Modelling biological nitrogen fixation in global natural terrestrial ecosystems

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Abstract. Biological nitrogen fixation plays an important role in the global nitrogen cycle. However, the fixation rate has been usually measured or estimated at a particular observational site. To quantify the fixation amount at the global scale, process-based models are needed. This study develops a biological nitrogen fixation model to quantitatively estimate nitrogen fixation rate by plants in natural environment. The revised nitrogen module better simulates the nitrogen cycle in comparison with our previous model that has not considered the fixation effects. The new model estimates that tropical forests have the highest fixation rate among all ecosystem types, which decreases from the equator to the polar region. The estimated nitrogen fixation in global terrestrial ecosystems is 61.5 Tg N yr^{-1} with a range of 19.8 - 107.9 Tg N yr^{-1} in the 1990s. Our estimates are relatively low compared to some early estimates using empirical approaches, but comparable to more recent estimates that involve more detailed processes in their modelling. Furthermore, the contribution of nitrogen made by biological nitrogen fixation depends on ecosystem type and climatic conditions. This study highlights that there are relatively large effects of biological nitrogen fixation on ecosystem nitrogen cycling and the large uncertainty of the estimation calls for more comprehensive understanding of biological nitrogen fixation. More direct observational data for different ecosystems to improve future quantification of fixation and its impacts.

1. Introduction

In most terrestrial ecosystems, nitrogen (N) available for plants is generally limited although it is the most abundant element in the atmosphere (LeBauer and Tresder, 2008). Nitrogen usually enters terrestrial ecosystems through processes of nitrogen deposition and from biological N fixation (BNF). Nitrogen deposition is a physical process, representing the direct input of reactive nitrogen including organic N, ammonia, and nitrogen oxides (NO_y) including nitric oxide (NO), nitrogen dioxide (NO_2), nitric acid (HNO_3) and organic nitrates from the atmosphere to biosphere. BNF, a biochemical process that converts nonreactive nitrogen (N_2) to reactive nitrogen, provides a liaison between the atmosphere and biological systems. Lightning is also a way to convert N_2, adding 3-5 Tg N yr^{-1} to terrestrial ecosystems (Levy and Moxim, 1996). Nitrogen input via rock weathering is another important source for terrestrial ecosystems, adding 3-10 kg N ha^{-1} yr^{-1}(Morford et al., 2011; Houlton et al., 2018). BNF is significantly greater than lightening induced N fixation (Galloway et al., 1995). On a global scale, anthropogenic nitrogen to the environment could be more than 160 Tg N yr-1 (Gruber and Galloway, 2008), which is even greater than terrestrial N fixation (~110 Tg N yr^{-1}). However, taken together, natural N fixation is the primary source in the absence of human activities to global terrestrial ecosystems. For natural terrestrial ecosystems, the amount of N added is approximately balanced by the nitrogen converted back to the atmosphere (Stedman and Shetter, 1983) and lost into ocean and other aquatic systems.

Once entering terrestrial ecosystems, N can be taken up by plants and microbes, and converted into other oxidized forms through mineralization, nitrification and denitrification. In terrestrial ecosystems, N fixation generally affects the nitrogen cycle and nutrient level to constrain plant productivity. Any change of nitrogen input to terrestrial ecosystems will influence their soil nitrogen content.

In the process of BNF, N_2 is converted to ammonia by certain soil microorganisms which can then be utilized by and incorporated into plants. In natural environment, N fixation is conducted by two types of microorganisms:

asymbiotic organisms, including blue-green algae, lichens and free-living soil bacteria (Belnap, 2002; Granhall & Lid-Torsvik, 1975), and symbiotic organisms, such as fungi and nodule forming Rhizobium species. Among them, the most dominant fixers are leguminous plants and their N fixation mechanisms are also best known (Sullivan et al., 2014; Vitousek et al., 2013). To date, the amount of N fixation by legumes is estimated in the range of 11.3-33.9 kg N ha^{-1} yr^{-1} (2.8~8.4 g m^{-2} yr^{-1}) in natural terrestrial ecosystems. A symbiotic relationship exists between legume plants and bacteria. In the process, legume plants provide the bacteria energy through photosynthesis, meanwhile, the bacteria around the rhizobia ~~-~~supply the legume N in the form of ammonia.

This study first models the BNF from the symbiotic relationship between legume plants and bacteria, then analyzes factors influencing the fixation rate from different terrestrial ecosystem soils.

2. Methods

2.1 Overview

We first develop a BNF model and then couple the model with an earlier version of biogeochemistry model quantifying soil carbon and nitrogen dynamics (Yu and Zhuang, 2019). The revised model is then used to quantify the BNF at regional and global scales in natural terrestrial ecosystems. The BNF rate estimates consider the effects of environmental conditions including temperature, soil moisture, soil mineral nitrogen content and soil carbon content. The modified model is calibrated and evaluated with observed N fixation rate data from published studies for various natural terrestrial ecosystems from the Arctic to tropical ecosystems. The model sensitivity to model input is analyzed. The model is then extrapolated to the global terrestrial ecosystems at a monthly step and a spatial resolution of 0.5° by 0.5° for the final decade of the 20th century. The effects of physical conditions on BNF are then analyzed.

2.2 Model description

The Terrestrial Ecosystem Model (TEM) is a process-based model that simulates carbon and nitrogen dynamics, hydrological and thermal processes for terrestrial ecosystems. Although many efforts were made to incorporate more details of the N cycle, the N input from the atmosphere to ecosystems has not fully been incorporated to date, especially the BNF as input. Here we improve the N dynamics within TEM by considering N fixation by legumes. The model schematic and other calculation~~s~~ including carbon cycle and nitrogen cycle are inherited from an earlier version of TEM (Zhuang et al., 2003; Yu and Zhuang, 2019).

BNF is the most significant process in either symbiotic or non-symbiotic forms converting stable molecular N_2 into N chemical compounds that are available to plants. For most terrestrial ecosystems, N fixer could be in many forms, such as free-living bacteria, lichens, and blue algae. But among them, symbiotic BNF is a dominant process to provide biologically accessible N, and most systematical BNF is regulated by legume plants, especially in croplands and semi-natural environment (Mus et al., 2016). In natural environment, contribution from legume can be significant but with large uncertainties, which is greatly determined by various environmental conditions

(Lindemann and Glover, 1996). In this study, the N fixation via legume plants is modeled considering (1) the accessible N concentration in soils, (2) the limitation of temperature, (3) soil water status, (4) the carbon demand for N fixation, and (5) the percentage of N fixing plants for each ecosystem type as:

$$N_{fix} = N_{fixpot} \, f_t \, f_W \, f_N \, f_c \, f_{plant} \tag{1}$$

where N_{fix} is the nitrogen fixation rate, N_{fixpot} is the potential N fixation rate (g N day^{-1}) , f_t is the influence function of soil temperature, f_W is the soil water function, f_N is the function of root substrate N concentration, f_C is the function of plant carbon availability, and f_{plant} is the function of legume plant coverage. Please refer to Table 4 for value range of related parameters.

The potential N fixation is highly related to the total N demand of plants and the available nitrogen in soils. Theoretically, the definition of potential N fixation rate should be the difference between the demand and supply of N. Both of them vary with plant types, stages of growth and soil conditions. For large spatial-scale simulations for various ecosystem types, it is impossible to derive potential N fixation because of data availability. N_{fixpot} can be estimated based on dry matter of root, nodule or plant dry matter (Voisin et al, 2003, 2007). However, root biomass is also difficult to measure directly. In most published studies, the potential nitrogen fixation rate was measured using an acetylene reduction array (ARA) method (Hardy et al, 1968, 1973), and some used ^{15}N methods (Shearer and Kohl, 1986). In our simulation, N_{fixpot} is assumed to be a constant for each ecosystem type. The N_{fixpot} range is determined from literature and specific values for various ecosystem types are obtained through model parameterization.

Soil temperature is a controlling factor for both microbial activities and plant growth. A large number of studies show that different plants have slightly different preferences for temperature (Montanez et al, 1995; Breitbarth et al., 2007; Gundale et al., 2012). For soybean, 20-35 °C is optimal (Boote et al., 2008), and for white clover the optimal temperature can be 13-26 °C (Wu and McGechan, 1999). The activity of microbes responds slightly differently to temperature among species. For most of them, the optimum temperature is 20-25 °C, and at 12-35 °C the activity is not limited. Generally, BNF increases as the temperature rises from minimum temperature (0-5 °C) for N fixation to optimal temperature, maximum rate occurs within an optimal range (15-25°C), and decreases from optimal to maximum temperature above which BNF will stop at 35-40 °C:

$$f_t = \begin{cases} 0 & when \ \ (t < t_{min} \ or \ t > t_{max}) \\ \frac{t - t_{min}}{t_{optL} - t_{min}} & when \ \ (t_{min} \le t < t_{optL}) \\ 1 & when \ \ (t_{optL} \le t \le t_{optH}) \\ \frac{t_{max} - t}{t_{max} - t_{optH}} & when \ \ (t_{optH} < t \le t_{max}) \end{cases} \tag{2}$$

where the upper limit (t_{max}) is set to 45 °C. There is no lower limit, but when t is low enough, f_t will be close to zero (Wu and McGechan, 1999; Boote et al., 2008; Holzworth et al., 2014) (Table 1). For the convenience in computing, a lower limit is set in our model. When the temperature goes beyond its upper or lower limit, f_t is assumed to be 0.

Water stress has a direct effect on nitrogen fixing system (Sprent, 1972). With proper temperature, soil moisture condition is the major factor controlling nitrogen fixation rate (Srivastava and Ambasht, 1994). Soil water deficit and flood dramatically inhibits N fixation because of drought stress and oxygen deficit, respectively (Omari et al., 2004; Mario et al., 2007). In our model, the water factor is linearly related with soil water content (Williams, 1990; Wu and McGachan, 1999):

$$f_w = \begin{cases} 0 \ when \ (W_f \leq W_a) \\ \varphi_1 + \varphi_2 \ when \ (W_a < W_f < W_b) \\ 1 \ when \ (W_f \geq W_b) \end{cases} \tag{3}$$

where W_f (J kg $^{-1}$) is the available soil water, which is defined as the ratio of water content to that at the field capacity. In soils, water potential generally includes osmotic and matrix potentials, ranging from -0.1 to -0.3 bar for typical soils, which has little effects on the N fixation. But when the soil gets very dry, the potential can be up to -100 to -200 bar and increases rapidly. W_a is the bottom threshold below which N fixation is totally restricted by soil moisture. W_b is the upper threshold above which nitrogen fixation is not limited by soil moisture. φ_1 and φ_2 are parameters representing the linear relationship between soil water content and its effect on N fixation, respectively (Table 1).

It is generally thought that more substrate N in soils will slow down the N fixation, because plants can uptake N directly from soil with less energy (Vitousek and Field, 1999). By comparison, N fixation needs more energy and consumes more carbon than plant N uptake does. Thus, the N fixation is only considered to occur when the direct N uptake from soil cannot meet the plant N demand. In our model, the inhibition effect of N is defined as (Wu and McGehan, 1999):

$$f_N = \begin{cases} 1 - f_{Nup} ln(1000 - N_s) & when \ (N_s \geq 0.001) \\ 1 & when \ (N_s < 0.001) \end{cases} \tag{4}$$

Where f_{Nup} is a parameter related to legume biological N fixation and soil N. N_S is the soil mineral N (g N m^{-2}). BNF efficiency shows a natural logarithmic relation with the soil mineral N.

N fixers get photosynthetic carbohydrate support from plants. Because the product of every unit of nitrogen fixed consumes a certain amount of carbon, the lack of carbon supply will inhibit the N fixation. The carbon cost for per unit of fixed N varies widely depending on environmental conditions and ecosystem types. For example, the consumption of carbon is only 1.54 times of fixed N for cowpea (Layzell et al., 1979), and it can be 6.3 to 6.8 times for soybeans (Ryle et al., 1979). It is also related to the life cycle of plants. The carbon effect is modeled following a Michaelis-Menten equation (Boote et al., 1998):

$$f_C = \frac{1}{1 + K_c / C_r} \tag{5}$$

where C_r is the soil carbon content (g C m^{-2}) to represent carbon availability from plants to N fixers. K_c is the Michaelis-Menten constant, which is plant species dependent.

2.3 Data

The classification of land cover and leguminous biomes were derived from the combination of the International Geosphere and Biosphere (IGP) land-cover classification system and the study of Schrire et al (2005). The experimental N_2 fixation data for model calibration were collected for 7 major ecosystem types. Nitrogen fixation rates were determined with acetylene reduction assay (ARA) method in most published studies (Table 2, data were from Cleveland et al. (1999)), expressed in kg N m^{-2} yr^{-1}. Some of them were measured with the ^{15}N natural abundance technique.

The parameters for N_2 fixation module were initialized with a priori values (Table 2). Ecosystem-specific and microbe guild-specific parameters were inherited from previous TEM model (Zhuang et al., 2003; Yu and Zhuang, 2019). The global simulations were conducted at a spatial resolution of 0.5 by 0.5 degree and at a monthly time step. Historical climate data including temperature, precipitation, cloudiness and water vapor pressure were derived from the Climate Research Unit (CRU) (Mitchell and Jones, 2005). Soil texture data were from Melillo et al. (1993) and Zhuang et al. (2003). Other initial conditions including vegetation properties, soil carbon content and soil nitrogen contents were from Chen and Zhuang (2013) and Zhuang et al. (2012).

For regional simulations, the total amount of fixed N was also influenced by legume coverage. For each ecosystem type, we estimated the coverage according to the distribution of legume plants and field studies (Table 3, the coverage data are compiled from Cleveland et al. (1999)), where the minimum and maximum values were derived from the abundance of N-fixers.

2.4 Model calibration and site-level validation

Most model parameters are legume-specific or vegetation-specific and are adjusted based on value ranges from previous studies (Table 1). Model is parameterized for 7 representative natural terrestrial ecosystems (Table 2). Root mean square error (RMSE) and coefficient of determination ($0 \leq R^2 \leq 1$) were used for model calibration. RMSE was calculated to show the mean difference between simulated data and observational values. The model is iterated with changing parameters until the RMSE reached a certain value for each site. Most parameters in the model driving nitrogen cycle in the soil have been defined and calibrated in previous studies (Yu and Zhuang, 2019).

2.5 Model sensitivity and uncertainty Analysis

The response of N fixation of different biomes to input data and parameters was analyzed using sensitivity testing. Four major input variables were selected, including air temperature, precipitation, soil nitrogen content and soil organic carbon content. The monthly average input variables were changed by ±10% of the original level for each site and each grid. The variables were changed at 6 levels, respectively, and the rest of input variables were kept at their original values. The sensitivity was calculated by comparing the simulated annual nitrogen fixation to the simulations with the original input values.

3. Results

3.1 Model evaluation

To evaluate the model, thirty-five observational sites were selected for 7 major ecosystem types across the globe, representing different climate and soil conditions. The experimental data of N fixation have a mean value of 12.9 kg N ha^{-1} yr^{-1}, with a standard deviation of 17.7 kg N ha^{-1} yr^{-1}. The maximum observed fixation occurred in temperate forest in New Zealand, while the minimum rate was also for temperate forest in Idaho State of the US. Our simulations are comparable with the observed data for all major ecosystem types with the coefficient of determination (R^2) of 0.44 and with a slope of 0.46 (Figure 2). The regression results are mainly influenced by some observed data greater than 30 kg N ha^{-1} yr^{-1}. By removing the outliers of observational data, the slope of regression increases to 0.72. Observational data for temperate forests show the greatest variation among all major ecosystem types, with a maximum value reaching 800 times of the minimum one. Simulations are closer to the observations across sites in temperate forests with R^2 of 0.26 and slope of 0.42. Our model underestimated nitrogen fixation rate in temperate forests. The large variation in observations may be due to the distribution of legume plants, different sampling time periods (e.g., growing and non-growing seasons), and varying climate conditions. For tropical forests, our model estimates of N fixation are higher than observations with the slope of 0.75 and R^2 of 0.44.

3.2 Model sensitivity analysis

The model sensitivity analysis quantifies the impact of changes in forcing data on nitrogen fixation rate. Climate conditions including air temperature and precipitation, and soil characteristics of nitrogen content and carbon content varied at 3 levels to examine the sensitivity. The response of nitrogen fixation rate emissions is quantified for each ecosystem type. The sensitivity test was conducted for all observational sites (Table 2). Temperature is the most sensitive variable (Figure 1). Nitrogen fixation is more sensitive to the change of all forcing conditions. Increasing soil nitrogen results in a lower N fixation. Abundant soil nitrogen content inhibits BNF activity, but stimulates nitrification and denitrification processes.

3.3 Biological nitrogen fixation in global terrestrial ecosystems

Tropical forests in South America, Central Africa and South Asia show a wide range of N fixation rate between 1 and 200 kg N ha^{-1} yr^{-1} (Bruijnzeel et al, 1991). Here all plants in tropical rainforest are assumed to fix nitrogen and one set of parameters are applied for all tropical forests. The coverage for tropical forests in the landscape was assumed to be 15% (Cleveland et al., 1999), ranging from 5% to 25%. The N_2 fixation rate was estimated to be 18.2 kg N ha^{-1} yr^{-1}, which is the highest among all vegetation types. Our simulations show that the total fixed nitrogen ranges from 10.8 Tg N yr^{-1} to 54 Tg N yr^{-1}, with the average value of 32.5 Tg N yr^{-1}(Table 3). Nitrogen fixation in tropical forests is almost half of the global total amount and a principal contributor of BNF in natural ecosystems. Tropical forests have the largest potential to fix nitrogen given that the optimal temperature and soil moisture for BNF is relatively easy to have under tropical climatic conditions.

Temperate forests cover the largest land area from 30°N to 60°N, including temperate coniferous forest, temperate deciduous forest and temperate evergreen forest. Temperate areas have the majority of legumes and many temperate ecosystems are considered to be N limited. Comparing to other ecosystem types in temperate regions, conifers are likely to limit the reproduction of legumes (Wheatley et al, 2010). In general, plant species carrying nitrogen fixers are only distributed in a small percentage of natural temperate forests, like clear-felled areas and pastures (Boring and Swank, 1984). Cleveland et al. (1999) indicated that the legume coverage ranges from 1% to 10% of the land area only. Consequently, our simulations indicate that N fixation by temperate forests was 12.7 kg N ha^{-1} yr^{-1}. The estimates of the total nitrogen fixation were between 1.9 and 19.14 Tg N yr^{-1} (Table 3). Nitrogen fixation in temperate areas contributes 12.5% of the global total amount.

Savanna covers over a half of African continent, Australia and large areas of South America. It is an important biome in the Southern Hemisphere. There is a great variation in native legume species. Only in humid savanna, legumes may significantly contribute to the increase of soil nitrogen (Cech et al., 2008). On average, 15% of the vegetation in savanna is considered as legume grass and biological nitrogen fixation occurs when precipitation is greater than 10 mm per month. Generally, nitrogen fixation in savanna is restricted by soil moisture, while temperate grassland is limited by both temperature and soil moisture (Bustamante et al, 1970). Nitrogen fixers are not abundant for these biomes (Woodmansee et al., 1981). The coverage of nitrogen fixers was assumed to be from 5% to 25%, (Cleveland et al., 1999). Our simulation assumed that nitrogen fixers cover 15% of the land, resulting in 1.9 kg N ha^{-1} yr^{-1} fixation, representing a much smaller fraction compared to forest ecosystems. Total fixed nitrogen in grasslands appeared to range from 0.62 to 3.1 Tg N yr^{-1}, with an average of 1.86 Tg N yr^{-1}. For savanna, the total contribution was less due to its relatively small area. The minimum, average and maximum values were estimated to be 0.45, 1.34 and 2.23 Tg N yr^{-1}, respectively.

In tundra and boreal forest regions, both host plants and their rhizobia are adapted to the environment with low temperature. Nitrogen fixation rate is extremely variable for boreal ecosystems. For tundra, the coverage was assumed to be 3-15%, and for boreal forest, the coverage was 4-18%. But in general, the low temperature and permafrost conditions limit the activity of nitrogen fixers (Alexander, 1981). We estimated that tundra ecosystems fix nitrogen at 3.2 kg N ha^{-1} yr^{-1}. Their total BNF was between 0.51 to 2.55 Tg N yr^{-1} with average of 1.54 Tg N yr^{-1}. In boreal forests, the fixation rate was much lower (2.1 kg N ha^{-1} yr^{-1}) compared to temperate forests.

In deserts, although some legumes may exist in extremely dry conditions, and some species may grow rapidly after rainfall, their fixation could be neglected. However, in semi-arid areas, legumes are common plants with several species, their N fixation is lower than tropical and temperate forests (5.7 kg N ha^{-1} yr^{-1}).

Mediterranean ecosystems such as in southern California and some areas in southern Australia are characterized with mild rainy winter and hot dry summer, containing both evergreen and deciduous shrublands, in which nodulated legumes are prominent (Sprent et al., 2017). These legumes are more active in comparatively wet season than in dry season (Sánchez-Diaz, 2001). The ability to fix nitrogen is considered to be one of the most important features that enable legumes and plants to survive under severe environments (Crisp et al., 2004). We estimated that the N fixation rate of these legume species is similar to that in grasslands (2.7 kg N ha^{-1} yr^{-1}).

Spatially, the highest rate of N fixation occurred in the tropical and sub-tropical areas, as a result of proper climate and soil characteristics for fixers (Figure 3). N fixation from tropical forests and xeric shrubland contributes to nearly half of the global terrestrial amount (Table 3). A lower N fixation rate was in high latitudes of eastern China, North America and Europe, which were mainly covered with temperate forests. Compared to tropical areas, N fixation in temperate regions shows a larger variability depending on vegetation types. The spatial variation could be attributed to the distribution of legume plants, in addition to the difference of humidity and temperature conditions. N fixation in temperate regions accounts for 35% of the total fixed N.

Our model estimated that high BNF rates in growing season is consistent with other regional and global estimates (Cleveland et al., 1999, 2013; Lee and Son, 2005; Lett and Michelsen, 2014). The energetic cost for active N uptake becomes lowest when soil temperature is around 25°C (Fisher et al. 2010). Similarly, our estimates of high BNF rates also occur at similar temperature conditions in spring and summer. The global soil nitrogen mineralization rate was estimated to be 696 Tg N yr^{-1}while 15% of plant N demand was provided by BNF (Cleveland et al., 2013). Our estimates of BNF were lower than the estimates by Cleveland et al. (2013) and fell within 10% of the total soil mineralization rate. This result also indicates that about 10% of the mineralized N was induced by BNF.

During 1990-2000, our simulations show that BNF in natural terrestrial ecosystems is 61.5 Tg N yr^{-1}, but anthropogenic N fixation was much higher at 140 Tg N yr^{-1} (Galloway et al., 2002). This large amount of anthropogenic N input to terrestrial ecosystems is expected to inhibit the natural BNF and might lead to less BNF in the future.

4. Discussion

4.1 Comparison with other estimates of biological nitrogen fixation

There is a large uncertainty in estimating the N input into terrestrial ecosystems, especially from BNF (Sutton et al., 2014) (Table 3). In our study, a calibrated process-based model was applied to estimate site-level and global BNF in natural terrestrial ecosystems. Empirical models provide reasonable estimation based on relationships between fixation rate and environmental factors (e.g. evapotranspiration) (Cleveland et al, 1999), while process-based approaches consider processes in BNF affected by multiple controlling factors (Fisher et al., 2010; Gerber et al., 2008; Meyerholt et al., 2016). Our estimated BNF in the global terrestrial ecosystems is 61.5 Tg N yr^{-1} with an uncertainty ranging from 19.8 to 107.9 Tg N yr^{-1}, which is lower than most existing studies. Cleveland et al. (1999) provided a central value of 195 Tg N yr^{-1} by scaling up field-based experimental data, with a range of 100 - 289 Tg

N yr^{-1}. This range represents potential distribution of nitrogen fixation. In reality, N fixation is also affected by climate and soil conditions, making the actual terrestrial BNF smaller than the potential one. In a more recent study of Cleveland et al. (2013), a total of 127.5 Tg N yr^{-1} was estimated to be related to BNF, based on the relationship between BNF and evapotranspiration (ET). Galloway et al (2002b) also provided several estimates for global BNF. Galloway et al. (2004) further suggested a range of 100 - 290 Tg N yr^{-1} and implied that the true rate of BNF would be at the low end of this range without large-scale human disturbance. In an earlier study (Galloway et al., 2002b), the mean annual global BNF was estimated to be 89-100 Tg N yr^{-1}. By assuming a steady state between N input to and loss from ecosystems, Vitousek et al. (2013) estimated the BNF to be 58 Tg N yr^{-1} with a plausible range of 40 - 100 Tg N yr^{-1}, which is similar to our estimates. However, Xu-Ri and Prentice (2017) estimated that the N fixation was about 340 Tg N yr^{-1} which is almost 5 times larger than our estimates. In their study, BNF was determined by plant N requirement across all biome types.

In our estimation, tropical forests significantly contribute to the total BNF, which is up to 18 kg N $ha^{-1}yr^{-1}$. This result is highly related to the density of leguminous plants, and the physical conditions in tropical areas (Crews, 1999). Our simulated results are comparable to the estimates of symbiotic N_2 fixation from tropical evergreen (5.5-16 kg N ha^{-1} yr^{-1}) and deciduous forests (7.5-30 kg N ha^{-1} yr^{-1}) (Reed et al., 2011). Barron et al. (2010) directly measured N_2-fixing root nodules across lowland tropical forests and their observations also showed a large variation among individual trees. For a mature forest matrix, the average value was around 10 kg N ha^{-1} yr^{-1}, but it could be as high as 200 kg N ha^{-1} yr^{-1} for some areas. Cleveland (2013) provided a similar estimate to ours (around 12 kg N ha^{-1} yr^{-1}), but higher values (20-30 kg N ha^{-1} yr^{-1}) in their earlier studies (Cleveland et al., 1999). Sullivan et al (2014) analyzed human's impact on tropical N fixation and found, depending on forest ages, fixation was 5.7 kg N ha^{-1} yr^{-1} with a range from 1.2 to 14.4 kg N ha^{-1} yr^{-1}, which is lower than our estimates.

For temperate and boreal forests, we estimated that BNF fixation is 2.1-18 kg N ha^{-1} yr^{-1}. The existing BNF estimates from literature also show a large uncertainty for those forest ecosystems. For instance, LM3V-N model (Gerber et al., 2009) suggested that the N input to forests to be less than 5 kg N ha^{-1} yr^{-1}. But their model also estimated that, in moist forests, the uptake of N could be 30-80 kg N ha^{-1} yr^{-1}. Deluca et al. (2002) reported that cyanobacterium and feather moss could act as a supplement to N fixation in boreal forests (0.5 kg N ha^{-1} yr^{-1}) while the organic N accumulation could be 3 kg N ha^{-1} yr^{-1}. For the forests in northwest Rocky Mountain, N fixation amount is on average between 0.5 and 2 kg N ha^{-1} yr^{-1} (Clayton and Kennedy, 1985; Fahey et al., 1988) while Kou-Giesbrecht and Menge's model (2019) estimated the N fixation rate to be 0 -10 kg N ha^{-1} yr^{-1} for temperate forests, and 0 to 6 kg N ha^{-1} yr^{-1} for boreal forests.

There could be a number of reasons for our comparatively lower estimates. The most important one is that there is a considerable uncertainty in estimating the coverage of N fixing plants. High diversity in the distribution of legume plants highly influences the estimation of total plant coverage, because our estimation was based on site-level experimental data. In order to improve our understanding, more investigation on legume plant distribution and associated data for N fixers is needed, especially in the Middle Asia, South America and Africa.

Large variations of BNF rates exist across terrestrial ecosystems spatially (Figure 3). The global BNF spatial pattern is similar to other estimates (Cleveland et al., 1999; Xu-Ri and Prentice, 2017). The highest N fixation rate in tropical regions (more than 50% of the global terrestrial N fixation) is primarily due to their warm and moist soil conditions. Further, N fixed by human activities became increasingly influential in the past century (Galloway et al., 2002), especially in temperate regions due to their large human population. The anthropogenic N deposition contributed more to soil N than BNF did. As a result, soils became N rich, inhibiting BNF in temperate soils. This could explain why the potential N fixation rate was high in temperate ecosystems, but only contributed to 20% of the total fixation.

4.2 Major controls on biological nitrogen fixation

In our simulations, the N fixation was primarily influenced by soil temperature, moisture and soil nitrogen content. The highest N fixation rate in tropical ecosystems is consistent with our sensitivity analysis for temperature and soil moisture. The sensitivity analysis indicated that a 1-3°C increase of temperature led to 7% increase in N fixation rate. Nitrogen cycle responds differently between different biomes and legume types. But in general, increasing temperature will accelerate processes in the N cycle. Soil moisture correlates with BNF in a similar way with temperature. A slightly increase of precipitation (10%) increased the nitrogenase activity. However, the response of N fixation to soil water stress is not as sensitive as that to the change in temperature. Xeric shrubland and savanna in dry tropical areas still contribute greatly to the global N fixation, while the contribution of boreal forests, with low temperature, is much lower.

BNF is highly regulated by soil nitrogen content. N-deficiency conditions usually favor BNF activities, for example, in xeric shrubland and savanna. Enhancing soil N content will decrease the N fixation rate, which is also consistent with our sensitivity analysis. It costs less energy for plants to take up N directly from soils rather than biologically fixing it from the atmosphere (Cannell and Thornley, 2000). However, there is an exception for some areas in tropical ecosystems. Many tropical soils are comparatively rich in nitrogen, but N-fixing plants are still active to compensate the nitrogen depletion due to the rapid N cycling (Pons et al., 2007). This explains why N fertilization inhibits the BNF in temperate ecosystems, but BNF is still active in N-rich soils in tropical ecosystems. In areas where the energetic cost succeeds the demand of N, the BNF rate will be comparatively lower. Sullivan et al. (2014) suggested that there were lower rates of BNF in undisturbed mature forests and higher rate in secondary forests, depending on the balance between N-demand and energy consumption.

4.3 Model limitation and future work

The incorporation of BNF into TEM allows us to more adequately simulate nitrogen cycle from natural terrestrial ecosystems. However, there are several limitations in this study.

First, the current model ignores the effect of free-living BNF. Although symbiotic BNF is critical for most natural and semi-natural ecosystems, asymbiotic organisms play an important role in extreme environments such as waterlogged soils and deserts. The importance of symbiotic BNF or fixation by leguminous plants may not be as significant as previously thought. Elbert et al. (2012) suggested that cryptogam contributed nearly half BNF in terrestrial ecosystems, which was up to 49 Tg N yr^{-1}. In some tropical areas, the spatial N input from free-living bacteria even exceeds symbiotic input (Sullivan et al., 2014). In addition, legumes are not the only source of symbiotic BNF. Some fungi species have the ability to actively fix atmospheric nitrogen. But in most existing models, fungi or mycorrhizae symbioses are not considered due to the limited knowledge about their mechanisms of fixing N (Fisher et al., 2010). A more comprehensive model that covers various types of nitrogen fixation is needed.

Second, the BNF process in our model is calibrated with a limited amount of data, imposing a general set of parameters to all plant species and soil conditions within an ecosystem type. More observational data from natural terrestrial ecosystems is desirable to improve our model.

Third, it is difficult to isolate the N addition via natural processes from human activities. In the US, 20-35% of annual N input into terrestrial ecosystems are human-related (Sobata et al., 2013). As a result, the quality of observational data varies from site to site, some BNF data are only semi-natural. The observational data are imperfect, which might have also biased our estimates through model parameterization process.

5. Conclusions

This study developed a process-based biological nitrogen fixation model and coupled it with an extant biogeochemistry model. The model was evaluated with observed data for N fixation. The model was then extrapolated to the global natural terrestrial ecosystems. Our model estimates that biological nitrogen fixation in natural terrestrial ecosystems was 61.5 Tg N yr^{-1} during the last decade of the 20th century and the greatest fixation rate occurred in tropical regions. Soil temperature, rather than soil moisture and nutrient content, is the most dominant control to the fixation. Lacking the knowledge about the distribution of N fixing plants and their physiological features might have biased our estimates of both biological nitrogen fixation at the global scale.

Data availability

Climate data including monthly cloudiness, precipitation, temperature, and water vapor pressure are from the Climate Research Unit (CRU) http://www.cru.uea.ac.uk/data (last access: May 2017). Global vegetation data and soil data are available in Zhuang et al. (2003) and McGuire et al. (2001). The explicit spatial data on soil water pH from the ORDL gridded soil properties product (https://daac.ornl.gov/cgi-bin/dsviewer.pl?ds_id=546, last access: March ,2017) are based on the World Inventory of Soil Emission Potentials (WISE) database (Batjes, 2000). The global average carbon dioxide concentration is observed at NOAA's Mauna Loa Observatory. N deposit data are from NADP monitor and CASTNET. The initial values of soil microbial carbon and nitrogen, and the ratio of C/ V / N at the global scale, were from a compilation of global soil microbial biomass carbon, nitrogen, and phosphorus data (https://doi.org/10.3334/ORNLDAAC/1264, last access: May, 2017). *The data presented in this paper can be accessed through our research website (http://www.eaps.purdue.edu/ebdl/).*

Author contribution

Q. Zhuang and T. Yu designed the research. T. Yu performed model simulations and data analysis. Both authors contributed to the paper writing.

Competing interests

The authors declare that they have no conflict of interest.

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Table 1. Description of parameters used in the model

Parameters	Description	Unit	Reference Value	reference
N_fix	nitrogen fixation rate	g N m^{-2} day^{-1}		
N_fixpot	potential nitrogen fixation rate	g N m^{-2} day^{-2}	0.01-1×10-3	Thornley (2001); Eckertsten et al.(2006); Corre-Hellou et al. (2007); Corre-Hellou et al. (2009);
ft	soil temperature factor	°C		
t_min	the minimum temperature for the start of N fixation	°C	0.5~5	Boote et al. (2008)
t_max	the maximum temperature for the stop of N fixation	°C	40~45	Boote et al. (2008)
t_optL	lower threshold of optimal temperature	°C	10~20	Boote et al. (2008)
t_optH	upper threshold of optimal temperature	°C	25~35	Boote et al. (2008)
fw	soil water factor			
φ1	coefficient for soil moisture		0	
φ2	coefficient for soil moisture		2	APSIM, EPIC (Sharpley and Williams, 1990; Bouniols et al., 1991; Cabelguenne et al., 1999); SOILN (Wu and McGechan, 1999)
Wa	lower threshold of water content below which N fixation is totally restrict by rhe defict of soil water		0	APSIM, EPIC (Sharpley and Williams, 1990; Bouniols et al., 1991; Cabelguenne et al., 1999); SOILN (Wu and McGechan, 1999)
Wb	upper threshold of water content above which N fixation is not limited by rhe defict of soil water		0.5	APSIM, EPIC (Sharpley and Williams, 1990; Bouniols et al., 1991; Cabelguenne et al., 1999); SOILN (Wu and McGechan, 1999)
Wf	available soil water content to that at field capacity			
f_Nup	parameter relating legume biological nitrogen fixation and soil nitrogen content		0.01~0.1	SOILN model (Wu and McGehan, 1999)
Ns	Soil mineral nitrogen content	g N m^{-2}		
f_N	Soil mineral N effect			
f_C	Soil carbon effect			
Cr	Carbon concentration in the soil	g C g^{-1} soil		
Kc	Michaelis-Menten Constant for carbon	g C m^{-2}	0.001~0.01	Thornley (2001); Eckertsten et al. (2006)

Table 2. Calibration Sites of Biological Nitrogen Fixation Rate for Representative Ecosystems

Site name	Ecosystem Type	Lon.	Lat.	Experimental method	Reference	N Fixation Rate kg N ha^{-1} yr^{-1}	Simulation, kg N ha^{-1} yr^{-1}
Stordalen, Sweden	Tundra	18	68	ARA	Christie (1987); Sonesson et al. (1980)	2	2.5
Truelove Lowland, Canada	Tundra	-84.5	75.5	ARA	Chapin et al. (1990)	3	2.8
Niwot Ridge, Colorado, US	Tundra	-105.5	40	^{15}N	Bowman et al. (1996)	4.9	5.1
Central Sweden	Boreal Forest	18	60	ARA	Nohrstedt (1985)	0.93	0.9
PNFI, Ontario, Canada	Boreal Forest	-77	45.5	ARA	Hendrickson (1990)	0.25	1.2
Southern British Columbia, Canada	Boreal Forest	-119	49	ARA	Hendrickson and Burgess (1989)	2.8	2.2
Robson moralines, British Columbia	Boreal Forest	-119	53	ARA	Blundon and Dale (1990)	1.1	1.2
Umea, Sweden	Boreal Forest	19.5	64	ARA	Huss-Danell (1976)	1	1.5
Coweeta Basin,	Temperate Forest	-83	35	N accumulation	Boring and Swank (1984)	48	19.5
Hoh River, Washington, US	Temperate Forest	-123.5	48	ARA	Luken and Fonda (1983)	40	13
Tom Swamp, Massachusetts, US	Temperate Forest	-75	42.5	unspecified	Schwinzer (1983)	35	25.7
Big Creek Basin, Melbourne, Austrilia	Temperate Forest	145.5	38	ARA	Adams and Attiwill (1984)	24	23.2
Jebo Creek, Utah, US	Temperate Forest	-112	42	^{15}N	Skujins et al. (1987)	10.2	12.5
Karri Forest, south-western Austrilia	Temperate Forest	116	-34.5	ARA	Grove and Malajczuk (1992)	7.93	8.5
Woodhill Forest, New Zealand	Temperate Forest	174.5	-37	N accumulation	Baker et al. (1986)	80	23.5
Gainesville, Florida, US	Temperate Forest	-82	30	N accumulation	Permar and Fisher (1983)	10.6	12.8
Fox park, Wyoming, US	Temperate Forest	-106	41	ARA	Fahey et al. (1985)	13	12.5
Mount Robson, Canada	Temperate Forest	-119	53.1	ARA	Blurdon and Dale (1990)	1.65	3.2
Dwellingup, South-western Austrilia	Temperate Forest	116	33	ARA	O'Connel and Grove (1987)	2.5	3.1
Adair, Oregon, US	Temperate Forest	-123	44.6	ARA	Heath et al (1988)	0.74	2.4

Priest River Experimental Forestry, Idaho, US	Temperate Forest	-116	48	ARA	Harvey et al (1989)	0.1	1.7
Arapaho Prarie, Nebraska, US	Grassland	-100	42	unspecified	Kaputsa and DuBois (1987)	0.2	0.7
Lynx Prairie Preserve, Ohio, US	Grassland	-83.5	39	ARA	DuBois and Kaputsa(1983)	8.2	1.9
Konza Prarie Research Natural Area, Kansas, US	Grassland	-96	39.5	nitrogenase activity	Eisele et al (1989)	21	3.3
Buso, Papua New Guinea	Tropical Forest	147	-7.5	ARA	Goosem and Lamb (1986)	0.5	5.2
Reserve Ducke, Manaus, Brazil	Tropical Forest	-59	-3	ARA	Sylvester-Bradley et al. (1980)	2.45	3.5
Sinharaja Man and Biosphere reserve	Tropical Forest	80.5	6.5	ARA	Maheswaran and Gunatilleke (1990)	8	8.5
Amazon Territory of Venezuela	Tropical Forest	-67	2	ARA	Jordan et al (1983)	32	20.3
Kilauea, Hawaii, US	Tropical Forest	-155	19	ARA	Vitousek (1994)	2.8	18.5
Volcano La Soufriere, Guadeloupe	Tropical Forest	-61.5	16	ARA	Sheridan (1991)	4.02	7.2
Hawaii Volcanoes National Park, US	Tropical Forest	-155	19.5	ARA	Ley and D'Antonio (1998)	4.9	9.3
Santa Ynez Mountain, California, US	Mediterranean Shrubland	-120	34.5	ARA	Schlesinger et al.(1982)	1	2.4
San Bernardino Mountains, California, US	Mediterranean Shrubland	-116.5	34	ARA	Lepper and Fleschner (1977)	6.9	3.7
Harpers Well, California, US	Xeric Shrubland	-116	33.5	N accumulation	Rundel et al.(1982)	30	18.5
Sonoran Desert, Arizona, US	Xeric Shrubland	-112.5	33	cation accumulation	Jarrell and Virginia (1990)	40	23.5

Table 3. Model estimated biological nitrogen fixation in global natural terrestrial ecosystems

Ecosystem	Average coverage of N fixing plants	Coverage range	Reference	N Fixation Rate (kg N ha^{-1} yr^{-1})	Total_Min (Tg N yr^{-1})	Total_Max (Tg N yr^{-1})	Total_Avg (Tg N yr^{-1})	Area (10^8 ha)
wet tundra	9%	3%~15%	May and Webber (1982)	3.2	0.51	2.55	1.54	5.37
alpine tundra & wet tundra	9%	3%~15%	May and Webber (1982)	3.2	0.51	2.55	1.54	5.36
boreal forest	9%	4%~18%	Alexander and Billington (1986); weber and Van Cleve (1981)	2.1	2.01	9.06	4.53	19.3
temperate coniferous forest	5%	1%~10%	Cleveland et al (1999)	12.7	0.71	7.15	3.5	5.51
temperate deciduous forest	5%	1%~10%	Cleveland et al (1999)	12.7	0.76	7.65	3.75	5.89
temperate evergreen forest	5%	1%~10%	Cleveland et al (1999)	12.7	0.43	4.34	2.13	3.35
grassland	15%	5%~25%	Woodmansee et al (1981); Robertson and Rosswall (1986)	1.9	0.61	3.1	1.86	8.4
tropical forest	15%	5%~25%	Cleveland et al (2001)	18.2	10.8	54	32.6	17.8
xeric shrubland	15%	10%~20%	Johnson and Mayeux (1990)	5.7	2.92	14.6	8.35	14.8
Mediterranean shrubland	15%	10%~20%	Johnson and Mayeux (1990)	2.7	0.13	0.66	0.4	1.47
savanna	15%	5%~25%	Stewart et al (1978); Bate and Gunton (1982)	1.9	0.45	2.23	1.34	7.05
Total					**19.84**	**107.89**	**61.54**	**94.3**

Table 4. Model parameters for various natural terrestrial ecosystems

		N _pot **(g N fixed day $^{-1}$)**	**t_optL** **(°C)**	**t_optH** **(°C)**	**W_upH** **(J kg $^{-1}$)**	**fNup**	**Kc** **(g C m^{-2})**
1	**wet tundra**	0.028	10	25	0.8	65	0.002
2	**alpine tundra & wet tundra**	0.028	10	25	0.8	65	0.002
3	**boreal forest**	0.032	12	25	0.8	70	0.008
4	**temperate coniferous forest**	0.55	16	35	0.6	80	0.01
5	**temperate deciduous forest**	0.55	18	35	0.6	80	0.01
6	**temperate evergreen forest**	0.55	18	35	0.6	80	0.01
7	**grassland**	0.05	18	35	0.5	60	0.012
8	**tropical forest**	0.8	20	35	0.8	100	0.005
9	**xeric shrubland**	0.7	15	35	0.4	65	0.016
10	**Mediterranean shrubland**	0.08	19	35	0.5	65	0.016
11	**savanna**	0.05	20	35	0.5	60	0.012

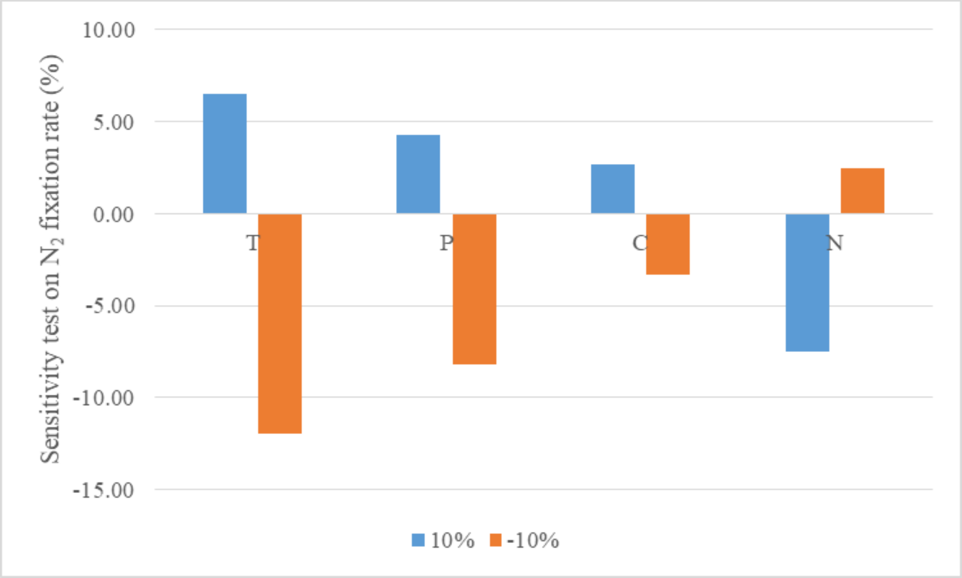
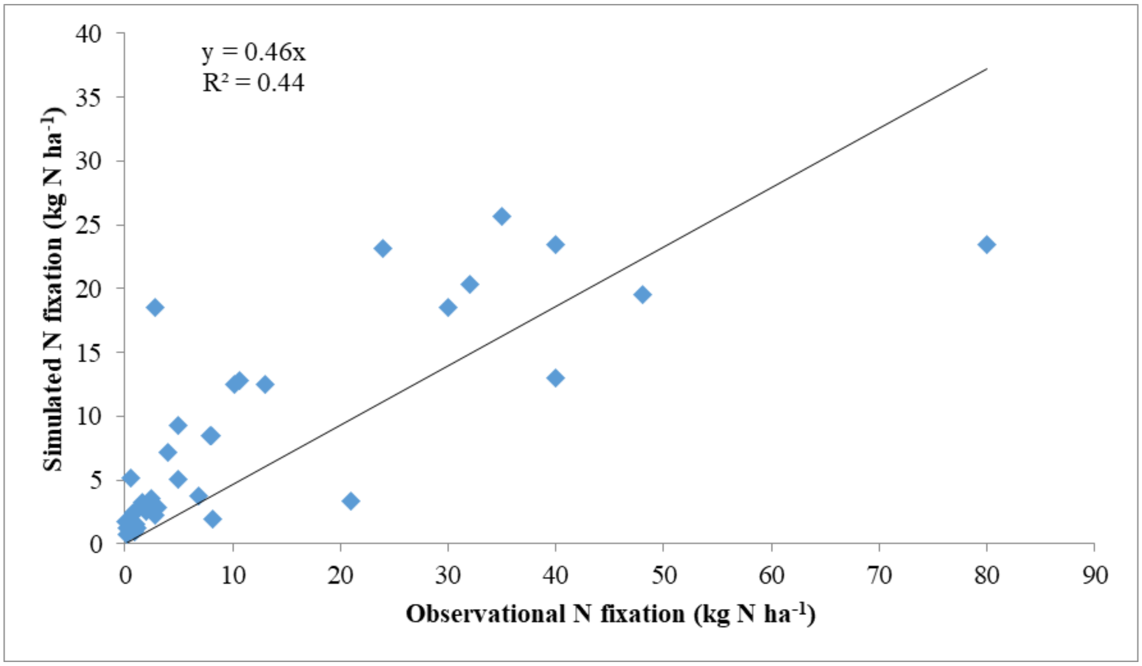
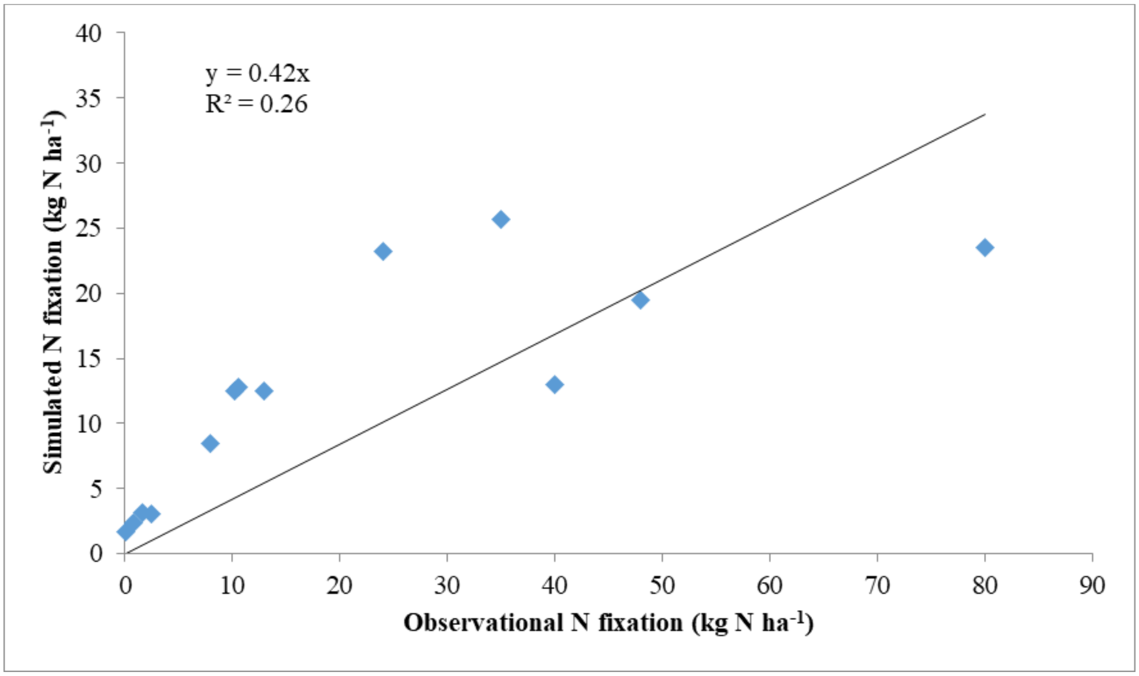


Figure 1. Model **s**ensitivity of N fixation in natural terrestrial ecosystems to changing model input data: Increasing or decreasing each variable by 10% for air temperature (T), precipitation (P), soil carbon content (C), soil nitrogen content (N) for N fixation rate.

(a)


y = 0.46x
R^2 = 0.44
Simulated N fixation (kg N ha^{-1})
Observational N fixation (kg N ha^{-1})
0
5
10
15
20
25
30
35
40
0
10
20
30
40
50
60
70
80
90

(b)


y = 0.42x
R^2 = 0.26
Simulated N fixation (kg N ha^{-1})
Observational N fixation (kg N ha^{-1})
0
5
10
15
20
25
30
35
40
0
10
20
30
40
50
60
70
80
90

(c)

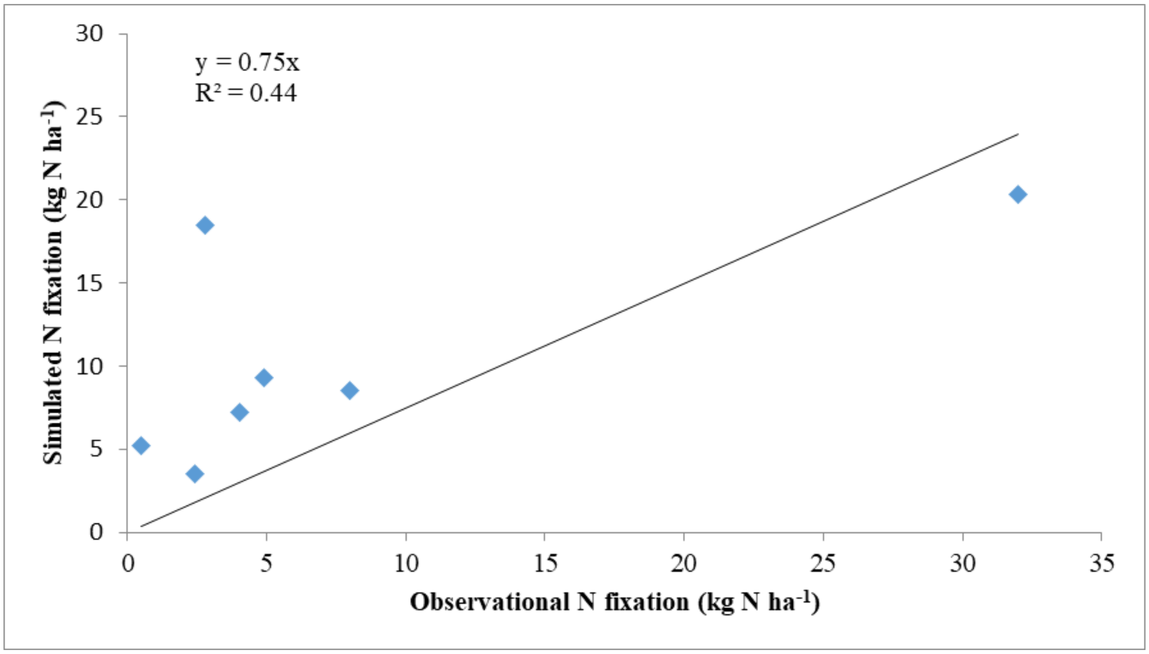


Figure 2. Comparison between Modeled and Observed Nitrogen Fixation Rate at site level: (a) All sites, (b) Temperate Forest, (c) Tropical Forest (data listed in Table 2). Y is simulated N fixation while X represents the observational N fixation.

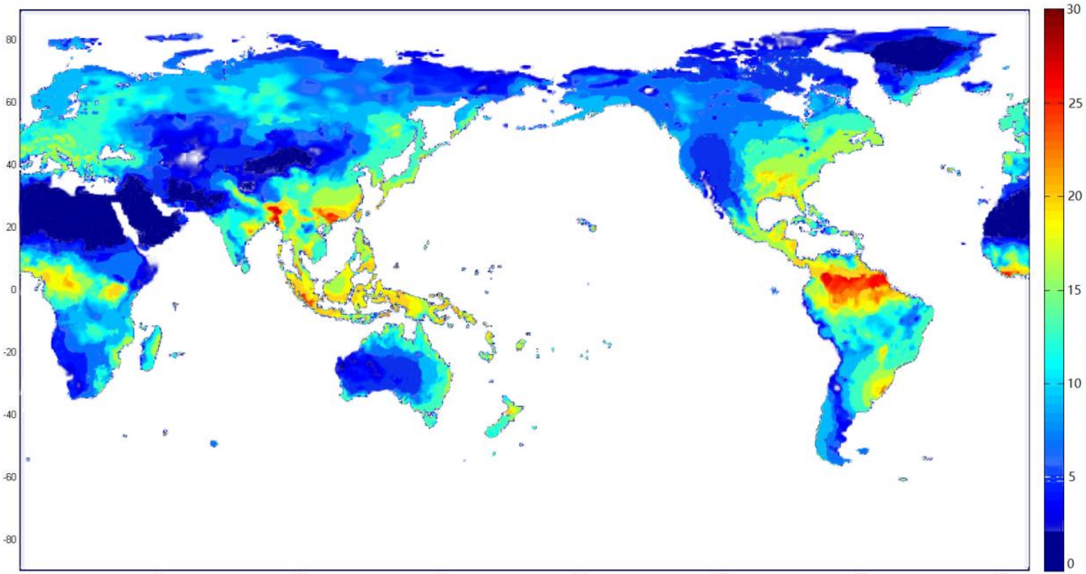


Figure 3. Simulated spatial distribution of BNF rates (kg N ha^{-1} yr^{-1}) in natural terrestrial ecosystems during 1990-2000 by considering the BNF effects.