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Global land-atmosphere exchange of methane and nitrous oxide: magnitude and spatiotemporal patterns

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Abstract

Methane (CH_4) and nitrous oxide (N_2O) are two most important greenhouse gases after carbon dioxide, but their regional and global budgets are far from certain, which is largely owing to uncertainties in scaling up field measurements as well as the poor model representation of processes and factors governing CH_4 and N_2O exchange between the terrestrial biosphere and atmosphere. In this study, we applied a process-based, coupled biogeochemical model (DLEM – the Dynamic Land Ecosystem Model) to estimate the magnitudes, spatial and temporal patterns of CH_4 and N_2O fluxes as driven by multiple environmental changes including climate variability, rising atmospheric CO_2, increasing nitrogen deposition, tropospheric ozone pollution, land use change and nitrogen fertilizer use. The estimated CH_4 and N_2O emissions from global land ecosystems were $169.43 \pm 32.92\,Tg\,C\,yr^{-1}$ and $12.52 \pm 1.52\,Tg\,N\,yr^{-1}$, respectively. Our simulations have indicated a significant ($P < 0.01$) increasing trend for CH_4 ($0.75 \pm 0.08\,Tg\,C\,yr^{-1}$) and N_2O ($0.14 \pm 0.02\,Tg\,N\,yr^{-1}$) during 1981–2010. CH_4 and N_2O emissions increased significantly in most climatic zones and continents, especially in tropical region and Asia. The most rapid increase in CH_4 emission was found in wetlands (including rice fields and natural wetlands) owing to increased rice field area and climate change; N_2O emission increased substantially for all the biome types and the largest increase occurred in upland crops owing to increasing air temperature and nitrogen fertilizer use. Given large increase in CH_4 and N_2O emission at global scale, we suggest that these two gases together with CO_2 have to be simultaneously considered when evaluating if a policy is effective or efficient to reduce global warming in the future.

1 Introduction

As two important greenhouse gases (GHG) contributing to climate warming, methane (CH_4) and nitrous oxide (N_2O) are receiving more and more attention (Prather and

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Hus, 2010; Montzka et al., 2011; Kirschke et al., 2013; Saikawa et al., 2013; Tian et al., 2012a). Recent analyses indicated that the atmospheric CH_4 has increased by more than 100 % since 1800 (Forster et al., 2007). Methane has a relative global warming potential 25 times that of CO_2 at a 100 yr time horizon, and contributes approximately 20 % to the global radiative forcing (Forster et al., 2007). Net CH_4 fluxes from terrestrial ecosystems are primarily determined by two distinct microbial processes: methanogenesis (production) and methanotrophy (consumption) (Walter and Heimann 2000; Bloom et al., 2010). Methanogenesis occurs under anaerobic conditions, while methanotrophy occurs in aerobic or anaerobic condition. Many factors, such as water table depth, substrate quality and quantity, soil pH, soil moisture, presence of permafrost, oxygen concentration, and ratio of methanogenic to methanotrophic bacteria, directly influence CH_4 production and consumption (Tian et al., 2010; Xu et al., 2010; Banger et al., 2012; Dijkstra et al., 2012). The dominant sources of CH_4 are natural wetlands, anthropogenic activities, and biomass burning (Dlugokencky et al., 2009; Ito and Inatomi et al., 2012), while the major CH_4 sink is the upland soils. Recent CH_4 budget estimation showed a global source of 548–678 $Tg\,CH_4\,yr^{-1}$ during 2000–2009, nearly 50–60 % from anthropogenic origin (Kirschke et al., 2013).

Atmospheric N_2O has increased by 18 % compared to the pre-industrial level, with a linear increasing rate of 0.26 % per year during the recent few decades (Forster et al., 2007). Nitrous oxide has a relative global warming potential 298 times that of CO_2 at a 100 yr time horizon, and contributes approximately 7 % to the radiative forcing (Forster et al., 2007). Therefore, although its concentration is much less than that of CO_2 and CH_4 in the atmosphere, it is drawing more attention in recent years. The observed increase in atmospheric N_2O concentration was primarily attributed to reactive nitrogen inputs from synthetic nitrogen fertilizer and animal manure applications, cropland expansion, and processes associated with fossil-fuel combustion and biomass burning. The production and consumption of N_2O in soils involves both biotic and abiotic processes. Many microbial groups contribute to the production and consumption

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of soil N_2O, but biological nitrification and denitrification are the dominant processes (Ussiri and Lal, 2012).

A number of studies using different modeling and statistical approaches were conducted to estimate regional or global CH_4 (e.g., Bousquet et al., 2006; Potter et al., 1996; Bloom et al., 2010; Tian et al., 2010; Xu et al., 2012; Zhuang et al., 2004; Schulze et al., 2009; Ito and Inatomi et al., 2012; Meng et al., 2012; Kirschke et al., 2013) and N_2O fluxes (e.g., Xu et al., 2008; Liu, 1996; Hirsch et al., 2006; Davidson 2009; US EPA 2010; Bouwman et al., 2002; Zhuang et al., 2012). However, the estimates are far from certain due to the complexity of the terrestrial ecosystems and the shortage of field and laboratory measurements to validate the modeled processes controlling gas emissions (Ito and Inatomi et al., 2012; Tian et al., 2012a; Melton et al., 2013). To simulate large-scale CH_4 and N_2O fluxes, two approaches are generally used during past decades: bottom-up (e.g., inventory and process-based modeling, Cao et al., 1995; Walter et al., 2001; Zhuang et al., 2004; Xu et al., 2008; Tian et al., 2010) and top-down (e.g., atmospheric inversion, Frankenberg et al., 2005; Bergamaschi et al., 2007, 2009; Kort et al., 2008; Bousquet et al., 2011; Spahni et al., 2011). Process-based modeling, as one of the bottom-up approaches, can simulate the spatial heterogeneity of ecosystem properties at relatively high resolution and identify the underlying mechanisms that control the magnitudes and variations of GHG fluxes (Spahni et al., 2011; Tian et al., 2010; Xu et al., 2010; Bousquet et al., 2011).

Many global change factors could affect the production and consumption of CH_4 and N_2O (Hutchin et al., 1995; Conrad, 1996; Huang et al., 2010; Toet et al., 2011). For instance, global warming was reported to increase both CH_4 and N_2O emissions (Dijkstra et al., 2012); elevated atmospheric CO_2 increased CH_4 emission (Dacey et al., 1994; Dijkstra et al., 2012), while decrease or increase N_2O emission (Ineson et al., 1998; Kettunen et al., 2005); ozone (O_3) pollution reduced CH_4 emission (Toet et al., 2011), while stimulated or reduced N_2O emission (Kanerva et al., 2008); nitrogen fertilization could dramatically decrease CH_4 consumption in grassland and forest (Mosier et al., 1991; Steudler et al., 1989), while increase N_2O emissions (Del Grosso et al., 2006;

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Mosier et al., 1998). Meanwhile, interactions among multiple factors also played an important role. For example, nitrogen deposition and elevated atmospheric CO_2 were reported to interactively reduce CH_4 emission from wetland (Pancotto et al., 2010); and another study concluded that temperature and elevated atmospheric CO_2 might interactively change seasonal variation of CH_4 emission (Blankinship et al., 2010). Previous studies addressed the impacts of only one or a subset of environmental factors on either CH_4 or N_2O fluxes; however, few of them were conducted to fully estimate the effects of multiple environmental changes (such as climate, land use, land management practices, atmospheric CO_2, nitrogen deposition and tropospheric O_3, etc.) on both gases. In addition, given the tight linkage between these two gases in both terrestrial origin (Raghoebarsing et al., 2006; Kort et al., 2008; Ettwig et al., 2010; Song et al., 2009; Dijkstra et al., 2012) and atmospheric chemistry (Prather and Hus, 2010), it is important to simultaneously estimate the CH_4 and N_2O fluxes at continental or global scale. In this study, we used a process-based model (the Dynamic Land Ecosystem Model, DLEM; Tian et al., 2010) to estimate CH_4 and N_2O fluxes driven by multiple environmental changes in climate, atmospheric CO_2, tropospheric O_3 concentration, nitrogen deposition, land use and nitrogen fertilizer use. The DLEM model has been applied to quantify and attribute the responses of non-CO_2 GHG fluxes to multiple environmental factors in China, North America and globe (Tian et al., 2010, 2011a, 2012a; Xu et al., 2010, 2012a; Melton et al., 2013; Wania et al., 2013). Built upon our previous work, the specific objectives in this study were to: (1) estimate the magnitude of CH_4 and N_2O fluxes from terrestrial biosphere and (2) explore the spatial and temporal variations in terrestrial CH_4 and N_2O fluxes as influenced by multiple environmental factors on regional and global scales.

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2 Methodology

2.1 Data description

In this study, we developed a series of spatiotemporal data sets to represent environmental changes at a spatial resolution of 0.5° × 0.5° during 1901–2010. These data include daily O_3 AOT40 index (an index of the accumulated hourly ozone concentrations above a threshold of 40 ppb), annual nitrogen deposition rate, daily climate conditions (minimum, average and maximum temperature, precipitation, and solar radiation), annual land use and land management practices (including nitrogen fertilizer use and irrigation). The climate data was downloaded from CRUNCEP website. O_3 AOT40 data were derived from the monthly AOT40 data developed by Felzer et al. (2005). EDGAR-HYDE 1.3 nitrogen emission data (Van Aardenne et al., 2001) were used to interpolate the three-time period nitrogen deposition data (1860, 1993, 2050; Dentener, 2006) to generate annual nitrogen deposition data set during 1900–2010 (Wei et al., 2013). Land use data (Cropland and urban area distribution) were from History Database of the Global Environment (HYDE 3.0) (Klein and van Drecht, 2006). Nitrogen fertilizer use rate for China and the United States were developed from county-level census data (Tian et al., 2010, 2011b), while information in other regions were based on Food and Agriculture Organization (FAO) country-level statistical data (www.fao.org). Soil property data, including soil texture, pH, and bulk density, were from Global Soil Data Task (www.daac.ornl.gov). The annual atmospheric CO_2 concentration data before 1959 were from the Vegetation/Ecosystem Modeling and Analysis Project (VEMAP), and the data thereafter were from the National Oceanic and Atmospheric Administration (NOAA) (www.esrl.noaa.gov). The potential vegetation (i.e., natural vegetation) map was developed from different data sources, including global land-cover data derived from Landsat imageries (De Fries et al., 1998), National Land Cover Dataset 2000 (www.usgs.gov), and global database of lakes, reservoirs, and wetland (Lehner and Döll, 2004). The contemporary global vegetation map in 2000 was shown in Fig. 1. All the datasets were transformed and re-projected to the same projection system. Due

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to the lack of data after 2005, the land use and land cover pattern, O_3 AOT40, and nitrogen fertilizer use rate after 2005 were assumed the same as the level of 2005.

2.2 Model description

The DLEM model is designed to estimate carbon, nitrogen, and water fluxes and pool sizes in terrestrial ecosystems by coupling major physiological, biogeochemical and hydrological processes, and vegetation dynamics with spatial coverage ranging from site to region and globe, and time step from daily to monthly and yearly (Tian et al., 2010, 2011a, b, 2012a, b). One of important DLEM's features is that it can simultaneously estimate the responses of land-atmosphere exchange of CO_2, CH_4 and N_2O to multiple environmental changes in climate, atmospheric CO_2 concentration, tropospheric O_3 pollution, nitrogen deposition, land use and land management practices. Each plant functional type defined in DLEM (i.e., types shown in Fig. 1) was calibrated against various field observations collected from the Chinese Ecological Research Network (CERN), US Long-Term Ecological Research (LTER) network, AmeriFlux network, and other independent research sites. Here we briefly described the CH_4 and N_2O module in the DLEM. The more detailed information could be referred to Tian et al. (2010, 2011a, b, 2012b) and Xu et al. (2010, 2012).

In DLEM, CH_4 production, consumption, and transport processes are considered to estimate land-atmosphere gas exchange. Dissolved organic carbon (DOC) is the only CH_4 production substrate considered in DLEM. The DOC comes from gross primary productivity (GPP) allocation, and decomposition byproducts from soil organic matter and litterfall, which indirectly controlled by environmental factors including soil pH, temperature and soil moisture content. CH_4 oxidation is determined by CH_4 concentrations in the air or pore space of soil, as well as soil moisture, pH, and temperature. We consider three pathways for CH_4 transport from soil to the atmosphere (i.e., ebullition, diffusion, and plant-mediated transport) (Tian et al., 2010). It is assumed that methane-related biogeochemical processes only occur in the top 50 cm of soil profile. Overall, the net CH_4 exchange between the atmosphere and soil is calculated by the following

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equation:

$$F_{CH_4} = F_P + F_D + F_E - F_{air,\,oxid} - F_{trans,\,oxid} - F_{soil,\,oxid} \quad (1)$$

where F_{CH_4} is the flux of CH_4 between soil and the atmosphere ($g\,C\,m^{-2}\,day^{-1}$); F_P is plant-mediated transport from soil pore water to the atmosphere ($g\,C\,m^{-2}\,day^{-1}$); F_D is the diffusive flux of CH_4 from water surface to the atmosphere ($g\,C\,m^{-2}\,day^{-1}$); F_E is the ebullitive CH_4 emission to the atmosphere; $F_{air,\,oxid}$ is atmospheric CH_4 oxidation rate ($g\,C\,m^{-2}\,day^{-1}$); $F_{trans,\,oxid}$ is the CH_4 oxidation during plant-mediated transport ($g\,C\,m^{-2}\,day^{-1}$); $F_{soil,\,oxid}$ is the CH_4 oxidation rate in soil pore water.

Terrestrial nitrogen cycling processes considered in the DLEM model include nitrogen input from the atmospheric deposition, agricultural fertilizer use, and biological fixation, plant nitrogen uptake, nitrogen transformation (e.g., mineralization and immobilization, nitrification and denitrification, adsorption/desorption), and nitrogen loss (e.g., nitrous gas emissions, nitrogen leaching, and other nitrogen loss pathways through fire, grazing, or other disturbances). The estimation of N_2O emissions (primarily from nitrification and denitrification) are closely interlinked with model representation of above processes.

In DLEM, all the products of nitrification and denitrification that leave the system are nitrogen-containing gases. We used the empirical equation reported by Davidson et al. (Davidson et al., 2000) to separate N_2O from other gases (mainly NO and N_2). The equations for calculating nitrification, denitrification and N_2O fluxes are:

$$F_{N_2O} = (0.001 \times N_{nit} + N_{denit}) \times \frac{10^{\left(\frac{vwc}{\emptyset} \times \times 0.026 - 1.66\right)}}{\left(1 + 10^{\left(\frac{vwc}{\emptyset} \times \times 0.026 - 1.66\right)}\right)} \quad (2)$$

where F_{N_2O} is the N_2O flux from soil to the atmosphere ($g\,N\,m^{-3}\,day^{-1}$), which can be converted to the unit of $g\,N\,m^{-2}\,day^{-1}$ by multiplying the soil depth (default: 0.5 m);

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N_{nit} and N_{denit} are the nitrification and denitrification rates ($g N m^{-3} day^{-1}$), respectively; 0.001 is the proportion of nitrification product released as gaseous nitrogen (Lin et al., 2000); $\emptyset$ is the soil porosity.

2.3 Model implementation and experimental design

The implementation of DLEM simulation includes three steps: (1) equilibrium run, (2) spin-up run, and (3) transient run. In this study, we first used land use and land cover map in 1900, long-term mean climate during 1901–1930, the concentration levels of nitrogen deposition, O_3 pollution, atmospheric CO_2 in 1900 to run the model to an equilibrium state (i.e., the inter-annual variations during 20 continuous years are less than $0.1\,g C m^{-2}$, $0.1\,g N m^{-2}$ and 0.1 mm for carbon, nitrogen and water pools, respectively). After model reached equilibrium state, a spin-up run was implemented for 900 yr using de-trended climate data during 1901–1930 (i.e., 30 spins with 30 yr data each time). Finally, the model was run in transient mode with daily or annual environmental data from 1901 to 2010 to simulate CH_4 and N_2O fluxes. Model results from 1981 to 2010 were used to analyze the spatial and temporal patterns of global CH_4 and N_2O fluxes in this study.

Two simulation experiments were designed to achieve the study goals: (1) Baseline experiment: transient input data during 1901–1980 for all environmental factors (i.e., climate, land use, nitrogen deposition, ozone pollution, atmospheric CO_2 concentration, and nitrogen fertilizer use) were fed into the DLEM model and then kept constant at the level of 1980 thereafter; (2) combined experiment: all environmental factors changed with time during 1901–2010. The effects of multiple environmental changes on CH_4 and N_2O fluxes were the difference between these two experiments. With this experiment design, the legacy effects of environmental factors before 1981 were excluded.

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2.4 Model evaluation and uncertainty analysis

In this study, we estimated the modeled uncertainty ranges for CH_4 and N_2O resulting from variations in the most sensitive parameters. First, we conducted sensitivity analysis to identify the most sensitive parameters that affect terrestrial CH_4 and N_2O fluxes. We selected about 10 parameters related to CH_4 and N_2O fluxes, respectively and assigned a variation range of ±20 % for each parameter. According to the simulation results driven by these individual parameters, we selected the top 5 most sensitive parameters to conduct the uncertainty simulation. Second, we assumed that each parameter follows normal distribution. Combined with the prior knowledge of parameter range, we used improved Latin Hypercube Sampling (LHS) approach to randomly select an ensemble of 100 sets of parameter values to do model simulations. Finally, the outliers for the simulation results were excluded before analyzing the uncertainty range. The 95 % confidence intervals were calculated and reported.

The evaluations of DLEM simulation results against field investigations, regional inventory, and other modeling results for both CH_4 and N_2O fluxes were conducted in our previous studies (Tian et al., 2010, 2011a, 2012b, c; Xu et al., 2010, 2012; Ren et al., 2011 and Melton et al., 2013). These evaluations indicated that DLEM is able to capture the daily, annual, and spatial variation patterns of the observed CH_4 and N_2O fluxes. We showed the DLEM model evaluations (some were from our previous publications) against field experiments or observations worldwide in Table 1. The observed CH_4 and N_2O fluxes showed large variability within and among research sites, indicating the complex impacts of multiple environmental factors on CH_4 and N_2O fluxes. We found that most of the DLEM-simulated CH_4 and N_2O fluxes fell in the observation range for different land cover types (i.e., forest, grassland, wetland, and cropland). We further added some regional comparisons with previous studies (see Discussion).

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2.5 Statistical analysis

The multiple linear regression analyses were conducted to explore the long-term changing trends of input data and CH_4 and N_2O fluxes. The Pearson correlation was conducted to evaluate the correlations between input data and CH_4 and N_2O fluxes. All the statistical analyses were conducted by using the software R-program (www.r-project.org); and the figures and maps were generated by Origin 8.0 and ArcGIS 9.2.

3 Results

3.1 Global environmental change over past three decades

From 1981 to 2010, air temperature and precipitation significantly increased by $0.02\,^{\circ}C\,yr^{-1}$ and $1.37\,mm\,yr^{-1}$, respectively (Table 2). Nitrogen fertilizer use, nitrogen deposition, atmospheric CO_2 concentration, cropland and urban land area continuously increased during the study period. The tropospheric O_3 concentration showed a slight increase before 1995 and a fast increase thereafter. Relative to a 30 yr (1961–1990) average, precipitation increased in most areas during the period 1981–2010. The highest increase occurred in the South America, whereas large precipitation decrease was found in the southern Africa, the northern India, and the southeastern Australia (Fig. 2a). Air temperature increased in most areas, with the highest increases in the northern high-latitude region and a slight decrease in the South America and Oceania (Fig. 2b). The highest nitrogen deposition rates were shown in the East and South Asia, while the highest nitrogen fertilizer use rates were shown in the eastern China. These regions experienced fast growth in population, urbanization, and industrialization during the past three decades. The highest tropospheric ozone concentrations in 2005 were distributed along the northern mid-latitude region, such as the southeastern United States, the southern Europe, the central Asia, and the western China.

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At continental scales, air temperature increased in all continents except for Oceania, with the most rapid increase in Europe and Asia (Table 2). Precipitation significantly increased for Africa and South America, while no significant trends were found for other continents. Nitrogen deposition and O_3 concentration (AOT40 index) increased significantly for all continents, with the highest increasing rates in Asia over past three decades. Cropland area significantly changed for all continents, with a decreasing trend found in Europe and North America and rapid increase in Asia and Africa. Cropland fertilizer use significantly increased in the continents other than Europe and the most rapid increase occurred in Asia. Air temperature increased for all climate zones except the southern temperate zone, and the northern polar zone showed the most rapid increase (Table 3). Precipitation increased significantly in the northern polar, northern tropical and southern tropical zones. Nitrogen deposition increased significantly for all continents, with the fastest increase in the northern tropical zone. Cropland nitrogen fertilizer use amounts increased significantly for most climate zones except that the northern polar zone showed a significant decrease. Cropland area decreased significantly in the northern polar and northern temperate zones, while increased significantly in other climate zones, with the most rapid growth in the northern tropical zone. Ozone concentration increased significantly in the tropical and northern temperate zones.

3.2 Global budget of terrestrial CH_4 and N_2O fluxes

The estimated terrestrial CH_4 emission during 1981–2010 was averaged $169.43 \pm 32.92\,Tg\,C\,yr^{-1}$ ($1\,Tg = 10^{12}\,g$), ranging from $155.36 \pm 30.05\,Tg\,C\,yr^{-1}$ in 1982 to $187.80 \pm 36.19\,Tg\,C\,yr^{-1}$ in 2010. Among which, the upland soil uptake of CH_4 was $17.10 \pm 0.17\,Tg\,C\,yr^{-1}$. The terrestrial N_2O emission was averaged $12.52 \pm 1.89\,Tg\,N\,yr^{-1}$, ranging from $10.53 \pm 1.52\,Tg\,N\,yr^{-1}$ in 1982 to $16.65 \pm 2.80\,Tg\,N\,yr^{-1}$ in 2010 (Fig. 3). Both CH_4 and N_2O showed a significant ($P < 0.01$) increasing trend of $0.75 \pm 0.08\,Tg\,C\,yr^{-1}$ for CH_4 and $0.14 \pm 0.02\,Tg\,N\,yr^{-1}$

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for N_2O over the study period. The interannual variations in CH_4 and N_2O fluxes were roughly consistent ($R^2 = 0.88$; $P < 0.01$), indicating a close relationship between CH_4 and N_2O fluxes, which was found to be primarily controlled by climate variability. Significantly positive correlations between air temperature and the model-estimated CH_4 ($R^2 = 0.73$, $P < 0.01$) and N_2O emissions ($R^2 = 0.71$, $P < 0.01$) was observed, implying a strong positive feedback between climate warming and CH_4/N_2O emissions.

In terms of global warming potential (100 yr horizon), the DLEM-estimated global CH_4 and N_2O emissions (i.e., $5.65 \pm 1.10\,Pg\,CO_2\,eq\,yr^{-1}$ and $5.86 \pm 0.89\,Pg\,CO_2\,eq\,yr^{-1}$, respectively) could completely offset the terrestrial CO_2 sink ($8.3 \pm 4.3\,Pg\,C\,yr^{-1}$ estimated by multiple process-based models, Le Quéré et al., 2012) over the recent three decades. It implies that global land ecosystem acted as a positive contributor to climate warming.

3.3 Spatial distribution of terrestrial CH_4 and N_2O fluxes

As expected from the distribution of wetlands and rice paddy fields, the South and Southeast Asia, tropical wetlands and river flood plains (e.g., Amazonia and the Pantanal) were dominant hot-spots of CH_4 emission with values as high as $30\,g\,C\,m^{-2}\,yr^{-1}$ (Fig. 4). In these regions, more CH_4 production precursor-DOC was produced due to the higher biomass, primary production, and litter decomposition rate. In contrast, the northern high-latitude region (e.g., Alaska, the northern Canada, West Siberia and the northern Eurasia) that have large wetland areas, were less substantial CH_4 emission sources ($\sim 10\,g\,C\,m^{-2}\,yr^{-1}$). Larger CH_4 sinks (i.e., $> 0.15\,g\,C\,m^{-2}\,yr^{-1}$) were found in the tropical and subtropical uplands due to more substrate and favorable climate conditions for methanotrophy. Similar to CH_4, N_2O released at rates of larger than $0.3\,g\,N\,m^{-2}\,yr^{-1}$ in most areas of the tropical region as well as intensively fertilized cropland, while smaller emissions occurred in the high-latitude and sparse vegetated area. In general, N_2O emissions decreased with air temperature from the low- to high-

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latitude region, indicating air temperature was one of the most important factors controlling N_2O emissions. To more clearly identify the spatial distribution patterns of CH_4 and N_2O fluxes, we further divided the globe into different latitudinal belts, biome types and continents.

3.4 Terrestrial CH_4 and N_2O fluxes and temporal trends along a latitudinal gradient

Along the latitudinal gradient, CH_4 emissions peaked (8.06 Tg C yr^{-1}) at the latitudinal zones of 2 ~ 3° S and 6 ~ 7° S (Fig. 4), primarily due to large wetland area in Amazon river basin. Another smaller peak for CH_4 emissions displayed at the northern high-latitude region (around 60° N). N_2O emissions peaked at the latitude zone of 6–7° N (0.43 Tg N yr^{-1}), which is primarily due to large area of fertilized cropland. We further grouped the global terrestrial ecosystems into five major climate zones: the northern polar zone (> 60° N), the northern temperate zone (30°–60° N), the northern tropical zone (0°–30° N), the southern tropical zone (0°–30° S), and the southern temperate zone (30°–60° S). All climate zones were net sources of CH_4 and N_2O emissions to the atmosphere (Table 4). The largest CH_4 emissions occurred in the southern tropical zone, followed by the northern tropical zone, and the least in the southern temperate zone. The tropical zone (both southern and northern) totally accounted for about 80 % of the total CH_4 emissions. The largest N_2O emissions occurred in the northern tropical zone, followed by the southern tropical zone, and the least was found in the southern temperate zone. The tropical zone also accounted for about 75 % of the total N_2O emissions.

Our simulation experiments indicated that temporal trends in the CH_4 and N_2O fluxes varied significantly among various climate zones (Fig. 5). Over the study period, the most rapid increase in CH_4 emission was observed in northern and southern tropical zone (0.22–0.37 Tg C yr^{-1}), followed by northern temperate zone (0.12 Tg C yr^{-1}) and northern polar zone (0.046 Tg C yr^{-1}) while no significant changing trend was found

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for the southern temperate zone. Similarly, faster increase in the N_2O emissions has occurred in northern tropical and southern tropical zones (0.039–0.078 $Tg N yr^{-1}$) than other zones. No significant changing trend in N_2O emission was observed for the southern temperate zone; however, larger interannual variations were found in this zone compared to other zones. The highest CH_4 and N_2O emissions occurred in 2010 for all the climate zones except for the northern polar and southern temperate zones.

3.5 Terrestrial CH_4 and N_2O fluxes and temporal trends for different biome types

We further compared the magnitude of CH_4 and N_2O fluxes and their temporal trends during 1981–2010 for different biome types (Table 5; Fig. 6). The largest CH_4 emissions were from natural wetland (130.74 $Tg C yr^{-1}$), followed by cropland (54.37 $Tg C yr^{-1}$). The highest CH_4 uptake occurred in forest (−6.43 $Tg C yr^{-1}$) and shrubland (4.38 $Tg C yr^{-1}$). Multiple environmental changes had caused wetland CH_4 emissions increased from 129.01 $Tg C yr^{-1}$ in 1981 to 137.67 $Tg C yr^{-1}$ in 2010. From 1981 to 2010, forest and shrubland showed a significant ($p < 0.05$) increasing trend in CH_4 uptake, with a rate of 0.014 $Tg C yr^{-1}$ and 0.0061 $Tg C yr^{-1}$, respectively (Fig. 6). Wetlands and rice field showed a more significant ($P < 0.01$) increasing trend in CH_4 emission, with a rate of 0.25 $Tg C yr^{-1}$ ($P < 0.01$) and 0.54 $Tg C yr^{-1}$ ($P < 0.01$), respectively, while no significant change trend was found for grassland. Rice field expansion (15 % as shown in Table 2) contributed to 12 % of cropland CH_4 emissions increase and the remaining 88 % increase resulted from other environmental changes including climate, atmospheric composition, and land management practices. It is notable that CH_4 uptake in upland ecosystems (i.e., forest, shrubland and grassland) exhibited a faster increase from 2000 to 2010 compared to previous two decades.

The largest emissions of N_2O were from forest (4.28 $Tg N yr^{-1}$), followed by cropland (3.36 $Tg N yr^{-1}$), and the least was from grassland (0.82 $Tg N yr^{-1}$). Most of the forest N_2O emissions came from tropical region. Compared to forests in other climatic zones,

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tropical forests had larger area and higher nitrogen transformation rate. Nitrous oxide emissions significantly increased for all the biome types from 1981 to 2010 (Fig. 6). N_2O emissions kept a slight ($P > 0.1$) increase from 1981 to 2000 for forest, shrubland and grassland, but a significantly faster increase was found during 2000–2010. Cropland displayed the largest increase in N_2O emission ($0.08\,Tg\,N\,yr^{-1}$; $R^2 = 0.93$; $P < 0.01$), which was primarily due to rapid cropland expansion and increasing nitrogen fertilizer uses (Table 2). Cropland area has expanded by 7.8 % (~ 11 million ha) since 1981, and nitrogen fertilizer uses have increased by 66 % (~ $33\,Tg\,N\,yr^{-1}$). Increased nitrogen deposition and air temperature were the major causes of increased N_2O emissions in the non-managed biomes.

3.6 Change trends in terrestrial CH_4 and N_2O fluxes for different continents

At continental scale, significant increasing trends ($p < 0.01$) in CH_4 emissions were found for all continents except Africa and Europe (Fig. 7). Asia showed the highest CH_4 emission and the most rapid increase ($0.56\,Tg\,C\,yr^{-1}$, $p < 0.01$). CH_4 emissions from Europe showed a large decrease since the early 1990s. Significant increasing trends in N_2O emissions were found in all continents except Europe and Oceania. N_2O emissions had no obvious change trend before 2000 in Europe, but a rapid increase occurred since then. The most rapid N_2O emission increases in Asia ($0.056\,Tg\,N\,yr^{-1}$, $P < 0.01$) was caused by larger rises in nitrogen fertilizer uses and nitrogen deposition compared to other continents (Table 2). Asia and South America acted as the largest CH_4 and N_2O sources with the highest increasing trends, suggesting that these two continents will continue to play a major role in greenhouse gas budget in the future. It is also notable that the interannual variations of CH_4 and N_2O emissions were very similar ($R^2 = 0.89$; $P < 0.01$) in Asia, implying a close linkage between these two gases across this continent.

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4 Discussion

4.1 Comparisons with other estimation of global CH_4 and N_2O budgets

The simultaneous estimation of global CH_4 and N_2O fluxes shown in this study were in-line with previous estimates (Table 6). The DLEM-estimated global CH_4 flux was 157–187 $Tg C yr^{-1}$ for the 1990s, which fell in the range of 86–200 $Tg C yr^{-1}$ estimated by other investigators (Fung et al., 1991; Houweling et al., 1999; Ridgwell et al., 1999). This study estimated that the global natural wetland emitted 130.98 $Tg C yr^{-1}$ in the 1990s, which is higher than 92 $Tg C yr^{-1}$ estimated by Cao et al. (1998), but similar to the estimate by Liu (1996) and slightly lower than the average value from multiple process-based models (142.5 $Tg C yr^{-1}$; Melton et al., 2013). Our study reported that 128.19 $Tg CH_4 - C yr^{-1}$ was released from global natural wetland in the 1980s, slightly higher than a previous estimate of 110 $Tg C yr^{-1}$ (Matthews and Fung, 1987). Our estimated CH_4 emission from rice field (51.42–62.4 $Tg C yr^{-1}$) was higher than most previous estimates. It might be due to the expanding rice field and other input drivers (e.g., nitrogen fertilizer amounts and nitrogen deposition rate) considered in this study, but omitted in previous work (Cao et al., 1998; Chen and Prinn 2005; Patra et al., 2009). DLEM-estimated upland CH_4 uptake during 1996–2005 ($-15.01 \sim -13.45$ $Tg C yr^{-1}$) was slightly lower than the previous estimates.

In addition to divergent model representation of biogeochemical processes and parameter values (Meng et al., 2012), different input data might be another reason responsible for the discrepancies among these CH_4 estimates (Meng et al., 2012; Ito and Inatomi et al., 2012; Zhu et al., 2011; Melton et al., 2013). For example, knowledge of wetland distribution and area was highly uncertain. Through multiple models comparisons, Melton et al. (2013) reported that the estimate of global wetland area ranged from 7.1×10^6 to 26.9×10^6 km^2. Even the inventory data largely varied among different data sources ranging from 4.3×10^6 to 12.9×10^6 km^2. In the future, there is a necessity to integrate all data sources into a most accurate data set for model use. In addition,

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large uncertainty might be also resulted from various climate data. Several series of climate data were previously used by the global models to simulated GHG fluxes. For example, CRU (Mitchell and Jones, 2005), CRUNCEP (Wei et al., 2013) and NCEP reanalysis (Kalnay et al., 1996) data were used by different model simulations (e.g., Bousquet et al., 2011; Ito and Inatomi et al., 2012; Melton et al., 2013).

This study reported global N_2O emission in a range of 10.49–14.30 $Tg N yr^{-1}$ during 1981–2000, which was close to an empirical estimate of 13.31 $Tg N yr^{-1}$ (Xu et al., 2008). The estimated N_2O flux during 1990–1994 (11.51–12.15 $Tg N yr^{-1}$) was slightly higher than a process-based modeling result (11.33 $Tg N yr^{-1}$; Liu, 1996) and an inverse modeling result (9.5 $Tg N yr^{-1}$; Nevison et al., 1996). In our study, we considered the manure application (no livestock management), nitrogen fertilizer use, nitrogen deposition, nitrogen fixation and agricultural biomass burning, making our estimated cropland N_2O flux slightly higher than the estimates from FAO/IFA (2001), Davison (2009) and Bouwman et al. (2002), which did not consider the impacts of nitrogen deposition and fixation. DLEM-estimated N_2O emissions from cropland were close to most of previous estimates (e.g., Davison 2009; Bouwman et al., 2002; US EPA 2010). Our estimates for natural land N_2O emissions were slightly higher than those from Bowden (1986), Saikawa et al. (2013), and US EPA (2010), and significantly higher than the estimate from the statistical extrapolation (Zhuang et al., 2012). DLEM estimated N_2O emission from global grassland and forest were consistent with the empirical extrapolations by Xu et al. (2008) and Zhang et al. (2010). The discrepancies in input datasets and estimation methods might be the major causes of these differences. DLEM used multiple environmental factors, including climate, nitrogen deposition, atmospheric CO_2, tropospheric ozone, nitrogen fertilization and land use change, as model input, while most previous studies only considered one or a subset of these factors.

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4.2 Regional difference in CH_4 and N_2O fluxes

Based on measurements and extrapolation, Bartlett and Harriss (1993) and US EPA (1993) estimated that 60.55 % of CH_4 emissions were from tropical wetland. Matthews and Fung (1987) estimated that only about 28.83 % was from tropical region, instead, over 58.56 % was from the high-latitude region. However, with the same methods but more observations, Fung et al. (1991) updated their early estimate and reported that about 69.57 % of CH_4 emissions were from tropical region. Based on eight process-based ecosystem models, Melton et al. (2013) estimated that about 66.32 % of CH_4 emissions were from tropical region. The inverse modeling approach estimated an even higher emission from tropical region (81.01 %, Hein et al., 1997; 83.52 %, Wang et al., 2004; 70.75 %, Bousquet et al., 2006). Combining all these estimates, US EPA (2010) reached a consensus that about 75 % of CH_4 emissions were from the tropical region. Our estimate of 73.92 % (upland CH_4 uptake is not separated) is close to this value. The increasing CH_4 emission from the northern high latitude and tropical region is also consistent with the estimate from Dlugokencky et al. (2009). We estimated that about 80 % of N_2O emissions were from the tropical region. Based on observations and inverse methods, Prinn et al. (1990) also reported that 52–68 % of the N_2O emissions were from the tropical region during 1978–1988. In contrast, using an inversing modeling approach, Hirsch et al. (2006) showed a lower estimate of N_2O emissions from 30° S to 90° S (0 to 4 %) and a higher N_2O emission from 0° to 30° N (50 to 64 %) during 1998 to 2001.

Due to different spatial and temporal change patterns of multiple environmental factors, the change trends of CH_4 and N_2O fluxes were greatly different among continents and climate zones. In Asia, land cover changed dramatically due to fast-growing population and industrialization during the past three decades and was primarily characterized by conversions of upland natural ecosystems to rice fields, which was the major contributor to the increasing CH_4 emission. In addition, nitrogen fertilizer use amount in Asia increased the most rapidly compared to other continents (Table 2) and led to

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substantial increase in N_2O emission. Based on inventory data, Kurokawa et al. (2013) found that CH_4 and N_2O increased by 32 % and 18 %, respectively from 2000 to 2008 in Asia. By using inversion modeling, Saikawa et al. (2013) also reported that N_2O emissions from agricultural soil increased by 56 % (0.055 $Tg\,yr^{-1}$) in Asia (i.e., Northern Asia + Southern Asia) from 1995 to 2008. In North America, land use kept relatively stable and climate variability became the major contributing factors for CH_4 emissions, while both climate and nitrogen fertilizer uses contributed the most to N_2O emissions as indicated by our previous studies (i.e., Tian et al., 2010, 2012b; Xu et al., 2010, 2012a). DLEM simulations in Europe showed a slight decreasing trend in CH_4 emission, and a less significant increase in N_2O compared to other continents. Over shorter period (1990–2011), Saikawa et al. (2013) and EEA (2013) reported that both agricultural and natural soil N_2O emissions had no significant changing trends in Europe.

4.3 Climate change and CH_4 and N_2O emissions

Previous studies indicated that climate variability and change determined inter-annual variations in terrestrial CH_4 and N_2O fluxes (Frolking and Crill, 1994; King, 1997; Xu et al., 2008, 2010, 2012a; Tian et al., 2010; Dijkstra et al., 2012). A field experiment found the hierarchical control on N_2O emission in forest, and concluded that precipitation controls the instant N_2O flux pattern while air temperature determines the relatively long-term regime (Brumme et al., 1999). Based on field experiments of 74 plots, Gundersen et al. (2012) found that increased air temperature and precipitation greatly increased CH_4 and N_2O emissions, with stronger impacts from increasing air temperature compared to increasing precipitation. A significant correlation was also found between air temperature and atmospheric CH_4 concentration during pre-industrial period (Wuebbles and Hayhoe 2002; Chappellaz et al., 1993a, b). An even stronger positive feedback between N_2O emissions and air temperature (about 1 $Tg\,N\,yr^{-1}\,°C^{-1}$) was found by Xu et al. (2012b). In this study, model simulation also revealed a high correlation ($R^2 = 0.73$ for CH_4 and 0.71 for N_2O; $P < 0.01$) between CH_4/N_2O and

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air temperature, implying a strong positive feedback between climate warming and CH_4/N_2O emissions. Global air temperature significantly increased (0.024°C^{-1}; Tables 2 and 3) from 1981 to 2010, with the highest increase in the northern polar region (0.039 °C yr^{-1}). Climate warming in the northern high-latitude region was reported as the most important stimulating factor for increases in atmospheric CH_4 and N_2O (e.g., Mosier et al., 1998; Cantarel et al., 2011; Koven et al., 2011). In our study, we also found that the increases in both CH_4 and N_2O emissions were primarily (> 50 %) attributed to air temperature change in the > 60° N climatic zone.

4.4 Temporal trends of CH_4 and N_2O fluxes for different biomes

In this study, we found that the increase in CH_4 emissions was primarily due to the effects of multiple environmental changes on wetland and rice paddy land. Multiple environmental changes had led to an increase in natural wetland CH_4 emission from 129.01 Tg C yr^{-1} in 1981 to 137.67 Tg C yr^{-1} in 2010. Through data synthesis, Kirschke et al. (2013) also found a steady increase in wetland CH_4 emission from 1985 to 2010 based on the bottom-up estimation approach, with the highest increase of about 15 Tg C yr^{-1} from 1985 to 2010. We found that rice field area has increased by 0.28×10^6 km^2 from 1981 to 2010, and CH_4 emission from paddy land increased by about 20 Tg C yr^{-1} (Fig. 6c). Many previous studies attributed the increased atmospheric CH_4 concentration to increased natural wetland emissions (e.g., Chen and Prinn, 2006; Kirschke et al., 2013; Saikawa et al., 2013), but our study suggested that cropland was among the most important contributors. US EPA (2006) predicted rapid increases in CH_4 emissions from rice cultivation from 1990 (17.64 Tg C yr^{-1}) to 2010 (20.37 Tg C yr^{-1}), with a 15.5 % increase. Kirschke et al. (2013) found an even more rapid increase in natural wetland CH_4 emissions from 2005 to 2010, which was also found in our study (Figs. 4a and 7c). For all the upland biomes (i.e., forest, shrubland and grassland), our model estimation showed CH_4 uptake increase was larger after 2000, indicating an acceleration of both CH_4 emission and uptake in recent decade.

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We found that cropland N_2O emissions constantly increased during 1981–2010, while natural biomes showed a more rapid increase during 2000–2010. By synthesizing most recent studies on global agricultural N_2O emissions, Reay et al. (2012) and US EPA (2011) indicated that N_2O emissions from agricultural soil increased by about $1\,Tg\,N\,yr^{-1}$ from 1990 to 2010. We had a similar finding with agricultural N_2O emission increasing by $1.4\,Tg\,N\,yr^{-1}$ during the same period (Fig. 6e). By using inversion modeling, Saikawa et al. (2013) also estimated an N_2O emission increase of $1.27\,Tg\,N\,yr^{-1}$ (from $2.65\,Tg\,N\,yr^{-1}$ in 1995 to $3.92\,Tg\,N\,yr^{-1}$ in 2008) in global agricultural land. The increases in global cropland area and nitrogen fertilizer amounts, as well as climate change were the major causes for rapid N_2O emissions increase in cropland (Mosier et al., 1998). Davidson (2009) combined both top-down and bottom-up approaches and estimated that about 2.5 % fertilized nitrogen was released during 1860 to 2005. Considering nitrogen fertilizer amount increasing from $\sim 50\,Tg\,N\,yr^{-1}$ in 1981 to $\sim 84\,Tg\,N\,yr^{-1}$ in 2005, we can conclude that during 1981–2005, increase of about $0.85\,Tg\ N_2O\text{-}N\ yr^{-1}$ was directly derived from the rising nitrogen fertilizer use. The increases in N_2O emissions for natural biomes might be primarily due to climate warming and nitrogen deposition. Based on manipulative field experiments, Dijkstra et al. (2012) and Cantarel et al. (2011) found that climate warming significantly increase N_2O emissions in grassland. Nitrogen deposition could greatly increase carbon storage in the terrestrial ecosystems and in the meanwhile increasing N_2O emissions for natural biomes (Liu et al., 2009; Lu et al., 2013).

4.5 Uncertainties and implications

Due to the complexity of the biogeochemical processes related to CH_4 and N_2O fluxes (Conrad, 1996; Xu et al., 2008; Tian et al., 2012a), some uncertainties need to be considered when interpreting the modeling results. First, uncertainties might be resulted from the simplification of modeling mechanisms for CH_4 and N_2O production and consumption. DLEM runs at daily time step and might miss pulses in CH_4 and N_2O fluxes

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at sub-daily scale. These high pulses may substantially contribute to the annual fluxes (Brumme et al., 1999). Studies have found that the actual ebullition process may be different from the mechanisms applied in most current process-based models (Baird et al., 2004; Kellner et al., 2005; Strack et al., 2005). Although these studies pointed out the possible drawbacks of current modeling representations for this process, no more reliable method has been put forward yet. Additional field or experimental investigations are needed to improve model representation of CH_4 ebullition. Second, the parameters uncertainties might lead to estimation biases. For example, it is important to take into account differences of CH_4 production and oxidation in tropical and northern wetlands, which have not been well documented yet (Blais et al., 2005). Third, the uncertainties in input data also need to be considered. For example, CH_4 and N_2O fluxes have been reported at an order of magnitude difference among different wetland classes (Barlett and Harriss, 1993; Song et al., 2009), thus the small discrepancy in wetland area and wetland classification could lead to a substantial difference in regional estimation. Meanwhile, the varied wetland extent along the study period is one of the major factor influencing inter-annual variation in CH_4 fluxes (Ringeval et al., 2010; Melton et al., 2013). As pointed out in a recent model intercomparison project (Wania et al., 2013; Melton et al., 2013), to more accurately simulate CH_4 fluxes, current process-based models need to be improved in several aspects including the wetland extent dynamics, hydrological cycle, etc. N_2O emission from pasture management (i.e., irrigation, fertilization, grazing rotation, etc.) may contribute a great portion to the global N_2O flux (Ambus and Robertson, 2006; Li et al., 1996); however, it is not considered in our study.

These findings have important implications for mitigation strategies, which include increasing efficiency of nitrogen use in crop production. In our previous studies (Tian et al., 2011a, 2012a; Lu et al., 2012), we found that some regions in the world are suffering from excessive nitrogen input. Less nitrogen input will maintain the same food productivity but reduce the risks for higher N_2O emissions, as well as soil and water nitrogen pollutions. To slow down future global warming, policy makers should

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pay special attention to reducing CH_4 and N_2O emissions in the meanwhile increasing carbon sequestration. Currently, many management practices or governmental policies were implemented either for increasing carbon sequestrations or for reducing carbon emissions, such as intensive management in cropland and planted forests, as well as expanding the area of energy crops to produce "clean" energy. These measures may effectively increase carbon sequestration or reduce carbon emission; however, their impacts on CH_4 and N_2O fluxes are still uncertain (Melillo et al., 2009; Murdiyarso et al., 2010; Tian et al., 2012b). Comprehensive considerations for the three most potent gases are necessary before putting forward any management practices or policies over large area. Future studies are also called for simultaneously investigating net fluxes of CO_2, CH_4 and N_2O in both field experiments and regional estimations.

5 Conclusions

Based on a process-based ecosystem model, this study concurrently estimated CH_4 and N_2O fluxes in the terrestrial biosphere during 1981–2010. As most of previous studies addressed only one of these gases and include one or a subset of all these environmental factors, our study might significantly contribute to the accurate estimation of global CH_4 and N_2O budgets and spatiotemporal patterns. We found that both CH_4 and N_2O emissions greatly increased from 1981 to 2010, indicating global environmental changes could greatly increase CH_4 and N_2O emission to the atmosphere, which would in turn speed climate warming. Most of the CH_4 and N_2O emissions and the highest increasing rate were found in the tropical zone, suggesting this region could be the hot spots for future climate change research. Although climate change also enhanced CH_4 and N_2O emissions in the northern high-latitude region, the increasing rate was much less than that of the tropical region. Methane uptakes slightly increased for the upland ecosystems (e.g., forest, dry cropland, shrubland and grassland) while CH_4 emission greatly increased in the lowland ecosystems (e.g., natural wetland and rice paddy land). N_2O emission increased in all the ecosystems with the highest increasing

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rate found in cropland, which is primarily resulted from cropland management practices (e.g., fertilizer use, irrigation and manure application). Our estimates for global CH_4 and N_2O budgets as well as temporal and spatial variations were consistent with previous field experimental, regional and global studies. High correlations between air temperature and CH_4/N_2O emissions indicate a positive feedback between climate warming and terrestrial emissions of CH_4 and N_2O. Given large increase in CH_4 and N_2O emission at global scale, we suggest that these two gases together with CO_2 have to be simultaneously considered when evaluating if a policy is effective or efficient to reduce global warming in the future.

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Table 1. DLEM validation against site-level measurement of CH_4 and N_2O fluxes.

Location	Land cover type	DLEM	Observation	Reference
		CH_4 fluxes ($g C m^{-2} yr^{-1}$)		
19° E, 68° N	Mire	6.1737	0.13–30.5	Svensson et al. (1984)
147.85° W, 64.8667° N	Tundra	7.2921	0.47–8.05	Whalen et al. (1988)
47°32′ N, 93°28′ W	Peatland	19.15	3.5–65.7	Dise et al. (1993)
113° W, 55° N	Sedge meadow	18.92	21.7	Vitt et al. (1990)
66° W, 54° N	Fen	9.34	1.3–9.8	Moore et al. (1990)
4° W, 55° N	Bog	9.29	1.3–9.3	Clymo et al. (1971)
21° W, 74.5° N	Bog	7.44	0–66.96	Christensen et al. (2000)
2.90° S, 54.95° W	Forest	−0.17	−0.12 to −0.37	Davidson et al. (2008)
105° W, 53° N	Forest	−0.041	−0.063	Hatson et al. (2009)
5° E, 52° N	Peatland	28.23	14.5–68.9	Hendriks et al. (2007)
133°31′ E, 47°35′ N	Freshwater marsh	14.5 ± 3.0 [a]	11.8 ± 2.6 [a]	Lu et al. (2012)
112°11′ E, 23°11′ N	Tropical forest	−0.115[a]	-0.145 ± 0.02 [a]	Lu et al. (2012)
90.11° W, 29.8°N	Woody wetland	21.18	−2.63–24.9	Yu et al. (2008)
84.67° W,45.57° N	Trembling aspen	-0.1 ± 0.07	0.271	Ambus et al. (1999)
122.23° W, 37.4° N	Grassland	-0.30 ± 0.01 to -0.38 ± 0.05[b]	-0.13 ± 0.01 to -0.45 ± 0.14[b]	Blankinship et al. (2010)
95.167° W, 47.25° N	Rice	342.18 ± 38.05	369.75	Harriss et al. (1985)
118° E, 32° N	Rice paddies	22.99	18.14 ± 21.17	Xiong et al. (2007)
91.55° W, 29.87° N	Wetland	52–57	4.3–160	DeLaune et al. (1983)
		N_2O fluxes ($g N m^{-2} yr^{-1}$)		
63° W, 10° S	Forest	0.1	0.05–0.322	Garcia (2003)
94° W, 3° S	Forest	0.13	0.08–0.22	Matson (1987)
48° E, 1° N	Agriculture	0.17	0.08–0.21	Dambreville (2008)
79° E, 9° N	Forest	0.17	0.09–0.73	Koehler (2009)
84° W, 10° N	Forest	0.17	0.08–0.58	Keller (1994)
76° W, 10° N	Forest	0.16	0.15–0.54	Matson (1987)
84° W, 10° N	Agriculture	0.38	0.029–0.75	Veldkamp (1997); Weitz (1998, 2001)
65° W, 17° N	Fallow	0.22	0.007–1.92	Ericson (2001)
65° W, 18° N	Forest	0.22	0.002–0.077	Erickson (2002)
98° W, 19° N	Agriculture	0.262	0.14–0.29	Luqueno (2009)
83° W, 19° N	Forest	0.03	0–0.022	Matson (1987)
105.03° W, 19.3° N	Forest	0.06	0.009–0.048	Davidson (1993)
100° W, 21° N	Agriculture	0.1	0.08–0.19	Perez (2004)

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Table 1. Continued.

Location	Land cover type	DLEM	Observation	Reference
157° W, 21° N	Forest	0.04	0.002–0.007	Vitousek (1989)
91° W, 35° N	Agriculture	0.07	0.01–0.05	Cochran (1997)
104° W, 38.5° N	Agriculture	0.24	0.082–0.25	Bronson (1992); Delago (1996); Mosier (1986)
107° W, 39° N	Agriculture	0.02	0.198	Mosier and Hutchinson (1981)
104° W, 40° N	Agriculture	0.03	0.026–0.27	Mosier (1982)
102° W, 41° N	Agriculture	0.022	0.009–0.011	Kessavalou (1998)
72.5° W, 41.5° N	Forest	0.02	0.001–0.002	Bowden (1990)
72.5° W, 43.5° N	Forest	0.05	0.003–0.006	Castro (1993)
89.5° W, 43.5° N	Forest	0.08	0.019–0.21	Goodroad (1984)
71.5° W, 44° N	Deciduous	0.04	0.091	Keller (1983)
68.5° W, 44.5° N	Forest	0.03	0.001	Castro (1993)
11° E, 48° N	Agriculture	0.11	0.13–0.96	Flessa (1995); Ruser et al. (2001)
112° W, 49° N	Agriculture	0.02	0.025–0.157	Chang (1998); Hao (2001)
14° E, 51° N	Forest	0.06	0.004–0.12	Butterbach (1997)
106° W, 52° N	Agriculture	0.007	0.007	Corre (1999)
114° W, 53° N	Agriculture	0.026	0.011–0.071	Lemke (1998)
24° E, 60° N	Agriculture	0.03	0.03	Parela (2006)
26° E, 66° N	Agriculture	0.06	0.62–0.73	Regina (2004)
13° E, 100° N	Agriculture	0.26	0.033–0.051	Towprayoon (2005)
28.55° E, 111.3° N	Agriculture	0.6	0–0.21	Shang (2011)
1° E, 114° N	Agriculture	0.17	−5.03–2.9	Hadi (2010)
31° E, 118° N	Agriculture	0.03	0.003–0.18	Liu (2010); Wang (2011); Zou (2005)
31° E, 120° N	Agriculture	0.43	0.18–2.53	Hou (2012); Peng (2011)
41° E, 122° N	Agriculture	0.09	0.046–0.057	Jiao (2005)
31° E, 136° N	Agriculture	0.229	0.34–1.2	Hadi (2010)

[a] Unit is $g C m^{-2}$ growingseason^{-1}.
[b] Unit is $mg C m^{-2} day^{-1}$.

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Table 2. Annual change rates of multiple environmental factors for different continents from 1981 to 2010.

Environmental factors	Asia	North America	Europe	Africa	South America	Oceania	Global
Air temperature ($^{\circ}C\,yr^{-1}$)	0.035^{b}	0.030^{b}	0.039^{b}	0.032^{b}	0.016^{b}	0.0079	0.026^{b}
Precipitation ($mm\,yr^{-1}$)	0.78	0.51	0.56	2.26^{b}	3.50^{b}	2.01	0.97^{a}
Shortwave radiation ($W\,m^{2}\,yr^{-1}$)	0.002	−0.017	0.002	0.014	0.11^{b}	0.15^{a}	0.048^{b}
N deposition ($mg\,N\,m^{2}\,yr^{-1}$)	8.70^{b}	2.20^{b}	4.70^{b}	5.20^{b}	7.20^{b}	0.50^{b}	5.60^{b}
N fertilizer use ($g\,N\,m^{2}\,yr^{-1}$)	0.27^{b}	0.05^{a}	-0.13^{a}	0.01^{a}	0.11^{b}	0.13^{b}	0.093^{b}
Ozone ($ppb - hr\,month^{-1}$)	58.6^{b}	40.71^{b}	28.86^{b}	47.11^{b}	7.97^{b}	0.05^{a}	32.87^{b}
Cropland ($10^{4}\,km^{2}\,yr^{-1}$)	2.52^{b}	-0.091^{b}	-1.45^{b}	1.96^{b}	0.72^{b}	0.17^{b}	4.01^{b}

Note: [a] indicates significant change trend at $P < 0.05$ and [b] indicates significant trend at $P < 0.01$.

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Table 3. Annual change rates of multiple environmental factors for different climate zones from 1981 to 2010.

Environmental factors	Northern polar	Northern temperate	Northern tropics	Southern tropics	Southern temperate
Air temperature ($°C\,yr^{-1}$)	0.039^{b}	0.034^{b}	0.033^{b}	0.016^{b}	0.007
Precipitation ($mm\,yr^{-1}$)	0.42^{a}	0.066	2.68^{b}	3.42^{b}	−0.68
N deposition ($mg\,N\,m^{2}\,yr^{-1}$)	0.26^{b}	5.07^{b}	8.57^{b}	2.75^{b}	0.72^{b}
N fertilizer use ($g\,N\,m^{2}\,yr^{-1}$)	-0.082^{b}	0.068^{b}	0.10^{b}	0.040^{b}	0.02^{b}
Ozone ($ppb - hr\,month^{-1}$)	−1.67	32.80^{a}	50.78^{b}	3.89^{a}	0.0008
Cropland ($10^{4}\,km^{2}\,yr^{-1}$)	-0.01^{a}	-0.42^{a}	2.72^{b}	0.83^{b}	0.70^{b}

Note: [a] indicates significant change trend at $P < 0.05$ and [b] indicates significant trend at $P < 0.01$.

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Table 4. Average terrestrial CH_4 ($TgC\,yr^{-1}$; $1\,Tg = 10^{12}\,g$) and N_2O ($TgN\,yr^{-1}$) fluxes and their change rates (%) for different climate zones during 1981–2010 as simulated by the DLEM.

Variables	Time period[a]	Northern polar zone	Northern temperate zone	Northern tropical zone	Southern tropical zone	Southern temperate zone
CH_4 fluxes	1980s	9.89 ± 0.24[b]	20.62 ± 0.44	54.55 ± 0.92	77.03 ± 0.92	0.16 ± 0.08
	2000s	10.80 ± 0.26	22.98 ± 0.57	61.31 ± 1.42	81.16 ± 1.12	0.23 ± 0.12
	Mean	10.30 ± 0.22	21.89 ± 0.51	58.15 ± 1.32	78.88 ± 0.92	0.21 ± 0.06
	Change rate (%)	9.22	11.44	12.39	5.37	47.53
N_2O fluxes	1980s	0.34 ± 0.01	2.47 ± 0.05	4.43 ± 0.19	3.93 ± 0.15	0.14 ± 0.01
	2000s	0.38 ± 0.02	2.88 ± 0.11	5.96 ± 0.36	4.68 ± 0.19	0.17 ± 0.01
	Mean	0.35 ± 0.01	2.63 ± 0.08	5.11 ± 0.28	4.27 ± 0.15	0.15 ± 0.01
	Change rate (%)	14.37	16.54	34.67	19.03	23.96

[a] Mean annual fluxes in the period 1981–1990, 2001–2010 and 1981–2010. [b] The values are denoted as mean ± 2 SE (standard error).

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Table 5. Average terrestrial CH_4 ($Tg\,C\,yr^{-1}$) and N_2O ($Tg\,N\,yr^{-1}$) fluxes for different biome types during 1981–2010 as simulated by the DLEM model.

Variables	Forest	Shrubland	Wetland	Grassland	Cropland
CH_4 fluxes	-6.43 ± 0.11	-4.38 ± 0.04	130.74 ± 1.11	-1.90 ± 0.01	54.37 ± 1.98
N_2O fluxes	4.28 ± 0.10	2.82 ± 0.11	0.97 ± 0.03	0.82 ± 0.03	3.36 ± 0.28

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Table 6. Comparison of global terrestrial CH_4 and N_2O fluxes estimated from multiple sources.

CH_4 flux ($TgCyr^{-1}$)	Time period	This study	Others	Reference
Global	1990s	163.86–182.51	86–195	Fung et al. (1991); Houweling et al. (1999)
Global	1993–2004	129.70–135.92	106–198	Melton et al. (2012)
natural	1990s	126.47–135.92	69.0–202.5	Cao et al. (1998); Bousquet
wetland				et al. (2011); Riley et al. (2011)
	1990s	126.47–135.92	127	Liu (1996)
	2000s	130.90–137.67	78.75–208.50	US EPA (2011)
Global upland	1996–2005	−17.76 to −16.13	−19.5 to −25.0	Ito and Inatomi (2012); Potter et al. (1996); Ridgwell et al. (1999); Del Grosso et al. (2000)
Rice paddy land	1990s	51.42–62.40	19.2–84.0	Cao et al. (1998); Chen and Prinn (2005); Patra et al. (2009); Ito and Inatomi (2012); Yan et al. (2009)
High-latitude wetland (> 60° N)	1980s	9.47–10.56	15 (3–27)	Christensen et al. (1996)
High-latitude (> 60° N)	1990s	8.89–10.89	15.75	Zhuang et al. (2004)

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Table 6. Continued.

N_2O flux ($Tg C yr^{-1}$)	Time period	This study	Other	Reference
Global	1980–2000	10.49–14.30	13.31 (8.19–18.43)	Xu et al. (2008)
	1990–1994	11.51–12.15	11.33	Liu, 1996
	1990–1994	11.51–12.15	9.5	Nevison et al. (1996)
	1997–2001	12.62–14.30	12.55[a]	Huang et al. (2008)
	1998–2001	12.62–14.30	10.7–12.7	Hirsch et al. (2006)
	1990s	11.51–14.30	3–39	Mosier et al. (1998); Banin (1986); Kroeze et al. (1999); Seiler and Conrad (1987)
Global natural land	2000	8.67	1.96–4.56[b]	Zhuang et al. (2012)
	1990	8.76	6.6 (3.3–9.0)[c]	FAO/IFA 2001; US EPA 2010
	Early 1980s	7.97–8.48	7–16	Bowden (1986)
	1995–2008	8.67–10.02	5.27–8.28	Saikawa et al. (2013)
Global cropland	1990	3.00	2.9[d]	FAO/IFA 2001
	1995	3.29	2.8	Bouwman et al. (2002)
	2000	3.50	4.4[e]	US EPA 2011
	2000	3.50	2.6–3.5[f]	Davison (2009)
	1995–2008	2.86–4.39	2.65–3.96	Saikawa et al. (2013)
Global grassland	2000–2007	0.85	0.92	Zhang et al. (2010)
	2000	0.78	1.31	Zhuang et al. (2012)
	2000–2008	0.85	1.52	Xu et al. (2008)
Global forest	2000–2008	4.44	6.99	Xu et al. (2008)
	2000	4.18	1.30	Zhuang et al. (2012)

[a] The emission from ocean (23 %) is excluded.
[b] Cropland is not included.
[c] Natural wetland emission (~ 0.97 $Tg N yr^{-1}$ as estimated by DLEM) is not included.
[d] Including emissions from fertilizer use, crops, decomposition of crops, and biomass burning.
[e] Including both direct and indirect emissions from fertilizer use.
[f] Including fertilizer use, biomass burning and manure application.

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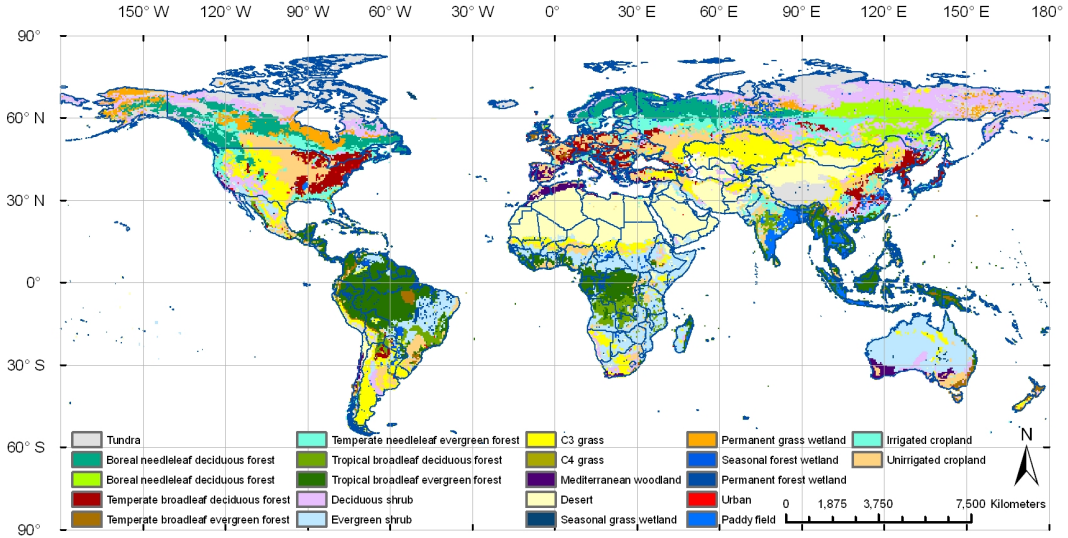

Fig. 1. Distribution of different land use and land cover types in the global terrestrial ecosystems in 2000.

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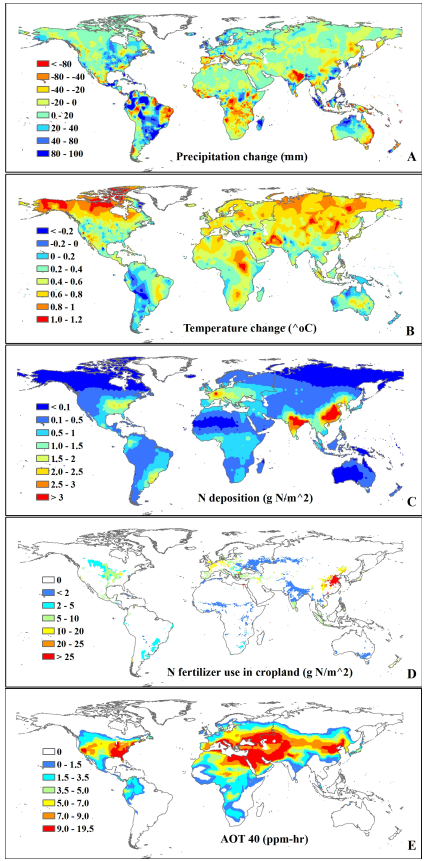

Fig. 2. Spatial distribution of environmental factors in the terrestrial ecosystems. **(A)** Precipitation anomaly (1981–2010mean – 1961–1990mean); **(B)** air temperature anomaly; **(C)** nitrogen deposition rate in 2010; **(D)** cropland distribution and nitrogen fertilizer amount in 2005; **(E)** cumulative tropospheric ozone concentration above 40 ppb – hr (AOT40: ppm – hr) in July 2005.

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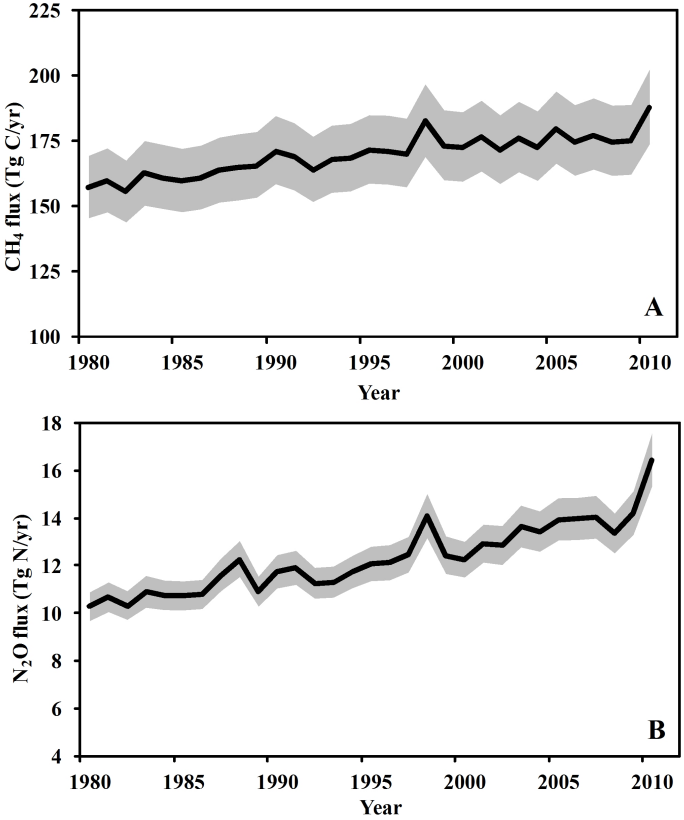

Fig. 3. Interannual variations in CH_4 (**A**, $Tg\,C\,yr^{-1}$) and N_2O (**B**, $Tg\,N\,yr^{-1}$) fluxes in the global terrestrial ecosystems during 1981–2010. Note: the gray area is the estimated uncertainty range with a mean ±95 % confidence interval.

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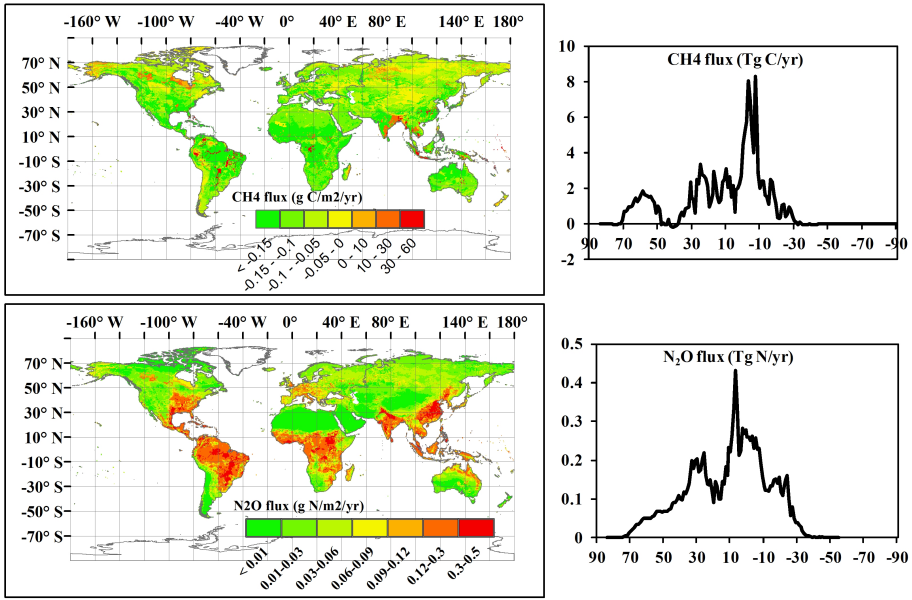

Fig. 4. DLEM-simulated mean global distributions of CH_4 (top, $gCm^{-2}yr^{-1}$) and N_2O (bottom, $m^{-2}yr^{-1}$) fluxes during 1981–2010. Note: The latitudinal (per degree latitude) distribution ($TgCyr^{-1}$ and $TgNyr^{-1}$ for CH_4 and N_2O, respectively) patterns are shown at the right.

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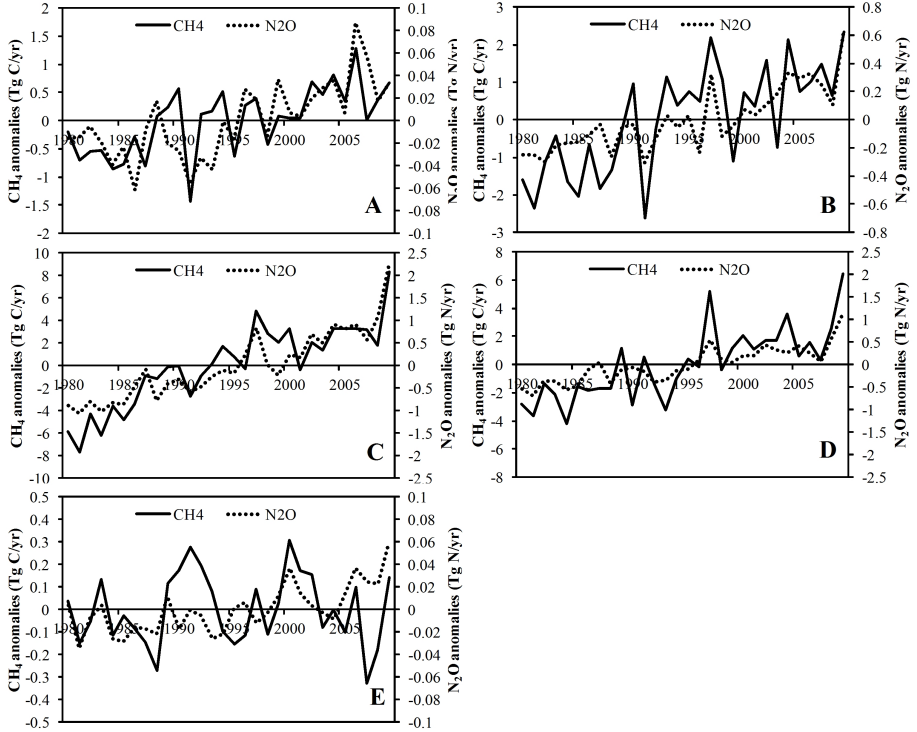

Fig. 5. Interannual variations of CH_4 ($TgCyr^{-1}$) and N_2O ($TgNyr^{-1}$) anomalies (relative to the mean value for the period 1981–2010) along a latitudinal gradient during 1981–2010. **(A)** The northern polar zone (60–90° N); **(B)** the northern temperate zone (30–60° N); **(C)** the northern tropical zone (0–30° N); **(D)** the southern tropical zone (0–30° S); **(E)** the southern temperate zone (30–60° S).

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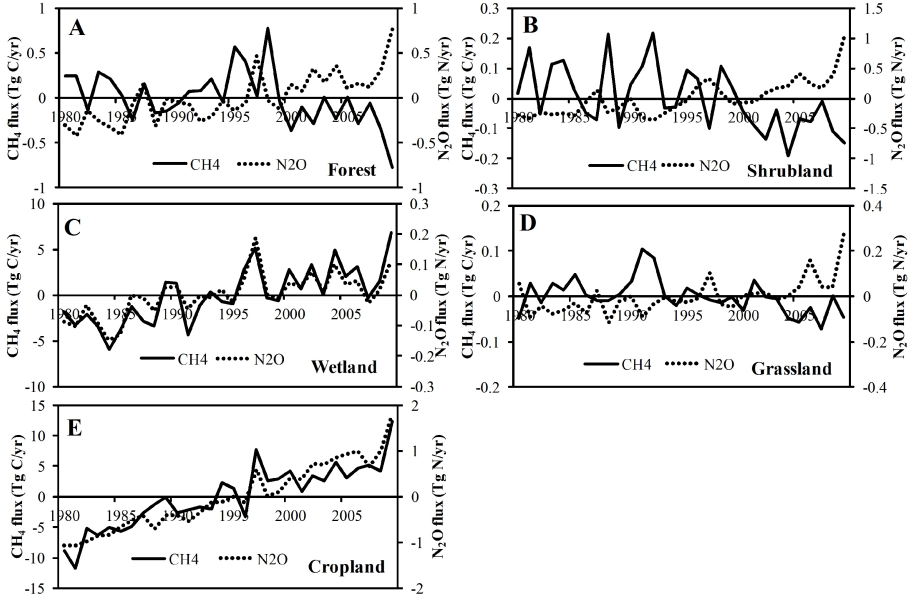

Fig. 6. Interannual variations of CH_4 ($Tg\,C\,yr^{-1}$) and N_2O ($Tg\,N\,yr^{-1}$) anomalies (relative to the mean value for the period 1981–2010) for different biome types during 1981–2010. **(A)** Forest; **(B)** wetland; **(C)** shrubland; **(D)** grassland; **(E)** cropland.

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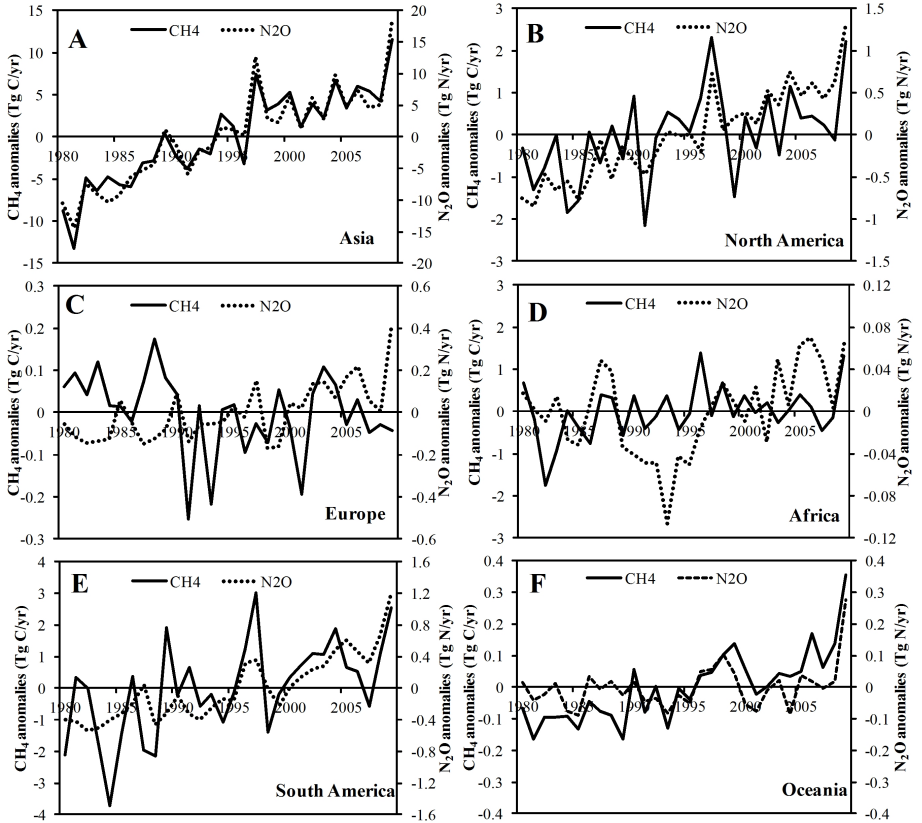

Fig. 7. Interannual variations in CH_4 ($Tg\,C\,yr^{-1}$) and N_2O ($Tg\,N\,yr^{-1}$) anomalies (relative to the mean value for the period 1981–2010) for different continents during 1981–2010.

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