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Article type : Primary Research Articles

Global soil nitrous oxide emissions since the pre-industrial era estimated by an ensemble of Terrestrial Biosphere Models: Magnitude, attribution and uncertainty

Running Head: Model ensemble of global soil N_2O emissions

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Key words: Global nitrogen cycle, Greenhouse gas emission, Nitrous oxide, Process-based modeling, Soil N_2O emission

Paper Type: Primary Research Articles

Global Change Biology (revised on 11/04/2018)

Abstract: Our understanding and quantification of global soil nitrous oxide (N_2O) emissions and the underlying processes remain largely uncertain. Here we assessed the effects of multiple anthropogenic and natural factors, including nitrogen fertilizer (N) application, atmospheric N deposition, manure N application, land cover change, climate change and rising atmospheric CO_2 concentration, on global soil N_2O emissions for the period 1861–2016 using a standard simulation protocol with seven process-based terrestrial biosphere models. Results suggest global soil N_2O emissions have increased from 6.3 ± 1.1 Tg N_2O-N yr^{-1} in the pre-industrial period (the 1860s) to 10.0 ± 2.0 Tg N_2O-N yr^{-1} in the recent decade (2007-2016). Cropland soil emissions increased from 0.3 Tg N_2O-N yr^{-1} to 3.3 Tg N_2O-N yr^{-1} over the same period, accounting for 82% of the total increase. Regionally, China, South Asia and Southeast Asia underwent rapid increases in cropland N_2O emissions since the 1970s. However, US cropland N_2O emissions had been relatively flat in magnitude since the 1980s, and EU cropland N_2O emissions appear to have decreased by 14%. Soil N_2O

emissions from predominantly natural ecosystems accounted for 67% of the global soil emissions in the recent decade but showed only a relatively small increase of 0.7 ± 0.5 Tg N_2O-N yr^{-1} (11%) since the 1860s. In the recent decade, N fertilizer application, N deposition, manure N application and climate change contributed 54%, 26%, 15% and 24%, respectively, to the total increase. Rising atmospheric CO_2 concentration reduced soil N_2O emissions by 10% through the enhanced plant N uptake, while land cover change played a minor role. Our estimation here does not account for indirect emissions from soils and the directed emissions from excreta of grazing livestock. To address uncertainties in estimating regional and global soil N_2O emissions, this study recommends several critical strategies for improving the process-based simulations.

Introduction

Nitrous oxide (N_2O) is a long-lived greenhouse gas (GHG) with an atmospheric lifetime of ~116 years (Prather *et al.*, 2015), which traps heat in the earth system (Butterbach-Bahl *et al.*, 2013, Ciais *et al.*, 2014, Tian *et al.*, 2016) and also contributes to ozone depletion in the stratosphere (Ravishankara *et al.*, 2009). The concentration of atmospheric N_2O has increased from 270 ppb in the pre-industrial period to 330 ppb in recent years as a result of anthropogenic industrial and agricultural activities. The fastest increases in the atmospheric N_2O concentration were seen in recent decades with an average of 0.73 ppb yr^{-1} (Ciais *et al.*, 2014).

Since the onset of industrialization, the global nitrogen (N) cycle has been continuously disturbed by human activities, especially after the invention of industrial N_2 fixation (Gruber & Galloway, 2008). With human population growth, the demand for more food production requires a substantial addition of reactive N (chemical fertilizer and manure) into the

cropland soils including land areas used for row crop cultivation. Over recent decades, the enhanced fertilizer use in conjunction with the expansion of legume crops lead to higher soil N_2O emissions (Ciais *et al.*, 2014, Montzka *et al.*, 2011, Zaehle *et al.*, 2011). Meanwhile, soils from predominantly natural systems, especially the tropical forests, which are N rich and contain many N-fixing tree species, contributed a large portion to the global N_2O emissions (Ciais *et al.*, 2014, van Lent *et al.*, 2015). In recent decades, N_2O emissions from terrestrial soils are the primary source for atmospheric N_2O (Butterbach-Bahl *et al.*, 2013, Davidson & Kanter, 2014, Smith, 1997), accounting for ~60% of all global N_2O emission sources (Syakila & Kroeze, 2011, Werner *et al.*, 2007). A comprehensive assessment of soil N_2O emissions, therefore, is of particular importance for understanding climate-ecosystem interaction and future climate change.

Investigation of N_2O emissions from soils has been a key research topic for decades (e.g. Bouwman *et al.*, 1993, Eichner, 1990, Gruber & Galloway, 2008, Syakila & Kroeze, 2011, Xu *et al.*, 2017). Field measurements have been extensively implemented across various biome types and climate zones (e.g. Bouwman *et al.*, 2002, Liu & Greaver, 2009, Smith & Dobbie, 2001). However, up-scaling soil N_2O emissions from sites to regional and global scales is still a challenge, mainly because of the variable characteristics and complicated mechanisms of N_2O emissions controlled by multiple biotic and abiotic factors (Butterbach-Bahl & Dannenmann, 2011). Current methods in estimating large-scale N_2O emissions can be separated into two broad categories: bottom-up and top-down approaches. Bottom-up approaches estimate N_2O emissions according to inventories, statistical extrapolation of field measurements, and/or Terrestrial Biosphere Models (TBMs), while top-down approaches estimate emissions by integrating atmospheric measurements and atmospheric inversion models (Davidson & Kanter, 2014, Saikawa *et al.*, 2014, Thompson *et al.*, 2014, Tian *et al.*, 2016). Results of both approaches have considerable uncertainties, and estimates of soil N_2O

emissions show significant divergences across studies. For example, in the Intergovernmental Panel on Climate Change fifth assessment (IPCC AR5) (Ciais *et al.*, 2014), global natural and anthropogenic N_2O emissions from land and ocean estimated by bottom-up approaches ranged between 8.1 and 30.7 Tg N_2O-N yr^{-1}.

The use of Emission Factors (EF) is one common bottom-up approach to quantify N_2O emissions from synthetic N fertilizer. The IPCC Tier 1 Protocol 2006 (De Klein *et al.*, 2006) recommended assuming by default that 1% of synthetic N fertilizer use in land ecosystems is directly emitted to the atmosphere in the form of N_2O gas (i.e., EF equals 1%). Based on the EF in the IPCC Tier 1 Protocol 2006, FAOSTAT (FAOSTAT, 2016) and the Emissions Database for Global Atmospheric Research (EDGAR) (Janssens-Maenhout *et al.*, 2017) provided sectoral estimates of N_2O emissions. However, the assumption of constant EF has been questioned because of an inability to depict spatial and temporal variations of N_2O emissions and to reflect the impacts of changing environments over time. For example, Shcherbak *et al.* (2014) found a faster growing nonlinear response of N_2O emissions to N inputs; additionally, soil N_2O emissions are affected by soil moisture, temperature conditions, and pH value (Del Grosso & Parton, 2012, Schindlbacher *et al.*, 2004, Wang *et al.*, 2018), as well as freeze/thaw events and livestock management (Wagner-Riddle *et al.*, 2017, Wolf *et al.*, 2010).

Model simulation is another important bottom-up approach to quantify soil N_2O emissions at regional, sectorial, and global scales (e.g. Del Grosso *et al.*, 2000, Li *et al.*, 2000, Parton *et al.*, 2001, Tian *et al.*, 2015, Tian *et al.*, 2018, Xu-Ri *et al.*, 2012). One notable advantage of the modeling approach is that it tends to describe the overall N cycle within the land systems and can integrate various driving factors (such as fertilizer application, atmospheric N deposition, and climate change) controlling soil N_2O production and emissions. Due to differences in model structure, parameterization schemes, and input data, simulated soil N_2O

emissions diverged considerably in previous estimates. For example, Xu-Ri *et al.* (2012) reported that global N_2O emissions from natural soil were 8.3 ~ 10.3 Tg N_2O-N yr^{-1} over the 20^{th} century, while Huang and Gerber (2015) simulated lower natural soil N_2O emissions of 6.7 Tg N_2O-N yr^{-1} in recent decades.

Nitrification and denitrification are two key processes for soil N_2O production, which are regulated by soil temperature, water content, oxygen levels, pH value, and substrate (NO_3^- and NH_4^+) availability (Bouwman *et al.*, 2002, Butterbach-Bahl *et al.*, 2013, Davidson *et al.*, 2000). These two processes have been parameterized in models by using different mathematical algorithms (e.g. Firestone & Davidson, 1989, Li *et al.*, 2000, Parton *et al.*, 2001, Potter *et al.*, 1996, Xu-Ri & Prentice, 2008). Besides, other N-related processes affecting mineral N concentration and N_2O production in soils (such as biological N fixation, plant N uptake, soil N mineralization and immobilization, and N leaching) are also parameterized differently in TBMs (Tian *et al.*, 2018). Pasture and rangeland (grassland used for grazing animals) emit sizeable N_2O gas from livestock excreta deposition, manure and mineral fertilizer application (Davidson, 2009, Steinfeld *et al.*, 2006). Although some models such as DNDC (Giltrap & Ausseil, 2016, Li *et al.*, 1992, Saggar *et al.*, 2004), the daily version of the Century ecosystem model (DayCent) (Abdalla *et al.*, 2010, Del Grosso *et al.*, 2005, Parton *et al.*, 1998), and the Soil and Water Assessment Tool (SWAT) (Arnold *et al.*, 2012, Shrestha *et al.*, 2018) simulated N_2O emission from pasture soils, most global-level model simulations have not considered the impacts of pasture management practices and livestock excreta deposition, which leads to the underestimation of soil N_2O emissions from world's grasslands (Dangal *et al.*, submitted). The differences in model input datasets (such as climate data, land use and land cover, and N deposition) can be another important source of uncertainty in model simulations. Thus, consistent and accurate input datasets are

particularly necessary to narrow the uncertainty range of the simulated soil N_2O emissions across the process-based models.

The global N_2O Model Inter-Comparison Project (NMIP) has been initiated under the umbrella of the Global Carbon Project (GCP) and the International Nitrogen Initiative (INI) and aims to quantify long-term N_2O emissions from global soils and determine the contributions of multiple environmental factors to emissions (Tian *et al.*, 2018). In the NMIP, participating TBMs simulated global soil N_2O emissions during the period of 1861 – 2016, following the same simulation protocol and driven by consistent input datasets, including direct human input of reactive N, and climate and atmospheric composition fields. The specific objectives of our study are to: (1) quantify the magnitude and spatiotemporal patterns of global soil N_2O emissions from the pre-industrial period to the contemporary period; (2) identify the critical regions making significant contributions to increased soil N_2O emissions; (3) attribute the changed N_2O emissions to natural and anthropogenic factors; and (4) discuss uncertainties of estimated emissions and provide insights for model improvement and future research directions.

Materials and Methods

The global N_2O Model Intercomparison Project (NMIP)

Currently, ten process-based Terrestrial Biosphere Models (TBMs) participate in NMIP, all of which explicitly consider terrestrial carbon (C), N, and water cycling processes, and simulate soil N_2O emissions (Tian *et al.*, 2018). Driven by consistent input datasets (i.e., climate, atmospheric CO_2 concentration, land cover change, atmospheric N deposition, mineral N fertilization, and manure N application), each model team implemented seven simulation experiments (SE0 – SE6, Table 1) at the spatial resolution of 0.5° globally

covering the period of 1861 – 2016. In SE0, all driving factors were kept constant at the levels in 1860 over the entire simulation period, while in SE1 all factors changed over time. Each modeling group was requested to simulate and submit terrestrial C- and N-related variables at grid- and biome-scales for model ensemble analysis.

NMIP model input datasets were collected from various sources. Climatic conditions were acquired from the CRU-NCEP v8 climate dataset (https://vesg.ipsl.upmc.fr); atmospheric CO_2 concentration was from the NOAA GLOBALVIEW-CO2 dataset (https://www.esrl.noaa.gov); gridded cropland area was from the HYDE 3.2 dataset (ftp://ftp.pbl.nl/hyde/); N deposition was obtained from the IGAC/SPARC Chemistry-Climate Model Initiative (CCMI); and N fertilization and manure N application datasets were developed specifically for the NMIP project (Lu & Tian, 2017, Zhang *et al.*, 2017). NMIP did not provide consistent crop type and rotation datasets because (1) the participating models have different crop classification schemes, and some models consider crop rotation while others do not; and (2) global-level cropping system datasets covering the entire simulation period (1860-2016) are not available. NMIP models, therefore, have the flexibility to use their default strategies to represent crop type and rotation. In this study, NMIP simulation protocol considered external manure inputs to cropland but did not include manure deposition and application in pasture and manure management as most NMIP models do not simulate N_2O emissions from animal excreta in grassland, which may lead to uncertainties (see Discussion section). The detailed NMIP simulation protocol, including model spin-up strategies, benchmarking, output variables and quality control approaches, is given in Tian *et al.* (2018).

In this study, we set four criteria to screen the participating models: (1) SE0 and SE2-SE6 were implemented; (2) grid-level N_2O emissions and N_2O emissions from cropland and natural soils were provided; (3) no significant trend (increasing or decreasing) of the

simulated global N_2O emissions in SE0 to ensure no drift in model simulation; and (4) the simulated N_2O emissions should be responsive to each of the environmental drivers. Finally, seven of the ten participating models were selected to estimate N_2O emissions from both cropland and natural soils and to quantify the relative contributions of each driving factor. These seven models are: (1) the Dynamic Land Ecosystem Model (DLEM) (Tian *et al.*, 2015, Xu *et al.*, 2017), (2) Lund-Potsdam-Jena - General Ecosystem Simulator (LPJ-GUESS) (Olin *et al.*, 2015, Xu-Ri & Prentice, 2008), (3) Land Processes and eXchanges model - Bern (LPX-Bern v1.4) (Lienert & Joos, 2018, Stocker *et al.*, 2013, Xu-Ri & Prentice, 2008), (4) O-CN (Zaehle *et al.*, 2011), (5) Organising Carbon and Hydrology In Dynamic Ecosystems (ORCHIDEE), (6) Organising Carbon and Hydrology In Dynamic Ecosystems-Carbon Nitrogen Phosphorus (ORCHIDEE-CNP) (Goll *et al.*, 2017), and (7) Vegetation Integrated SImulator for Trace gases (VISIT) (Inatomi *et al.*, 2010, Ito & Inatomi, 2012) (See more model information in Table S1). Five models (DLEM, LPJ-GUESS, ORCHIDEE, ORCHIDEE-CNP, and VISIT) considered the effects of manure use in cropland and ran all the seven simulation experiments (S0-S6), while the other two models (LPX-Bern and O-CN) did not include manure effects and ran six model experiments (all except SE1). Note that N_2O emissions from ground water and rivers are not included in these models.

Assessment of soil N_2O emissions and attribution analysis

The SE1 includes all driving factors for models with manure addition, and the SE2 is the experiment including all the driving factors for models except manure N. We, therefore, used the SE1 results of five models with manure considered and SE2 results of two models without manure considered as the "best estimate" of soil N_2O emissions. The "best estimate" of the seven models was further averaged to estimate the pre-industrial and contemporary

patterns of soil N_2O emissions. In the SE0 simulation, driving forces were kept constant at the level in 1860 over the entire simulation period (1861-2016). SE0 is the "control run" with no trend in any input data and shows no increasing or decreasing trend in the simulated soil N_2O emissions if there is no long-term model drift. The simulations in SE0 by all seven models were used to define the pre-industrial level of soil N_2O emissions. By comparing results from different model scenarios (Table 1), it is possible to attribute the changed spatiotemporal variations of soil N_2O emissions to the variations of six natural and anthropogenic factors, namely, climate (CLIM, including precipitation, humidity, temperature and photosynthetic active radiation changes), atmospheric CO_2 concentration (CO_2), land cover change (LCC), atmospheric N deposition (NDEP), mineral N fertilizer use (NFER), and manure N use in cropland (MANN). Note that the MANN effect was calculated based on the results of five models, while the effects of other driving factors (NFER, NDEP, LCC, CO_2, and CLIM) were estimated based on the results of seven models. The estimated contributions were relative to the levels of driving factors in pre-industrial period. In order to understand soil N_2O emissions dynamics caused by crop cultivation, we further separated the global and regional N_2O emissions into those derived from cropland soils and those from soils of other land ecosystems. Except for cropland, the current NMIP simulations do not include management practices (such as grazing and forest logging) for other managed ecosystems such as pasture, planted forests and urban. All soils in other land ecosystems except cropland were treated as "natural soils" while model simulations were implemented in this study.

Regional boundaries

While analysis in each model was performed at a 0.5° grid, display of results also took advantage of specific global regions. A new map was designed in this study to divide the global ice-free land area (Greenland and Antarctic excluded) into 18 regions (see Figure S1) for reporting soil N_2O emissions at the regional level. The 18 regions are USA (9.4 Million km^2), Canada (CAN, 9.8 Million km^2), Central America (CAM, 2.7 Million km^2), Northern South America (NSA, 2.7 Million km^2), Brazil (BRA, 8.5 Million km^2), Southwest South America (SSA, 6.4 Million km^2), Europe (EU, 5.7 Million km^2), Northern Africa (NAF, 15.2 Million km^2), Equatorial Africa (EQAF, 8.2 Million km^2), Southern Africa (SAF, 6.6 Million km^2), Russia (RUS, 16.8 Million km^2), Central Asia (CAS, 5.5 Million km^2), Middle East (MIDE, 6.1 Million km^2), China (CHN, 9.4 Million km^2), Korea and Japan (KAJ, 0.6 Million km^2), South Asia (SAS, 5.1 Million km^2), Southeast Asia (SEAS, 4.8 Million km^2), and Oceania (OCE, 8.0 Million km^2).

Results

Pre-industrial soil N_2O emissions

As indicated by HYDE 3.2 land cover data, cropland area was 6.2×10^6 km^2 in 1860, equivalent to ~40% of the cropland area in the recent decade. Manure use in cropland was estimated as 2.9 Tg N yr^{-1}, and no mineral fertilizer was applied to cropland. Soil N_2O emissions in the pre-industrial period provides a baseline for understanding how intensified human activities have disturbed the patterns of soil N_2O emissions. We found that global soil N_2O emissions in the pre-industrial period were 6.3 ± 1.1 Tg N_2O-N yr^{-1} (model ensemble mean ± 1 std. dev., same hereafter, Table 2) and the average emission density was 0.05 ± 0.01 g N m^{-2} ice-free land area yr^{-1} (Figure 1). The lowest and highest N_2O emissions were simulated by ORCHIDEE-CNP (5.1 Tg N_2O-N yr^{-1}) and LPJ-GUESS (8.6 Tg N_2O-N yr^{-1}),

respectively (see spatial patterns of the simulated soil N_2O emissions by individual models in Figure S2). N_2O emissions along a latitudinal gradient (Figure 2a) showed a single peak in the tropics (23.5 °N – 23.5 °S in this study), which contributed 69% to the global total soil N_2O emissions. Soil N_2O emission density in the tropics (0.09 ± 0.02 g N m^{-2} yr^{-1}) was also 85% higher than that of the global average. Of all the 18 regions, the four tropical regions (EQAF, BRA, NSA, and SEAS) were associated with the highest N_2O emission density (Figure 3): 0.13 g N m^{-2} yr^{-1}, 0.12 g N m^{-2} yr^{-1}, 0.12 g N m^{-2} yr^{-1}, and 0.10 g N m^{-2} yr^{-1}, respectively.

Due to less intensive land management in the pre-industrial period, soil N_2O emission density in cropland (0.04 ± 0.02 g N m^{-2} yr^{-1}) was comparable to that in other ecosystems (0.05 ± 0.01 g N m^{-2} yr^{-1}). Cropland soil N_2O emissions were 0.3 ± 0.1 Tg N_2O-N yr^{-1} and contributed only ~4% to the global total soil N_2O emissions. N_2O emissions from natural soil were estimated to be 6.0 ± 1.1 Tg N_2O-N yr^{-1} (see spatial patterns in Figure 4).

Soil N_2O emissions in the recent decade

In the recent decade (2007-2016), global soil N_2O emissions were estimated to be 10 ± 2.0 Tg N_2O-N yr^{-1}, and the average emission density was 0.07 ± 0.01 g N m^{-2} yr^{-1} according to the "best estimate" of model simulations (Figure 1 and Table 2). ORCHIDEE simulated the lowest global soil N_2O emissions (7.8 Tg N_2O-N yr^{-1}), while LPJ-GUESS simulated the highest global soil N_2O emissions (13.6 Tg N_2O-N yr^{-1}) (see spatial patterns of the simulated soil N_2O emissions by individual models in Figure S3). A latitudinal gradient of N_2O emissions displays two peaks, one in the tropics and the other in the temperate region of the Northern Hemisphere (Figure 2b). Tropical soil N_2O emissions were 5.3 ± 0.9 Tg N_2O-N yr^{-1}, accounting for ~53% of global total soil N_2O emissions. N_2O emission density in the tropics

was 0.11 ± 0.02 g N m^{-2} yr^{-1}, which is 51% higher than the global average soil N_2O emission density. Regionally, the four tropical regions (EQAF, BRA, NSA, and SEAS) had high soil N_2O emission densities (0.14 g N m^{-2} yr^{-1}, 0.14 g N m^{-2} yr^{-1}, 0.13 g N m^{-2} yr^{-1}, and 0.15 g N m^{-2} yr^{-1}, respectively). In addition, SAS and CHN also had high soil N_2O emission density (0.16 g N m^{-2} yr^{-1} and 0.14 g N m^{-2} yr^{-1}, respectively), indicating that regions with high soil N_2O emissions rate expanded from the tropical forest regions to cropland-dominant regions.

Soil N_2O emissions were 3.3 ± 1.1 Tg N_2O-N yr^{-1} in cropland, and 6.7 ± 1.4 Tg N_2O-N yr^{-1} in other ecosystems (see spatial patterns of soil N_2O emissions from cropland and other ecosystems in Figure 4). Soil N_2O emission density of cropland (0.21 ± 0.08 g N m^{-2} yr^{-1}) was more than three times that of other ecosystems (0.06 ± 0.01 g N m^{-2} yr^{-1}). Although global cropland area was only ~13% of the ice-free land area, the contribution of cropland soil N_2O emissions to global total soil N_2O emissions reached as much as 33% (Figure 5). For cropland, DLEM simulated the highest soil N_2O emissions (5.0 Tg N_2O-N yr^{-1}), and LPX-Bern and ORCHIDEE simulated the lowest (1.7 and 1.8 Tg N_2O-N yr^{-1}). For other ecosystems, the highest soil N_2O emissions were simulated by LPJ-GUESS (9.5 Tg N_2O-N yr^{-1}), and the lowest was simulated by OCN (5.5 Tg N_2O-N yr^{-1}).

Spatial and temporal changes in soil N_2O emissions

From the pre-industrial period to the recent decade, global soil N_2O emissions increased by 59% (3.7 ± 1.1 Tg N_2O-N yr^{-1}), and the emission density increased on average by 0.03 ± 0.01 g N m^{-2} yr^{-1}. Of the seven models, LPJ-GUESS simulated the largest increases (5.1 Tg N_2O-N yr^{-1}), while LPX-Bern and ORCHIDEE simulated the smallest (2.4 Tg N_2O-N yr^{-1}) (Figure 1, also see spatial patterns of the changed soil N_2O emissions simulated by individual models in Figure S4). The model-ensemble mean showed that the most significant increases occurred

in the mid-latitudes of the Northern Hemisphere (Figure 2). Soil N_2O emissions increased across all the 18 regions (Table 2). Particularly, CHN, SAS, and EU were the three regions showing the fastest increases in N_2O emission density (Figure 3). The increased emission density in the three regions (0.11 g N m^{-2} yr^{-1} for CHN, 0.09 g N m^{-2} yr^{-1} for SAS, and 0.07 g N m^{-2} yr^{-1} for EU) was 2 ~ 4 times that of global average increase rate.

Global cropland soil N_2O emissions increased by 3.0 ± 1.1 Tg N_2O-N yr^{-1} (~11 times) from the pre-industrial period to the recent decade, contributing the majority (82%) to the increased global soil N_2O emissions (Figure 5). DLEM simulated the largest increase (4.7 Tg N_2O-N yr^{-1}), while LPX-Bern and ORCHIDEE simulated the smallest increase (1.6 Tg N_2O-N yr^{-1}) (Figure S5). At the regional level, the largest increases in cropland N_2O emissions were found in CHN, SAS, USA, EU, and SEAS, which accounted for 27%, 15%, 11%, 9% and 8% of the increased global cropland N_2O emissions, respectively. CHN, SAS, and SEAS underwent rapid increases of cropland N_2O emissions since the 1970s (Figure 6). Cropland N_2O emissions in these three regions increased by more than 200% from the 1970s to the recent decade (Table S2). In contrast, US cropland N_2O emissions were relatively stable since the 1980s, and EU cropland N_2O emissions even decreased by an estimated 14% from the 1980s to the recent decade.

From the pre-industrial period to the recent decade, global N_2O emissions from natural soils showed a relatively small increase of 0.7 ± 0.5 Tg N_2O-N yr^{-1} (11%) although total area decreased (Figure 5). Of all the regions, N_2O emissions from natural soil increased by more than 50% in CHN, KAJ, and EU (Figure 6 and Table S3).

Contributions of factors responsible for the increased N_2O emissions

Variations in temporal and spatial patterns of soil N_2O emissions were attributed to multiple land use-related factors, climate, as well as atmospheric composition factors (Figure 7 and 8). Over the study period, manure and N fertilizer addition (MANN and NFER), N deposition (NDEP) and climate change (CLIM) were found to increase soil N_2O emissions, in part through increases in mineral N in soils that serves as a substrate for the N_2O-producing nitrification and denitrification processes (Figure 7). During the period of 2007-2016, the MANN effect on soil emission was 0.6 ± 0.4 Tg N_2O-N yr^{-1}. The NFER effect resulted in continuous increases of soil N_2O emissions, and thus turned croplands into the dominating factor behind the increasing global soil N_2O emissions since the 1970s. During the period of 2007-2016, NFER effect contributed 2.0 ± 0.8 Tg N_2O-N yr^{-1}, accounting for 54% of the increased terrestrial N_2O emissions compared to the pre-industrial situation. During the period of 2007-2016, NDEP was the second largest contributing factor, contributing 26% to increased soil N_2O emissions. The seven models in this study agreed that the effects of direct (MANN and NFER) and indirect (NDEP) anthropogenic N additions to land ecosystems enhanced global soil N_2O emissions, although the magnitudes of contributions varied considerably (Figure S6). LPJ-GUESS was the most sensitive to N addition. The lowest contributions of MANN, NFER, and NDEP were simulated by VISIT, ORCHIDEE, and LPX-Bern, respectively. From the pre-industrial period to the recent decade (2007-2016), the increased amount of N additions (including MANN, NFER, and NDEP) was 174 Tg N yr^{-1}, which enhanced soil N_2O emissions by 3.5 ± 0.9 Tg N_2O-N yr^{-1}, accounting for 96% of the increased global soil N_2O emissions.

The effect of climate (CLIM) was found to stimulate soil N_2O emissions by an estimated 0.9 ± 0.3 Tg N_2O-N yr^{-1} during 2007-2016, ranging between 0.6 Tg N_2O-N yr^{-1} (LPX-Bern) and 1.2 Tg N_2O-N yr^{-1} (LPJ-GUESS) (Figure S6). The regression of the CLIM effect on soil

N_2O emissions simulated by each NMIP model against global average temperature (Figure S7) indicated the strong linear correlation with a transient temperature sensitivity of global soil N_2O emissions by 0.6 ± 0.2 Tg N_2O-N yr^{-1} per unit warming (°C). This sensitivity is higher than that estimated by Zaehle (2013) (0.5 Tg N yr^{-1} $°C^{-1}$), but lower than the estimate of Xu-Ri *et al.* (2012) (1 Tg N yr^{-1} $°C^{-1}$). Rising CO_2 concentration reduced global soil N_2O emissions with its effect increasing through time. In the recent decade, the CO_2 effect reduced global soil N_2O emissions by 0.6 ± 0.6 Tg N_2O-N yr^{-1}, equivalent to 10% of the N_2O emissions in pre-industrial level. The linear regression of CO_2 concentration against soil N_2O emissions (Figure S7) indicated that the transient sensitivity of global soil N_2O emissions to rising CO_2 concentration was -4.6 Gg N_2O-N yr^{-1} ppm^{-1}. The negative CO_2 effect is likely caused by the increased vegetation N use efficiency and higher N uptake from soils. LPJ-GUESS simulated the strongest CO_2 reduction effect on global soil N_2O emissions (-1.8 Tg N_2O-N yr^{-1} in the recent decade). ORCHIDEE-CNP is the only model simulating a small positive CO_2 effect (0.1 Tg N_2O-N yr^{-1} in the recent decade) likely due to the simulated CO_2 effect on soil moisture and substrate availability. LCC had minor impacts on soil N_2O emissions when manure and mineral fertilizer use were not used. Model ensemble results showed that LCC effects on emissions were close to neutral (-0.0 ± 0.5 Tg N_2O-N yr^{-1} in the recent decade), indicating that LCC can either increase or decrease soil N_2O emissions over different regions. Model results of LCC effect diverged, ranging between -0.9 Tg N_2O-N yr^{-1} (LPJ-GUESS) and 0.4 Tg N_2O-N yr^{-1} (ORCHIDEE). Four models (LPX-Bern, ORCHIDEE, DLEM, and ORCHIDEE-CNP) simulated positive effect, two models (OCN and LPJ-GUESS) simulated negative effect, and one model (VISIT) simulated a nearly neutral effect.

Significant changes were found in the driving factors between the pre-industrial period and the contemporary period over all of the 18 regions (Table S4). The top five regions for increases in N addition (NDEP+MANN+NDEP) were CHN (47 Tg N yr^{-1}), SAS (26.6 Tg N

yr^{-1}), EU (18.4 Tg N yr^{-1}), USA (16.3 Tg N yr^{-1}), and SEAS (11.9 Tg N yr^{-1}). The contributions of driving factors to changes in soil N_2O emissions varied across regions (Figure 8). Spatially, in the recent decade, MANN effects were stronger in East CHN and EU. NFER had the strongest effects in stimulating soil N_2O emissions between 20 °N and 50 °N, particularly in East US, EU, SAS, East CHN, and SEAS. The impacts of NDEP, CO_2, and CLIM were more uniformly distributed across the global land area, although NDEP impacts were stronger in EAST US, EU, SAS, East CHN, and SEAS. Rising CO_2 concentration reduced soil N_2O emissions over most of the global land areas, with stronger impacts in the tropics.

At the regional level, NFER was the key factor in enhancing soil N_2O emissions over seven regions, namely, USA, EU, MIDE, CHN, KAJ, SAS, and SEAS (Figure 9). For each of the seven regions, NFER effect contributed at least 40% of the increased soil N_2O emissions. Particularly, the contributions of NFER reached up to 63% in CHN and 74% in SAS. In contrast, CLIM was the dominant factor affecting soil N_2O emissions in ten regions with less intensive human management activities, including CAN, CAM, NSA, BRA, SSA, NAF, EQAF, SAF, RUS, and OCE (Figure 9).

Discussion

Pre-industrial soil N_2O emissions

Pre-industrial N_2O emissions were recently estimated from ice core and marine N_2O measurements (Battaglia & Joos, 2018). The pre-industrial atmospheric lifetime of N_2O (Prather *et al.*, 2015) in combination with the ice core N_2O concentration measurements (MacFarling Meure *et al.*, 2006) yields a net global preindustrial N_2O source to the atmosphere of 10.5 ±1.0 Tg N_2O-N yr^{-1}. Marine N_2O emissions were constrained in a

Bayesian model framework by water column and surface water N_2O measurements and preindustrial emission was estimated to be between 3.1 and 6.1 Tg-N yr^{-1}. This implies by difference with the total source a preindustrial soil N_2O source of about 5.9 (4.1 to 7.7) Tg N_2O--N yr^{-1} (Battaglia & Joos, 2018). This value is consistence with our multi-model estimate of 6.3 ± 1.1 Tg N_2O-N yr^{-1} in the pre-industrial era. Our average pre-industrial soil N_2O emissions from other ecosystems was estimated to be 6.0 Tg N_2O-N yr^{-1}, which is 0.3 Tg N_2O-N yr^{-1} higher than the estimate of Xu *et al.* (2017), but 0.7 Tg N_2O-N yr^{-1} lower than the estimate of Bouwman *et al.* (1993). Direct N_2O emissions from global cropland soil in this study (0.3 Tg N_2O-N yr^{-1}) is consistent with the EF-based estimate of 0.3 Tg N_2O-N yr^{-1} by (Syakila & Kroeze, 2011).

The simulated soil N_2O emissions showed significant spatial variations across global land areas. In this study, soil N_2O emission density (N_2O emission per land area) was higher in the tropics, especially in the Congo and Amazon rainforests (Figure 2a). The lowland tropical ecosystems have higher biological N fixation (BNF) rate (Hedin *et al.*, 2009, Vitousek *et al.*, 2013), are often phosphorus-limited relative to N (Reich & Oleksyn, 2004, Vitousek, 1984), and are associated with optimum temperatures and soil moistures for microbial decomposition organic matter and produce soil N_2O (Butterbach-Bahl *et al.*, 2013). According to Bouwman *et al.* (1993) and Xu *et al.* (2017), the tropics accounted for more than half of the global soil N_2O emissions, which is consistent with the result in this study. Smaller soil N_2O emission density was found in the boreal areas (Figure 2a), which is mainly caused by the fact that boreal and temperate ecosystems are more limited by N availability and the cold weather suppressed microbial activities (Alexander & Billington, 1986).

Contemporary soil N_2O emissions

The NMIP model-ensemble mean indicates that global N_2O emissions from natural soils were 6.5 ± 1.2 Tg N_2O-N yr^{-1} during 1981-2016, which is consistent with rates reported by the IPCC AR5 [6.6 (3.3-9.0) Tg N_2O-N yr^{-1}] (Ciais *et al.*, 2014). Saikawa *et al.* (2014) estimated N_2O emissions from natural soils during 1995-2008 by using a top-down inversion approach, and reported that the average emissions were 7.1 Tg N_2O-N yr^{-1} (lowest value: 4.7 ± 0.65 Tg N_2O-N yr^{-1} in 1995; highest value: 8.4 ± 0.47 Tg N_2O-N yr^{-1} in 2001). NMIP models estimated comparable natural soil N_2O emissions of 6.6 Tg N_2O-N yr^{-1} in the same 14 years period.

For soil N_2O emissions in cropland, significant divergences emerge between previous estimates and this study. Global agricultural N_2O emission in the contemporary period was reported as 4.1 (1.7-4.8) Tg N_2O-N yr^{-1} in the IPCC AR5 (Ciais *et al.*, 2014), including both direct soil N_2O emissions and N_2O emissions from animal production. In this study, we only included direct soil N_2O emissions, which was 3.3 ± 1.2 Tg N_2O-N yr^{-1} in the recent decade. By using IPCC 2006 EF, Del Grosso *et al.* (2008) estimated N_2O from direct soil emissions of 3.8 Tg N_2O-N yr^{-1} in 2000 with emissions from livestock excreta included, which was 46% higher than our result in 2000 (2.6 Tg N_2O-N yr^{-1}). Syakila and Kroeze (2011) estimated the direct cropland N_2O emissions in 2006 as 2.2 Tg N_2O-N yr^{-1}, which was 24% lower than our result in 2006 (2.9 Tg N_2O-N yr^{-1}). FAOSTAT provided long-term estimates of N_2O emission from different sectors in agriculture based on the IPCC Tier1 guideline (FAOSTAT, 2016). Combining emissions from synthetic N fertilizer and manure application, crop residues and cultivated organic soils, FAOSTAT reported the 10-year average cropland soil N_2O emissions to be 1.9 Tg N_2O-N yr^{-1} during 2007-2016. Our estimate (3.3 Tg N_2O-N yr^{-1}) is 74% larger than FAOSTAT estimate in the same period. EDGAR (Janssens-Maenhout *et al.*, 2017) provided global N_2O emissions during 1970-2012 with the global direct cropland

soil emissions of 1.3 Tg N_2O-N yr^{-1} in the 1980s, 1.5 Tg N_2O-N yr^{-1} in the 1990s, and 1.7 Tg N_2O-N yr^{-1} in the 2000s. Our estimates in these three decades (2.2 in the 1980s, 2.5 in the 1990s, and 2.8 Tg N_2O-N yr^{-1} in the 2000s) are higher than EDGAR estimates. The differences highlight the large uncertainties in the previously estimated N_2O emissions and the possible caveats of the NMIP models.

Soil N_2O emissions in response to natural and anthropogenic factors

At the global scale, the EF for direct N_2O emissions caused by fertilizer use has been established at ~ 1% but is considered highly uncertain (Bouwman *et al.*, 2002, De Klein *et al.*, 2006, Gerber *et al.*, 2016, Shcherbak *et al.*, 2014, Smith *et al.*, 2012). Davidson (2009) derived, from global top-down estimates, an emission factor of 2.5%, which however covers both direct and indirect emissions. According to the results of this study, the ratio of NFER effect (2.0 ± 0.8 Tg N_2O-N yr^{-1}) to global total mineral N fertilizer use (113 Tg N_2O-N yr^{-1}) was 1.8 ± 0.7% in the recent decade. The model-derived N_2O emissions in this study consider NFER direct effect as well as the legacy effect resulting from historical soil N accumulation that was not accounted by IPCC. For natural ecosystems, Liu and Greaver (2009) conducted a meta-data analysis by collecting field observations and found that N enrichment significantly enhanced soil N_2O emissions. NMIP models simulated considerable effects of N addition on soil N_2O emissions, which was consistent with Liu and Greaver (2009). However, the simulated soil N_2O emissions in response to N addition (fertilizer and manure N applications, and N deposition) show large divergence among the participated NMIP models (Figure S6). These divergences are primarily caused by the differences in model representation of N processes and parameterization schemes among models (Tian et al. 2018).

Regarding the CLIM effect, experiment-based studies illustrated that warming generally enhanced soil N_2O emissions (Smith, 1997) and the denitrifying bacteria community may adapt to higher temperature (Pärn *et al.*, 2018), which is in line with the results simulated by the NMIP models. The positive feedbacks between soil N_2O emissions and climate warming should be seriously taken into consideration when designing national climate change policies. Variations in precipitation affect microbial processes and soil N dynamics and the corresponding soil N_2O effluxes (Austin, 2011, Dijkstra *et al.*, 2012). The soil drying and wetting cycles caused by rainfall can trigger a considerable N_2O emission pulse (Barton *et al.*, 2008, Davidson *et al.*, 1993, Van Haren *et al.*, 2005). It is important to better simulate soil N_2O emissions associated with the precipitation regimes of extreme dry and wet events. In addition, the freeze-thaw induced N_2O emissions have recently been reported to make up 17-28% of global agriculture N_2O emissions (Wagner-Riddle *et al.*, 2017), which need to be considered in future model development.

For CO_2 effect, previous studies based on model simulations (Kanter *et al.*, 2016, Xu-Ri *et al.*, 2012, Xu *et al.*, 2012) reported that the rising CO_2 concentration suppressed soil N_2O emissions from land ecosystems through stimulating vegetation N uptake and possibly N use efficiency and reducing soil inorganic N concentration. However, observation-based results diverged among ecosystem types. For example, Phillips *et al.* (2001) reported a reduced N_2O emissions during growing season at forest site; while Moser *et al.* (2018) and Regan *et al.* (2011) suggested that atmospheric CO_2 enrichment stimulated soil N_2O emissions in grasslands by increasing soil moisture content, and enhancing root biomass and soil biological activity. CO_2 effect on N_2O emissions could depend on the availability of soil mineral N for plant uptake (Kanter *et al.*, 2016), and then diverges among ecosystems with varied N limitation strength (Xu *et al.*, 2012). However, the magnitude of CO_2 effect on N_2O emissions is still poorly understood at the global level. Of the seven NMIP models,

ORCHIDEE-CNP was the only one model that simulated a positive CO_2 effect, consistent with the meta-analysis of Van Groenigen *et al.* (2011).

Without the effects of fertilizers and manure added to agricultural lands converted from natural lands, the vegetation types prior to and after land conversion determined LCC effect on soil N_2O emissions. Humid forests usually have a higher N_2O emission density, while grassland and shrub have lower N_2O emission density (Potter *et al.*, 1996). In South and Central America, soil N_2O emissions increased following deforestation but then declined with cropland age, resulting in lower N_2O emissions from secondary forests and cropland compared to the mature forests that they replaced (Keller & Reiners, 1994, Melillo *et al.*, 2001, Verchot *et al.*, 1999). In the past several decades, large areas of tropical and temperate forests had been converted to cropland. If cropland management practices were not included, cropland soils would emit less N_2O than mature forests. NMIP simulations show large divergence in LCC effects on soil N_2O emissions among the seven models.

Uncertainties associated with model structure, parameters and drivers

Although all the models used consistent input data, model structure and parameterizations diverged considerably, leading to the different spatial and temporal patterns of the simulated N_2O emissions by models (Figure S2 – Figure S6). In the pre-industrial period, larger standard deviation of the simulated soil N_2O emissions occurred in the tropical areas (Figure S2). This is mainly caused by the large N fluxes in tropical ecosystems and the divergences in model representations of these processes. For example, LPJ-GUESS parameterizes BNF based on evapotranspiration (ET) rate, while ORCHIDEE-CNP estimates BNF based on ecosystem Net Primary Productivity (NPP) (Cleveland *et al.*, 1999). This difference could lead to large divergences in the simulated BNF (Meyerholt *et al.*, 2016), and the NPP-based

algorithm tends to estimate higher BNF in the tropics than ET-based algorithm (Wieder *et al.*, 2015). In addition, N losses (e.g. N denitrification, NH_3 volatilization, and N leaching) have also been parameterized differently in NMIP models (Tian *et al.*, 2018), which could result in large uncertainties in the simulated organic N storage and mineral N availability in the tropical region (Meyerholt & Zaehle, 2018).

Over the recent decades, regions with large standard deviation of the simulated soil N_2O emissions shifted to East CHN, US Midwest, and India (Figure S3), where cropland is extensively distributed. This is likely caused by model uncertainties in representing crop cultivation and management, such as manure use, mineral fertilizer application, and crop type. For example, DLEM (Zhang *et al.*, 2018) and LPJ-GUESS (Lindeskog *et al.*, 2013) parameterize cropland processes based on crop species (e.g. wheat, corn, rice, soybean, cotton); while crops in VISIT are grouped into paddy, generic C_3 crop (e.g. wheat), and warm C_4 crop (e.g. maize). Manure use in cropland is another important uncertainty source. For example, ORCHIDEE simply assumes that all manure N applied to cropland is mineralized and added into the soil mineral N pool; while ORCHIDEE-CNP considers organic manure N addition in urine and feces forms and adds manure into litter and soil organic pools. Manure N, unlike fertilizer N, is obtained from the terrestrial processes, and thus, without removing this N from other places in the terrestrial biosphere, applying the N manure fertilizer could create a non-existing source of N in these models, which could cause more soil N_2O emissions than actually implied.

It should be noted that some factors influencing soil N_2O emissions were not represented by the participating models. In pasture and rangeland, a large amount of N_2O is emitted from livestock excreta deposition, manure and mineral fertilizer application (Davidson, 2009, Steinfeld *et al.*, 2006). However, this study did not consider the impacts of pasture management practices and livestock excreta deposition, which could result in underestimated

soil N_2O emissions in managed grasslands. According to Oenema *et al.* (1997), the contribution of grazing animals was about 1.6 Tg N_2O-N yr^{-1} in the 1990s. Except for fertilization and manure use, other cropland management practices (such as irrigation, tillage, legumes and straw management) were not included in the current NMIP. Previous studies found that cropland management practices could change soil physical conditions, microbial activities, and then soil N_2O emissions (Bouwman *et al.*, 2002, Liu *et al.*, 2011, Smith & Conen, 2004). The missing cropland management in models could lead to larger model uncertainty range. In addition, the impacts of ecosystem disturbances (such as wildfires, hurricane, and logging) were not well parameterized in the NMIP models, largely due to our current knowledge gap regarding the impacts of land disturbances on soil N_2O emissions. Observational results in previous studies diverged: some studies reported increased N_2O emissions after burning (Ishizuka *et al.*, 2002, Karhu *et al.*, 2015, Levine *et al.*, 1990, Levine *et al.*, 1988), while other studies found no significant differences in N_2O emissions between burned site and unburned site (Hao *et al.*, 1988, Takakai *et al.*, 2006). Additionally, the effect of freeze-thaw on N_2O emissions has not been fully represented by NMIP models, although some models simulating the freeze-thaw cycle could presumably represent the trapped N_2O under the frozen soil layer.

Recommendation and Outlook

This study provided a processed-based modeling assessment of global and regional soil N_2O emissions during 1861-2016 and disentangled the underlying mechanisms of the changed patterns by factorial analysis. However, large uncertainties existed not only in the emission magnitude, spatial pattern, temporal trend, but also the contributions of natural and anthropogenic factors. According to the discussions on model uncertainties and the

contributing factors, here we offer the following recommendations for model improvement and future research needs.

Improving model representation of key processes responsible for N_2O fluxes

Soil N_2O formation takes place as a result of nitrification and denitrification processes. Responses of nitrification and denitrification processes to soil conditions and substrate availability are particularly important to accurately simulate soil N_2O emissions. N_2O emissions in these processes need to be validated against observations at different water-filled pore space levels (Bateman & Baggs, 2005, Bollmann & Conrad, 1998, Diem *et al.*, 2017), soil and vegetation types (Ambus, 1998, Maag & Vinther, 1996), temperature (Maag & Vinther, 1996), and substrate levels (Weier *et al.*, 1993). In addition, other N-related fluxes and processes such as BNF, N leaching, and ammonia (NH_3) volatilization also introduced large uncertainties in the simulated soil N concentration (Meyerholt & Zaehle, 2018, Wieder *et al.*, 2015). Explicit representation of these processes is in a critical need for enhancing model simulation accuracy. Cropland manure application accounted for ~15% of the increased global soil N_2O emissions. The transformation of manure organic N to inorganic N could alter soil NH_4^+ and NO_3^- concentrations. The decomposition of manure organic components and N mineralization are in need to be better parameterized by models. It is also necessary to ensure mass conservation for manure addition in cropland by removing C and N from other ecosystems. Field-based studies found that tillage, irrigation, legumes cultivation could alter soil physical and chemical characteristics and N concentration (Jangid *et al.*, 2008, Smith & Conen, 2004), and should be parameterized to reduce uncertainty in cropland emissions. N addition in pasture and rangeland (such as livestock excreta deposition, manure and mineral fertilizer application) is an important source of global soil N_2O emissions

(Davidson, 2009, De Klein & Eckard, 2008), accounting for more than half of the global agriculture N_2O emissions (Dangal *et al.*, submitted). However, these processes were not included in the current NMIP simulation protocol. The consideration of N addition in managed grasslands is an essential task for NMIP to estimate grassland soil N_2O emissions accurately. Other processes, such as peatland drainage (Inubushi *et al.*, 2003), wildfires occurrence (Levine *et al.*, 1990), and freeze-thaw cycle (De Bruijn *et al.*, 2009, Mørkved *et al.*, 2006, Wagner-Riddle *et al.*, 2017, Wolf *et al.*, 2010) have also been found to influence soil N_2O emissions, and need to be considered by future model development. The indirect N_2O emissions from soils could be estimated according to the model-simulated N leaching and volatilization in combination with the IPCC Tier 1 emission factors (De Klein et al., 2006). In addition, terrestrial models could be linked with hydrological models to simulate lateral N_2O emission from the land-aquatic continuum (Klatt et al. 2017).

Improving simulations of soil N_2O response to individual and combined factors

Large divergence among models existed in attributing soil N_2O emissions to different driving factors (Figure S6). As these driving factors would change dramatically in the future, it is impossible to correctly project future soil N_2O emissions if the contributions of these factors were not well understood. Field observations of single factor effect are important for validating model responses. For example, N addition experiments have been widely conducted in natural ecosystems, pasture and croplands (e.g. Bouwman *et al.*, 2002, Liu & Greaver, 2009, Meng *et al.*, 2005, Shcherbak *et al.*, 2014, Zou *et al.*, 2005). These datasets are valuable for understanding the impacts of fertilizer use and N deposition, and validating model-simulated magnitude of soil N_2O emissions due to N enrichment. In addition, the measurements of N_2O emissions in FACE experiments (Ineson *et al.*, 1998, Regan *et al.*,

2011, Reich *et al.*, 2006) can be used to explain the effects of rising CO_2 concentration and climate warming on soil N_2O emissions. Precipitation manipulation experiment (Beier *et al.*, 2012) may provide an opportunity to better parameterize models for the effect of soil drying and wetting cycles caused by changed precipitation regimes. Better representation of the multi-factor effects represents a critical challenge for reducing model uncertainties in projecting future soil N_2O emissions.

Improving the quality of input data sets

Input datasets of land management practices are large sources of uncertainties in the N_2O emissions reported here. For NMIP at the current stage, fertilizer data provided the total amount of inorganic N, rather than the amount of N in different forms (i.e., NH_4^+ and NO_3^-). Nishina *et al.* (2017) reported that, although global N fertilizer amount increased continuously between 1961 and 2010, the fraction of NO_3^- in N fertilizer reduced from 35% to 13%. Fertilizer form could affect soil NH_4^+ and NO_3^- concentrations, and then nitrification and denitrification processes (Hénault *et al.*, 1998). The frequency and timing of fertilizer application could affect the daily and monthly variations of soil N_2O emissions (Smith & Dobbie, 2001). Fertilizer use rate varied considerably among crop types, which contributed to the large uncertainties in estimating cropland N_2O emissions (Ruser *et al.*, 2001). Thus, detailed information of fertilizer application (such as fertilizer form, frequency, timing, and crop-specific application rate) is necessary to be included in the development of fertilizer datasets. In addition, time series of cropland management practices, including legume cultivation, tillage and irrigation, should be developed to drive NMIP models. For cropland area, NMIP used HYDE 3.2 dataset, the quality of which was questionable in some regions, such as continental US (Yu & Lu, 2018) and Asia (Calle *et al.*, 2016, Tian *et al.*, 2014). To

better represent cropland area, regional long-term cropland area datasets with higher accuracy (e.g. Liu & Tian, 2010, Tian *et al.*, 2014, Yu & Lu, 2018) need to be incorporated into HYDE dataset to drive models.

Enhancing data-model integration for improving model performance at multiple scales

Soil N_2O emissions have been measured across various land ecosystem types and regions by using multiple measurement approaches, including chamber measurements (Smith & Dobbie, 2001), eddy covariance (Jones *et al.*, 2011), and laboratory incubation (Miller *et al.*, 2008). To understand soil N_2O emissions at a large scale, national and global N_2O flux measurement networks were established such as the Long-Term Agroecosystem Research Network (LTAR) (Walbridge & Shafer, 2011), International Long-Term Ecological Research Network (ILTER) (Vanderbilt & Gaiser, 2017), Greenhouse Gas Reduction through Agriculture Carbon Enhancement Network (GRACEnet) (Jawson *et al.*, 2005), and the National Agricultural Nitrous Oxide Research Program (NANORP) (Dalal *et al.*, 2003). A recent study of data-model comparison and benchmarking effort has been conducted to estimate site-level N_2O emissions in cropland and grassland (Ehrhardt *et al.*, 2018). Global terrestrial N_2O emissions have also been reconstructed from ice core isotope data over periods of past abrupt climate change allowing for N_2O model evaluation on the century time scale (Schilt *et al.*, 2014). These observational data could be used to validate model performance and constrain large-scale model simulations. More effect in data assimilation and data-model integration at multiple spatial and temporal scales is clearly needed for improving model accuracy in estimating global and regional N_2O fluxes.

Acknowledgements

This study has been supported partially by US National Science Foundation (1210360, 1243232), National Key Research and Development Program of China (# 2017YFA0604702), CAS Grants (KFJ-STS-ZDTP-0; SKLURE2017-1-6), AU-OUC Joint Center Program and Auburn University IGP Program. A. Arneth acknowledges support from the Helmholtz Association (Impulse and Networking fund, and ATMO programme) and EU FP grant LUC4C (603542). S. Olin acknowledges support from the strong research areas BECC and MERGE together with support from LUCCI (Lund University Centre for studies of Carbon Cycle and Climate Interactions). F. Joos and S. Lienert acknowledge support by the Swiss National Science Foundation (#200020_172476). C. Peng acknowledges the support from the Natural Sciences and Engineering Research Council of Canada (NSERC) Discovery Grant. W. Winiwarter acknowledges the support by the Austrian Science Fund (FWF, project number P29130-G27). A. Ito acknowledges support from grant-in-aid by the Japan Society for Promotion of Science (17H01867). The authors declare no conflict of interest.

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Table 1. Simulation experiments in the Global N_2O Model Intercomparison Project (modified from Figure 4 in Tian *et al.* (2018))

	CLIM	**CO_2**	**LCC**	**NDEP**	**NFER**	**MANN**
SE0	1901-1920*	1860	1860	1860	1860	1860
SE1	1901-2016	1860-2016	1860-2016	1860-2016	1860-2016	1860-2016
SE2	1901-2016	1860-2016	1860-2016	1860-2016	1860-2016	1860
SE3	1901-2016	1860-2016	1860-2016	1860-2016	1860	1860
SE4	1901-2016	1860-2016	1860-2016	1860	1860	1860
SE5	1901-2016	1860-2016	1860	1860	1860	1860
SE6	1901-2016	1860	1860	1860	1860	1860

Note: CLIM: climate condition; CO_2: atmospheric CO_2 concentration; LCC: land cover change; NDEP: atmospheric N deposition; NFER: mineral N fertilizer use; and MANN: manure N use in cropland. SE0: baseline and control run with repeated climate forcing from 1901-1920; SE1: CLIM+CO_2+LCC+NDEP+NFER+MANN; SE2: CLIM+CO_2+LCC+NDEP+NFER; SE3: CLIM+ CO_2+LCC+NDEP; SE4: CLIM+ CO_2+LCC; SE5: CLIM+ CO_2; SE6: CLIM. "1901-1920*" denotes that variable is constant at the level of 20-year average; "1860" denotes that variable is constant at the level of 1860; and "1860-2016" denotes that variable changes with time over the study period.

Table 2. Global and regional soil N_2O emissions (Tg N_2O-N yr^{-1}) (mean ± SD)

	Pre-industrial	1960s	1970s	1980s	1990s	2000s	2007-2016
Global	6.3 ± 1.1	7.2 ± 1.2	7.5 ± 1.5	8.4 ± 1.6	9.0 ± 1.8	9.5 ± 1.8	10.0 ± 2.0
1. USA	0.4 ± 0.2	0.6 ± 0.2	0.7 ± 0.3	0.7 ± 0.3	0.8 ± 0.3	0.8 ± 0.3	0.8 ± 0.3
2. CAN	0.1 ± 0.1	0.1 ± 0.1	0.1 ± 0.1	0.1 ± 0.1	0.1 ± 0.1	0.2 ± 0.1	0.2 ± 0.1
3. CAM	0.2 ± 0.1	0.2 ± 0.1	0.2 ± 0.1	0.2 ± 0.1	0.3 ± 0.1	0.3 ± 0.1	0.3 ± 0.1
4. NSA	0.3 ± 0.1	0.4 ± 0.1	0.3 ± 0.1	0.4 ± 0.1	0.4 ± 0.1	0.4 ± 0.1	0.4 ± 0.1
5. BRA	1.0 ± 0.3	1.1 ± 0.3	1.0 ± 0.2	1.0 ± 0.2	1.1 ± 0.3	1.1 ± 0.2	1.2 ± 0.3
6. SSA	0.4 ± 0.2	0.5 ± 0.2	0.4 ± 0.2	0.4 ± 0.2	0.5 ± 0.2	0.5 ± 0.2	0.5 ± 0.2
7. EU	0.2 ± 0.1	0.4 ± 0.2	0.5 ± 0.2	0.6 ± 0.3	0.6 ± 0.3	0.6 ± 0.3	0.6 ± 0.3
8. NAF	0.4 ± 0.1	0.4 ± 0.1	0.5 ± 0.2	0.5 ± 0.2	0.5 ± 0.2	0.5 ± 0.1	0.5 ± 0.1
9. EQAF	1.1 ± 0.3	1.1 ± 0.2	1.1 ± 0.2	1.2 ± 0.2	1.2 ± 0.3	1.2 ± 0.2	1.2 ± 0.2
10. SAF	0.4 ± 0.1	0.4 ± 0.1	0.5 ± 0.1	0.5 ± 0.1	0.5 ± 0.1	0.5 ± 0.1	0.5 ± 0.1
11. RUS	0.3 ± 0.2	0.4 ± 0.3	0.4 ± 0.3	0.5 ± 0.3	0.4 ± 0.3	0.4 ± 0.3	0.4 ± 0.3
12. CAS	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0
13. MIDE	0.0 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.1	0.1 ± 0.1	0.1 ± 0.1	0.1 ± 0.1
14. CHN	0.3 ± 0.1	0.4 ± 0.1	0.4 ± 0.1	0.6 ± 0.2	0.9 ± 0.4	1.1 ± 0.5	1.4 ± 0.8
15. KAJ	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0
16. SAS	0.4 ± 0.1	0.4 ± 0.1	0.5 ± 0.1	0.6 ± 0.2	0.7 ± 0.2	0.7 ± 0.2	0.9 ± 0.3
17. SEAS	0.5 ± 0.2	0.5 ± 0.2	0.5 ± 0.2	0.6 ± 0.2	0.7 ± 0.2	0.7 ± 0.2	0.7 ± 0.2
18. OCE	0.3 ± 0.1	0.3 ± 0.1	0.3 ± 0.2	0.4 ± 0.1	0.4 ± 0.1	0.4 ± 0.2	0.4 ± 0.1

Note: The letters stand for 18 regionals, including USA, Canada (CAN), Central America (CAM), Northern South America (NSA), Brazil (BRA), Southwest South America (SSA), Europe (EU), Northern Africa (NAF), Equatorial Africa (EQAF), Southern Africa (SAF), Russia (RUS), Central Asia (CAS), Middle East (MIDE), China (CHN), Korea and Japan (KAJ), South Asia (SAS), Southeast Asia (SEAS), and Oceania (OCE).

Figure 1. Global soil N_2O emissions in the pre-industrial period and the recent decade (2007-2016), and the change between the two periods simulated by seven models. Error bars denote the ± 1 standard deviation of the seven models.

Figure 2. Model ensemble mean of soil N_2O emission density across global land surface in the pre-industrial period (a) and the recent decade (b, 2007-2016), and the difference between the two periods (c). Right panels are the emission density along latitudinal gradient by 0.5°, and the shaded areas denote the ± 1 standard deviation.

Figure 3. Comparison of regional soil N_2O emissions between the pre-industrial period and the recent decade (2007-2016). (a) Regional total soil N_2O emissions (Tg N_2O-N yr^{-1}), and (b) regional soil N_2O emission density (g N m^{-2} yr^{-1}). The inserted figures are the differences of N_2O emissions between the two periods. Error bars denote the ± 1 standard deviation of the seven models.

Figure 4. Model ensemble mean of soil N_2O emissions from cropland (a) and other ecosystems (b) in the pre-industrial period (first row) and the recent decade (second row), and the differences between the two periods (third row).

Figure 5. Global N_2O emissions from cropland and other ecosystems during 1861-2016: (a) Long-term trend and variations and (b) relative change.

Figure 6. Decadal changes and variations of regional soil N_2O emissions from cropland and other ecosystems between the 1860s and the 2010s (Tg N_2O-N yr^{-1}). The last decade (the 2010s) in each panel refers to the period of 2010-2016.

Figure 7. Contributions of natural and anthropogenic factors [Manure use (MANN), N fertilizer application (NFER), N deposition (NDEP), Land cover change (LCC), CO_2 concentration rising, and climate conditions (CLIM)] to global soil N_2O emissions from the 1860s to the 2010s. The 2010s refers to the period of 2010-2016. Black line is the sum of the contribution of all factors.

Figure 8. Spatial and latitudinal patterns of the contributions of natural and anthropogenic factors [Manure use (MANN), N fertilizer application (NFER), N deposition (NDEP), Land cover change (LCC), CO_2 concentration rising, and climate conditions (CLIM)] on soil N_2O emissions in the recent decade (2007-2016). The latitudinal pattern is calculated at 0.5° interval. Shaded areas denote the ± 1 standard deviation.

Figure 9. Contributions of natural and anthropogenic factors [Manure (MANN), N fertilizer (NFER), N deposition (NDEP), Land cover change (LCC), CO_2 concentration rising (CO_2), and climate conditions (CLIM)] to the changed regional soil N_2O emissions in the recent decade (2007-2016).