Global methan and nitrous oxide emissions from terrestrial ecosystems due to multiple environmental changes

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Abstract. Greenhouse gas (GHG)-induced climate change is among the most pressing sustainability challenges facing humanity today, posing serious risks for ecosystem health. Methane (CH4) and nitrous oxide (N2O) are the two most important GHGs after carbon dioxide (CO2), but their regional and global budgets are not well known. In this study, we applied a process-based coupled biogeochemical model to concurrently estimate the magnitude and spatial and temporal patterns of CH4 and N2O fluxes as driven by multiple environmental changes, including climate variability, rising atmospheric CO2, increasing nitrogen deposition, tropospheric ozone pollution, land use change, and nitrogen fertilizer use. The estimated CH4 and N2O emissions from global land ecosystems during 1981–2010 were 144.39 ± 12.90 Tg C/yr (mean ± 2 SE; 1 Tg = 1012 g) and 12.52 ± 0.74 Tg N/yr, respectively. Our simulations indicated a significant (P < 0.01) annually increasing trend for CH4 (0.43 ± 0.06 Tg C/yr) and N2O (0.14 ± 0.02 Tg N/yr) in the study period. CH4 and N2O emissions increased significantly in most climatic zones and continents, especially in the tropical regions and Asia. The most rapid increase in CH4 emission was found in natural wetlands and rice fields due to increased rice cultivation area and climate warming. N2O emission increased substantially in all the biome types and the largest increase occurred in upland crops due to increasing air temperature and nitrogen fertilizer use. Clearly, the three major GHGs (CH4, N2O, and CO2) should be simultaneously considered when evaluating if a policy is effective to mitigate climate change.

Key words: coupled biogeochemical cycles; global warming potential; greenhouse gas; methane; nitrous oxide; terrestrial ecosystem.

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Introduction

As two important greenhouse gases (GHGs) contributing to climate warming, methane (CH4) and nitrous oxide (N2O) are receiving more and more attention (Montzka et al. 2011, Tian et al. 2012b, Kirschke et al. 2013, Saikawa et al. 2013a). Methane, with a relative global warming potential 25 times that of CO2 at a 100-yr time horizon, has increased by more than 100% since 1800 and contributes approximately 20% to the global radiative forcing (Forster et al. 2007). The dominant sources of CH4 are natural wetlands, anthropogenic activities, and biomass burning (Dlugokencky et al. 2009, Ito and Inatomi 2012), while upland soils are the major CH4 sink. Nitrous oxide has a relative global warming potential 298 times that of CO2 at a 100-yr time horizon, and contributes approximately 7% to the radiative forcing (Forster et al. 2007). Atmospheric N2O has increased by 18% compared to the preindustrial level, with a linear increasing rate of 0.26% per year during the recent few decades (Forster et al. 2007). The observed increase in atmospheric N2O 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 N₂O in soils involves both biotic and abiotic processes. Comparing to global CH₄ emission estimate, global N₂O emission remains largely uncertain, ranging from 6.7 to 36.6 Tg N/yr (IPCC 2001).

Many environmental factors influence the production and consumption of CH₄ and N₂O (e.g., Hutchin et al. 1995, Conrad 1996, Huang et al. 2010, Toet et al. 2011). As summarized by Dijkstra et al. (2012), global warming might increase net CH₄ uptake in upland ecosystems due to stimulation of microbial CH₄ oxidation and higher CH₄ diffusivity with lower soil moisture. Global warming results in increased N₂O emissions in most land ecosystems due to stimulations of nitrifiers and denitrifiers activity and nitrogen supply through mineralization; however, global warming may also reduce N₂O emissions through soil drying and stimulation of plant growth and nitrogen uptake (Dijkstra et al. 2012, Luo et al. 2013). Elevated atmospheric CO₂ might either increase or decrease CH₄ uptake in the upland ecosystems, but increase CH₄ emissions in water-saturated ecosystems through increased photosynthesis and carbon input to the soil, which stimulate the methanogenic bacteria growth (Dacey et al. 1994, Dijkstra et al. 2012). Under the condition of high nitrogen supply, elevated atmospheric CO₂ could significantly increase N₂O emission due to the increase of soil moisture and soil labile carbon (Ineson et al. 1998, van Groenigen et al. 2011, Dijkstra et al. 2012), while the increasing effect could be small or even negative in nitrogen-limited ecosystems since increased plant growth may result in less nitrogen availability for nitrifiers and denitrifiers (Mosier et al. 2002). Tropospheric ozone (O₃) pollution could cause losses of photosynthesis, and thus reduce CH₄ emissions (Toet et al. 2011), while either stimulate or reduce N₂O emissions by influencing the litter mass and quality (Kanerva et al. 2008). Nitrogen fertilization could dramatically decrease CH₄ consumption in grassland and forest (Steudler et al. 1989, Mosier et al. 1991) and either decrease or increase CH₄ emissions in rice fields, depending on nitrogen fertilizer amount (Banger et al. 2012), while increasing N₂O emissions (Mosier et al. 1998, Del Grosso et al. 2006). In addition, interactions among multiple factors also play an important role. For example, nitrogen deposition and elevated atmospheric CO₂ were reported to interactively reduce CH₄ emission from wetland and increase CH₄ uptake in upland soil (Pancotto et al. 2010); and another study concluded that temperature and elevated atmospheric CO₂ interactively changed seasonal variation of CH₄ emissions (Blankinship et al. 2010). Atmospheric CO₂ and nitrogen deposition can interactively increase soil available nitrogen and labile carbon, thus greatly increase soil N₂O emissions (Dijkstra et al. 2012).

It has not yet been investigated well about how these two gases have been affected by multiple environmental changes. The specific objectives of this study were to (1) provide a new and simultaneous estimate of global CH₄ and N₂O budgets; (2) explore interannual and decadal variations of global CH₄ and N₂O as influenced by multiple environmental factors, including climate, atmospheric CO₂, tropospheric O₃ pollution, nitrogen deposition, land use and land cover, and nitrogen fertilizer use; and (3) compare the regional, biome-level and latitudinal differences in the magnitudes and variations of CH₄ and N₂O fluxes.

**Methodology**

**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° latitude and longitude during 1901–2010. These data include daily O₃ AOT40 index (an index of the accumulated hourly ozone concentrations above a threshold of 40 ppb-h), 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 from the CRUNCEP data set (Wei et al. 2013). Monthly O₃ AOT40 data were derived from the monthly AOT40 data developed by Felzer et al. (2005), and further interpolated to daily data (Ren et al. 2007). EDGAR-HYDE 1.3 nitrogen emission data at 1° × 1° (Van Aardenne et al. 2001) were used to interpolate the three time-period nitrogen deposition data (1860, 1993, 2050; data available online)¹ into an annual nitrogen deposition data set during 1901–2010 (Wei et al. 2013). Land use data were from the History Database of the Global Environment (HYDE 3.1; Goldewijk et al. 2010). 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 (available online).² Soil property data, including soil texture, pH, and bulk density, were from Global Soil Data Task (available online).³ The annual atmospheric CO₂ 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) (available online).⁴ The potential vegetation (i.e., natural vegetation) map was

¹ http://www.esdis.orl.gov/cgi-bin/dsviewer.pl?ds_id=830
² http://faostat.fao.org/Site/677/Default.aspx#ancor
³ http://daac.ornl.gov/SOILS/guides/igbp.html
⁴ http://www.esrl.noaa.gov/gmd/ccgg/trends/global.html
developed from different data sources, including global land-cover data derived from Landsat imageries (De Fries et al. 1998), National Land Cover Dataset 2001 (available online),5 and a global database of lakes, reservoirs, and wetland (Lehner and Döll 2004). The contemporary global vegetation map in 2000 is shown in Fig. 1. Due to the limited data availability, the land use and land cover pattern, O₃ (AOT40), and nitrogen fertilizer use rate after 2005 were assumed to be the same as the level in 2005.

Model description

The Dynamic Land Ecosystem Model (DLEM) is a process-based coupled biogeochemical model for simulating 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 steps from daily to monthly to yearly (e.g., Tian et al. 2010, Xu et al. 2010, 2012, Ren et al. 2011, Lu et al. 2012, Zhang et al. 2012, Chen et al. 2013). One of the important features in the DLEM is to simultaneously estimate the responses of land–atmosphere exchange of CO₂, CH₄, and N₂O to multiple environmental changes in climate, atmospheric CO₂ concentration, tropospheric O₃ pollution, nitrogen deposition, land use, and land management practices. Each plant functional type defined in the DLEM (Fig. 1) was calibrated against various field data collected from individual sites and observational networks such as the Chinese Ecological Research Network (CERN), U.S. Long-Term Ecological Research (LTER) network, AmeriFlux network, and other independent research sites. Here we briefly described the CH₄ and N₂O module in the DLEM. More detailed information can be found in Tian et al. (2010, 2011a, b, 2012a) and Xu et al. (2010, 2012).

In the DLEM, CH₄ production, consumption, and transport processes are considered to estimate land–atmosphere gas exchange. Dissolved organic carbon (DOC) is the only CH₄ production substrate considered in the DLEM. DOC comes from gross primary productivity (GPP) and decomposition byproducts from soil organic matter and litter, which are indirectly controlled by environmental factors including soil pH, temperature, and moisture content. CH₄ oxidation is determined by CH₄ concentrations in the air, pore space of soil, and soil moisture, pH, and temperature. We consider three pathways for CH₄ transport from soil to the atmosphere (i.e., ebullition, diffusion, and plant-mediated transport). It is assumed that CH₄-related biogeochemical processes only occur in the top 50 cm of the soil profile. Overall, the net CH₄ exchange between the atmosphere and soil is calculated by the following equation:

\[
F_{\text{CH}_4} = F_P + F_D + F_E - F_{\text{air, oxid}} - F_{\text{trans, oxid}} - F_{\text{soil, oxid}}
\]

where \(F_{\text{CH}_4}\) is the flux of CH₄ between soil and the atmosphere (g C·m⁻²·d⁻¹); \(F_P\) is plant-mediated transport from soil pore water to the atmosphere (g C·m⁻²·d⁻¹); \(F_D\) is the diffusive flux of CH₄ from soil to the atmosphere (g C·m⁻²·d⁻¹); \(F_E\) is the ebullitive CH₄ emission to the atmosphere; \(F_{\text{air, oxid}}\) is atmospheric CH₄ oxidation rate (g C·m⁻²·d⁻¹); \(F_{\text{trans, oxid}}\) is the CH₄ oxidation during plant-mediated transport (g C·m⁻²·d⁻¹); \(F_{\text{soil, oxid}}\) is the CH₄ oxidation rate in soil pore water.

In the 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. (2000) to separate N₂O from other gases (mainly NO and N₂). The equation for calculating nitrification, denitrification, and N₂O fluxes is

\[
F_{\text{N}_2\text{O}} = 0.001 \times N_{\text{nit}} + N_{\text{denit}} \times \left( \frac{10^{(\frac{m}{\text{air, oxid}} \times 0.026 - 1.66)}}{1 + 10^{(\frac{m}{\text{air, oxid}} \times 0.028 - 1.66)}} \right)
\]

where \(F_{\text{N}_2\text{O}}\) is the N₂O flux from soil to the atmosphere (g N·m⁻³·d⁻¹), which can be converted to units of g N·m⁻³·d⁻¹ by multiplying the soil depth (default 0.5 m); \(N_{\text{nit}}\) and \(N_{\text{denit}}\) are the nitrification and denitrification rates (g N·m⁻³·d⁻¹), respectively; 0.001 is the proportion of nitrification product released as gaseous nitrogen (Lin et al. 2000); \(\Psi\) is the soil porosity; and vwc is the soil volumetric water content (m³/m³).

In the DLEM, multiple environmental factors indirectly or directly influence CH₄ and N₂O fluxes. For example, the effect of O₃ pollution on CH₄ and N₂O fluxes is through its influences on GPP and litter decomposition; the effects of air temperature are through the influences of GPP, litter decomposition, nitrogen mineralization, soil temperature, and other factors.

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 the land use and land cover map for 1900, long-term mean climate during 1901–1930, and the levels of nitrogen deposition, O₃ pollution, and atmospheric CO₂ 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/m², 0.1 g N/m², and 0.1 mm for carbon, nitrogen, and water pools, respectively). After the model reached equilibrium state, a spin-up run was implemented for 900 years using detrended 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$_2$O fluxes. Model results from 1981 to 2010 were used to analyze the spatial and temporal patterns of global CH$_4$ and N$_2$O fluxes in this study.

Two simulation experiments were designed to achieve the study goals. In the baseline experiment, transient input data during 1901–1980 for all environmental factors (i.e., climate, land use, nitrogen deposition, O$_3$ pollution, atmospheric CO$_2$ concentration, and nitrogen fertilizer use) were fed into the DLEM and then kept constant at the level of 1980 thereafter. In the combined experiment, all environmental factors changed with time during 1901–2010. The effects of multiple environmental changes on CH$_4$ and N$_2$O fluxes were the difference between these two experiments. With this experiment design, the legacy effects of environmental factors before 1981 were excluded.

**Estimate of uncertainty**

In this study, we estimated uncertainty in simulated fluxes of CH$_4$ and N$_2$O resulting from variations in the most sensitive parameters (see Xu et al. 2010 and Tian et al. 2011b for details). First, we conducted a sensitivity analysis to identify the most sensitive parameters that affect terrestrial CH$_4$ and N$_2$O fluxes. We selected the top five most sensitive parameters (i.e., for CH$_4$, maximum CH$_4$ production and consumption rates, half-saturation coefficients of CH$_4$ oxidation and production, and DOC allocation rate from litter decomposition; for N$_2$O, maximum nitrification and denitrification rates, half-saturation coefficients of soil ammonium and nitrates, and maximum nitrogen uptake rate; see Tian et al. 2010 for more details) to conduct the uncertainty simulation. Second, we assumed that each parameter follows a normal distribution. Combined with the prior knowledge of the parameter range, we used an improved Latin Hypercube Sampling (LHS) approach to randomly select an ensemble of 100 sets of parameter values to conduct model simulations. Finally, the outliers for the simulation results were excluded before analyzing the uncertainty range. The 95% confidence intervals were calculated and reported.

**Model evaluation**

Our previous studies had evaluated DLEM-simulated CH$_4$ and N$_2$O fluxes against field observations, regional inventory, and other modeling results (e.g., Tian et al. 2010, 2011a, 2012a, c, 2014, Xu et al. 2010, 2012, Ren et al. 2011, Wang et al. 2012, Melton et al. 2013). These evaluations indicated that DLEM is able to capture the daily, annual, and spatial variations in the observed CH$_4$ and N$_2$O fluxes. In this study, we further evaluated the daily patterns of DLEM-simulated CH$_4$ and N$_2$O against the observational data at eight sites for CH$_4$ and three sites for N$_2$O fluxes (Fig. 2). The fitted lines between the simulated and observed values were close to the 1:1 line and had a high $R^2$ (0.63 for
CH₄ and 0.88 for N₂O; \( P < 0.01 \), showing that DLEM can effectively capture the daily patterns of CH₄ and N₂O at the site level. In addition, at the global scale, our estimates of CH₄ fluxes in wetlands and upland soils were compared with the inverse modeling results from CarbonTracker (Bruhwiler et al. 2014), and the simulated N₂O emissions from agricultural and natural soils were compared with the inversing modeling results from MOZART v4 (Saikawa et al. 2013a) (Fig. 3). The year-by-year comparison indicated that the DLEM
simulation results were quite close to top-down estimates in magnitude, with a small RMSE (square root of the mean square error of predictions; $\text{RMSE} = \sqrt{\frac{1}{n} \sum (x_i - y_i)^2}$) of 10.61 Tg CH$_4$-C/yr and 2.08 Tg N$_2$O-N/yr, respectively, which is pretty small relative to the observed CH$_4$ and N$_2$O emissions. However, the temporal trend of DLEM simulation results was not consistent with CarbonTracker ($R^2$, 0.2; $P$, 0.05). DLEM results showed a significant ($P$, 0.01) increasing trend while CarbonTracker results did not have an obvious trend. Both results from DLEM and MOZART v4 showed a significant increasing trend for cropland N$_2$O emissions ($R^2$ > 0.66; $P$ < 0.01) and no obvious trend for natural ecosystems. The performance of DLEM in simulating the effects of individual and combined environmental factors on carbon, nitrogen and GHGs has been well evaluated in our previous studies (e.g., Ren et al. 2007, Zhang et al. 2007, Xu et al. 2010, Tian et al. 2011b, 2012c, Lu and Tian 2013).

Statistical analysis
We used multiple linear regression analyses to explore the long-term changing trends of input data and CH$_4$ and N$_2$O fluxes, and the Pearson correlation to evaluate the correlations between input data and CH$_4$ and N$_2$O fluxes. All the statistical analyses were conducted using R 3.0.3 (R Development Core Team 2014).

Results
Global environmental change over past three decades

From 1981 to 2010, air temperature and precipitation significantly increased by 0.024°C/yr and 1.37 mm/yr, respectively (Table 1). 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-year (1961–1990) average, precipitation increased in most areas during the period 1981–2010. The largest increase occurred in South America, whereas annual precipitation decreased in southern Africa, northern India, and southeastern Australia (Fig. 4A). The largest increase in air temperature has occurred in the northern high-latitude region and a slight decrease in South America and Oceania (Fig. 4B). The highest nitrogen deposition rates were shown in east and south Asia (Fig. 4C), while the highest nitrogen fertilizer use rates were located in eastern China (Fig. 4D). These regions experienced fast growth in population, urbanization, and industrialization during the past three decades.

At continental scales, air temperature increased in all continents except for Oceania, with the most rapid increase in Europe and Asia (Table 1). Precipitation significantly increased for Africa and South America, while no significant trends were found for other continents. Nitrogen deposition and O3 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. Nitrogen fertilizer use significantly increased in all the continents, with a decreasing trend in Europe and North America (i.e., 5.39 ± 0.48 Pg CO2 eq/yr and 5.21 ± 0.31 Pg CO2 eq/yr, respectively) could completely offset the terrestrial CO2 sink (7.55 ± 2.93 Pg CO2 eq/yr estimated by multiple process-based models; Le Quéré et al. 2014) over the recent three decades. It suggested that global land ecosystem acted as a positive contributor to climate warming during the past decades.

Global budget of terrestrial CH4 and N2O fluxes

The estimated mean terrestrial CH4 emission during 1981–2010 was 144.39 ± 12.90 Tg C/yr (mean ± 2 SE), ranging from 136.45 ± 11.78 Tg C/yr in 1982 to 156.45 ± 14.19 Tg C/yr in 2010. Upland soil uptake of CH4 was −17.10 ± 0.17 Tg C/yr. Terrestrial N2O emission averaged 12.52 ± 0.74 Tg N/yr, ranging from 10.53 ± 0.59 Tg N/yr in 1982 to 16.65 ± 1.10 Tg N/yr in 2010 (Fig. 5). Our analysis showed a significant (P < 0.01) annually increasing trend of 0.43 ± 0.06 Tg C/yr for CH4 and 0.14 ± 0.02 Tg N/yr for N2O over the study period. The interannual variations in CH4 and N2O fluxes were roughly consistent (R2 = 0.87; P < 0.01), indicating a close relationship between CH4 and N2O fluxes. Significantly positive correlations between air temperature and the model-estimated CH4 (R2 = 0.73, P < 0.01) and N2O emissions (R2 = 0.71, P < 0.01) were observed, implying a strong positive feedback between climate warming and CH4/N2O emissions.

In terms of global warming potential (100-yr horizon), the DLEM-estimated global CH4 and N2O emissions (i.e., 5.39 ± 0.48 Pg CO2 eq/yr and 5.21 ± 0.31 Pg CO2 eq/yr, respectively) could completely offset the terrestrial CO2 sink (7.55 ± 2.93 Pg CO2 eq/yr estimated by multiple process-based models; Le Quéré et al. 2014) over the recent three decades. It suggested that global land ecosystem acted as a positive contributor to climate warming during the past decades.

Spatial distribution of terrestrial CH4 and N2O fluxes

As expected from the distribution of wetlands and rice paddy fields, south and southeast Asia, tropical wetlands and river flood plains (e.g., Amazonia and the Pantanal) were dominant hot-spots of CH4 emission with values as high as 30 g C m−2 yr−1 (Fig. 6). In contrast, the northern high-latitude regions (e.g., Alaska, northern Canada, West Siberia, and northern Eurasia) that have large wetland area, were less substantial CH4 emission sources (~10 g C m−2 yr−1). Larger CH4 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 CH4

| Environmental factors | Asia       | North America | Europe    | Africa      | South America | Oceania     | Global    |
|-----------------------|------------|---------------|-----------|-------------|---------------|------------|-----------|
| Air temperature (°C/yr) | 0.035**    | 0.030**       | 0.039**   | 0.032**     | 0.016**       | 0.0079     | 0.024**   |
| Precipitation (mm/yr)  | 0.78       | 0.51          | 0.56      | 2.26**      | 3.50**        | 2.01       | 1.37*     |
| Shortwave radiation (W m−2 yr−1) | 0.002     | −0.017        | 0.002     | 0.014       | 0.11**        | 0.15*      | 0.041**   |
| N deposition (mg N m−2 yr−1) | 8.70**   | 2.20**        | 4.70**    | 5.20**      | 7.20**        | 5.00**     | 5.60**    |
| N fertilizer use (g N m−2 yr−1) | 0.27**   | 0.05*         | −0.13*    | 0.01*       | 0.11**        | 0.13**     | 0.074**   |
| O3 pollution (ppb-h/month) | 58.6**  | 40.71**       | 28.86**   | 47.11**     | 7.97**        | 0.05*      | 32.87**   |
| Cropland (104 km2/yr)  | 2.52**     | −0.091**      | −1.45**   | 1.96**      | 0.72**        | 0.17**     | 4.01**    |

* P < 0.05; ** P < 0.01.
Fig. 4. Spatial distribution of environmental factors in the terrestrial ecosystems. (A) Precipitation anomaly ([1981–2010 mean] – [1961–1990 mean]); (B) air temperature anomaly; (C) nitrogen deposition rate in 2010; (D) cropland distribution and nitrogen fertilizer amount in 2005; (E) cumulative tropospheric O₃ concentration above 40 ppb-h (AOT40: ppm-h) in July 2005.
N₂O 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, N₂O emissions decreased with air temperature from the low- to high-latitude regions, indicating air temperature was one of the most important factors controlling N₂O emissions. To more clearly identify the spatial distribution patterns of CH₄ and N₂O fluxes, we further divided the globe into different latitudinal belts, biome types, and continents.

**Terrestrial CH₄ and N₂O fluxes and their temporal trends along a latitudinal gradient**

Along the latitudinal gradient, CH₄ emissions peaked (8.06 Tg C/yr) at the latitudinal zones of 2°–3° S and 6°–7° S (Fig. 6), primarily due to large wetland area in Amazon river basin. Another smaller peak for CH₄ emissions displayed at the northern high-latitude region (around 60° N). N₂O emissions peaked at the latitude zone of 6°–7° N (0.43 Tg N/yr), 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 climatic zones were net atmospheric sources of CH₄ and N₂O emissions (Table 3). The largest CH₄ emissions occurred in the southern tropical zone, followed by the northern tropical zone, and the least in the southern temperate zone. The tropical zone also accounted for about 80% of the total CH₄ emissions.

Our simulation experiments indicated that trends in the CH₄ and N₂O fluxes varied significantly among various climatic zones (Fig. 7). The most rapid increase in CH₄ emission was observed in the northern and southern tropical zones (0.16 Tg C/yr), followed by the northern temperate zone (0.07 Tg C/yr) and northern polar zone (0.046 Tg C/yr) while no significant changing trend was found for the southern temperate zone. Similarly, N₂O emissions increased more rapidly in the northern tropical and southern tropical zones (0.039–0.078 Tg N/yr) than other zones. No significant changing trend in N₂O emission was observed in the southern temperate zone; however, larger interannual variations were found in this zone compared to other zones. The highest CH₄ and N₂O emissions occurred in 2010 for all the climate zones except for the northern polar and southern temperate zones.

**Terrestrial CH₄ and N₂O fluxes and their temporal trends for different biome types**

We further compared the magnitude of CH₄ and N₂O fluxes and their change trends during 1981–2010 for

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**Table 2. 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) | 0.039**       | 0.034**            | 0.033**          | 0.016**          | 0.007             |
| Precipitation (mm/yr)   | 0.42*          | 0.066              | 2.68**           | 3.42**           | –0.68             |
| N deposition (mg N m⁻² yr⁻¹) | 0.26**        | 5.07**             | 8.57**           | 2.75**           | 0.72**            |
| N fertilizer use (g N m⁻² yr⁻¹) | –0.082**     | 0.068**            | 0.10**           | 0.040**          | 0.02**            |
| O₃ pollution (ppb-h/month) | –1.67        | 32.80*             | 50.78**          | 3.89*            | 0.0008            |
| Cropland (10⁴ km²/yr)   | –0.01*         | –0.42*             | 2.72**           | 0.83**           | 0.70**            |

* P < 0.05; ** P < 0.01.
different biome types (Table 4; Fig. 8). The largest CH$_4$ emissions were from natural wetland (130.74 Tg C/yr), followed by cropland (30.82 Tg C/yr). The highest CH$_4$ uptake occurred in forest (–6.43 Tg C/yr) and shrubland (–4.38 Tg C/yr). Multiple environmental changes had increased wetland CH$_4$ emissions from 129.01 Tg C/yr in 1981 to 137.67 Tg C/yr in 2010. From 1981 to 2010, forest and shrubland showed a significant ($P < 0.05$) increasing trend in CH$_4$ uptake, with an annual rate of 0.014 Tg C/yr and 0.0061 Tg C/yr, respectively (Fig. 8). Wetlands and rice field showed a more significant ($P < 0.01$) increasing trend in CH$_4$ emission, with an annual rate of 0.25 Tg C/yr and 0.54 Tg C/yr, respectively, while no significant change trend was found for grassland. Interestingly, rice field expansion (15% as shown in Table 1) contributed to only 12% of the increased cropland CH$_4$ emissions and the remaining 88% increase resulted from other environmental changes including climate, atmospheric composition, and land management practices.

### Table 3. Average terrestrial CH$_4$ and N$_2$O fluxes and their change rates (%) for different climate zones during 1981–2010.

| Flux and time period   | Northern polar zone | Northern temperate zone | Northern tropical zone | Southern tropical zone | Southern temperate zone | Global     |
|------------------------|---------------------|-------------------------|------------------------|------------------------|------------------------|------------|
| CH$_4$ (Tg C/yr)       |                     |                         |                         |                        |                        |            |
| 1980s                  | 9.89 ± 0.67         | 18.44 ± 1.46            | 39.08 ± 3.44           | 72.55 ± 4.82           | 0.16 ± 0.03            | 140.12 ± 12.20 |
| 2000s                  | 10.80 ± 0.73        | 19.76 ± 1.62            | 41.65 ± 3.94           | 75.27 ± 5.10           | 0.23 ± 0.04            | 147.71 ± 13.46 |
| 1981–2010              | 10.30 ± 0.70        | 19.20 ± 1.55            | 40.71 ± 3.73           | 73.80 ± 4.96           | 0.21 ± 0.04            | 144.39 ± 12.90 |
| Change rate (%)        | 9.22                | 7.14                    | 6.56                   | 3.75                   | 47.53                  | 5.39       |
| N$_2$O (Tg N/yr)       |                     |                         |                         |                        |                        |            |
| 1980s                  | 0.34 ± 0.02         | 2.47 ± 0.13             | 4.43 ± 0.23            | 3.93 ± 0.21            | 0.14 ± 0.01            | 11.30 ± 0.63 |
| 2000s                  | 0.38 ± 0.02         | 2.88 ± 0.15             | 5.96 ± 0.32            | 4.68 ± 0.27            | 0.17 ± 0.01            | 14.08 ± 0.87 |
| 1981–2010              | 0.35 ± 0.02         | 2.63 ± 0.14             | 5.11 ± 0.28            | 4.27 ± 0.24            | 0.15 ± 0.01            | 12.52 ± 0.74 |
| Change rate (%)        | 14.37               | 16.54                   | 34.67                  | 19.03                  | 23.96                  | 24.53      |

Notes: Mean annual fluxes are for the periods 1981–1990, 2001–2010, and 1981–2010. Values for fluxes are means ± 2 SE.
The largest emissions of N₂O were from forest (4.28 Tg N/yr), followed by cropland (3.36 Tg N/yr), and the least was from grassland (0.82 Tg N/yr). Most of the forest N₂O emissions came from tropical region. Compared to forests in other climatic zones, tropical forests had larger area and higher nitrogen transformation rate. The N₂O emissions significantly increased for all the biome types from 1981 to 2010 (Fig. 8). N₂O 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 N₂O emission (annual increase rate of 0.08 Tg N/yr; \(R^2 = 0.93\); \(p < 0.01\)), which was primarily due to rapid cropland expansion and increasing nitrogen fertilizer uses (Table 1). Cropland area has expanded by 7.8% (~11 million ha) since 1981, and nitrogen fertilizer uses have increased by 66% (~33 Tg N/yr). Increased nitrogen deposition and air temperature were the major causes of increased N₂O emissions in the non-managed biomes.

![Fig. 7. Interannual variations of CH₄ (Tg C/yr) and N₂O (Tg N/yr) 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).](image)

| Flux         | Forest        | Shrubland    | Wetland      | Grassland    | Cropland      |
|--------------|---------------|--------------|--------------|--------------|---------------|
| CH₄ (Tg C/yr)| −6.43 ± 0.47  | −4.38 ± 0.28 | 130.74 ± 9.94| −1.90 ± 0.13 | 30.82 ± 4.02  |
| N₂O (Tg N/yr)| 4.28 ± 0.36   | 2.82 ± 0.13  | 0.97 ± 0.07  | 0.82 ± 0.05  | 3.36 ± 0.17   |

*Note: Values for fluxes are means ± 2 SE.*
Temporal trends in terrestrial CH₄ and N₂O fluxes for different continents

At the continental scale, significant increasing trends \((P < 0.01)\) in CH₄ emissions were found for all continents except Africa and Europe (Fig. 9). Asia showed the highest CH₄ emission and the most rapid increase \((0.56 \text{Tg C/yr}, P < 0.01)\) while CH₄ emissions from Europe decreased since the early 1990s. Significant increasing trends in N₂O emissions were found in all continents except Europe and Oceania. Similar to CH₄, N₂O emission increases in Asia \((0.056 \text{Tg N/yr}, P < 0.01)\) was caused by larger increases in nitrogen fertilizer amounts and nitrogen deposition rates compared to other continents (Table 1). Overall, Asia and South America acted as the largest CH₄ and N₂O 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 CH₄ and N₂O emissions were very similar \((R^2 = 0.89; P < 0.01)\) in Asia, implying a close linkage between these two gases across this continent.

Discussion

Comparisons with other estimation of global CH₄ and N₂O budgets

The simultaneous estimation of global CH₄ and N₂O fluxes shown in this study were in line with previous estimates. The DLEM-estimated global CH₄ flux was 139.57–153.69 Tg C/yr for the 1990s (Table 5), which fell in the range of 86–200 Tg C/yr 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 CH₄ by 126.47–135.92 Tg C/yr in the 1990s, which is higher than 92 Tg C/yr 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; Melton et al. 2013). Our study reported that 128.19 Tg C/yr was released from global natural wetlands in the 1980s, slightly higher than a previous estimate of 110 Tg C/yr (Matthews and Fung 1987). Our estimated CH4 emission from rice fields (29.30–35.17 Tg C/yr) was similar to most previous estimates. DLEM-estimated upland CH4 uptake during 1996–2005 ($C_0$ 16.13 to $C_0$ 17.76 Tg C/yr) was slightly lower than the previous estimates ($C_0$ 19.5 to $C_0$ 25.0 Tg C/yr).

This study reported global N2O emission in a range of 10.49–14.30 Tg N/yr during 1981–2000 (Table 6), which was close to an empirical estimate of 13.31 Tg N/yr (Xu et al. 2008). The estimated N2O flux during 1990–1994 (11.51–12.15 Tg N/yr) was slightly higher than a process-based modeling result (11.33 Tg N/yr; Liu 1996) and an inverse modeling result (9.5 Tg N/yr; 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 N2O flux slightly higher than the estimates from FAO/IFA (2001), Davidson (2009) and Bouwman et al. (2002), which did not consider the impacts of nitrogen deposition and fixation. DLEM-estimated N2O emissions from cropland were close to most of previous estimates (e.g., Bouwman et al. 2002, Davidson 2009, U.S. EPA 2010). Our estimates for natural land N2O emissions were comparable to those from Xu et al. (2008), Bowden (1986), Saikawa et al. (2013a, b), and U.S. EPA (2010), and significantly higher than the estimate from the statistical extrapolation (Zhuang et al. 2012). DLEM-estimated N2O emission from global grassland was consistent with that from Zhang et al. (2010), but

**Fig. 9.** Interannual variations in CH4 (Tg C/yr) and N2O (Tg N/yr) anomalies (relative to the mean value for the period 1981–2010) for different continents during 1981–2010.
Table 5. Comparison of global terrestrial CH$_4$ fluxes estimated from multiple sources.

| Location and time period | CH$_4$ flux (Tg C/yr) | Other studies | Sources |
|--------------------------|-----------------------|---------------|---------|
| Global                   |                       |               |         |
| 1990s                    | 139.57 to 153.69      | 86 to 195     | Fung et al. (1991), Houweling et al. (1999) |
| Global natural soils     |                       |               |         |
| 1993–2004                | 129.70 to 135.92      | 106 to 198    | Melton et al. (2013) |
| 1990s                    | 126.47 to 135.92      | 69.0 to 202.5 | Cao et al. (1998), Bousquet et al. (2011), Riley et al. (2011), Liu (1996) |
| 2000s                    | 130.90 to 137.67      | 78.8 to 208.5 | U.S. EPA (2011) |
| Global upland            |                       |               |         |
| 1996–2005                | −16.13 to −17.76      | −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                    | 29.30 to 35.17        | 19.2 to 84.0  | Cao et al. (1998), Chen and Prinn (2005), Patra et al. (2009), Yan et al. (2009), Ito and Inatomi (2012) |
| High-latitude wetland (>60° N) | 9.47 to 10.56    | 3 to 27       | Christensen et al. (1996) |
| High-latitude (>60° N)   | 8.89 to 10.89         | 15.8          | Zhuang et al. (2004) |

Table 6. Comparison of global terrestrial N$_2$O fluxes estimated from multiple sources.

| Location and time period | N$_2$O flux (Tg N/yr) | Other studies | 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$^+$     | 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          |           |
| Global natural soils     |                       |               |           |
| 2000                     | 8.67                  | 1.96–4.56$^+$ | Zhuang et al. (2012) |
| 1990                     | 8.76                  | 6.6 (3.3–9.0)$^*$ | FAO/IFA (2001), U.S. EPA (2010) |
| 2000–2008                | 9.25                  | 9.34          | Xu et al. (2008) |
| Early 1980s              | 7.97–8.48             | 7–16          | Bowden (1986) |
| 1975–2000                | 10.49–14.30           | 7.42–10.6     | Saikawa et al. (2013a) |
| 1995–2008                | 8.67–10.02            | 5.27–8.28     | Saikawa et al. (2013a) |
| Global cropland          |                       |               |           |
| 1990                     | 3.00                  | 2.9#          | FAO/IFA (2001) |
| 1995                     | 3.29                  | 2.8           | Bouwman et al. (2002) |
| 2000                     | 3.50                  | 4.4||         | U.S. EPA (2011) |
| 2000                     | 3.50                  | 2.6–3.5$^{+1}$ | Davidson (2009) |
| 2000–2008                | 4.04                  | 3.97          | Xu et al. (2008) |
| 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) |

† The range in parentheses shows 95% CI.
‡ The emission from ocean (23%) is excluded.
§ Cropland is not included.
¶ Natural wetland emission (~0.97 Tg N/yr as estimated by DLEM) is not included. Range in parentheses shows estimated minimum and maximum values.
# Including emissions from fertilizer use, crops, decomposition of crops, and biomass burning.
|| Including both direct and indirect emissions from fertilizer use.
†† Including fertilizer use, biomass burning, and manure application.
slightly lower than that from the empirical extrapolations by Xu et al. (2008) and Zhuang et al. (2012), which were primarily attributed to the smaller grassland area in our estimate. For global forest, our estimate was higher than Zhuang et al. (2012) but smaller than Xu et al. (2008). We separated forested wetland from forest category and grouped into wetland, while Xu et al. (2008) treated it as a forest type, which could result in larger forest land area and thus higher N$_2$O fluxes in Xu et al. (2008).

In addition to divergent model representation of biogeochemical processes and parameter values, difference in input data might be a major cause of the large discrepancies among these estimates of CH$_4$ and N$_2$O fluxes (Zhu et al. 2011, Ito and Inatomi 2012, Meng et al. 2012, Melton et al. 2013, Saikawa et al. 2013b). For example, more model driving forces (e.g., climate, land use, atmospheric CO$_2$ concentration, nitrogen fertilizer amounts, tropospheric O$_3$ pollution, and nitrogen deposition) were considered in our study, while most other previous work only considered one or a subset of these environmental factors (e.g., Cao et al. 1998, Chen and Prinn 2005, Patra et al. 2009). Uncertainty in wetland area and distribution was another important factor leading to divergent estimates of CH$_4$ flux. Melton et al. (2013) reported that the estimate of global wetland area ranged from $7.1 \times 10^6$ to $26.9 \times 10^6$ km$^2$. Even the inventory data varied among different data sources, ranging from $4.3 \times 10^6$ to $12.9 \times 10^6$ km$^2$. Furthermore, large uncertainty may also result from the various data sets used in different models. For example, CRU, CRUNCEP, and NCEP reanalysis data sets were used by different model simulations (e.g., Bousquet et al. 2011, Ito and Inatomi 2012, Melton et al. 2013), and these climate data sets have large difference at both spatial and temporal scales. Based on a process-based model, Saikawa et al. (2013b) estimated that the global natural soil N$_2$O emission had a maximum difference of 3.5 Tg N/yr among the modeling results based on four different climate data sets.

**Regional difference in CH$_4$ and N$_2$O fluxes**

Based on measurements and extrapolation, Bartlett and Harris (1993) and U.S. 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 the tropical regions, instead, over 58.56% was from the high-latitude region. However, using the same methods with more observations, Fung et al. (1991) updated their early estimate and reported that about 69.57% of CH$_4$ emissions were from the tropical regions. Based on eight process-based ecosystem models, Melton et al. (2013) estimated that about 66.32% of CH$_4$ emissions were from the tropical regions. The inverse modeling approach estimated an even higher emission from the tropical regions (81.01% [Hein et al. 1997]; 83.52% [Wang et al. 2004]; 70.75% [Bousquet et al. 2006]). Combining all these estimates, U.S. EPA (2010) reached a consensus that about 75% of CH$_4$ emissions were from the tropical regions, which is close to our estimate of 79.22% (upland CH$_4$ uptake is not separated). The DLEM-simulated 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$_2$O emissions were from the tropical regions. Based on observations and inverse methods, Prinn et al. (1990) also reported that 52–68% of the N$_2$O emissions were from the tropical regions during 1978–1988. In contrast, using an inverse modeling approach, Hirsch et al. (2006) showed a lower estimate of N$_2$O emissions from 30° S to 90° S (0–4%) and a higher N$_2$O emission from 0° to 30° N (50–64%) during 1998 to 2001.

In Asia, cropland particularly rice fields have expanded dramatically due to fast-growing population, which was the major contributor to the increasing CH$_4$ emission. In addition, nitrogen fertilizer use amount in Asia increased the most rapidly (Table 1) and led to substantial increase in N$_2$O emission. Based on inventory data, Kurokawa et al. (2013) found that CH$_4$ and N$_2$O increased by 32% and 18%, respectively from 2000 to 2008 in Asia. By using inversion modeling, Saikawa et al. (2013a) also reported that N$_2$O emissions from agricultural soil increased by 56% (0.055 Tg/yr) in 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$_2$O emissions as indicated by our previous studies (i.e., Tian et al. 2010, 2012a, Xu et al. 2010, 2012a).

**Temporal trends of CH$_4$ and N$_2$O fluxes in different biomes**

We found that increase in CH$_4$ emissions was primarily due to the effects of multiple environmental changes on wetland and rice fields. Multiple environmental changes had led to an increase in natural wetland CH$_4$ emission from 129.01 Tg C/yr in 1981 to 137.67 Tg C/yr in 2010 (Fig. 8C). Through data synthesis, Kirschke et al. (2013) also found a steady increase in wetland CH$_4$ emission based on the bottom-up estimation approach, with the highest increase of about 15 Tg C/yr from 1985 to 2010. We found that rice field area has increased by $0.28 \times 10^6$ km$^2$ from 1981 to 2010, and CH$_4$ emission from rice fields increased by about 4 Tg C. Similarly, U.S. EPA (2006) predicted rapid increases in CH$_4$ emissions from rice cultivation from 1990 (17.64 Tg C/yr) to 2010 (20.37 Tg C/yr), with a 15.5% increase. 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 the recent decade.
We found that cropland N\textsubscript{2}O 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\textsubscript{2}O emissions, Reay et al. (2012) and U.S. EPA (2011) indicated that N\textsubscript{2}O emissions from agricultural soil increased by about 1 Tg N/yr from 1990 to 2010. We had a similar finding with agricultural N\textsubscript{2}O emission increasing by 1.4 Tg N/yr during the same period (Fig. 8E). By using inversion modeling, Saikawa et al. (2013) also estimated an N\textsubscript{2}O emission increase of 1.27 Tg N/yr from 1990 to 2010. We had a similar finding with agricultural N\textsubscript{2}O emission increasing by 1.4 Tg N/yr during the same period (Fig. 8E). By using inversion modeling, Saikawa et al. (2013a) also estimated an N\textsubscript{2}O emission increase of 1.27 Tg N/yr (from 2.65 Tg N/yr in 1995 to 3.92 Tg N/yr 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\textsubscript{2}O 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 1860–2005. Considering nitrogen fertilizer amount increasing from \(\sim 50 \text{Tg N/yr in 1981 to \sim 84 \text{Tg N/yr in 2005, we can conclude that during 1981–2005, increase of about 0.85 \text{Tg N}_\text{2}O-N/yr was directly derived from the rising nitrogen fertilizer use. The increases in N\textsubscript{2}O emissions for natural biomes might be primarily due to climate warming and nitrogen deposition. Based on field experiments, Dijkstra et al. (2012) and Cantarel et al. (2011) found that climate warming significantly increase N\textsubscript{2}O emissions in grassland. Nitrogen deposition could greatly increase carbon storage in the terrestrial ecosystems and in the meanwhile increasing N\textsubscript{2}O emissions for natural biomes (Liu and Greaver 2009, Lu et al. 2012).

Uncertainties and implications

Some uncertainties need to be considered when interpreting the simulated results. First, uncertainties might be resulted from the simplified representation of mechanisms controlling CH\textsubscript{4} and N\textsubscript{2}O production and consumption. The DLEM runs at a daily time step and may smooth pulses in CH\textsubscript{4} and N\textsubscript{2}O fluxes at a sub-daily scale, which may substantially contribute to annual fluxes (Brumme et al. 1999). Some 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 model representations for this process, no other reliable method has yet been put forward. Additional field or experimental investigations are needed to improve model representation of CH\textsubscript{4} ebullition. Second, parameter uncertainty might lead to estimation biases. For example, it is important to take into account the differences of CH\textsubscript{4} processes 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\textsubscript{4} and N\textsubscript{2}O fluxes have been reported at an order of magnitude difference among different wetland classes (Bartlett and Harris 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\textsubscript{4} fluxes (Ringeval et al. 2010, Wania et al. 2012, Melton et al. 2013). N\textsubscript{2}O emission from pasture management (i.e., irrigation, fertilization, grazing rotation, etc.) may contribute a great portion to the global N\textsubscript{2}O flux (Li et al. 1996, Ambus and Robertson 2006); however, it is not considered in this study.

Our findings have important implications for mitigation strategies. In our previous studies (Tian et al. 2011a, 2012h, Lu et al. 2012), we found that some regions in the world are experiencing excessive nitrogen input. Less nitrogen input will maintain the same food productivity but reduce the risks for higher N\textsubscript{2}O emissions, as well as soil and water nitrogen pollution. To slow down future global warming, policy makers should pay special attention to reducing CH\textsubscript{4} and N\textsubscript{2}O emissions and, 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\textsubscript{4} and N\textsubscript{2}O fluxes are still uncertain (Melillo et al. 2009, Murdiyarso et al. 2010, Tian et al. 2012a). 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\textsubscript{2}, CH\textsubscript{4} and N\textsubscript{2}O in both field experiments and regional estimations.

Conclusions

Most of previous studies addressed only one of non-CO\textsubscript{2} GHGs (CH\textsubscript{4} and N\textsubscript{2}O) and one or a subset of environmental factors controlling emissions of these gases. Here we provided the first concurrent estimation of global CH\textsubscript{4} and N\textsubscript{2}O budgets and spatiotemporal patterns in the past three decades (1981–2010) by using a process-based, coupled biogeochemical model driven by multiple environmental factors. We found that both global CH\textsubscript{4} and N\textsubscript{2}O emissions largely increased from 1981 to 2010 resulting from global environmental changes. Large proportion of the CH\textsubscript{4} and N\textsubscript{2}O emissions and the most rapid increase were found in the tropical zone, suggesting this region could be a hot spot for mitigating GHGs. Although climate change also enhanced CH\textsubscript{4} and N\textsubscript{2}O emissions in the northern high-
latitude region, its increasing rate was much lower than that of the tropical region. Methane uptakes slightly increased in the upland ecosystems (e.g., forest, dry cropland, shrubland, and grassland) while CH₄ emission largely increased in the lowland ecosystems (e.g., natural wetland and rice field). N₂O emission increased in all the ecosystems with the highest increasing rate found in cropland, primarily as a result of agronomic management practices (e.g., nitrogen fertilizer use, irrigation, and manure application). High correlations between air temperature and CH₄/N₂O emissions indicate a positive feedback between climate warming and terrestrial emissions of CH₄ and N₂O. Given large increase in CH₄ and N₂O emission at global scale, we suggest that these two non-CO₂ GHGs together with CO₂ have to be simultaneously considered when evaluating if a policy is effective or efficient to mitigate climate change.

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