly included. Historical chemical fertilizer and manure inputs into croplands were derived from datasets produced by Nishina et al. (2017) and Potter et al. (2010), respectively. In the fertilizer dataset, country statistics from the Food and Agriculture Organization were down-scaled to 0.5° meshes after filling data gaps. Notably, the dataset differentiated fertilizer N into NH_4^+ and NO_3^- and considered seasonal crop calendars of dominant crops. Historical trends of atmospheric N deposition were derived from Galloway et al. (2004); seasonal variability, based on the results of atmospheric chemistry model simulation studies (Sudo et al. 2002; Dentener et al. 2006), was included. Figure 1 shows the spatial distribution of N inputs to East Asian land areas in 2000 that was assumed in this study.

The baseline simulation experiment (called EX0) considered all driving factors, and the results were analyzed to assess the characteristics of the N budget in this region. In addition, to separate the contribution of individual factors, we conducted the following series of sensitivity simulations:

– EX1: fertilizer input was always fixed at the level in 1901

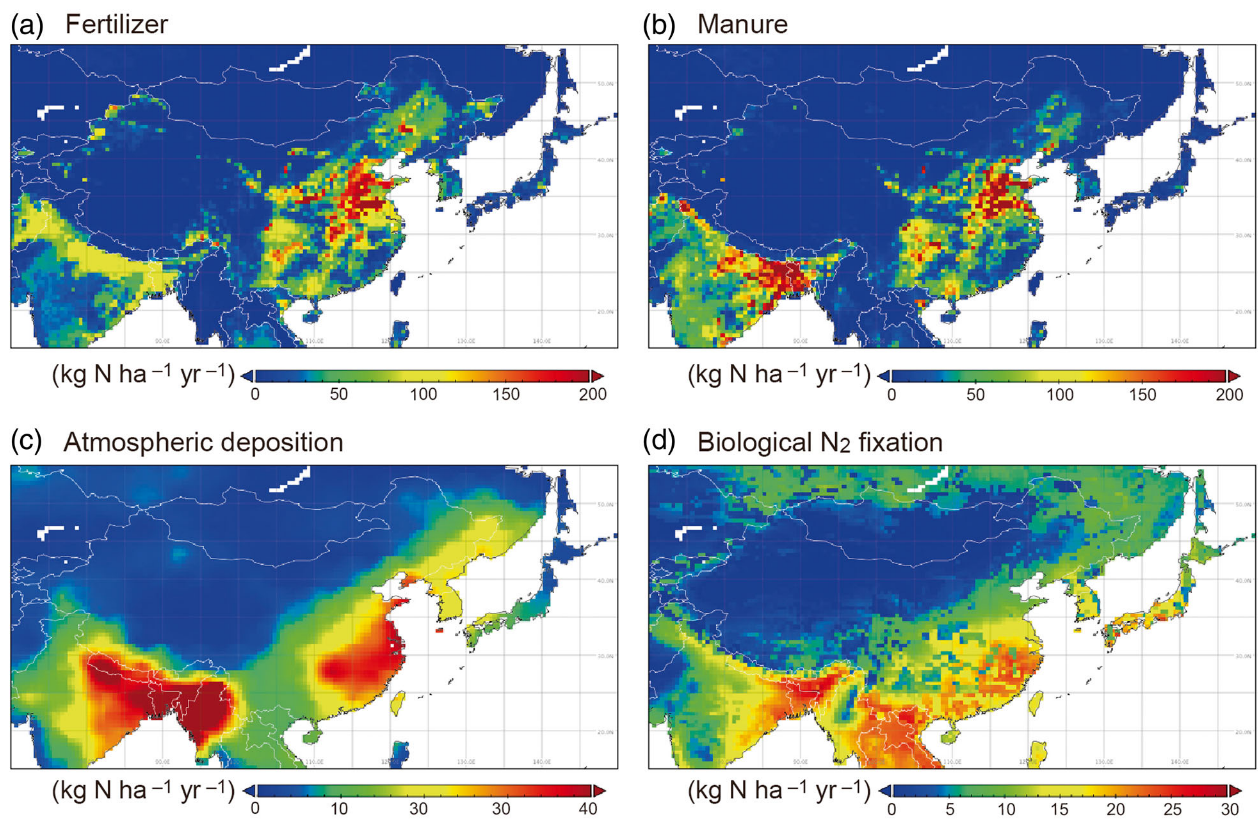

Fig. 1 Spatial distribution of nitrogen inputs in East Asia in 2000–2009. **a** Fertilizer, including both ammonium and nitrate, **b** organic nitrogen in manure, **c** atmospheric deposition, including both ammonium and nitrate and both dry and wet depositions, and **d** biological nitrogen fixation

– EX2: manure input was always fixed at the level in 1901
– EX3: atmospheric N deposition was always fixed at the level in 1901
– EX4: land use was always fixed at the level in 1901
– EX5: fertilizer and manure inputs were always fixed at the levels in 1901
– EX6: land use, fertilizer, and manure inputs were always fixed at the values in 1901
– EX7: manure was ignored (i.e., always zero) throughout the simulation

The EX1–4 simulations separated the effects of individual N inputs; EX5 separated the effect of direct N input to croplands; and EX6 separated the human impacts on land systems. EX3 separated the effect of N inputs from atmospheric deposition and indirectly included anthropogenic inputs. EX6 removed direct impacts of local human manipulations and showed the effects caused by elevated CO_2 concentration and climate change. EX7 was conducted to quantify the impact of ignoring manure inputs, which have been inconsistently considered in terrestrial N model studies (discussed later).

The emission factor, which is defined as the mean atmospheric emission per unit mass of substrate or input to a system, is a useful metric for greenhouse gas studies because it facilitates estimates of emissions and clarification of regional characteristics. Here, the emission factor for N_2O was defined as the N_2O emission per unit N input as chemical fertilizer, manure, and atmospheric deposition (Stehfest and Bouwman 2006) on a yearly basis. The model simulation enabled us to estimate a representative emission factor for East Asia during a time when N inputs were changing. Most results are shown on an N_2O weight basis (e.g., Tg N_2O year^{-1}) to facilitate conversion to a CO_2-equivalent weight basis with a global warming potential of 298 (IPCC 2013; 100-year horizon); the values can be converted to an N weight basis (e.g., Tg N year^{-1}) by multiplying by 28/44.

Results

Regional N_2O emissions

In the EX0 simulation (i.e., all inclusive and so presumably closest to reality), total N_2O emissions from the soil surface of East Asia in the 2000s (2000–2009) were estimated to be 2.03 Tg N_2O year $^{-1}$ (i.e., 1.29 Tg N year^{-1}).

Natural ecosystems and croplands contributed 32.3% and 67.7%, respectively, to the total emissions.

The spatial patterns of simulated N_2O emissions from natural and cropland soils differed markedly (Fig. 2). In most natural ecosystems (Fig. 2a, not weighted by land-use fractions), N_2O emissions occurred at low rates and land use patterns created a heterogeneous distribution of emissions. The relatively high rates in the middle of China were consistent with the high rates of N deposition in that area (Fig. 1c). The N_2O emissions from croplands were relatively high (Fig. 2b, not weighted by land-use fractions), especially in central China, western South Korea, and parts of Japan. The total emission map (Fig. 2c, weighted by land-use fractions) showed that high-N_2O emissions occurred in central to southern parts of China and parts of South Korea and Japan. In contrast, N_2O emissions were quite low in the interior regions covered mainly by grasslands and alpine meadows.

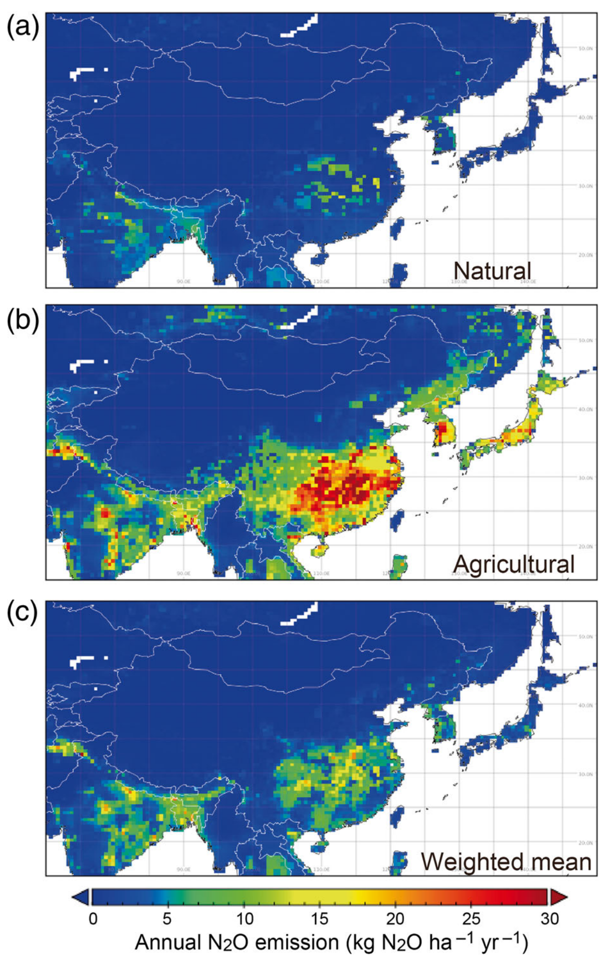

Fig. 2 Emissions of N_2O in East Asia in 2000–2009 in the EX0 simulation with VISIT. **a** Natural ecosystems per unit area, **b** croplands per unit area, and **c** grid-based emissions obtained as a cropland-fraction weighted mean

The simulated N_2O emissions showed clear seasonal patterns (Fig. 3). Regional N_2O emissions were high from June to September, the warm and rainy period of the monsoon climate cycle. Average regional N_2O emission rates varied from 0.03 Tg N_2O month^{-1} in January to 0.38 Tg N_2O month^{-1} in July during the 2000s. The period of high emissions clearly corresponded to the season of agricultural cultivation and fertilizer application.

Historical changes in N_2O emissions

In the EX0 experiment, regional soil N_2O emissions increased from 0.60 Tg N_2O year^{-1} (0.38 Tg N year^{-1}) in 1901 to 2.35 Tg N_2O year $^{-1}$ (1.50 Tg N year^{-1}) in 2015 (Fig. 4a). The increase was especially evident after 1960, i.e., the era of increased agricultural production stimulated by the Green Revolution. Between the 1960s and the 2000s of EX0, N_2O emissions from natural ecosystems increased by 0.01 Tg N_2O year^{-1} (+ 1.8%), while emissions from croplands increased by 1.05 Tg N_2O year $^{-1}$ (+ 331.2%). The dominant source therefore shifted from natural ecosystems (66.9% of the total in the1960s) to croplands (67.7% of the total in the 2000s). Spatially, an increase of N_2O emission was evident in cropland areas (Fig. 5 and Additional file 1: Figure S1). From 1901 to 2016, fertilizer and manure inputs to croplands in this region intensified from 5.8 Tg N year^{-1} to 55.5 Tg N year^{-1}, and atmospheric N deposition also increased from 2.49 Tg N year^{-1} to 4.4 Tg N year^{-1} (especially NH_4^+ deposition). Biological N fixation showed a small increase of 0.2 Tg N year^{-1}.

From the 1960s to the 2000s, temperatures over East Asia increased by 0.88 °C on average, but annual precipitation did not change significantly (from 561 mm year^{-1} to 564 mm year^{-1}). The very small increase of N_2O emissions driven by climate in the sensitivity simulation (EX6; Fig. 4), from 0.90 Tg N_2O year^{-1} in the 1960s to 0.97 Tg N_2O year^{-1} in the 2000s, indicated a weak influence of climate. When the effect of land use change was included (EX5), the result did not vary much from that of EX6 (i.e., from 0.91 Tg N_2O year^{-1} to 0.99 Tg N_2O year^{-1}), because N inputs from fertilizer and manure into croplands were kept at low levels.

In contrast, the sensitivity simulations for fertilizer, manure, and N deposition inputs clearly suggested strong influences from these driving factors. When N deposition was fixed (EX3), regional N_2O emissions in the 2000s were estimated to be 1.87 Tg N_2O year^{-1}, 92.2% of the emission in EX0. When manure inputs were fixed (EX2), regional N_2O emissions were estimated to be 1.69 Tg N_2O year^{-1}, 83.5% of the emissions in EX0. When chemical fertilizer input was fixed (EX1), regional N_2O emissions were estimated to be 1.42 Tg N_2O year^{-1}, 70.3% of the emissions in EX0.

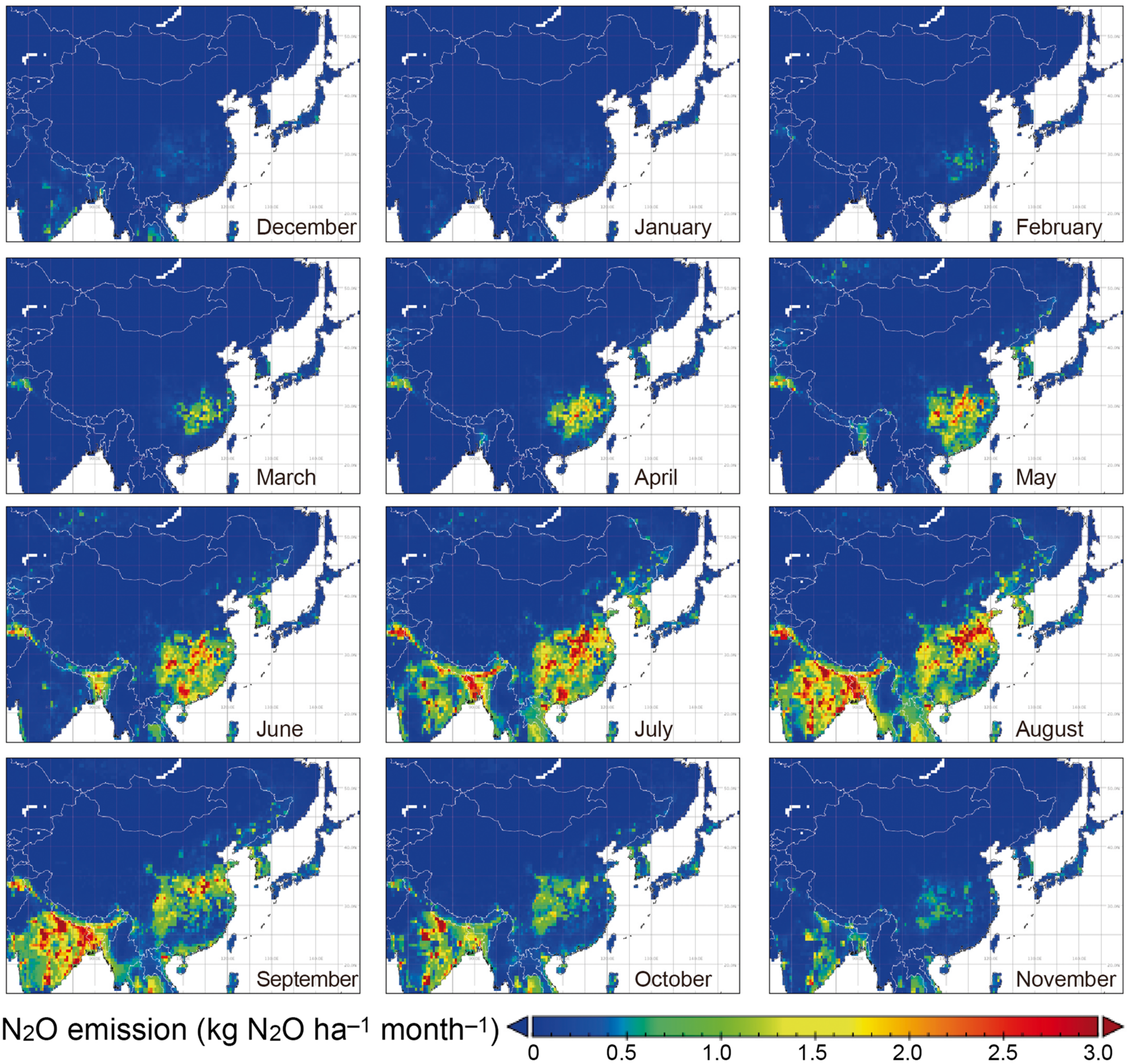

Fig. 3 Monthly maps of soil N_2O emissions in East Asia during 2000–2009 in the EX0 simulation with VISIT: from the top to bottom, northern winter, spring, summer, and autumn

Cumulatively, the increases of the N inputs since 1950 led to about 70 Tg of additional N_2O emissions to the atmosphere (Fig. 4b). Thus, the historical increase of soil N_2O emissions from this region was primarily (more than half) attributable to the increase of fertilizer input to croplands and secondarily to increments of manure use and atmospheric deposition.

Relationships with driving factors

The simulated annual soil N_2O emissions from East Asia were very weakly correlated with annual temperature anomalies (Fig. 6a) but more closely correlated with annual precipitation anomalies (Fig. 6b). Note that linear trends were removed from both the N_2O emissions and climate factors to focus on interannual variability (i.e., anomaly), defined as a year-to-year deviations from the long-term linear trend. This result therefore implies that interannual variability of soil N_2O emissions in this region is limited mainly by soil wetness, which is largely determined by the precipitation anomaly (and partly by evapotranspiration variability). Among the N_2O production processes, denitrification, which occurs under anaerobic conditions, was more closely related to precipitation anomalies (coefficient of determination [R^2] = 0.63) than was nitrification (R^2 = 0.08). This result is consistent with the positive response of total N_2O emissions to

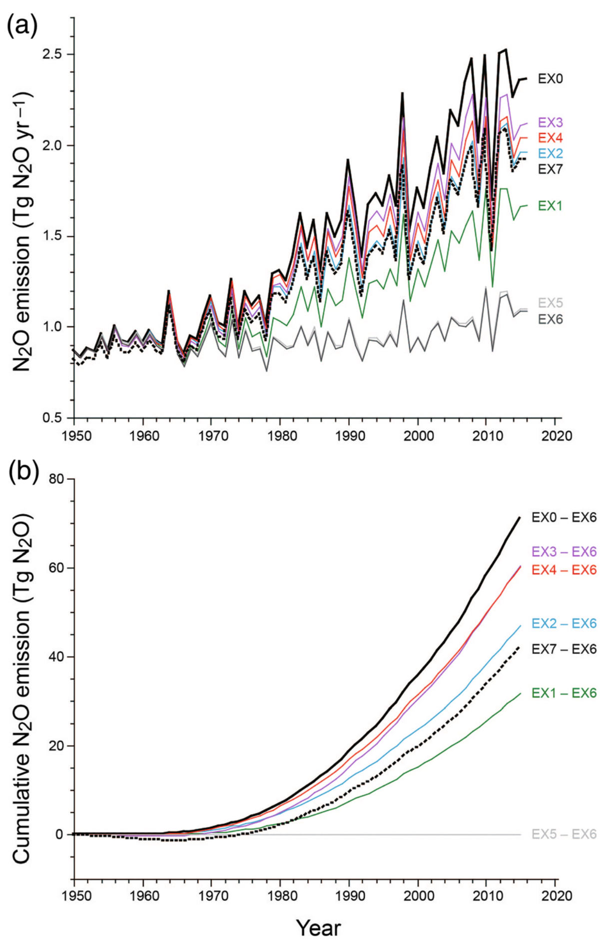

Fig. 4 Interannual variability of total N_2O emissions in East Asia simulated by the VISIT model. **a** Annual emissions in the EX0 to EX7 simulations: EX0, control; EX1, fixed fertilizer; EX2, fixed manure input; EX3, fixed atmospheric deposition; EX4, fixed land-use; EX5, fixed fertilizer and manure; EX6, fixed fertilizer, manure, and land-use; and EX7, no manure. **b** Cumulative differences from the EX6 result

precipitation anomalies. During the twentieth century, the highest annual N_2O emission rate (2.28 Tg N_2O year^{-1} in EX0) occurred in 1998, a very strong El Niño year with the warmest temperature and highest precipitation of the century.

The simulated regional N_2O emissions were strongly correlated with N inputs (Fig. 7 for EX0). The slope of a linear regression of N_2O emission versus N inputs is a metric of N_2O emission factor: in this study, 0.0216 Tg N_2O Tg^{-1} N or 0.0138 Tg N-N_2O Tg^{-1} N (i.e., 1.38%). The estimated emission factor for cropland, calculated on the basis of direct N inputs to cropland, was almost the same (1.38%) as the regional mean, which includes both natural and agricultural ecosystems. The relationship between N inputs and N_2O emissions during the period 1960–2015 was almost linear ($R^2 = 0.88$), and there were corresponding increases of residual N outflows by NO_3^- leaching and NH_3 volatilization with increasing N inputs (data not shown). The simulated N_2O emission factor was not uniform throughout the study region (Fig. 8b). The emission factor ranged from low values in interior dry regions and northern parts of China to high values in humid regions in southern parts of China, the Korean Peninsula, and Japan. The weak relationship between the distribution of emission factors and land use for agriculture (Fig. 8a, b) implied that emission factors were primarily affected by climatic and intrinsic soil conditions.

Discussion

The model simulation allowed us to evaluate soil N_2O emissions and their spatial and temporal variability in East Asia; the methodology we used may be applicable to other regions. Compared with global N_2O emissions (18.3 Tg N_2O year^{-1} in the 2000s; unpublished result of a global simulation), the East Asia region accounts for about 11% of the global total (5.9% of natural ecosystems and 18.9% of croplands). Note that the global result is comparable with previous studies (Syakila and Kroeze 2010; Wells et al. 2018): in 2002, 6–7 Tg N year^{-1} (9.4–11 Tg N_2O year^{-1}) from soils and 4.9 Tg N year^{-1} (7.7 Tg N_2O year^{-1}) from agriculture. Using the global warming potential value of 298, we estimated the N_2O emission of 2.03 Tg N_2O year^{-1} to be equivalent to 605 Tg CO_2-eq. year^{-1} (on a carbon basis, 165 Tg C year^{-1}). Soil N_2O emissions can be a substantial fraction of the total greenhouse-gas budget for this region. For example, an integrated synthesis of regional C budgets by Piao et al. (2012) conducted an integrated synthesis of regional C budgets and concluded that land ecosystems in East Asia are a net CO_2 sink of 224–413 Tg C year^{-1} at the present time. The range of numbers reflects differences in estimation methods such as bottom-up summation and top-down inversion. The estimated soil N_2O emission is therefore likely to offset 40–74% of the mitigation effect of CO_2 sequestration by terrestrial ecosystems in this region. Reducing soil N_2O emissions from the region should therefore contribute greatly not only to mitigation of global warming but also to amelioration of environmental pollution (e.g., eutrophication and acidification) by reducing agricultural N use. For example, the distribution of emission factors estimated in this study (Fig. 8b) may enable optimization of N fertilizer and manure use, which are related to both food production and environmental loads. Although not addressed in this study, the simulation of N leaching and of NH_3 and NO emissions by the model leads to reductions of water use and air pollution. In addition, changes of ecosystem N stocks are linked with the degree of N limitation, which can alter ecosystem structure (including biodiversity), functions, and services (Aber et al. 1995). Such co-benefits

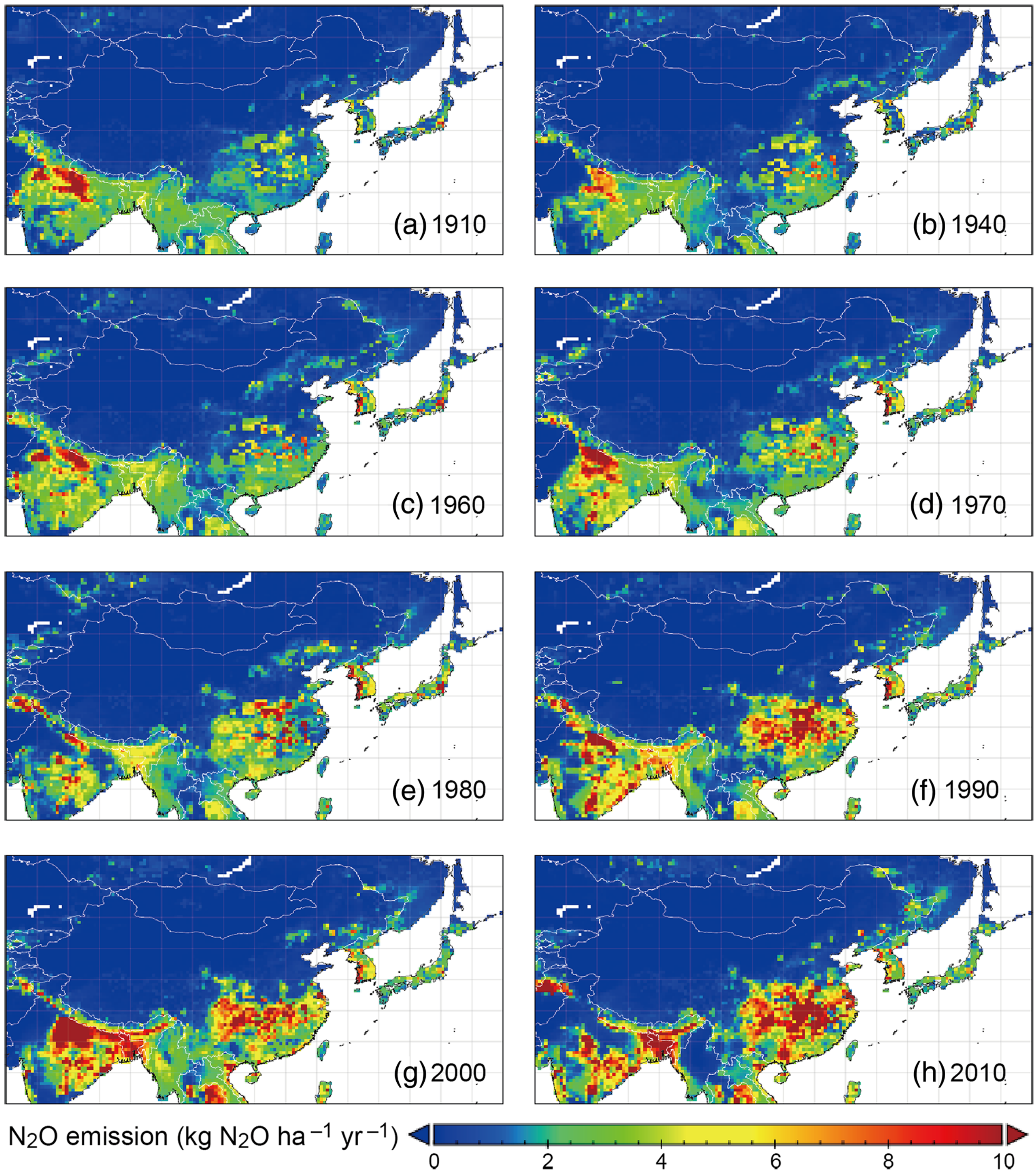

Fig. 5 Historical changes of N_2O emissions in the EX0 simulation with VISIT: **a** 1910, **b** 1940, **c** 1960, **d** 1970, **e** 1980, **f** 1990, **g** 2000, and **h** 2010

and trade-offs of N management are among the focal themes of the International Nitrogen Management System (INMS, http://www.inms.international/). The INMS aims at synthesizing the achievements and challenges on N research, and our research may therefore make some contribute to activities such as regional demonstration.

The large soil N_2O emissions from this region are attributable to extensive use of land for agriculture and high N inputs from chemical fertilizer, organic manure, and atmospheric deposition. Note that other anthropogenic emissions of N_2O from industrial and urban sectors (not included in this study) are likely substantial in this region (e.g., Ohara

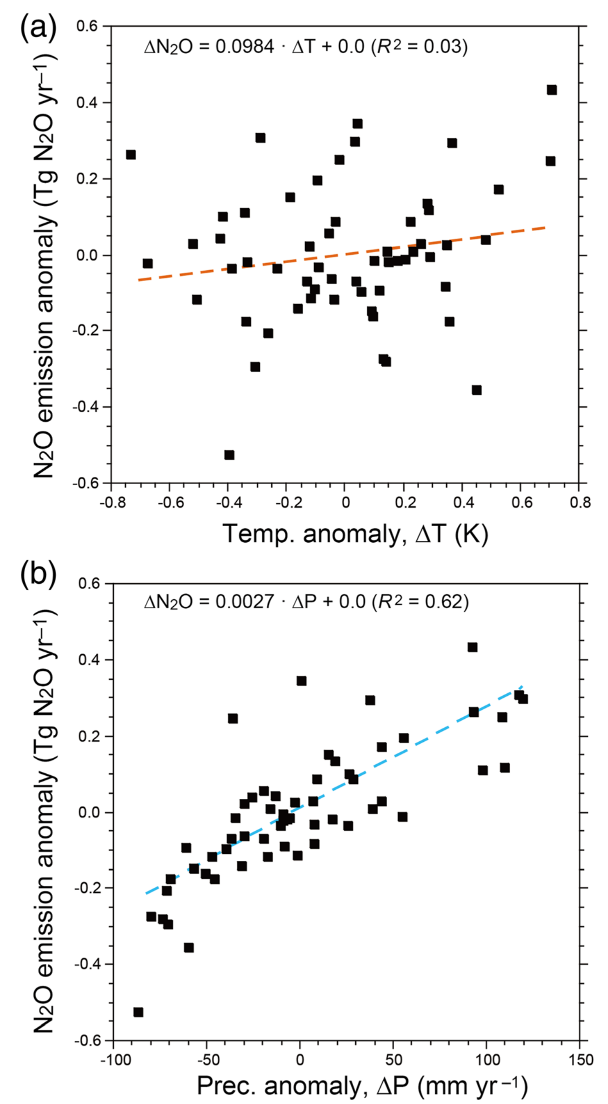

Fig. 6 Relationships between regional N_2O emissions in East Asia and anomalies of meteorological factors: **a** temperature and **b** precipitation. Annual values of the EX0 simulation results (i.e., standard run) by the VISIT model were used after removal of long-term linear trends

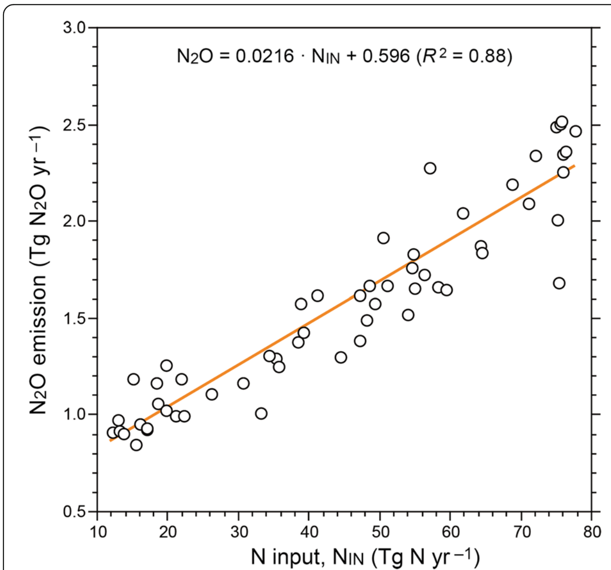

Fig. 7 Relationship between nitrogen inputs and N_2O emissions in East Asia in the EX0-simulation results of the VISIT model. Each circle represents an annual value. Nitrogen inputs include fertilizer, manure, atmospheric deposition, and biological fixation

et al. 2007). Emissions of N_2O from the soil surface have apparently increased during the last few decades in East Asia, mainly because of human interventions in the N cycle, as demonstrated by the sensitivity simulations. These simulation results, in conjunction with data for other factors such as food production and environmental pollution, should inform identification of strategies for mitigation of global warming and N management in this region (Shibata et al. 2015). The mean emission factor of 1.38% for N_2O in East Asia estimated in this study is slightly higher than the default value of the IPCC guideline, i.e., 1% for mineral fertilizer and organic amendments to cropland, but within the range of uncertainty (0.3–3.0%) (IPCC 2006). Several meta-analysis studies have indicated that emission factors derived from observations of soil N_2O emissions vary widely, from 0.03 to 10% (e.g., Kim et al. 2013; Charles et al. 2017). The mean value estimated in this study is within this range, but there was much variability of both observations and model estimates (e.g., inter-grid standard deviation, 2.09%). The simulated N_2O emission factor (Fig. 8b) varied between low values (~ 1%) in northern area and high values (> 1%) in the southern area, irrespective of agricultural contributions (Fig. 8a). Remarkably, higher emission factors were found in the Tibetan Plateau, where N_2O emissions were low because of low N inputs (Fig. 1) and cool temperatures. In contrast, emission factors were low in north-central China, which has been intensively cultivated (Additional file 1: Figure S1) and is receiving high N inputs (Fig. 1). Such regional heterogeneity in the emission factor implies a need for diverse regulatory mechanisms. In the model used in this study, N_2O emission rates from both nitrification and denitrification were sensitive to soil moisture conditions, which are affected by precipitation and soil water-holding capacity. In this regard, the model estimation of N_2O emissions from paddy fields could be improved by considering water management and associated characteristics of the N cycle specific to the land cover type. For example, an empirical study by Yan et al. (2003) obtained an N_2O emission factor of 0.93% (versus 1.38% in this study). As noted by Lesschen et al. (2011), variability of N_2O emission factors influences regional greenhouse gas accounting.

The model used in this study is one of the models used in the terrestrial N model intercomparison and global N_2O synthesis project (Tian et al. 2018). Comparison of soil N_2O emissions simulated by different biogeochemical

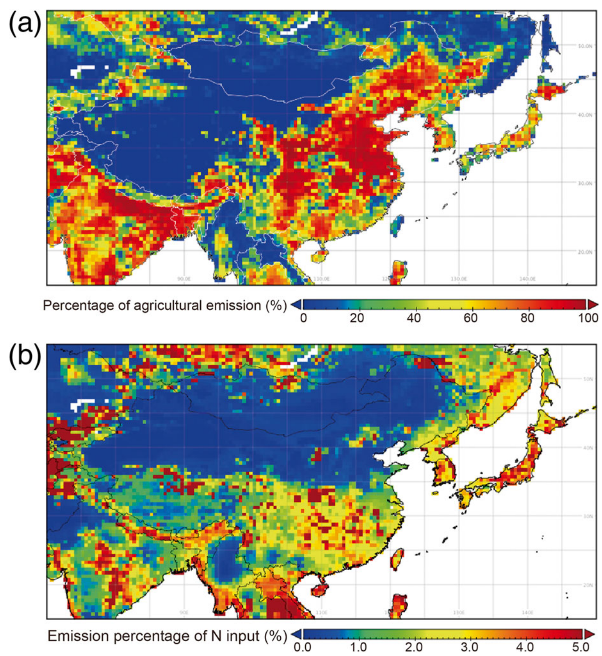

Fig. 8 Spatial characteristics of simulated soil N_2O emissions in the EX0-simulation results for 2000–2009 by VISIT. **a** Percentage contributions of agricultural emissions, and **b** emission factors (percentage of N_2O emissions per unit of nitrogen input)

models has shown that the VISIT model provides broad-scale estimates comparable to those of other contemporary models. However, it is difficult to relate the regional N_2O emissions simulated here with atmospheric measurements for validation because of the long atmospheric residence time of N_2O, urban emissions, and exchanges of N_2O with the stratosphere (Ishijima et al. 2010). The trends of the observed atmospheric N_2O concentrations in East Asia increased linearly with time, but seasonal patterns were unclear (Additional file 1: Figure S2). Comparison with the results of atmospheric inversion studies would provide further insights. For example, Thompson et al. (2014) have reported surface N_2O fluxes inferred using atmospheric inversion models based on chemistry transport models. Their estimates of N_2O emissions in East Asia have indicated a spatial distribution comparable to that simulated in the present study, e.g., high emissions in parts of southern China and low emissions in interior drylands and northern areas. The peak emission intensity in parts of southern China, around 8 kg N ha^{-1} yr^{-1}, is also comparable between the two studies. Recently, Wells et al. (2018) made a similar analysis using a different atmospheric model and data-assimilation techniques and obtained comparable results for total (i.e., including soil and other sources) N_2O emissions. Saikawa et al. (2014) have presented another model-based estimate of global and regional N_2O emissions derived from atmospheric observational data. In their results, Southern Asia (i.e., East, South, and Southeast Asia) was estimated to release N_2O at a rate of 3–5 Tg N $year^{-1}$ in 1995–2008; this estimate is consistent with our model estimate of 3.3 Tg N $year^{-1}$ for the corresponding area.

It is difficult, however, to compare simulated regional N_2O fluxes directly with field observations based on chamber methods, because of the large difference in spatial scales: i.e., square meter for chamber measurements vs 10^4–10^6 km^2 of model simulations. Recently, several numerical techniques have been developed to extrapolate (i.e., scale up) field measurements to the global scale (e.g., Jung et al. 2011; Tramontana et al. 2016). A more realistic strategy for validating model estimates would be a comparison with these up-scaled flux estimates. Hashimoto (2012) has provided a global dataset of soil greenhouse gas emissions extrapolated from field data using a statistical model based on extensive field data (e.g., Morishita et al. 2007). Spatial patterns of the estimated N_2O emissions (Additional file 1: Figure S3a, a revised estimate of Hashimoto 2012) is roughly comparable with those of the present study. Both studies capture higher emissions from parts of southern China and from hot spots in central China. In general, however, the intensities of the emissions estimated by Hashimoto (2012) in the study region are lower (Additional file 1: Figure S3b: − 25% on average), probably because his study used data obtained mainly from temperate forests (Morishita et al. 2007). As noted by Hashimoto (2012), statistical methods might capture background emissions from croplands. The VISIT-estimated emissions in natural ecosystems are closer to the regional estimates of Hashimoto (2012). Further validations using multiple observational data, similar to the validation conducted in this study, will clearly be needed to evaluate and reduce uncertainties in model-based assessments.

Several limitations of this study remain. First, this study used a single model (i.e., VISIT) and is therefore susceptible to model-specific bias. Such a study based on a small number of models is effective for conducting many simulations to separate the influence of driving factors, but a composite of multiple model results is likely to provide more robust outcomes. A recent model intercomparison project (Tian et al. 2018) has provided us with an opportunity to assess inter-model variability and to obtain less biased estimates. Moreover, systematic analyses of the formulations and parameters related to pivotal processes are required to scrutinize inter-model differences. Second, our incomplete understanding of N biogeochemical processes leads to insufficient formulations in N models. The complexity of biochemical processes mediated by microbes such as mineralization, nitrification, and denitrification (Butterbach-Bahl et al. 2013; Isobe and Ohte 2014) makes it difficult to formulate mechanisms accurately. Physical processes such as freezing and thawing and their impacts on the N cycle (Yanai et al. 2011; Hosokawa et al. 2017) are sufficiently complicated that most models do not adequately characterize them. Also, human manipulations by agricultural practices such as reduced tillage (Koga et al. 2004), enhanced-efficiency fertilizer (Akiyama et al. 2010), and inoculation of soils with microbes that process N more efficiently (Itakura et al. 2012) can affect N_2O emissions but have not been adequately incorporated into biogeochemical models. Third, there are still uncertainties in the forcing input data. In this study, we used popular datasets to drive the model. Recent studies have been providing updated datasets for fertilizer (e.g., Lu and Tian 2017), manure (e.g., Wolf et al. 2017; Zhang et al. 2017), and land use (Lawrence et al. 2016). Use of these recent and forthcoming datasets in future studies is expected to considerably improve the accuracy of the simulated results.

Conclusions

Our estimated N_2O emissions from the soil surface of East Asia of 2.03 Tg N_2O year^{-1} in the 2000s based on a process-based model showed that the region was a significant source of N_2O emissions to the atmosphere. The estimated mean emission factor, 1.38%, was robust through time but showed high-spatial heterogeneity. Compared to previous studies, this study used up-to-date forcing data, simulated longer periods, and provided detailed emission maps. Despite the uncertainty of the model estimates, this study demonstrated the effectiveness of model-based evaluations of regional N_2O budgets, which are difficult to obtain from direct methods by tower and satellite measurements. The capability of male long-term analyses is another advantage of the model-based approach. Our historical simulation results imply that there has been a significant increase of regional soil N_2O emissions, by a factor of three or more, mainly because of human activities. Our results also suggest opportunities for mitigation of climatic impacts through application of appropriate emission reduction measures such as optimization of fertilizer use and agricultural practices (e.g., tillage and water management). Because of the importance of N_2O as a greenhouse gas, contemporary climate policies such as the Paris Accord of the United Nations Framework Convention of Climate Change should pay more attention to N_2O emissions. Also, management of N_2O emissions from soil should provide other benefits in terms of environmental quality through reduction of excessive N inputs to water and air and losses from soil. Further studies are nevertheless required to elucidate the biogeochemical processes involved in N dynamics, the interactions between the N cycle and the water and C cycles, and the best way to enlarge the spatial scale of models.

Additional file

Additional file 1: Figure S1. Cropland fraction in East Asia derived from Hurtt et al. (2011). **Figure S2.** Comparison between the simulated monthly N_2O emissions and observed atmospheric N_2O concentrations in East Asia. The observational data were obtained from the World Data Center of Greenhouse Gases (https://gaw.kishou.go.jp/). **Figure S3.** Comparison of the VISIT estimations (EX0) with the estimates of Hashimoto (2012, updated). (a) Annual N_2O emissions and (b) grid-based scatter diagram. Orange line shows linear regression (y [N_2O emissions by VISIT] = 1.25 x [N_2O emissions by Hashimoto (2012)] + 0.0596, in kg N_2O ha^{-1} yr.$^{-1}$; $R_2 = 0.44$). (DOCX 1792 kb)

Abbreviations

CRU: Climate Research Unit; IGBP-DIS: International Geosphere-Biosphere Program Data and Information System; VISIT: Vegetation Integrative SImulator for Trace gases

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Availability of data and materials

Please contact author for data requests.

Authors' contributions

AI designed the research, conducted simulations and analyses, and drafted the manuscript. MI contributed to model development. KN, KI, and SH examined results and contributed to the manuscript. All authors read and approved the final manuscript.

Competing interests

The authors declare that they have no competing interests.

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