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

Hanqin Tian[1], Guangsheng Chen[1,2], Chaoqun Lu[1], Xiaofeng Xu[1,2], Wei Ren[1], Bowen Zhang[1], Kamaljit Banger[1], Bo Tao[1], Shufen Pan[1], Mingliang Liu[1,3] and Chi Zhang[1,4]

[1] International Center for Climate and Global Change Research, School of Forestry and Wildlife Sciences, Auburn University, Auburn, AL, 36849, USA;
[2] Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA;
[3] Department of Civil and Environmental Engineering, Washington State University, Pullman, WA, 99164, USA;
[4] Xinjiang Institute of Ecology and Geography, Chinese Academy of Sciences, Urumqi, 830011, China

Revised to Biogeosciences

March 12, 2014

Abstract

Methane (CH_4) and nitrous oxide (N_2O) are two most important greenhouse gases after carbon dioxide (CO_2), 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 concurrently 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 during 1981-2010 were 144.39 ± 25.54 Tg C/yr and 12.52 ± 1.52 Tg N/yr, respectively. Our simulations indicated a significant ($p < 0.01$) increasing trend for CH_4 (0.43 ± 0.06 Tg C/yr per year) and N_2O (0.14 ± 0.02 Tg N/yr per year) in the study period. 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 natural wetlands and rice fields due to increased rice field area and climate warming. Nitrous oxide emission increased substantially in all the biome types and the largest increase occurred in upland croplands due 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 should be simultaneously considered when evaluating if a policy is effective or efficient to reduce global warming in the future.

Keywords: global warming potential; methane; nitrous oxide; process-based ecosystem model; terrestrial ecosystem

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 Hus, 2010; Montzka et al., 2011; Kirschke et al., 2013; Saikawa et al., 2013; Tian et al., 2012a). 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). Recent analyses indicated that the atmospheric CH_4 has increased by more than 100% since 1800 (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 (Koven et al., 2011; 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 upland soils are the major CH_4 sink. Recent CH_4 budget estimation showed a global source of 548-678 Tg CH_4/yr during 2000-2009, nearly 50–60% from anthropogenic origin (Kirschke et al., 2013).

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. Atmospheric N_2O has increased by 18% compared to

68 the pre-industrial level, with a linear increasing rate of 0.26% per year during the recent few
69 decades (Forster et al., 2007). The observed increase in atmospheric N_2O concentration was
70 primarily attributed to reactive nitrogen inputs from synthetic nitrogen fertilizer and animal
71 manure applications, cropland expansion, and processes associated with fossil-fuel combustion
72 and biomass burning. The production and consumption of N_2O in soils involves both biotic and
73 abiotic processes. Many microbial groups contribute to the production and consumption of soil
74 N_2O, but biological nitrification and denitrification are the dominant processes (Ussiri and Lal,
75 2012). Due to more variable and fewer estimates as compared to CH_4, global N_2O emissions
76 were estimated ranging between 6.7 and 36.6 Tg N/yr (IPCC 2001), with a large uncertainty
77 range.

78 A number of studies using different modeling and statistical approaches were conducted
79 to estimate regional or global CH_4 (e.g., Bousquet et al., 2006; Potter et al., 1996; Bloom et al.,
80 2010; Tian et al., 2010; Xu et al., 2012; Zhuang et al., 2004; Schulze et al., 2009; Ito and Inatomi
81 et al., 2012; Meng et al., 2012; Kirschke et al., 2013; Bruhwiler et al., 2014; Miller et al. 2014)
82 and N_2O fluxes (e.g., Xu et al., 2008; Liu, 1996; Hirsch et al. 2006; Davidson, 2009; US EPA
83 2010; Bouwman et al., 2002; Zhuang et al., 2012). However, the estimates are far from certain
84 due to the complexity of the terrestrial ecosystems and the shortage of field and laboratory
85 measurements to validate the modeled processes controlling gas emissions (Ito and Inatomi et
86 al., 2012; Tian et al., 2012a; Melton et al., 2013). To simulate large-scale CH_4 and N_2O fluxes,
87 two approaches are generally used during past decades: bottom-up (e.g., inventory and process-
88 based modeling. Cao et al., 1995; Walter et al., 2001; Zhuang et al., 2004; Xu et al., 2008; Tian
89 et al., 2010) and top-down (e.g., atmospheric inversion. Frankenberg et al., 2005; Bergamaschi et
90 al., 2007; Kort et al., 2008; Bergamaschi et al., 2009; 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 have been reported to influence the production and consumption of CH_4 and N_2O (Hutchin et al., 1995; Conrad, 1996; Huang et al., 2010; Toet et al., 2011). As summarized by Dijkstra et al. (2012), global warming increases net CH_4 uptake in upland ecosystems due to stimulation of microbial CH_4 oxidation and higher CH_4 diffusivity with lower soil moisture, while had variable effects in water-saturated ecosystems (e.g., natural wetlands, rice fields, and peatlands). Global warming results in increased N_2O emissions in most land ecosystems due to stimulations of nitrifiers and denitrifiers activity and nitrogen supply through mineralization; however, global warming may also reduce N_2O emission through soil drying and stimulations of plant growth and nitrogen uptake (Dijkstra et al., 2012; Luo et al. 2013). Elevated atmospheric CO_2 may either increase or decrease CH_4 uptake in the upland ecosystems but increase CH_4 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 ecosystems with high nitrogen supply, elevated atmospheric CO_2 is reported to significantly increase N_2O 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 is small or even negative in nitrogen-limited ecosystems since elevated plant growth may result in less nitrogen availability for nitrifiers and denitrifiers (Mosier et al., 2002). Ozone (O_3) pollution caused losses of photosynthesis, and thus reduce CH_4 emissions (Toet et al., 2011), while either stimulating or reducing N_2O emission by influencing

114 the litter mass and quality (Kanerva et al., 2008). Nitrogen fertilization may dramatically
115 decrease CH_4 consumption in grassland and forest (Mosier et al., 1991; Steudler et al., 1989) and
116 either decrease or increase CH_4 emissions in rice fields depending on nitrogen fertilizer use
117 amount (Banger et al. 2012), while increase N_2O emissions (Del Grosso et al., 2006; Mosier et
118 al., 1998). In addition, interactions among multiple factors also played an important role. For
119 example, nitrogen deposition and elevated atmospheric CO_2 are reported to interactively reduce
120 CH_4 emission from wetland and increase CH_4 uptake in upland soil (Pancotto et al., 2010); and
121 another study concluded that temperature and elevated atmospheric CO_2 may interactively
122 change seasonal variation of CH_4 emissions (Blankinship et al., 2010). Atmospheric CO_2 and
123 nitrogen deposition can interactively increase soil available nitrogen and labile carbon, thus
124 greatly increase soil N_2O emissions (Dijkstra et al., 2012).

125 Most of previous experimental or modeling studies addressed the impacts of only one or
126 a subset of environmental factors on either CH_4 or N_2O fluxes. Few of them were conducted to
127 fully estimate the effects of multiple environmental changes (such as climate, land use, land
128 management practices, atmospheric CO_2, nitrogen deposition and tropospheric O_3, etc.) on both
129 gases. In addition, given the tight linkage between these two gases in both terrestrial origin
130 (Raghoebarsing et al., 2006; Kort et al., 2008; Ettwig et al., 2010; Song et al., 2009; Dijkstra et
131 al., 2012) and atmospheric chemistry (Prather and Hus, 2010), it is important to simultaneously
132 estimate the CH_4 and N_2O fluxes at continental or global scale. The specific objectives of this
133 study include: 1) providing a new and simultaneous estimate of global CH_4 and N_2O budgets,
134 which may help narrow down the estimate ranges of both gases in the terrestrial ecosystems; 2)
135 exploring interannual and decadal patterns of global CH_4 and N_2O as influenced by multiple
136 environmental factors including climate, atmospheric CO_2, tropospheric O_3 pollution, nitrogen

deposition, land use and land cover, and nitrogen fertilizer use; 3) evaluating the regional, biome-level and latitudinal differences in the impacts of multiple environmental factors on long-term CH_4 and N_2O patterns; and 4) identifying uncertainties associated with major model parameters, additional biogeochemical processes and data needed for future research.

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° latitude/longitude 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-hr), 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 (https://www.earthsystemgrid.org/dataset/ucar.cgd.ccsm4.CRUNCEP.v4.TPHWL6Hrly.html). Monthly O_3 AOT40 data were derived from the monthly AOT40 data developed by Felzer et al. (2005). We further interpolated this monthly data to daily data through simply dividing the monthly data by the number of days in each month since we did not have other auxiliary daily data for interpolation (Ren et al., 2007). EDGAR-HYDE 1.3 nitrogen emission data at 1 × 1 degree (Van Aardenne et al., 2001) were used to interpolate the three-time period nitrogen deposition data (1860, 1993, 2050; Dentener, 2006) into annual nitrogen deposition data set during 1901-2010, through assuming that the interannual variability of nitrogen deposition is consistent with that of NH_3 and NO_x emissions from 10 anthropogenic sources (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 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.

[Insert Fig 1 Here]

2.1 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 (Ren et al., 2007, 2011; Zhang et al., 2007, 2012; Tian et al., 2010, 2011a,b, 2012a,b; Xu et al., 2010, 2012a; Chen et al., 2012, 2013; Lu et

al., 2012, 2013). 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 equation:

$$F_{CH4} = F_P + F_D + F_E - F_{air,oxid} - F_{trans,oxid} - F_{soil,oxid} \quad (1)$$

where F_{CH4} is the flux of CH_4 between soil and the atmosphere (g C/m^2/day); F_P is plant-mediated transport from soil pore water to the atmosphere (g C/m^2/day); F_D is the diffusive flux

of CH_4 from water surface to the atmosphere (g C/m^2/d); 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); $F_{trans,\ oxid}$ is the CH_4 oxidation during plant-mediated transport (g C/m^2/day); $F_{soil,\ oxid}$ 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 (2000) to separate N_2O from other gases (mainly NO and N_2). The equations for calculating nitrification, denitrification and N_2O fluxes are:

$$F_{N2O} = (0.001 \times N_{nit} + N_{denit}) \times \frac{10^{(\frac{vwc}{\emptyset} \times \times 0.026 - 1.66)}}{(1+10^{(\frac{vwc}{\emptyset} \times \times 0.026 - 1.66)})} \quad (2)$$

Where F_{N2O} is the N_2O flux from soil to the atmosphere (g N/m^3/day), which can be converted to the unit of g N/m^2/day by multiplying the soil depth (default: 0.5 m); N_{nit} and N_{denit} are the nitrification and denitrification rates (g N/m^3/day), respectively; 0.001 is the proportion of nitrification product released as gaseous nitrogen (Lin et al., 2000); $\emptyset$ is the soil porosity.

2.2 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 years using de-trended climate data during 1901-1930 (i.e., 30 spins with 30-year 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.

2.3 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 observations, 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, 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_2O fluxes. In this study, we evaluated the DLEM-estimated CH_4 and N_2O fluxes against over 50 selected field observations worldwide with various land cover types including forest, grassland, wetland, and cropland (some field observation data were used in our previous publications, Table 1). We found that most of the DLEM-simulated CH_4 and N_2O fluxes fell in the observation range. 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. In addition, we compare DLEM- simulated CH_4 and N_2O fluxes with inversion estimates at global scale (Fig 2). Our estimates of CH_4 fluxes in wetlands and natural soils are compared with CarbonTracker results (Bruhwiler et al., 2014), while the simulated-N_2O emissions from

agricultural and natural soils area are compared with Saikawa et al., (2013). It showed that the DLEM simulation results are quite close to top-down estimates in both magnitude and temporal variability, with a small RMSE (square root of the mean square error of predictions; $RMSE = \sqrt{\frac{1}{n}\sum(x_i - y_i)^2}$) of 10.61 Tg CH_4-C/yr and 2.08 Tg N_2O-N/yr, which is pretty small relative to the observed CH_4 and N_2O emissions. In *Discussion* section, we further addressed additional regional comparisons between DLEM simulation and other studies from multiple approaches including statistical extrapolation, inverse modeling and process-based modeling.

[Insert Table1 and Fig. 2 here]

2.4 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 ^{o}C/yr and 1.37 mm/yr, 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-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 the southern Africa, the northern India, and the southeastern Australia (Fig. 3a). Air temperature increased in most areas, with the largest increase in the northern high-latitude region and a slight decrease in the South America and Oceania (Fig. 3b). The highest nitrogen deposition rates were shown in the East and South Asia, while the highest nitrogen fertilizer use rates were located 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.

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 in Europe and North America and an increasing trend 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 climatic 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 climatic zones except for the northern polar zone. Cropland area

316 decreased in the northern polar and northern temperate zones, while increased significantly in
317 other climatic zones, with the most rapid growth in the northern tropical zone. Ozone
318 concentration increased in the tropical and northern temperate zones, while no significant
319 changes in other climatic zones.

320 ***[Insert Fig. 3 Here]***

321 ***[Insert Tables. 2 and 3 Here]***

322

323 **3.2 Global budget of terrestrial CH_4 and N_2O fluxes**

324 The estimated terrestrial CH_4 emission during 1981-2010 was averaged 144.39 ± 32.92
325 Tg C/yr (1 Tg = 10^{12} g), ranging from 136.45 ± 30.05 Tg C/yr in 1982 to 156.45 ± 36.19 Tg C/yr
326 in 2010. Among which, the upland soil uptake of CH_4 was -17.10±0.17 Tg C/yr. The terrestrial
327 N_2O emission was averaged 12.52 ± 1.89 Tg N/yr, ranging from 10.53 ± 1.52 Tg N/yr in 1982
328 to 16.65 ± 2.80 Tg N/yr in 2010 (Fig. 4). Both CH_4 and N_2O showed a significant ($p < 0.01$)
329 increasing trend of 0.43 ± 0.06 Tg C/yr for CH_4 and 0.14 ± 0.02 Tg N/yr for N_2O over the study
330 period. The interannual variations in CH_4 and N_2O fluxes were roughly consistent ($R^2 = 0.87$; p
331 < 0.01), indicating a close relationship between CH_4 and N_2O fluxes. Significantly positive
332 correlations between air temperature and the model-estimated CH_4 ($R^2 = 0.73$, $p<0.01$) and N_2O
333 emissions ($R^2 = 0.71$, $p<0.01$) was observed, implying a strong positive feedback between
334 climate warming and CH_4/N_2O emissions.

335 In terms of global warming potential (100-yr horizon), the DLEM-estimated global CH_4
336 and N_2O emissions (i.e., 4.82 ± 1.10 Pg CO_2 eq/yr and 5.86 ± 0.89 Pg CO_2 eq/yr, respectively)
337 could completely offset the terrestrial CO_2 sink (8.3 ± 4.3 Pg C/yr estimated by multiple process-

based models, Le Quéré et al. 2012) over the recent three decades. It implies that global land ecosystem may act as a positive contributor to climate warming.

[Insert Fig. 4 Here]

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 (Fig. 5). 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 regions (e.g., Alaska, the northern Canada, West Siberia and the northern Eurasia) that have large wetland area, were less substantial CH_4 emission sources (~10 g C/m^2/yr). Larger CH_4 sinks (i.e., > 0.15 g C/m^2/yr) 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 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-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.

[Insert Fig. 5 Here]

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

Along the latitudinal gradient, CH_4 emissions peaked (8.06 Tg C/yr) at the latitudinal zones of 2–3^{o}S and 6–7^{o}S (Fig. 5), 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^{o}N). N_2O emissions peaked at the latitude zone of 6–7^{o}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°N–60°N), the northern tropical zone (0°–30°N), the southern tropical zone (0°–30°S), and the southern temperate zone (30°S–60°S). All climatic zones were net atmospheric sources of CH_4 and N_2O emissions (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 trends in the CH_4 and N_2O fluxes varied significantly among various climatic zones (Fig. 6). Over the study period, the most rapid increase in CH_4 emission was observed in the northern and southern tropical zone (0.16Tg 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, faster increase in the N_2O emissions occurred in northern tropical and southern tropical zones (0.039–0.078 Tg N/yr) than other zones. No significant changing trend in N_2O emission was observed in 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.

[Insert Fig. 6 Here]

[Insert Table 4 Here]

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

We further compared the magnitude of CH_4 and N_2O fluxes and their change trends during 1981-2010 for different biome types (Table 5; Fig. 7). 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 (1.90-4.38 Tg C/yr). Multiple environmental changes had caused wetland CH_4 emissions to increase 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 a rate of 0.014Tg C/yr and 0.0061 Tg C/yr, respectively (Fig. 7). 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 and 0.54 Tg C/yr, respectively, while no significant change trend was found for grassland. Rice field expansion (15% as shown in Table 2) contributed to 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. 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), followed by cropland (3.36 Tg N/yr), and the least was from grassland (0.82 Tg N/yr). Most of the forest N_2O emissions came from tropical region. Compared to forests in other climatic zones, tropical

forests had larger area and higher nitrogen transformation rate. The N_2O emissions significantly increased for all the biome types from 1981 to 2010 (Fig. 7). 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; 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). Increased nitrogen deposition and air temperature were the major causes of increased N_2O emissions in the non-managed biomes.

[Insert Fig. 7 Here]

[Insert Table 5 Here]

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. 8). Asia showed the highest CH_4 emission and the most rapid increase (0.56 Tg C/yr, 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, p < 0.01) was caused by larger increases in nitrogen fertilizer amounts and nitrogen deposition rates 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.

[Insert Fig. 8 Here]

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 139.57-153.69 Tg C/yr for the 1990s, 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 130.98 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 wetland in the 1980s, slightly higher than a previous estimate of 110 Tg C/yr (Matthews and Fung, 1987). Our estimated CH_4 emission from rice field (29.30-35.17 Tg C/yr) was similar with most previous estimates. We considered model driving forces (e.g., nitrogen fertilizer amounts, ozone pollution, and nitrogen deposition rate), which were not included in previous work (e.g., Cao et al., 1998; Chen and Prinn, 2005; Patra et al., 2009). DLEM-estimated upland CH_4 uptake during 1996-2005 (-15.01 ~ -13.45 Tg C/yr) was slightly lower than the previous estimates (-19.5 - -25.0 Tg C/yr).

In addition to divergent model representation of biogeochemical processes and parameter values, different input data might be a major cause of the large discrepancies among these estimates for CH_4 (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. Furthermore, large uncertainty might be also resulted from various climate data sources. Several series of climate data were previously used by the global models to simulated GHG fluxes. For example, CRU (Mitchell and Jones 2005), CRUNCEP 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) in which large differences exist at both spatial and temporal scales.

[Insert Table 6 Here]

This study reported global N_2O emission in a range of 10.49-14.30 Tg N/yr during 1981-2000, which was close to an empirical estimate of 13.31 Tg N/yr (Xu et al., 2008). The estimated N_2O 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 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.

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 79.22% (upland CH_4 uptake is not separated) is close to this value. The

494 increasing CH_4 emission from the northern high latitude and tropical region is also consistent
495 with the estimate from Dlugokencky et al. (2009). We estimated that about 80% of N_2O
496 emissions were from the tropical region. Based on observations and inverse methods, Prinn et al.
497 (1990) also reported that 52-68% of the N_2O emissions were from the tropical region during
498 1978-1988. In contrast, using an inversing modeling approach, Hirsch et al. (2006) showed a
499 lower estimate of N_2O emissions from 30°S to 90°S (0 to 4%) and a higher N_2O emission from
500 0° to 30°N (50 to 64%) during 1998 to 2001.

501 Due to different spatial and temporal change patterns of multiple environmental factors,
502 the change trends of CH_4 and N_2O fluxes were greatly different among continents and climate
503 zones. In Asia, land cover changed dramatically due to fast-growing population and
504 industrialization during the past three decades and was primarily characterized by conversions of
505 upland natural ecosystems to rice fields, which was the major contributor to the increasing CH_4
506 emission. In addition, nitrogen fertilizer use amount in Asia increased the most rapidly compared
507 to other continents (Table 2) and led to substantial increase in N_2O emission. Based on inventory
508 data, Kurokawa et al. (2013) found that CH_4 and N_2O increased by 32% and 18%, respectively
509 from 2000 to 2008 in Asia. By using inversion modeling, Saikawa et al. (2013) also reported that
510 N_2O emissions from agricultural soil increased by 56% (0.055 Tg/yr) in Asia (i.e., Northern Asia
511 + Southern Asia) from 1995 to 2008. In North America, land use kept relatively stable and
512 climate variability became the major contributing factors for CH_4 emissions, while both climate
513 and nitrogen fertilizer uses contributed the most to N_2O emissions as indicated by our previous
514 studies (i.e., Tian et al., 2010, 2012b; Xu et al., 2010, 2012a). DLEM simulations in Europe
515 showed a slight decreasing trend in CH_4 emission, and a less significant increase in N_2O
516 compared to other continents. Over shorter period (1990-2011), Saikawa et al. (2013) and EEA

517 (2013) reported that both agricultural and natural soil N_2O emissions had no significant changing
518 trends in Europe.
519
520 **4.3 Climate change and CH_4 and N_2O emissions**
521
522
523 Previous studies indicated that climate variability and change determined inter-annual
524 variations in terrestrial CH_4 and N_2O fluxes (Frolking and Crill, 1994; King, 1997; Xu et al.,
525 2008, 2010, 2012a; Tian et al., 2010; Dijkstra et al., 2012). A field experiment found the
526 hierarchical control on N_2O emission in forest, and concluded that precipitation controls the
527 instant N_2O flux pattern while air temperature determines the relatively long-term regime
528 (Brumme et al., 1999). Based on field experiments of 74 plots, Gundersen et al. (2012) found
529 that increased air temperature and precipitation greatly increased CH_4 and N_2O emissions, with
530 stronger impacts from increasing air temperature compared to increasing precipitation. A
531 significant correlation was also found between air temperature and atmospheric CH_4
532 concentration during pre-industrial period (Wuebbles and Hayhoe, 2002; Chappellaz et al.,
533 1993a, b). An even stronger positive feedback between N_2O emissions and air temperature
534 (about 1 Tg N/yr/^oC) was found by Xu et al. (2012b). In this study, model simulation also
535 revealed a high correlation between CH_4/N_2O and air temperature (R^2 = 0.73 for CH_4 and 0.71
536 for N_2O; p<0.01), implying a strong positive feedback between climate warming and CH_4/N_2O
537 emissions. Global air temperature significantly increased (0.024 ^oC/yr; Tables 2, 3) from 1981 to
538 2010, with the highest increase in the northern polar region (0.039 ^oC/yr). Climate warming in
539 the northern high-latitude region was reported as the most important stimulating factor for
540 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^o N climatic zone. We found a smaller positive correlation between CH_4/N_2O and precipitation (R^2 = 0.37 for CH_4 and 0.39 for N_2O), implying that increasing precipitation (Table 2) contributed to the increasing CH_4 and N_2O fluxes but the correlation was relatively weaker than that of temperature at global and continental scales.

4.4 Change 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 in 1981 to 137.67 Tg C/yr in 2010. 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 0.28×10^6 km^2 from 1981 to 2010, and CH_4 emission from paddy land increased by about 4 Tg C (Fig. 7C). Many previous studies attributed the increased atmospheric CH_4 concentration to increasing 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) to 2010 (20.37 Tg C/yr), 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. 5A and 8C). 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_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 from 1990 to 2010. We had a similar finding with agricultural N_2O emission increasing by 1.4 Tg N/yr during the same period (Fig. 7E). By using inversion modeling, Saikawa et al. (2013) also estimated an N_2O 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_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 ~50 Tg N/yr in 1981 to ~84 Tg N/yr in 2005, we can conclude that during 1981-2005, increase of about 0.85 Tg N_2O-N/yr 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

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

As most of previous studies addressed only one of non-CO_2 GHGs (CH_4 and N_2O) and one or a subset of environmental factors controlling emissions of these gases, here we provided the first concurrent estimation of global CH_4 and N_2O budgets and spatiotemporal patterns in the past three decades (1981-2010) by using a process-based, coupled biogeochemical model driven by multiple environmental factors, which includes climate variability, rising atmospheric CO_2,

632 increasing nitrogen deposition, tropospheric ozone pollution, land use change and nitrogen
633 fertilizer use. We found that both global CH_4 and N_2O emissions largely increased from 1981 to
634 2010 resulting from global environmental changes. Large proportion of the CH_4 and N_2O
635 emissions and the most rapid increase were found in the tropical zone, suggesting this region
636 could be a hot spot for mitigating greenhouse gases. Although climate change also enhanced CH_4
637 and N_2O emissions in the northern high-latitude region, its increasing rate was much lower than
638 that of the tropical region. Methane uptakes slightly increased in the upland ecosystems (e.g.,
639 forest, dry cropland, shrubland and grassland) while CH_4 emission largely increased in the
640 lowland ecosystems (e.g., natural wetland and rice field). N_2O emission increased in all the
641 ecosystems with the highest increasing rate found in cropland, which is primarily resulted from
642 agronomic management practices (e.g., nitrogen fertilizer use, irrigation and manure
643 application). High correlations between air temperature and CH_4/N_2O emissions indicate a
644 positive feedback between climate warming and terrestrial emissions of CH_4 and N_2O. Given
645 large increase in CH_4 and N_2O emission at global scale, we suggest that these two non-CO_2
646 GHGs together with CO_2 have to be simultaneously considered when evaluating if a policy is
647 effective or efficient to reduce global warming in the future.
648
649
650 **Acknowledgement**
651 This study has been supported by NASA Atmospheric Composition: Modeling and Analysis
652 Program (NNH10CC80C), NASA IDS Program (NNX10AU06G, NNG04GM39C), US National
653 Science Foundation Grants (AGS-1243220, CNS-1059376). This study contributes to the Non-
654 CO_2 Greenhouse Gases Synthesis of North American Carbon Program (NACP).

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References

Ambus, P. and Robertson, G. P.: Fluxes of CH_4 and N_2O in aspen stands grown under ambient and twice-ambient CO_2, Plant and Soil, 209(1), 1–8, 1999.

Baird, A. J., Beckwith, C. W., Waldron, S. and Waddington, J. M.: Ebullition of methane-containing gas bubbles from near-surface Sphagnum peat, Geophysical Research Letters, 31(21), 2004.

Banger, K., Tian, H. and Lu, C.: Do nitrogen fertilizers stimulate or inhibit methane emissions from rice fields? Global Change Biology, 18(10), 3259–3267, 2012.

Banin, A.: Global budget of N_2O: The role of soils and their change, Science of The Total Environment, 55, 27–38, 1986.

Bartlett, K. B. and Harriss, R. C.: Review and assessment of methane emissions from wetlands, Chemosphere, 26(1–4), 261–320, 1993.

Bergamaschi, P., Frankenberg, C., Meirink, J. F., Krol, M., Dentener, F., Wagner, T., Platt, U., Kaplan, J. O., Körner, S., Heimann, M., Dlugokencky, E. J. and Goede, A.: Satellite chartography of atmospheric methane from SCIAMACHY on board ENVISAT: 2. Evaluation based on inverse model simulations, Journal of Geophysical Research: Atmospheres, 112(2), doi:10.1029/2006JD007268, 2007.

Bergamaschi, P., Frankenberg, C., Meirink, J. F., Krol, M., Villani, M. G., Houweling, S., Dentener, F., Dlugokencky, E. J., Miller, J. B., Gatti, L. V., Engel, A. and Levin, I.: Inverse modeling of global and regional CH_4 emissions using SCIAMACHY satellite retrievals, Journal of Geophysical Research: Atmospheres, 114(22), doi:10.1029/2009JD012287, 2009.

Blais, A.M., Lorrain, S. and Tremblay, A.: Greenhouse Gas Fluxes (CO_2, CH_4 and N_2O) in Forests and Wetlands of Boreal, Temperate and Tropical Regions, in Greenhouse Gas Emissions — Fluxes and Processes, edited by D. A. Tremblay, D. L. Varfalvy, D. C. Roehm, and D. M. Garneau, pp. 87–127, Springer Berlin Heidelberg. Available from: http://link.springer.com/chapter/10.1007/978-3-540-26643-3_5, 2005.

Blankinship, J. C., Brown, J. R., Dijkstra, P. and Hungate, B. A.: Effects of interactive global changes on methane uptake in an annual grassland, Journal of Geophysical Research: Biogeosciences, 115(2), doi:10.1029/2009JG001097, 2010.

Bloom, A. A., Palmer, P. I., Fraser, A., Reay, D. S. and Frankenberg, C.: Large-Scale Controls of Methanogenesis Inferred from Methane and Gravity Spaceborne Data, Science, 327(5963), 322–325, 2010.

Bousquet, P., Ciais, P., Miller, J. B., Dlugokencky, E. J., Hauglustaine, D. A., Prigent, C., Van der Werf, G. R., Peylin, P., Brunke, E.-G., Carouge, C., Langenfelds, R. L., Lathière, J., Papa, F., Ramonet, M., Schmidt, M., Steele, L. P., Tyler, S. C. and White, J.: Contribution of anthropogenic and natural sources to atmospheric methane variability, Nature, 443(7110), 439–443, doi:10.1038/nature05132, 2006.

694 Bousquet, P., Ringeval, B., Pison, I., Dlugokencky, E. J., Brunke, E.-G., Carouge, C., Chevallier,
695 F., Fortems-Cheiney, A., Frankenberg, C., Hauglustaine, D. A., Krummel, P. B.,
696 Langenfelds, R. L., Ramonet, M., Schmidt, M., Steele, L. P., Szopa, S., Yver, C., Viovy,
697 N. and Ciais, P.: Source attribution of the changes in atmospheric methane for 2006–
698 2008, Atmos. Chem. Phys., 11(8), 3689–3700, doi:10.5194/acp-11-3689-2011, 2011.

699 Bouwman, A. F., Boumans, L. J. M. and Batjes, N. H.: Modeling global annual N_2O and NO
700 emissions from fertilized fields, Global Biogeochemical Cycles, 16(4), 28–1–28–9, 2002.

701 Bouwman, A. F., Drecht, G. V. and Hoek, K. W. van der: Surface N balances and reactive N loss
702 to the environment from global intensive agricultural production systems for the period
703 1970–2030, Sci. China Ser. C.-Life Sci., 48(2), 767–779, 2005.

704 Bowden, R. D., Steudler, P. A., Melillo, J. M. and Aber, J. D.: Annual nitrous oxide fluxes from
705 temperate forest soils in the northeastern United States, Journal of Geophysical Research:
706 Atmospheres, 95(9), 13997–14005, 1990.

707 Bowden, W. B.: Gaseous nitrogen emissions from undisturbed terrestrial ecosystems: An
708 assessment of their impacts on local and global nitrogen budgets, Biogeochemistry, 2(3),
709 249–279, doi:10.1007/BF02180161, 1986.

710 Bronson, K. F., Neue, H.-U., Abao,, E. B. and Singh, U.: Automated Chamber Measurements of
711 Methane and Nitrous Oxide Flux in a Flooded Rice Soil: I. Residue, Nitrogen, and Water
712 Management, Soil Science Society of America Journal, 61(3), 981, 1997.

713 Bruhwiler, L. M., Dlugokencky, E., Masarie, k., Ishizawa, M., Andrews, A., Miller, J., Sweeney,
714 C., Tans, P., and Worthy, D.: CarbonTracker-CH4: an assimilation system for estimating
715 emissions of atmospheric methane. Atmos. Chem. Phys. Discuss., 14, 2175–2233, 2014

716 Brumme, R., Borken, W. and Finke, S.: Hierarchical control on nitrous oxide emission in forest
717 ecosystems, Global Biogeochemical Cycles, 13(4), 1137–1148,
718 doi:10.1029/1999GB900017, 1999.

719 Butterbach-Bahl, K., Gasche, R., Breuer, L. and Papen, H.: Fluxes of NO and N_2O from
720 temperate forest soils: impact of forest type, N deposition and of liming on the NO and
721 N_2O emissions, Nutrient Cycling in Agroecosystems, 48(1-2), 79–90,
722 doi:10.1023/A:1009785521107, 1997.

723 Cantarel, A. A. M., Bloor, J. M. G., Deltroy, N., et al.: Effects of climate change drivers on
724 nitrous oxide fluxes in an upland temperate grassland. Ecosystems, 14, 223–33, 2011.

725 Cao, M., Gregson, K. and Marshall, S.: Global methane emission from wetlands and its
726 sensitivity to climate change, Atmospheric Environment, 32(19), 3293–3299, 1998.

727 Chen, G., Tian, H., Huang, C., Prior, S.A., Pan, S..: Integrating a process-based ecosystem model
728 with Landsat imagery to assess impacts of forest disturbance on terrestrial carbon
729 dynamics: Case studies in Alabama and Mississippi. J Geophys Res 118, 1208–1224,
730 2013

Chen, G., Tian, H., Zhang, C., Liu, M., Ren, W., Zhu W., Chappelka, A., Prior, S.A., Lockaby, G.B.: Drought in the Southern United States over the 20th century: variability and its impacts on terrestrial ecosystem productivity and carbon storage. Climatic Change 114, 379–397, 2012

Christensen, T. R., Friborg, T., Sommerkorn, M., Kaplan, J., Illeris, L., Soegaard, H., Nordstroem, C. and Jonasson, S.: Trace gas exchange in a high-Arctic valley: 1. Variations in CO_2 and CH_4 Flux between tundra vegetation types, Global Biogeochemical Cycles, 14(3), 701–713, doi:10.1029/1999GB001134, 2000.

Christensen, T. R., Prentice, I. C., Kaplan, J., Haxeltine, A. and Sitch, S.: Methane flux from northern wetlands and tundra, Tellus B, 48(5), 652–661, 1996.

Clymo, R. S. and Reddaway, E. J. F.: Productivity of Sphagnum (bog-moss) and peat accumulation, Hydrobiologia, 12(18), 1–192, 1971.

Cochran, V. L., Sparrow, E. B., Schlentner, S. F. and Knight, C. W.: Long-term tillage and crop residue management in the subarctic: fluxes of methane and nitrous oxide, Canadian Journal of Soil Science, 77(4), 565–570, doi:10.4141/S96-089, 1997.

Conrad, R.: Soil microorganisms as controllers of atmospheric trace gases (H_2, CO, CH_4, OCS, N_2O, and NO), Microbiol. Rev., 60(4), 609–640, 1996.

Corre, M. D., Pennock, D. J., Kessel, C. V. and Kirkelliott, D.: Estimation of annual nitrous oxide emissions from a transitional grassland-forest region in Saskatchewan, Canada, Biogeochemistry, 44(1), 29–49, doi:10.1007/BF00992997, 1999.

Dacey, J. W. H., Drake, B. G. and Klug, M. J.: Stimulation of methane emission by carbon dioxide enrichment of marsh vegetation, Nature, 370(6484), 47–49, 1994.

Dambreville, C., Morvan, T. and Germon, J.C.: N_2O emission in maize-crops fertilized with pig slurry, matured pig manure or ammonium nitrate in Brittany, Agriculture, Ecosystems & Environment, 123(1–3), 201–210, 2008.

Davidson, E. A., Matson, P. A., Vitousek, P. M., Riley, R., Dunkin, K., Garcia-Mendez, G. and Maass, J. M.: Processes Regulating Soil Emissions of NO and N_2O in a Seasonally Dry Tropical Forest, Ecology, 74(1), 130–139, doi:10.2307/1939508, 1993.

Davidson, E. A., Nepstad, D. C., Ishida, F. Y. and Brando, P. M.: Effects of an experimental drought and recovery on soil emissions of carbon dioxide, methane, nitrous oxide, and nitric oxide in a moist tropical forest, Global Change Biology, 14(11), 2582–2590, 2008.

Davidson, E. A.: The contribution of manure and fertilizer nitrogen to atmospheric nitrous oxide since 1860, Nature Geosci, 2(9), 659–662, doi:10.1038/ngeo608, 2009.

De Fries, R. S., Hansen, M., Townshend, J. R. G. and Sohlberg, R.: Global land cover classifications at 8 km spatial resolution: The use of training data derived from Landsat imagery in decision tree classifiers, International Journal of Remote Sensing, 19(16), 3141–3168, doi:10.1080/014311698214235, 1998.

De Laune, R. D., Smith, C. J. and Patrick, W. H.: Methane release from Gulf coast wetlands, Tellus B, 35(1), 8–15, 1983.

Del Grosso, S. J., Ogle, S. M., Parton, W. J. and Breidt, F. J.: Estimating uncertainty in N_2O emissions from U.S. cropland soils, Global Biogeochemical Cycles, 24(1), 2010.

Del Grosso, S. J., Parton, W. J., Mosier, A. R., Walsh, M. K., Ojima, D. S. and Thornton, P. E.: DAYCENT National-Scale Simulations of Nitrous Oxide Emissions from Cropped Soils in the United States, Journal of Environment Quality, 35(4), 1451, doi:10.2134/jeq2005.0160, 2006.

Denman, K. L., Brasseur, G., Chidthaisong, A., Ciais, P., Cox, P. M., Dickinson, R. E., Hauglustaine, D., Heinze, C., Holland, E. and Jacob, D.: Couplings between changes in the climate system and biogeochemistry, Climate change, 541–584, 2007.

Dentener, F. J.: Global maps of atmospheric nitrogen deposition, 1860, 1993, and 2050, Data set. Available from: https://www-eosdis.ornl.gov/cgi-bin/dsviewer.pl?ds_id=830, 2006.

Dijkstra, F. A., Prior, S. A., Runion, G. B., Torbert, H. A., Tian, H., Lu, C. and Venterea, R. T.: Effects of elevated carbon dioxide and increased temperature on methane and nitrous oxide fluxes: evidence from field experiments, Frontiers in Ecology and the Environment, 10(10), 520–527, doi:10.1890/120059, 2012.

Dise, N. B.: Methane emission from Minnesota peatlands: Spatial and seasonal variability, Global Biogeochemical Cycles, 7(1), 123–142, doi:10.1029/92GB02299, 1993.

Dlugokencky, E. J., Bruhwiler, L., White, J. W. C., Emmons, L. K., Novelli, P. C., Montzka, S. A., Masarie, K. A., Lang, P. M., Crotwell, A. M., Miller, J. B. and Gatti, L. V.: Observational constraints on recent increases in the atmospheric CH_4 burden, Geophysical Research Letters, 36(18), doi:10.1029/2009GL039780, 2009.

EEA (European Environment Agency): Annual European Union greenhouse gas inventory 1990-…2011 and inventory report 2013, Technical report No 8/2013, 2013. http://www.eea.europa.eu/publications/european-union-greenhouse-gas-inventory-2013.

Erickson, H., Davidson, E. A. and Keller, M.: Former land-use and tree species affect nitrogen oxide emissions from a tropical dry forest, Oecologia, 130(2), 297–308, 2002.

Ettwig, K. F., Butler, M. K., Le Paslier, D., Pelletier, E., Mangenot, S., Kuypers, M. M. M., Schreiber, F., Dutilh, B. E., Zedelius, J., de Beer, D., Gloerich, J., Wessels, H. J. C. T., van Alen, T., Luesken, F., Wu, M. L., van de Pas-Schoonen, K. T., Op den Camp, H. J. M., Janssen-Megens, E. M., Francoijs, K.-J., Stunnenberg, H., Weissenbach, J., Jetten, M. S. M. and Strous, M.: Nitrite-driven anaerobic methane oxidation by oxygenic bacteria, Nature, 464(7288), 543–548, doi:10.1038/nature08883, 2010.

FAO/IFA: Global Estimates of Gaseous Emissions of NH_3, NO and N_2O from Agricultural Land, Rome: Food and Agriculture Organization of the United Nations (FAO)/International Fertilizer Industry Association (IFA), 106, 2001

Flessa, H., Dörsch, P. and Beese, F.: Seasonal variation of N_2O and CH_4 fluxes in differently managed arable soils in southern Germany, Journal of Geophysical Research: Atmospheres, 100(11), 23115–23124, doi:10.1029/95JD02270, 1995.

Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Myhre, G., Nganga, J., Prinn, R., Raga, G., Schultz, M. and Van Dorland, R.: Changes in atmospheric constituents and in radiative forcing, Cambridge University Press, Cambridge, United Kingdom. Available from: http://www.ipcc.ch/pdf/assessment-report/ar4/wg1/ar4-wg1-chapter2.pdf, 2007.

Frankenberg, C., Meirink, J. F., Weele, M. van, Platt, U. and Wagner, T.: Assessing Methane Emissions from Global Space-Borne Observations, Science, 308(5724), 1010–1014, doi:10.1126/science.1106644, 2005.

Frolking, S. and Crill, P.: Climate controls on temporal variability of methane flux from a poor fen in southeastern New Hampshire: Measurement and modeling, Global Biogeochemical Cycles, 8(4), 385–397, doi:10.1029/94GB01839, 1994.

Fung, I., John, J., Lerner, J., Matthews, E., Prather, M., Steele, L. P. and Fraser, P. J.: Three-dimensional model synthesis of the global methane cycle, Journal of Geophysical Research: Atmospheres, 96(7), 13033–13065, doi:10.1029/91JD01247, 1991.

Garcia-Montiel, D. C., Steudler, P. A., Piccolo, M., Neill, C., Melillo, J. and Cerri, C. C.: Nitrogen oxide emissions following wetting of dry soils in forest and pastures in Rondônia, Brazil, Biogeochemistry, 64(3), 319–336, doi:10.1023/A:1024968802018, 2003.

Goodroad, L. L. and Keeney, D. R.: Nitrous oxide production in aerobic soils under varying pH, temperature and water content, Soil Biology and Biochemistry, 16(1), 39–43, 1984.

Haapanala, S., Rinne, J., Pystynen, K.-H., Hellén, H., Hakola, H. and Riutta, T.: Measurements of hydrocarbon emissions from a boreal fen using the REA technique, Biogeosciences, 3(1), 103–112, 2006.

Hadi, A., Inubushi, K. and Yagi, K.: Effect of water management on greenhouse gas emissions and microbial properties of paddy soils in Japan and Indonesia, Paddy Water Environ, 8(4), 319–324, doi:10.1007/s10333-010-0210-x, 2010.

Hao, X., Chang, C., Carefoot, J. M., Janzen, H. H. and Ellert, B. H.: Nitrous oxide emissions from an irrigated soil as affected by fertilizer and straw management, Nutrient Cycling in Agroecosystems, 60(1-3), 1–8, 2001.

Harriss, R. C., Gorham, E., Sebacher, D. I., Bartlett, K. B. and Flebbe, P. A.: Methane flux from northern peatlands, Nature, 315(6021), 652–654, doi:10.1038/315652a0, 1985.

Hendriks, D. M. D., van Huissteden, J., Dolman, A. J. and van der Molen, M. K.: The full greenhouse gas balance of an abandoned peat meadow, Biogeosciences Discuss., 4(1), 277–316, doi:10.5194/bgd-4-277-2007, 2007.

Hirsch, A. I., Michalak, A. M., Bruhwiler, L. M., Peters, W., Dlugokencky, E. J. and Tans, P. P.: Inverse modeling estimates of the global nitrous oxide surface flux from 1998–2001, Global Biogeochemical Cycles, 20(1) ,doi:10.1029/2004GB002443, 2006.

Hou, H., Peng, S., Xu, J., Yang, S. and Mao, Z.: Seasonal variations of CH_4 and N_2O emissions in response to water management of paddy fields located in Southeast China, Chemosphere, 89(7), 884–892, 2012.

Houweling, S., Kaminski, T., Dentener, F., Lelieveld, J. and Heimann, M.: Inverse modeling of methane sources and sinks using the adjoint of a global transport model, Journal of Geophysical Research: Atmospheres, 104(21), 26137–26160, 1999.

Huang, J., Golombek, A., Prinn, R., Weiss, R., Fraser, P., Simmonds, P., Dlugokencky, E. J., Hall, B., Elkins, J., Steele, P., Langenfelds, R., Krummel, P., Dutton, G. and Porter, L.: Estimation of regional emissions of nitrous oxide from 1997 to 2005 using multinetwork measurements, a chemical transport model, and an inverse method, Journal of Geophysical Research: Atmospheres, 113(17) ,doi:10.1029/2007JD009381, 2008.

Huang, Y., Sun, W., Zhang, W., Yu, Y., Su, Y. and Song, C.: Marshland conversion to cropland in northeast China from 1950 to 2000 reduced the greenhouse effect, Global Change Biology, 16(2), 680–695, 2010.

Hutchin, P. R., Press, M. C., Lee, J. A. and Ashenden, T. W.: Elevated concentrations of CO_2 may double methane emissions from mires, Global Change Biology, 1(2), 125–128, doi:10.1111/j.1365-2486.1995.tb00012.x, 1995.

Ineson, P., Coward, P. A. and Hartwig, U. A.: Soil gas fluxes of N_2O, CH_4 and CO_2 beneath Lolium perenne under elevated CO_2: The Swiss free air carbon dioxide enrichment experiment, Plant and Soil, 198(1), 89–95, doi:10.1023/A:1004298309606, 1998.

IPCC: Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change, edited by Houghton ,J. T., et al., Cambridge Univ. Press, New York, 2001.

Ito, A. and Inatomi, M.: Use of a process-based model for assessing the methane budgets of global terrestrial ecosystems and evaluation of uncertainty, Biogeosciences, 9(2), 759–773, doi:10.5194/bg-9-759-2012, 2012.

Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G., Woollen, J., Zhu, Y., Leetmaa, A., Reynolds, R., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J., Mo, K. C., Ropelewski, C., Wang, J., Jenne, R. and Joseph, D.: The NCEP/NCAR 40-Year Reanalysis Project, Bulletin of the American Meteorological Society, 77(3), 437–471, 1996.

Kanerva, T., Palojärvi, A., Rämö, K. and Manninen, S.: Changes in soil microbial community structure under elevated tropospheric O_3 and CO_2, Soil Biology and Biochemistry, 40(10), 2502–2510, 2008.

879 Keller, M. and Reiners, W. A.: Soil-atmosphere exchange of nitrous oxide, nitric oxide, and
880 methane under secondary succession of pasture to forest in the Atlantic lowlands of Costa
881 Rica, Global Biogeochemical Cycles, 8(4), 399–409, doi:10.1029/94GB01660, 1994.

882 Keller, M., Goreau, T. J., Wofsy, S. C., Kaplan, W. A. and McElroy, M. B.: Production of
883 nitrous oxide and consumption of methane by forest soils, Geophysical Research Letters,
884 10(12), 1156–1159, doi:10.1029/GL010i012p01156, 1983.

885 Kellner, E., Waddington, J. M. and Price, J. S.: Dynamics of biogenic gas bubbles in peat:
886 Potential effects on water storage and peat deformation, Water Resources Research,
887 41(8), 2005.

888 Kessavalou, A., Doran, J. W., Mosier, A. R. and Drijber, R. A.: Greenhouse Gas Fluxes
889 following Tillage and Wetting in a Wheat-Fallow Cropping System, Journal of
890 Environment Quality, 27(5), 1105, 1998.

891 Kim, D.-G., Mishurov, M. and Kiely, G.: Effect of increased N use and dry periods on N2O
892 emission from a fertilized grassland, Nutr Cycl Agroecosyst, 88(3), 397–410,
893 doi:10.1007/s10705-010-9365-5, 2010.
894 King, G.: Responses of atmospheric methane consumption by soils to global climate change,
895 Global Change Biology, 3(4), 351–362, 1997.

896 Kirschke, S., Bousquet, P., Ciais, P., Saunois M., et al.: Three decades of global methane sources
897 and sinks, Nature Geoscience, 6, 813-823, 2013.

898 Kirschke, S., Bousquet, P., Ciais, P., Saunois, M., Canadell, J. G., Dlugokencky, E. J.,
899 Bergamaschi, P., Bergmann, D., Blake, D. R., Bruhwiler, L., Cameron-Smith, P.,
900 Castaldi, S., Chevallier, F., Feng, L., Fraser, A., Heimann, M., Hodson, E. L., Houweling,
901 S., Josse, B., Fraser, P. J., Krummel, P. B., Lamarque, J. F., Langenfelds, R. L., Le
902 Quéré, C., Naik, V., O'Doherty, S., Palmer, P. I., Pison, I., Plummer, D., Poulter, B.,
903 Prinn, R. G., Rigby, M., Ringeval, B., Santini, M., Schmidt, M., Shindell, D. T.,
904 Simpson, I. J., Spahni, R., Steele, L. P., Strode, S. A., Sudo, K., Szopa, S., van der Werf,
905 G. R., Voulgarakis, A., van Weele, M., Weiss, R. F., Williams, J. E. and Zeng, G.: Three
906 decades of global methane sources and sinks, Nature Geosci, 6(10), 813–823,
907 doi:10.1038/ngeo1955, 2013.

908 Klein, G.K., van Drecht, G. (Eds.): HYDE 3: Current and historical population and land cover
909 MNP . Netherlands Environmental Assessment Agency (MN), Bilthoven, The
910 Netherlands, 2006

911 Koehler, B., Corre, M. D., Veldkamp, E., Wullaert, H. and Wright, S. J.: Immediate and long
912 term nitrogen oxide emissions from tropical forest soils exposed to elevated nitrogen
913 input, Global Change Biology, 15(8), 2049–2066, 2009.

914 Kort, E. A., Eluszkiewicz, J., Stephens, B. B., Miller, J. B., Gerbig, C., Nehrkorn, T., Daube, B.
915 C., Kaplan, J. O., Houweling, S. and Wofsy, S. C.: Emissions of CH_4 and N_2O over the
916 United States and Canada based on a receptor-oriented modeling framework and COBRA
917 NA atmospheric observations, Geophysical Research Letters, 35(18), 2008.

Le Quéré, C., Andres, R. J., Boden, T., Conway, T., Houghton, R. A., House, J. I., Marland, G., Peters, G. P., van der Werf, G. R., Ahlström, A., Andrew, R. M., Bopp, L., Canadell, J. G., Ciais, P., Doney, S. C., Enright, C., Friedlingstein, P., Huntingford, C., Jain, A. K., Jourdain, C., Kato, E., Keeling, R. F., Klein Goldewijk, K., Levis, S., Levy, P., Lomas, M., Poulter, B., Raupach, M. R., Schwinger, J., Sitch, S., Stocker, B. D., Viovy, N., Zaehle, S. and Zeng, N.: The global carbon budget 1959–2011, Earth System Science Data, 5(1), 165–185, 2013.

Lehner, B. and Döll, P.: Development and validation of a global database of lakes, reservoirs and wetlands, Journal of Hydrology, 296, 2004.

Lemke, R. l., Izaurralde, R. C., Malhi, S. S., Arshad, M. A. and Nyborg, M.: Division S-8 - nutrient management & soil & plant analysis - nitrous oxide emissions from agricultural soils of the boreal and parkland regions of Alberta, 62, 1096–1102, 1998.

Li, C., Narayanan, V. and Harriss, R. C.: Model estimates of nitrous oxide emissions from agricultural lands in the United States, Global Biogeochemical Cycles, 10(2), 297–306, doi:10.1029/96GB00470, 1996.

Lin, B. L., Sakoda, A., Shibasaki, R., Goto, N., and Suzuki, M.: Modelling a global biogeochemical nitrogen cycle in terrestrial ecosystems, Ecol. Model., 135, 89–110, 2000

Liu, L. L., and Greaver T. L.: A review of nitrogen enrichment effects on three biogenic GHGs: the CO2 sink may be largely offset by stimulated N_2O and CH_4 emission, Ecology Letters, 12, 1103-1117, 2009.

Liu, M., Tian, H., Chen, G., Ren, W., Zhang, C. and Liu, J.: Effects of Land-Use and Land-Cover Change on Evapotranspiration and Water Yield in China During 1900-2001, JAWRA Journal of the American Water Resources Association, 44(5), 1193–1207, 2008.

Lu, C. and Tian, H.: Net greenhouse gas balance in response to nitrogen enrichment: perspectives from a coupled biogeochemical model, Global Change Biology, 19(2), 571–588, doi:10.1111/gcb.12049, 2013.

Lu, C., Tian, H., Liu, M., Ren, W., Xu, X., Chen, G. and Zhang, C.: Effect of nitrogen deposition on China's terrestrial carbon uptake in the context of multifactor environmental changes, Ecol Appl, 22(1), 53–75, 2012.

Luo, G. J., Kiese, R., Wolf, B. and Butterbach-Bahl, K.: Effects of soil temperature and moisture on methane uptake and nitrous oxide emissions across three different ecosystem types, Biogeosciences, 10, 3205–3219, 2013.

Matson, A., Pennock, D. and Bedard-Haughn, A.: Methane and nitrous oxide emissions from mature forest stands in the boreal forest, Saskatchewan, Canada, Forest Ecology and Management, 258(7), 1073–1083, 2009.

Matson, P. A., Vitousek, P. M., Livingston, G. P. and Swanberg, N. A.: Sources of variation in nitrous oxide flux from Amazonian ecosystems, Journal of Geophysical Research: Atmospheres, 95(10), 16789–16798, 1990.

Matthews, E. and Fung, I.: Methane emission from natural wetlands: Global distribution, area, and environmental characteristics of sources, Global Biogeochemical Cycles, 1(1), 61–86, 1987.

Melillo, J. M., Reilly, J. M., Kicklighter, D. W., Gurgel, A. C., Cronin, T. W., Paltsev, S., Felzer, B. S., Wang, X., Sokolov, A. P. and Schlosser, C. A.: Indirect Emissions from Biofuels: How Important?, Science, 326(5958), 1397–1399, 2009.

Melton, J. R., Wania, R., Hodson, E. L., Poulter, B., Ringeval, B., Spahni, R., Bohn, T., Avis, C. A., Beerling, D. J., Chen, G., Eliseev, A. V., Denisov, S. N., Hopcroft, P. O., Lettenmaier, D. P., Riley, W. J., Singarayer, J. S., Subin, Z. M., Tian, H., Zürcher, S., Brovkin, V., van Bodegom, P. M., Kleinen, T., Yu, Z. C. and Kaplan, J. O.: Present state of global wetland extent and wetland methane modeling: conclusions from a model inter-comparison project (WETCHIMP), Biogeosciences, 10(2), 753–788, doi:10.5194/bg-10-753-2013, 2013.

Miller, S. M., Doug E. J. Worthy, D.E., Michalak, A.M., Wofsy, S.C., Kort, E.A., Havice, T.C., Andrews, A.E., Dlugokencky, E.J., Kaplan, J.O., Levi, P.J., Tian, H.Q., Zhang, B.: Observational constraints on the distribution, seasonality, and environmental predictors of North American boreal methane emissions, Global Biogeochem. Cycles, 28, doi:10.1002/2013GB004580, 2014

Mitchell, T. D. and Jones, P. D.: An improved method of constructing a database of monthly climate observations and associated high-resolution grids, International Journal of Climatology, 25(6), 693–712, doi:10.1002/joc.1181, 2005.

Molodovskaya, M., Singurindy, O., Richards, B. K., Warland, J., Johnson, M. S. and Steenhuis, T. S.: Temporal Variability of Nitrous Oxide from Fertilized Croplands: Hot Moment Analysis, Soil Science Society of America Journal, 76(5), 1728, doi:10.2136/sssaj2012.0039, 2012.

Montzka, S. A., Dlugokencky, E. J. and Butler, J. H.: Non-CO_2 greenhouse gases and climate change, Nature, 476(7358), 43–50, 2011.

Moore, T., Roulet, N. and Knowles, R.: Spatial and temporal variations of methane flux from subarctic/northern boreal fens, Global Biogeochemical Cycles, 4(1), 29–46, 1990.

Mosier, A. R. and Hutchinson, G. L.: Nitrous Oxide Emissions From Cropped Fields1, Journal of Environment Quality, 10(2), 169, 1981.

Mosier, A. R., Delgado, J. A. and Keller, M.: Methane and nitrous oxide fluxes in an acid Oxisol in western Puerto Rico: effects of tillage, liming and fertilization, Soil Biology and Biochemistry, 30(14), 2087–2098, 1998.

Mosier, A. R., Guenzi, W. D. and Schweizer, E. E.: Soil Losses of Dinitrogen and Nitrous Oxide from Irrigated Crops in Northeastern Colorado1, Soil Science Society of America Journal, 50(2), 344, 1986.

Mosier, A. R., Hutchinson, G. L., Sabey, B. R. and Baxter, J.: Nitrous Oxide Emissions from Barley Plots Treated with Ammonium Nitrate or Sewage Sludge1, Journal of Environment Quality, 11(1), 78, 1982.

Mosier, A. R., Morgan, J. A., King, J. Y., LeCain, D. and Milchunas, D. G.: Soil-atmosphere exchange of CH_4, CO_2, NOx, and N_2O in the Colorado shortgrass steppe under elevated CO_2, Plant and Soil, 240(2), 201–211, 2002.

Mosier, A., Schimel, D., Valentine, D., Bronson, K. and Parton, W.: Methane and nitrous oxide fluxes in native, fertilized and cultivated grasslands, Nature, 350(6316), 330–332, 1991.

Mukhopadhyay, S. K., Biswas, H., De, T. K., Sen, B. K., Sen, S. and Jana, T. K.: Impact of Sundarban mangrove biosphere on the carbon dioxide and methane mixing ratios at the NE Coast of Bay of Bengal, India, Atmospheric Environment, 36(4), 629–638, doi:10.1016/S1352-2310(01)00521-0, 2002.

Murdiyarso, D., Hergoualc'h, K. and Verchot, L. V.: Opportunities for reducing greenhouse gas emissions in tropical peatlands, PNAS, 107(46), 19655–19660, 2010.

Nicolini, G., Castaldi, S., Fratini, G. and Valentini, R.: A literature overview of micrometeorological CH4 and N2O flux measurements in terrestrial ecosystems, Atmospheric Environment, 81, 311–319, doi:10.1016/j.atmosenv.2013.09.030, 2013.

Pancotto, V. A., van Bodegom, P. M., van Hal, J., van Logtestijn, R. S. P., Blokker, P., Toet, S. and Aerts, R.: N deposition and elevated CO_2 on methane emissions: Differential responses of indirect effects compared to direct effects through litter chemistry feedbacks, Journal of Geophysical Research: Biogeosciences, 115(2) , doi:10.1029/2009JG001099, 2010.

Peng, S., Hou, H., Xu, J., Mao, Z., Abudu, S. and Luo, Y.: Nitrous oxide emissions from paddy fields under different water managements in southeast China, Paddy Water Environ, 9(4), 403–411, doi:10.1007/s10333-011-0275-1, 2011.

Pérez, M. V. A., Castañeda, J. G., Frías-Hernández, J. T., Franco-Hernández, O., Cleemput, O. V., Dendooven, L. and Olalde, V.: Trace gas emissions from soil of the central highlands of Mexico as affected by natural vegetation: a laboratory study, Biol Fertil Soils, 40(4), 252–259, 2004.

Potter, C. S., Matson, P. A., Vitousek, P. M. and Davidson, E. A.: Process modeling of controls on nitrogen trace gas emissions from soils worldwide, Journal of Geophysical Research: Atmospheres, 101(1), 1361–1377, doi:10.1029/95JD02028, 1996.

Prather, M. J. and Hsu, J.: Coupling of Nitrous Oxide and Methane by Global Atmospheric Chemistry, Science, 330(6006), 952–954, doi:10.1126/science.1196285, 2010.

Prinn, R., Cunnold, D., Rasmussen, R., Simmonds, P., Alyea, F., Crawford, A., Fraser, P. and Rosen, R.: Atmospheric emissions and trends of nitrous oxide deduced from 10 years of ALE–GAGE data, Journal of Geophysical Research: Atmospheres, 95(11), 18369–18385, 1990.

Raghoebarsing, A. A., Pol, A., van de Pas-Schoonen, K. T., Smolders, A. J. P., Ettwig, K. F., Rijpstra, W. I. C., Schouten, S., Damsté, J. S. S., Op den Camp, H. J. M., Jetten, M. S. M. and Strous, M.: A microbial consortium couples anaerobic methane oxidation to denitrification, Nature, 440(7086), 918–921, 2006.

Reay, D. S., Davidson, E. A., Smith, K. A., Smith, P., Melillo, J. M., Dentener, F., and Crutzen, P. J.: Global agriculture and nitrous oxide emissions, Nature Climate Change, 2, 410-416, 2012.

Regina, K., Syväsalo, E., Hannukkala, A. and Esala, M.: Fluxes of N_2O from farmed peat soils in Finland, European Journal of Soil Science, 55(3), 591–599, 2004.

Ren, W., Tian, H., Liu, M., Zhang, C., Chen, G., Pan, S., Felzer, B. and Xu, X.: Effects of tropospheric ozone pollution on net primary productivity and carbon storage in terrestrial ecosystems of China, Journal of Geophysical Research: Atmospheres, 112(22), 2007.

Ren, W., Tian, H., Xu, X., Liu, M., Lu, C., Chen, G., Melillo, J., Reilly, J. and Liu, J.: Spatial and temporal patterns of CO_2 and CH_4 fluxes in China's croplands in response to multifactor environmental changes, Tellus B, 63(2), 222–240, 2011.

Ridgwell, A. J., Marshall, S. J. and Gregson, K.: Consumption of atmospheric methane by soils: A process-based model, Global Biogeochemical Cycles, 13(1), 59–70, doi:10.1029/1998GB900004, 1999.

Ringeval, B., de Noblet-Ducoudré, N., Ciais, P., Bousquet, P., Prigent, C., Papa, F. and Rossow, W. B.: An attempt to quantify the impact of changes in wetland extent on methane emissions on the seasonal and interannual time scales, Global Biogeochemical Cycles, 24(2) , 2010.

Rinne, J., Riutta, T., Pihlatie, M., Aurela, M., Haapanala, S., Tuovinen, J.-P., Tuittila, E.-S. and Vesala, T.: Annual cycle of methane emission from a boreal fen measured by the eddy covariance technique, Tellus B, 59(3), 449–457, doi:10.1111/j.1600-0889.2007.00261.x, 2007.

Ruser, R., Flessa, H., Schilling, R., Beese, F. and Munch, J. C.: Effect of crop-specific field management and N fertilization on N_2O emissions from a fine-loamy soil, Nutrient Cycling in Agroecosystems, 59(2), 177–191, doi:10.1023/A:1017512205888, 2001.

Saikawa, E., Prinn, R. G., Dlugokencky, E., Ishijima, K., et al.: Global and regional emissions estimates for N_2O, Atmospheric Chemistry and Physics Discussion, 13, 19471-19525, 2013.

Saikawa, E., Schlosser, C. A. and Prinn, R. G.: Global modeling of soil nitrous oxide emissions from natural processes, Global Biogeochemical Cycles, 27(3), 972–989, doi:10.1002/gbc.20087, 2013.

Schulze, E. D., Luyssaert, S., Ciais, P., Freibauer, A., Al, I. A. J. et, Soussana, J. F., Smith, P., Grace, J., Levin, I., Thiruchittampalam, B., Heimann, M., Dolman, A. J., Valentini, R., Bousquet, P., Peylin, P., Peters, W., Rödenbeck, C., Etiope, G., Vuichard, N., Wattenbach, M., Nabuurs, G. J., Poussi, Z., Nieschulze, J. and Gash, J. H.: Importance of

methane and nitrous oxide for Europe's terrestrial greenhouse-gas balance, Nature Geosci, 2(12), 842–850, 2009.

Song, C., Xu, X., Tian, H. and Wang, Y.: Ecosystem–atmosphere exchange of CH_4 and N_2O and ecosystem respiration in wetlands in the Sanjiang Plain, Northeastern China, Global Change Biology, 15(3), 692–705, 2009.

Spahni, R., Wania, R., Neef, L., van Weele, M., Pison, I., Bousquet, P., Frankenberg, C., Foster, P. N., Joos, F., Prentice, I. C. and van Velthoven, P.: Constraining global methane emissions and uptake by ecosystems, Biogeosciences, 8(6), 1643–1665, doi:10.5194/bg-8-1643-2011, 2011.

Steudler, P. A., Bowden, R. D., Melillo, J. M. and Aber, J. D.: Influence of nitrogen fertilization on methane uptake in temperate forest soils, Nature, 341(6240), 314–316, 1989.

Strack, M., Kellner, E. and Waddington, J. M.: Dynamics of biogenic gas bubbles in peat and their effects on peatland biogeochemistry, Global Biogeochemical Cycles, 19(1) , doi:10.1029/2004GB002330, 2005.

Svensson, B. H. and Rosswall, T.: In situ Methane Production from Acid Peat in Plant Communities with Different Moisture Regimes in a Subarctic Mire, Oikos, 43(3), 341, doi:10.2307/3544151, 1984.

Tian, H., Chen, G., Zhang, C., Liu, M., Sun, G., Chappelka, A., Ren, W., Xu, X., Lu, C., Pan, S., Chen, H., Hui, D., McNulty, S., Lockaby, G. and Vance, E..: Century-scale response of ecosystem carbon storage to multifactorial global change in the Southern United States, Ecosystems, 15, 674–694, 2012b.

Tian, H., Lu, C., Chen, G., Tao, B., Pan, S., Del Grosso, S., Xu, X., Bruhwiler, L., Wofsy, S.C., Kort, E.A. and Prior, S.A.: Contemporary and projected biogenic fluxes of methane and nitrous oxide in North American terrestrial ecosystems. Frontiers in Ecology and the Environment 10: 528–536. http://dx.doi.org/10.1890/120057, 2012a

Tian, H., Lu, C., Melillo, J., Ren, W., Huang, Y., Xu, X., Liu, M., Zhang, C., Chen, G., Pan, S., Liu, J. and Reilly, J.: Food benefit and climate warming potential of nitrogen fertilizer uses in China, Environ. Res. Lett., 7(4), 2012c.

Tian, H., Melillo, J.M., Lu, C., Kicklighter, D.W., Liu, M., Ren, W., Xu, X., Chen, G., Zhang, C., Pan, S., Liu, J. and Running, S.: China's terrestrial carbon balance: contributions from multiple global change factors. Global Biogeochemical Cycles 25, 2011a.

Tian, H., Xu, X., Liu, M., Ren, W., Zhang, C., Chen, G. and Lu, C.: Spatial and temporal patterns of CH_4 and N_2O fluxes in terrestrial ecosystems of North America during 1979–2008: application of a global biogeochemistry model, Biogeosciences, 7(9), 2673–2694, 2010.

Tian, H., Xu, X., Lu, C., Liu, M., Ren, W., Chen, G., Melillo, J. and Liu, J.: Net exchanges of CO_2, CH_4, and N_2O between China's terrestrial ecosystems and the atmosphere and their

contributions to global climate warming, Journal of Geophysical Research: Biogeosciences, 116(2), doi:10.1029/2010JG001393, 2011b.

Tian, H., Chen, G., Lu, C., Xu, X., Hayes, D.J., Ren W., Pan, S., Huntzinger, D.N., Wofsy, S.C.: North American terrestrial CO_2 uptake largely offset by CH_4 and N_2O emissions: toward a full accounting of the greenhouse gas budget. Climatic Change, DOI 10.1007/s10584-014-1072-9, 2014

Toet, S., Ineson, P., Peacock, S. and Ashmore, M.: Elevated ozone reduces methane emissions from peatland mesocosms, Global Change Biology, 17(1), 288–296, 2011.

Towprayoon, S., Smakgahn, K. and Poonkaew, S.: Mitigation of methane and nitrous oxide emissions from drained irrigated rice fields, Chemosphere, 59(11), 1547–1556, 2005.

US EPA (Environmental Protection Agency): Global Anthropogenic Non-CO_2 Greenhouse Gas Emissions: 1990-2020. 2006. available at <http://www.epa.gov/nonco2/econ-inv/international.html>

US EPA (US Environmental Protection Agency): Inventory of US greenhouse-gas emissions and sinks: 1990–2009. Washington, DC: US Environmental Protection Agency. EPA 430-R-12-001. 2011.

USEPA (US Environmental Protection Agency): Methane and Nitrous Oxide Emissions from Natural Sources. U.S. Environmental Protection Agency, Washington, DC, USA. http://www.epa.gov/methane/sources.html. 2010.

USEPA: Global Anthropogenic Non-CO_2 Greenhouse Gas Emissions: 1990-2020 (Environmental Protection Agency, 2006); available at <http://www.epa.gov/nonco2/econ-inv/international.html>

Ussiri, D. and Lal, R.: Soil Emission of Nitrous Oxide and its Mitigation, New York. P378, 2012.

Van Aardenne, J. A., Dentener, F. J., Olivier, J. G. J., Goldewijk, C. G. M. K. and Lelieveld, J.: A 1°×1° resolution data set of historical anthropogenic trace gas emissions for the period 1890–1990, Global Biogeochemical Cycles, 15(4), 909–928, 2001.

van Groenigen, K. J., Osenberg, C. W. and Hungate, B. A.: Increased soil emissions of potent greenhouse gases under increased atmospheric CO_2, Nature 475, 214–216, 2011.

Veldkamp, E. and Keller, M.: Nitrogen oxide emissions from a banana plantation in the humid tropics, Journal of Geophysical Research: Atmospheres, 102(13), 15889–15898, 1997.

Vitt, D., Bayley, S., Jin, T., Halsey, L., Parker, B. and Craik, R.: Methane and carbon dioxide production from wetlands in boreal Alberta. Report on Contract No. 90-0270 to Alberta Environment, 35p, 1990

Wagner-Riddle, C., Thurtell, G. W., Kidd, G. K., Beauchamp, E. G. and Sweetman, R.: Estimates of nitrous oxide emissions from agricultural fields over 28 months, Canadian Journal of Soil Science, 77(2), 135–144, doi:10.4141/S96-103, 1997.

Walter, B. P. and Heimann, M.: A process-based, climate-sensitive model to derive methane emissions from natural wetlands: Application to five wetland sites, sensitivity to model parameters, and climate, Global Biogeochemical Cycles, 14(3), 745–765, 2000.

Wang, L., Tian, H., Song, C., Xu, X., Chen, G., Ren, W., and Lu, C.: Net exchanges of CO_2, CH_4 and N_2O between marshland and the atmosphere in Northeast China as influenced by multiple global environmental changes. *Atmospheric Environment,* 63, 77-85, 2012.

Wania, R., Melton, J. R., Hodson, E. L., Poulter, B., Ringeval, B., Spahni, R., Bohn, T., Avis, C. A., Chen, G., Eliseev, A. V., Hopcroft, P. O., Riley, W. J., Subin, Z. M., Tian, H., Brovkin, V., van Bodegom, P. M., Kleinen, T., Yu, Z. C., Singarayer, J. S., Zürcher, S., Lettenmaier, D. P., Beerling, D. J., Denisov, S. N., Prigent, C., Papa, F. and Kaplan, J. O.: Present state of global wetland extent and wetland methane modeling: methodology of a model intercomparison project (WETCHIMP), Geoscientific Model Development Discussions, 5(4), 4071–4136, 2012.

Wei, Y., Liu, S., Huntzinger, D.N., Michalak, A.M., Niovy, N., Post, W.M., Schwalm, C.R., Schaefer, K., Jacobson, A.R., Lu, C., Tian, H., Ricciuto, D.M., Cook, r.B., Mao, J., Shi, X. : The North American Carbon Program Multi-scale Synthesis and Terrestrial Model Intercomparison Project – Part 2: Environmental driver data. Geoscientific Model Development Discussion, 6, 5375-5422, 2013.

Weitz, A. M., Veldkamp, E., Keller, M., Neff, J. and Crill, P. M.: Nitrous oxide, nitric oxide, and methane fluxes from soils following clearing and burning of tropical secondary forest, Journal of Geophysical Research: Atmospheres, 103(21), 28047–28058, 1998.

Weitz, A., Linder, E., Frolking, S., Crill, P. . and Keller, M.: N_2O emissions from humid tropical agricultural soils: effects of soil moisture, texture and nitrogen availability, Soil Biology and Biochemistry, 33(7–8), 1077–1093, 2001.

Whalen, S. C. and Reeburgh, W. S.: A methane flux time series for tundra environments, Global Biogeochemical Cycles, 2(4), 399–409, 1988.

Xu, X. and Tian, H.: Methane exchange between marshland and the atmosphere over China during 1949–2008, Global Biogeochemical Cycles, 26(2), 2012b.

Xu, X., Tian, H. and Hui, D.: Convergence in the relationship of CO_2 and N_2O exchanges between soil and atmosphere within terrestrial ecosystems, Global Change Biology, 14(7), 1651–1660, 2008.

Xu, X., Tian, H., Chen, G., Liu, M, Ren, W., Lu, C. and Zhang, C.: Multifactor controls on terrestrial N_2O flux over North America from 1979 through 2010, Biogeosciences, 9(4), 1351–1366, 2012a.

Xu, X., Tian, H., Zhang, C., Liu, M. L., Ren, W., Chen, G., Lu, C. and Bruhwiler, L.: Attribution of spatial and temporal variations in terrestrial methane flux over North America, Biogeosciences Discussions, 7(4), 5383–5428, 2010.

Yan, X., Akiyama, H., Yagi, K. and Akimoto, H.: Global estimations of the inventory and mitigation potential of methane emissions from rice cultivation conducted using the 2006 Intergovernmental Panel on Climate Change Guidelines, Global Biogeochemical Cycles, 23(2) , 2009.

Yu, K., Faulkner, S. P. and Baldwin, M. J.: Effect of hydrological conditions on nitrous oxide, methane, and carbon dioxide dynamics in a bottomland hardwood forest and its implication for soil carbon sequestration, Global Change Biology, 14(4), 798–812, 2008.

Zhang, C., Tian, H., Chappelka, A. H., Ren, W., Chen, H., Pan, S., Liu, M., Styers, D. M., Chen, G. and Wang, Y.: Impacts of climatic and atmospheric changes on carbon dynamics in the Great Smoky Mountains National Park, Environmental Pollution, 149(3), 336–347, 2007.

Zhang, C., Tian, H., Chen, G., Chappelka, A., Xu, X., Ren, W., Hui, D., Liu, M., Lu, C., Pan, S., Lockaby, G.: Impacts of Urbanization on Carbon Balance in Terrestrial Ecosystems of the Southern United States. Environmental Pollution 164, 89-101. doi:10.1016/j.envpol.2012.01.020, 2012.

Zhang, F., Qi, J., Li, F. M., Li, C. S., and Li, C. B.: Quantifying nitrous oxide emissions from Chinese grasslands with a process-based model, Biogeosciences, 7, 2039-2050, 2010

Zhuang, Q., Lu, Y. and Chen, M.: An inventory of global N_2O emissions from the soils of natural terrestrial ecosystems, Atmospheric Environment, 47, 66–75, 2012.

Zhuang, Q., Melillo, J. M., Kicklighter, D. W., Prinn, R. G., McGuire, A. D., Steudler, P. A., Felzer, B. S. and Hu, S.: Methane fluxes between terrestrial ecosystems and the atmosphere at northern high latitudes during the past century: A retrospective analysis with a process-based biogeochemistry model, Global Biogeochemical Cycles, 18(3), 2004.

1208 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)				
19°E, 68°N	Mire	6.1737	0.13-30.5	Svensson et al., 1984
147.85W,64.8667N	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 - -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 *	11.8±2.6 *	Lu et al., 2012
112°11'E,23°11'N	Tropical forest	-0.115*	-0.145±0.02*	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 – -0.38±0.05 #	-0.13±0.01 - -0.45±0.14 #	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
24.15°E,61.8°N	Boreal Fen	26.67	0 - 65.54	Haapanala et al., 2006
24.02°E,61.83°N	Boreal Fen	29.5	6.55 - 22.93	Rinne et al., 2007
88.42°E,21.53°N	Flood Forest	35.85	-0.50 - 65.84	Mukhopadhyay et al., 2002

N_2O fluxes (g $N/m^2/yr$)				
63º 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; Kristell
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; Weitz 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
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
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
75°W,45°N	Cropland	0.041	-0.88 - 5.07	Desjardins et al., 2012
76.22°W,42.42°N	Cropland	0.074	0.079	Molodovskaya et al., 2012
80.58°W,43.62°N	Cropland	0.135±0.2	-0.04 - 1.99	Wagner-Riddle et al., 1997
8.45°W,51.58°N	Cropland	0.01 - 0.03	0.04 - 0.18	Kim et al., 2010

* unit is g C/m^2/growing season; # unit is mg C/m^2/day.

1212 Table 2 Annual change rates of multiple environmental factors for different continents from
1213 1981 to 2010

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

1214 Note: * indicates significant change trend at P<0.05 and ** indicates significant trend at P<0.01.

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1217 Table 3 Annual change rates of multiple environmental factors for different climate zones from
1218 1981 to 2010

Environmental factors	Northern polar	Northern temperate	Northern tropics	Southern tropics	Southern temperate
Air temperature (^{o}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)	0.26**	5.07**	8.57**	2.75**	0.72**
N fertilizer use (g N/m^2/yr)	-0.082**	0.068**	0.10**	0.040**	0.02**
Ozone (ppb-hr/month)	-1.67	32.80*	50.78**	3.89*	0.0008
Cropland (10^4 km^2/yr)	-0.01*	-0.42*	2.72**	0.83**	0.70**

1219 Note: * indicates significant change trend at P<0.05 and ** indicates significant trend at P<0.01.

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1225 Table 4 Average terrestrial CH_4 (Tg C/yr; 1 Tg = 10^{12} g) and N_2O (Tg N/yr) fluxes and their
1226 change rates (%) for different climate zones during 1981-2010 as simulated by the DLEM

Variables	Time period*	Northern polar zone	Northern temperate zone	Northern tropical zone	Southern tropical zone	Southern temperate zone
CH_4 fluxes	1980s	9.89±0.24[&]	18.44±0.36	39.08±0.81	72.55±0.80	0.16±0.08
	2000s	10.80±0.26	19.76±0.57	41.65±0.95	75.27±0.49	0.23±0.12
	Mean	10.30±0.22	19.20±0.38	40.60±0.64	73.80±0.65	0.21±0.06
	Change rate (%)	9.22	7.14	6.56	3.75	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

1227 * Mean annual fluxes in the period 1981-1990, 2001-2010 and 1981-2010. [&]The values are
1228 denoted as mean ± 2 SE (standard error).
1229

1230 Table 5 Average terrestrial CH_4 (Tg C/yr) and N_2O (Tg N/yr) fluxes for different biome types
1231 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	30.82 ± 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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1233 Table 6 Comparison of global terrestrial CH_4 and N_2O fluxes estimated from multiple sources

CH_4 flux (Tg C/yr)	**Time period**	**This study**	**Others**	**Reference**
Global	1990s	139.57-153.69	86-195	Fung et al. 1991; Houweling et al. 1999
Global natural wetland	1993-2004	129.70-135.92	106-198	Melton et al. 2012
	1990s	126.47-135.92	69.0-202.5	Cao et al., 1998; Bousquet 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 - -16.13	-19.5 - -25.0	Ito and Inatomi 2012; Potter et al. 1996; Ridgwell et al. 1999; Del Grosso et al. 2000
Rice paddy land	1990s	29.30-35.17	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
N_2O flux (Tg N/yr)	**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©	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*	Zhuang et al. 2012
	1990	8.76	6.6 (3.3-9.0)$	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#	FAO/IFA 2001
	1995	3.29	2.8	Bouwman et al. 2002

	2000	3.50	4.4§	US EPA 2011
	2000	3.50	2.6-3.5[&]	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

[©] 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; [#] 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.

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

Fig. 2 Comparison between the DLEM -simulated CH_4 and N_2O emissions with inversion estimates. (a) CH_4 fluxes in wetlands and soil (inversion result is from CarbonTracker, http://www.esrl.noaa.gov/gmd/ccgg/carbontracker-ch4/); (b), N_2O emission from agricultural and natural soils (inversion result is from Saikawa et al., 2013). Error bar denotes standard deviation.

Fig. 3 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

Fig. 4 Interannual variations in CH_4 (A, Tg C/yr) and N_2O (B, Tg N/yr) fluxes in the global terrestrial ecosystems during 1981-2010. Note: the gray area is the estimated uncertainty range with a mean ± 95% confidence interval.

Fig. 5 DLEM-simulated mean global distributions of CH_4 (top, g $C/m^2/yr$) and N_2O (bottom, g $N/m^2/yr$) fluxes during 1981-2010. Note: The latitudinal (per degree latitude) distribution (Tg C/yr and Tg N/yr for CH_4 and N_2O, respectively) patterns are shown at the right.

Fig. 6 Interannual variations of CH_4 (Tg C/yr) and N_2O (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^oN-90^oN); B: the northern temperate zone (30^oN-60^oN); C: the northern tropical zone (0^oN-30^oN); D: the southern tropical zone (0^oS-30^oS); E: the southern temperate zone (30^oS-60^oS)

Fig. 7 Interannual variations of CH_4 (Tg C/yr) and N_2O (Tg N/yr) 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

Fig. 8 Interannual variations in CH_4 (Tg C/yr) and N_2O (Tg N/yr) anomalies (relative to the mean value for the period 1981-2010) for different continents during 1981-2010.