Suppression of nitrogen deposition on global forest soil CH4 uptake depends on nitrogen status

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Abstract

Methane (CH4) is the second most important atmospheric greenhouse gas (GHG) and forest soils are a significant sink for atmospheric CH4. Uptake of CH4 by global forest soils is affected by nitrogen (N) deposition; clarifying the effect of N deposition helps to reduce uncertainties of the global CH4 budget. However, it remains an unsolved puzzle why N input stimulates soil CH4 flux (RCH4) in some forests while suppressing it in others. Combining previous findings and data from N addition experiments conducted in global forests, we proposed and tested a "stimulating-suppressing-weakening effect" ("three stages") hypothesis on the changing responses of RCH4 to N input. Specifically, we calculated the response factors (f) of RCH4 to N input for N-limited and N-saturated forests across biomes; the significant changes in f values supported our hypothesis. We also estimated the global forest soil CH4 uptake budget to be approximately 11.2 Tg yr–1. CH4 uptake hotspots were located predominantly in temperate forests. Furthermore, we quantified that current level of N deposition reduced global forest soil CH4 uptake by ~3%. This suppression effect was more pronounced in temperate forests than in tropical or boreal forests, likely due to differences in N status. The proposed "three stages" hypothesis in this study generalizes the diverse effects of N input on RCH4, which could help improve experimental design. Additionally, our findings imply that by regulating N pollution and reducing N deposition, soil CH4 uptake can be significantly increased in the N-saturated forests in tropical and temperate biomes.

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Key Points:

- A “three stages hypothesis” was developed to generalize the diverse responses of forest soil CH_4 flux to N input
- CH_4 uptake by global forest soils was estimated to be 11.2 Tg yr^{-1}, with N deposition suppressing ~3% of this uptake
- Effective regulation to reduce N deposition would promote CH_4 uptake by N-saturated forests and mitigate global warming

Abstract

Methane (CH_4) is the second most important atmospheric greenhouse gas (GHG) and forest soils are a significant sink for atmospheric CH_4. Uptake of CH_4 by global forest soils is affected by nitrogen (N) deposition; clarifying the effect of N deposition helps to reduce uncertainties of the global CH_4 budget. However, it remains an unsolved puzzle why N input stimulates soil CH_4 flux (R_{CH4}) in some forests while suppressing it in others. Combining previous findings and data from N addition experiments conducted in global forests, we proposed and tested a "stimulating-suppressing-weakening effect" ("three stages") hypothesis on the changing responses of R_{CH4} to N input. Specifically, we calculated the response factors (f) of R_{CH4} to N input for N-limited and N-saturated forests across biomes; the significant changes in f values supported our hypothesis. We also estimated the global forest soil CH_4 uptake budget to be approximately 11.2 Tg yr^{-1}. CH_4 uptake hotspots were located predominantly in temperate forests. Furthermore, we quantified that current level of N deposition reduced global forest soil CH_4 uptake by ~3%. This suppression effect was more pronounced in temperate forests than in tropical or boreal forests, likely due to differences in N status. The proposed "three stages" hypothesis in this study generalizes the diverse effects of N input on R_{CH4}, which could help improve experimental design. Additionally, our findings imply that by regulating N pollution and reducing N deposition, soil CH_4 uptake can be significantly increased in the N-saturated forests in tropical and temperate biomes.

Plain Language Summary

Methane is an important greenhouse gas. Forest soils can absorb methane from the atmosphere and mitigate its warming effect. Meanwhile, forests suffer from high atmospheric nitrogen

deposition, yet the effect of nitrogen on the methane uptake by forest soils remain unclear. Using data from global nitrogen addition experiments, we validated a “stimulating-suppressing-weakening effect” (“three stages”) hypothesis, which could explain the diverse responses of soil methane flux to nitrogen input observed in different forests. On the basis, we quantified that nitrogen deposition decreased global forest soil methane uptake by approximately 3%. Our findings also imply that that by regulating nitrogen pollution, soil methane uptake can be significantly increased in the nitrogen-saturated forests in tropical and temperate biomes, potentially mitigate global warming.

1 Introduction

Methane (CH_4) is the second most important greenhouse gas (GHG), responsible for approximately 20% of global warming since the industrial revolution (Kirschke et al., 2013; Saunois et al., 2020). Biological CH_4 absorption by soils contributes to 5–7% of total CH_4 removal from the atmosphere (Dlugokencky et al., 2011; Saunois et al., 2020). Soils, however, do not always function as net sinks of atmospheric CH_4. The net effect of two biological processes, namely CH_4 production ("methanogenesis", widespread in anoxic microsites and deep soils; Angel et al., 2012; Kotelnikova, 2002; Lacroix et al., 2023) and CH_4 oxidation ("methanotrophy", widespread in oxic surface soils; Le Mer & Roger, 2001), determines whether a soil is a source or sink of CH_4. The delicate, variable balance between soil CH_4 consumption and production depends on various changing environmental factors, which leads to uncertainties in soil-atmosphere CH_4 exchange dynamics and the potential feedback of soil CH_4 uptake to climate change (Bodelier & Steenbergh, 2014; Feng et al., 2020). Approximately 30% of the Earth's land surface are forests, which are significant for regulating global climate (Bonan, 2008). Recently, forests received much attention because forestland-based management practices, such as afforestation, are crucial for achieving net-zero emissions by mid-21st century and mitigating global warming (Griscom et al., 2017; IPCC, 2021). Mechanisms underlying forest GHG fluxes are fundamental to assessing and predicting the effectiveness of the practices. Therefore, it is important and urgent to understand global forest soil CH_4 flux variations under environmental changes.

Since the 19th century, following an exponential increase in the artificial production and anthropogenic emission of reactive nitrogen compounds (e.g., through fertilizer use, combustion processes), deposited nitrogen (N) to terrestrial ecosystems has increased by more than threefold

(Galloway et al., 2004). This exogenous N input impacts the structure and functioning of ecosystems by altering plant and microbial properties (Vitousek et al., 1997). Furthermore, enhanced N deposition has led to widespread "N saturation" of forests (Ågren & Bosatta, 1988), resulting in divergent responses of ecological processes (such as primary production and N mineralization) to N input in N-saturated as compared to N-limited forests (Aber et al., 1998). To quantify the effect of N deposition on forest system functioning, researchers have conducted N addition experiments in forests worldwide during the past half-century. Although the effects of N input on some ecosystem properties have been clarified by the experiments, the relationship between N input and soil CH_4 flux remained an unsolved puzzle. Some experiments revealed stimulating effects of N input on soil CH_4 uptake, while some others showed inhibited soil CH_4 uptake by N input (Veldkamp et al., 2013; Zhang et al., 2012). Currently, there is no universally applicable framework to explain the diverse responses of soil CH_4 flux to N input. This lack of understanding hinders the development of quantitative models and assessment of the change in global forest soil CH_4 budget caused by N deposition.

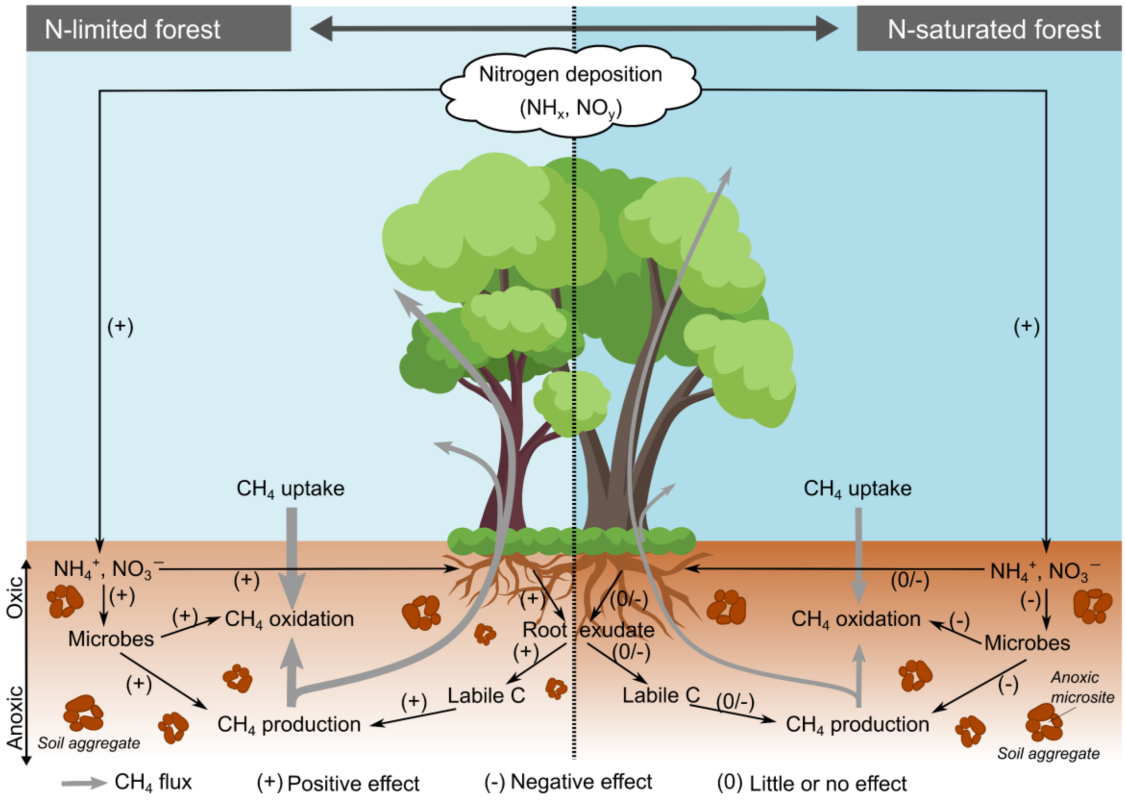

Fig. 1. Soil CH$_4$ fluxes exhibit varying responses to N deposition as forests transition from a N-limited status to a N-saturated status (or vice versa) due to human activities.

The response of soil CH$_4$ flux to N deposition is influenced by the rate and persistence of N input and the N availability in forests (Aronson & Helliker, 2010; Chang et al., 2021). In N-limited forests, a low N input rate can stimulate plant and microbial activities. Methanotrophs, which are more active in near-surface soils (Butterbach-Bahl & Papen, 2002), may benefit from the external N supply, with increased abundance and activity (see Fig. 1; Bodelier & Laanbroek, 2004), causing more CH$_4$ to be oxidized. However, CH$_4$ oxidation can be suppressed by high N input, as a result of the inhibitory effect of excessive N on methanotrophs (Agathokleous et al., 2020; Chen et al., 2021; Peng et al., 2019). In N-saturated forests, the N supply surpasses the demands of plants and microbes. Consequently, suppression of soil CH$_4$ uptake has been observed even under a low N input rate (Mochizuki et al., 2012). Moreover, methanogenesis and methanotrophy can both be suppressed under high N input rates, resulting in a weak response of soil CH_4 flux to N input (Keiluweit et al., 2018; Steinkamp et al., 2001). Therefore, there appear to be distinct stages in the response of soil CH_4 fluxes to N input, with N-limited and N-saturated forests experiencing different stages under elevated N input rates. In light of these observations, we have developed a stimulating-suppressing-weakening effect (referred to below as "three stages") hypothesis (see Fig. 2b) to offer a unified framework that generalizes the response of soil CH_4 flux to N input.

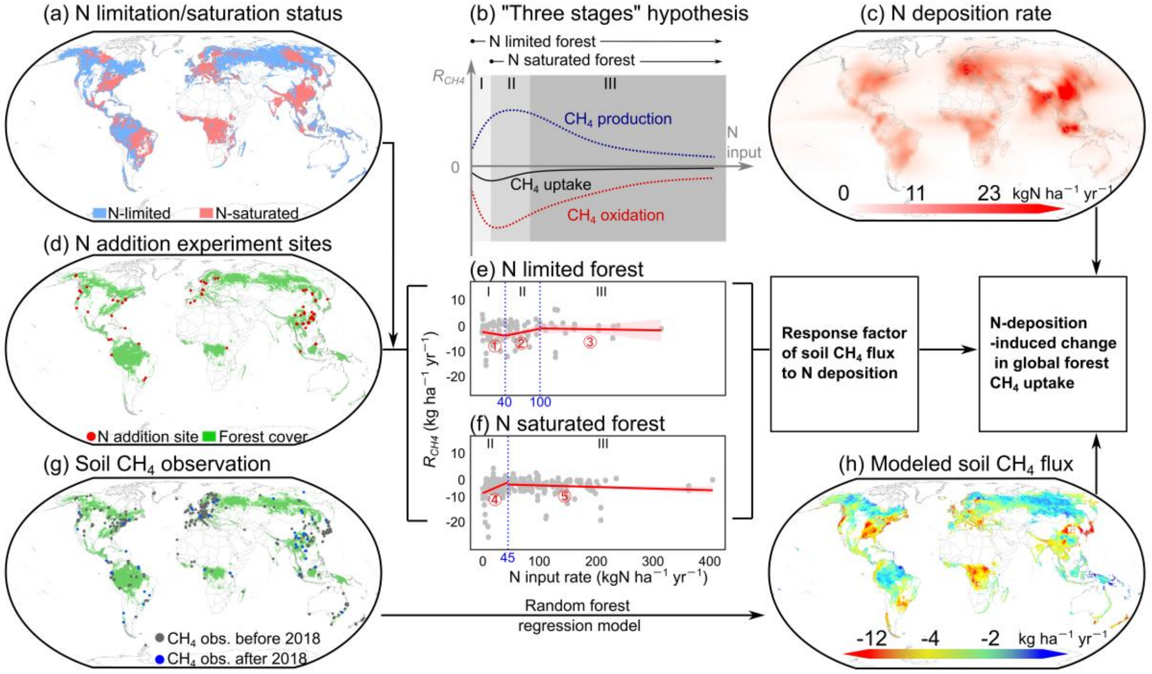

Fig. 2. Workflow for quantifying the impact of N deposition on forest soil CH_4 flux (R_{CH4}) in global forests. (a) N limitation or saturation status of global forests, indicated by the sensitivity of soil N_2O emissions to N deposition (See Supporting Text S1 for details). (b) Proposed "three stages" hypothesis on the response of R_{CH4} to N input. (c) Global map of N deposition rates, data from Ackerman et al. (2019). (d) Forest sites where N addition experiments was conducted and

R_{CH4} was observed (CH_4_exp dataset). (e) Segmented regression models on R_{CH4} and N input rate, using data from N-limited forests where N addition experiments lasted for no more than 3 years. (f) Segmented regression models on R_{CH4} and N input rate, using data from N-saturated forests and forests with N addition experiments lasting more than 3 years (see Supporting Table S1 for model parameters). (g) Forest sites where no experiment was conducted and R_{CH4} was observed under natural conditions (CH_4_obs dataset). (h) Estimated R_{CH4} in global forests.

In this study, we gathered data from N addition experiments conducted in forests worldwide. We aimed to examine the validity of the "three stages" hypothesis by comparing the alterations in soil CH_4 flux (R_{CH4}) caused by each unit of N input, known as "response factors", in N-limited and N-saturated forests. Furthermore, we aimed to estimate the global budget for CH_4 uptake by forest soils and determine the specific contribution of N deposition to this budget.

2 Methods

2.1 Data source

We conducted a systematic compilation of soil CH_4 flux data observed in N addition experiments by searching relevant literature published prior to 1/1/2022 in the Web of Science Core Collection (www.webofscience.com) and the China National Knowledge Infrastructure Theses and Dissertations Database (https://oversea.cnki.net/kns?dbcode=CDMD). The search utilized keywords "forest" AND ("greenhouse gas" OR "CH_4" OR "methane"). Subsequently, we manually refined the obtained 8702 papers and theses based on the following criteria: (i) N addition experiments were conducted in forest ecosystems with recorded site locations and N addition doses and (ii) field observations of soil CH_4 flux were measured using gas chromatograph technique (Holland et al., 1999). The resulting compiled dataset, named "CH_4_exp", comprises 465 observations from 85 sites (refer to Fig. 2; Supporting Information Data Set S1).

Additionally, we compiled data on soil CH_4 flux observed under natural conditions. Soil CH_4 flux observations before 2018 were obtained from three published datasets by Dutaur and Verchot (2007), L. J. Yu et al. (2017), and Ni and Groffman (2018), while data observed after 2018 were gathered from the abovementioned 8702 literature using a different set of criteria: (i) forest soil CH_4 fluxes were observed in the field and measured using gas chromatograph technique (Holland et al., 1999) and (ii) no N or other nutrient addition experiments were conducted at the forest sites. The compiled dataset, referred to as "CH_4_obs", consists of 1946 observations from 652 forest sites worldwide (see Fig. 2; Supporting Information Data Set S2).

We also collected supplementary information on environmental factors, including climate, N deposition, and soil properties. In cases where not all required information was provided for a particular site, we extracted data from spatial datasets based on the coordinates of the sites. Specifically, temperature and precipitation data were sourced from the Climatic Research Unit, University of East Anglia (https://crudata.uea.ac.uk/cru/data/hrg/cru_ts_4.03/); soil texture data were obtained from the Harmonized World Soil Database (https://www.fao.org/soils-portal/data-hub/soil-maps-and-databases/harmonized-world-soil-database-v12/en/); global N deposition data were derived from a published dataset by Ackerman et al. (2019); forest cover data were obtained from the GLASS-GLC project (Liu et al., 2020); and forest biome boundaries were sourced from the Global Forest Monitoring project (Hansen et al., 2010).

2.2 Calculating response factor of soil CH_4 flux to N input

Considering the distinct responses of R_{CH4} to N input in N-limited and N-saturated forests, we partitioned the CH_4_exp dataset into two subsets based on whether the experimental sites were N-limited or N-saturated. Since the N limitation or saturation status was largely unknown at most experimental sites, we initially predicted the N limitation or saturation status of global forests, using the sensitivity of soil N_2O emissions to N deposition as an indicator; the accuracy of prediction exceeded 70% on both regional and global levels (see Supporting Text S1 for details about determining the N status of global forests). On the basis, we determined the N limitation or saturation status of forest sites in CH_4_exp dataset; the dataset was then divided into two sub-datasets, CH_4_exp_NL and CH_4_exp_NS, consisting of data from N-limited and N-saturated forests, respectively (Supporting Fig. S1). Moreover, recognizing that long-term N addition can lead to the transition of N-limited forests into N-saturated forests, we also included data from experimental sites where N addition had been implemented for more than three years in the CH_4_exp_NS sub-dataset.

Using data from the CH_4_exp_NL sub-dataset, we constructed segmented linear regression models to account for the changing relationship between soil CH_4 flux and N input (see Fig. 2e,f, Supporting Table S1). In accordance with the segmented regression models, we further divided the sub-dataset into several groups based on N input levels (low, medium, and high). Using data from each group, we computed the change in soil CH_4 flux per unit of N input on site level (Eq. 1), which we referred to as the response factor of soil CH_4 flux to N input (f). Similarly, we calculated f for N-saturated forests using data from the CH_4_exp_NS sub-dataset.

$$f = \frac{R_2 - R_1}{N_2 - N_1} \qquad \text{(Eq. 1)}$$

where f is the response factor of the soil CH_4 flux to N input, N_1 and N_2 are the two different N input rates (kg N ha^{-1} yr^{-1}), R_1 and R_2 are the corresponding soil CH_4 fluxes (kgCH_4 ha^{-1} yr^{-1}).

2.3 Estimating global forest soil CH_4 flux using random forest regression method

Using soil methane flux (R_{CH4}) observed under natural conditions (CH_4_obs dataset), we predicted R_{CH4} of global forests on grid level with random forest regression models (Breiman, 2001). In practice, R_{CH4} observed at the same site were aggregated by taking the mean value. After excluding one outlier that is approximately two times lower than the second lowest R_{CH4} value, we randomly sampled 20% of the 872 entries of data to form a testing dataset (n = 175). 75% of the remaining data (i.e., 60% of all data) were randomly chosen to form a training dataset (n = 523), and the rest data were to allow for the variation of training dataset (n = 174). Climate, N deposition, soil texture, and soil N status variables were used as predictors (Supporting Table S2).

Because the constructed models can vary depending on which data were used to train the models, the random sampling of training data was repeated for 1000 times, which derived 1000 models. When estimating R_{CH4} on grid level, each grid had 1000 predicted R_{CH4} values from the 1000 models. The mean R_{CH4} of the 1000 values were used as the estimated R_{CH4} of the grid, and the standard error of the estimation was also calculated from the 1000 values.

Estimated R_{CH4} for grids in the test dataset (which were never used in model construction) were then compared with observed values to measure the accuracy of prediction.

Also, we randomly sampled a different 80% of data to form different training datasets, repeated the above processes, and checked the robustness of our prediction on grid level.

2.4 Quantifying the contribution of N deposition to global forest soil CH_4 budget

By summarizing the grid-level R_{CH4} data (Eq. 2), we obtained soil CH_4 budgets for forests in various regions. Combining the N deposition rate with the previously quantified response factor (*f*), we determined the N-deposition-induced changes in the soil CH_4 budget. This allows us to quantify the contribution rate of N deposition to the global forest soil CH_4 budget (Eq. 3).

$$Budget = \sum_i (R_{CH4,i} \times A_i) \qquad \text{(Eq. 2)}$$

$$Contribution\ rate = \frac{\sum_i (N_{depo,i} \times f_i)}{Budget} \times 100\% \qquad \text{(Eq. 3)}$$

where $R_{CH4,i}$ is the soil CH_4 flux in grid *i* (kgCH_4 ha^{-1} yr^{-1}), A_i is the forest area in grid *i* (ha), $N_{depo,i}$ denotes the N deposition rate in grid *i* (kgN ha^{-1} yr^{-1}), and f_i is the response factor determined based on the N deposition rate and the N limitation/saturation status of the forests in grid *i* (kgCH_4 kgN^{-1}).

Additionally, we employed the bootstrap method (Davison & Hinkley, 1997) to compare the mean R_{CH4} values among forests in different biomes. Furthermore, we conducted an analysis to determine the relative importance of environmental factors in explaining the spatial variation in R_{CH4} (Grömping, 2006). Data analyses were performed using R software (R Core Team, 2020), with a significance level set at $p < 0.05$. The production of maps was accomplished using ArcGIS software (ESRI, 2011).

3 Results

3.1 Response of forest soil CH_4 fluxes to N input

Locally weighed regression models showed different patterns in the responses of soil CH_4 flux (R_{CH4}) to N input in N-limited and N-saturated forests (Supporting Fig. S1; see Supporting Text S1 for determination of N limitation or saturation status of global forests), and the responses changed with N input level. Segmented linear regression models were separately fitted to data from N-limited and N-saturated forests to detect the thresholds in the phased responses of R_{CH4} to N input. In accordance with the detected N input thresholds (40 and 100 kgN ha^{-1} yr^{-1} for N-limited forests, and 45 kgN ha^{-1} yr^{-1} for N-saturated forests; refer to Fig. 2e,f), data obtained in N-limited forests were then divided into three groups based on N input levels (low, medium, and high), whereas data from N-saturated forests were divided into two groups (low and high N inputs). The quantified response factor (f) for N-limited forests under low, medium, and high N inputs represented the response of R_{CH4} to N input in Stages I, II, and III, respectively (Fig. 3). Similarly, the quantified f for N-saturated forests under low and high N inputs represented the response of R_{CH4} to N input in Stages II and III. The observed changes in f values across different stages and different biomes provide support for our "three stages" hypothesis (Fig. 3).

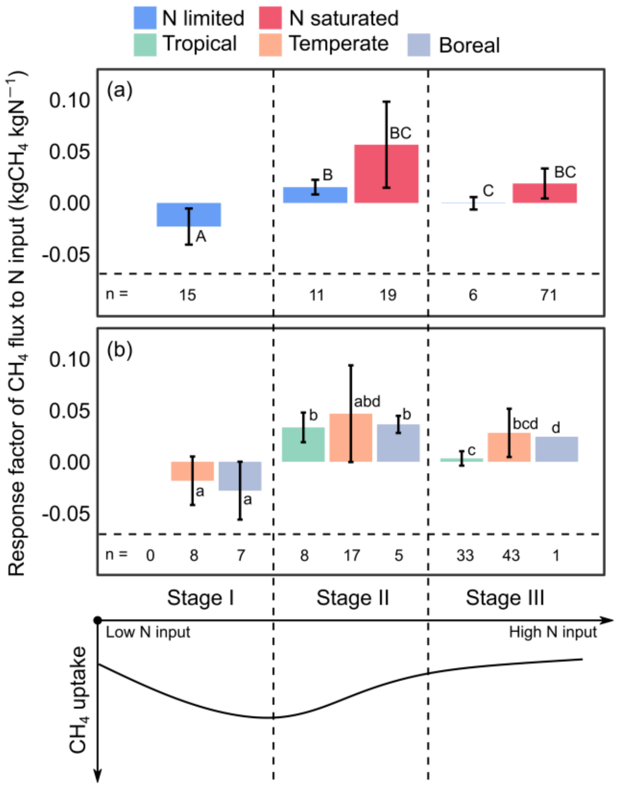

Fig. 3. Changes in the response factors (*f*) of soil CH_4 flux to N input at three stages. (a) Mean response factors of N-limited and N-saturated forests; (b) Mean response factors of forests in different biomes. The error bars represent the standard errors of the mean values. Different letters beside each column indicate that the mean values of *f* were significantly different ($p < 0.05$). Numbers below each column are the number of *f* values derived from CH_4_exp dataset.

At Stage I, mean value of *f* in N-limited forests was significantly lower than 0 ($p <$ 0.005), indicating that low N input stimulated soil CH_4 uptake (or suppressed soil CH_4 emissions). At Stage II, the mean *f* values for both N-limited and N-saturated forests were significantly higher than 0 ($p < 0.005$), signifying that medium N input suppressed soil CH_4 uptake in N-limited forests, while low N input had a suppressing effect on R_{CH4} in N-saturated forests. At Stage III, the mean *f* value for N-limited forests approached zero, and the mean *f* value for N-saturated forests at this stage was lower than the mean *f* value for N-saturated forests in Stage II. These findings suggested that the response of R_{CH4} generally diminished under high N input in both N-limited and N-saturated forests. Furthermore, we observed a decrease in the standard deviation of R_{CH4} under high N input (Supporting Fig. S2), indicating the weakening of at least one process underlying soil CH_4 flux (methanogenesis or methanotrophy). This in combination with the relatively stable mean R_{CH4} values under high N input (refer to Supporting Fig. S1), suggested that both CH_4 production and oxidation rates declined (see Supporting Text S2 for inference processes), which agreed with our hypothesis (Fig. 2b).

On biome level, the significant changes in *f* values at different stages were as well consistent with our hypothesis (Fig. 3).

3.2 Global forest soil CH_4 fluxes, and the underlying environmental variables

We built random forest regression models using natural R_{CH4} observations (R_{CH4}_obs dataset; Supporting Table S2), and predicted R_{CH4} in global forests on grid level. Estimated R_{CH4} was compared with observations from the testing dataset not used in model construction. The correlation coefficient of 0.6 proved the reliability of this method (Supporting Fig. S3). Also, we randomly sampled data to form different training datasets and built a different set of random forest models. Estimated R_{CH4} were in good agreement with our initial estimations (r = 0.91; Supporting Fig. S4), showing the robustness of grid-level estimations of R_{CH4}.

The average R_{CH4} for global forests was estimated to be -2.95 kg ha^{-1} yr^{-1}. Mean R_{CH4} of temperate forests was significantly more negative than those of tropical and boreal forests ($p < 0.001$; refer to Fig. 4).

Environmental factors influencing the spatial variation in R_{CH4} differed across biomes. In tropical forests, approximately 50% of the explainable variation could be attributed to N deposition and its annual fluctuations. In temperate forests, precipitation played a dominant role in explaining the spatial variation in R_{CH4}. Both precipitation and temperature emerged as the main factors influencing R_{CH4} in boreal forests.

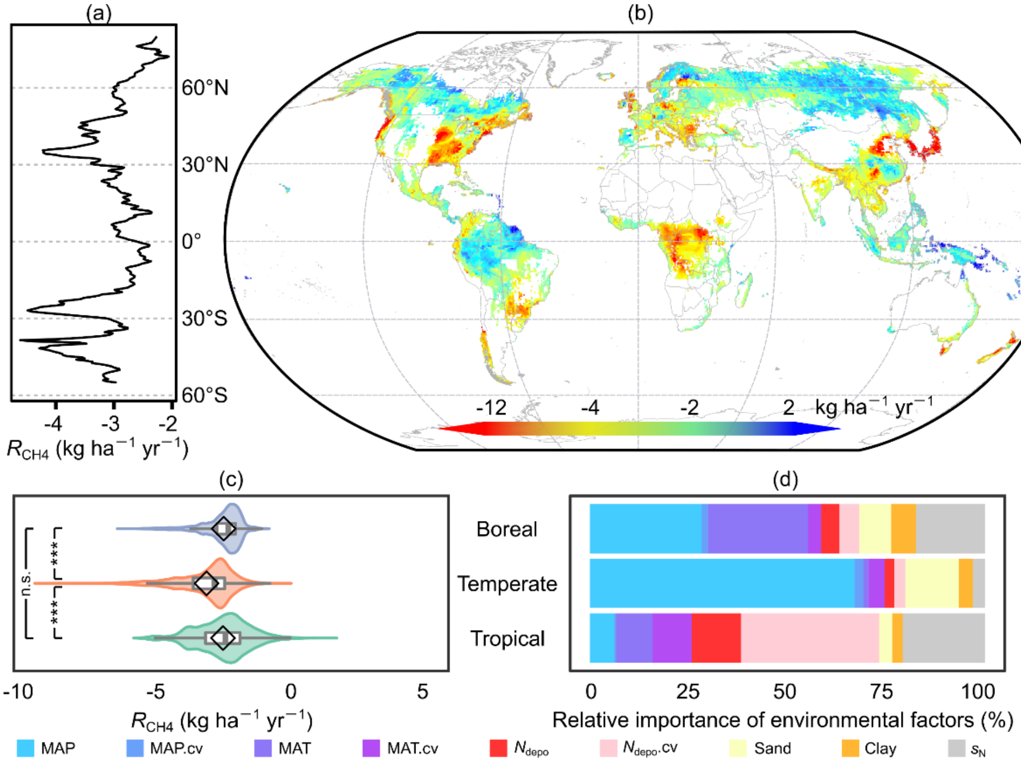

Fig. 4. Estimation of soil CH_4 fluxes (R_{CH4}) in global forests using the random forest method. (a) Latitudinal gradient in soil CH_4 flux. The black line represents the average R_{CH4} values across latitudes. (b) Global map illustrating soil CH_4 fluxes in forests. Negative values indicate net CH_4 uptake. (c) Violin plots and boxplots displaying the statistical distribution of R_{CH4} values in

different biomes. (d) Assessment of the relative importance of environmental factors in the spatial variation of R_{CH4} across different biomes. The factors include mean annual precipitation (MAP), mean annual temperature (MAT), atmospheric N deposition rate (N_{depo}), soil sand content (Sand), soil clay content (Clay), and sensitivity of soil N_2O emission to N deposition (s_N), which serves as an indicator of the N limitation/saturation status of forests.

3.3 Contribution of N deposition to global forest soil CH_4 budget

By summarizing the grid-level R_{CH4} data, the CH_4 uptake by global forest soils was estimated to be approximately 11.2 TgCH$_4$ yr^{-1}. Currently, N deposition reduced global forest soil CH_4 uptake by 0.29 TgCH$_4$ yr^{-1}, representing a global suppression of 2.6%. The overall effect of N deposition on forest soil CH_4 uptake varied among different biomes (see Table 1). N deposition suppressed soil CH_4 uptake by 3–6% in tropical and temperate forests, whereas it stimulated boreal forest soil CH_4 uptake by 1.1%.

Table 1. Contribution of N deposition to soil CH_4 budget in global forests.

	Area (10^8 ha)	N deposition rate (kgN ha^{-1} yr^{-1})	Response factor (kgCH_4 kgN^{-1})	N deposition induced change in CH_4 emission (TgCH_4 yr^{-1})	CH_4 emission rate (kgCH_4 ha^{-1} yr^{-1})	CH_4 emission budget (TgCH_4 yr^{-1})	Contribution rate of N deposition to CH_4 budget (%)
Tropical forest							
N limited	8.4	3.87	–0.019 (0.024)*	–0.06 (0.08)	–2.33 (0.011)	–1.77 (0.01)	3.4
N saturated	9.6	7.23	0.034 (0.015)	0.24 (0.11)	–3.20 (0.012)	–3.07 (0.01)	–7.8
Subtotal	18.0	5.66		0.18 (0.13)	–2.81 (0.012)	–4.84 (0.02)	–3.7
Temperate forest							
N limited	3.6	5.40	–0.019 (0.024)	–0.04 (0.05)	–3.41 (0.011)	–1.34 (0.004)	3.0
N saturated	3.8	10.50	0.047 (0.047)	0.19 (0.19)	–3.48 (0.012)	–1.34 (0.004)	–14.2
Subtotal	7.4	7.99		0.15 (0.20)	–3.44 (0.012)	–2.68 (0.01)	–5.6
Boreal forest							
N limited	10.0	2.08	–0.028 (0.028)	–0.07 (0.07)	–2.65 (0.012)	–2.70 (0.01)	2.6
N saturated	3.0	2.53	0.037 (0.008)	0.03 (0.01)	–3.32 (0.016)	–1.00 (0.01)	–3.0
Subtotal	13.0	2.18		-0.04 (0.07)	–2.79 (0.013)	–3.70 (0.02)	1.1
Total	38.4	4.64		0.29 (0.25)	–2.95 (0.012)	–11.22 (0.05)	–2.6

* No observations were available for N-limited tropical forests; hence, the mean response factor of N-limited temperate forests was used instead.

Values in parentheses represent standard errors of the mean.

4 Discussion

4.1 “Three stages” hypothesis generalizes response of forest soil CH_4 flux to N input

Both the exogenous N input level and the internal properties of forest ecosystems (such as N availability) can influence the response of soil CH_4 flux to N input. Manipulative experiments and meta-analyses have been conducted to examine changes of R_{CH4} in response to different N input levels (Aronson & Helliker, 2010; Chen et al., 2021). However, the spatially varying responses of R_{CH4} to N input in different forests remained unresolved. The absence of a comprehensive framework for the effect of N input on R_{CH4} has impeded the integration of site-level observations and identification of a global pattern. In this study, we proposed a "three stages" hypothesis to elucidate the relationship between R_{CH4} and N input. It not only accounts for the varying responses of R_{CH4} to different levels of N input, but also explains the divergent effects of N input on R_{CH4} in N-limited and N-saturated forests.

The "three stages" concept is primarily determined by the biphasic dose-response relationship between N input and biotic processes, exhibiting a stimulating effect at low doses and a suppressing effect at high doses (referred to as the "hormesis" effect; (Agathokleous et al., 2020). Additionally, the asynchronous responses of methane production and oxidation processes to N input play a role; the hormesis effect leads to the transition from Stage I to subsequent stages, and the transition from Stage II to Stage III occurs due to the lower tolerance of methanotrophs to N input as compared to methanogens (Li et al., 2021). While methanotrophs are generally sensitive to nitrogen addition (Nyerges & Stein, 2009), at least some methanogens (such as hydrogenotrophic methanogens) are tolerant to high N and low soil pH (Horn Marcus et al., 2003).

More generally, We postulate that although the responses of many biological processes (such as net primary production or N mineralization) across a wide range of N input is unimodal (Aber et al., 1998), response patterns may differ for variables controlled by the interactions of multiple functional groups (Fig. 5), such as soil CH_4 flux and soil respiration rate. Therefore, experiments testing the response of forest soil CH_4 fluxes to N input may need to test at least three levels of N additions, so as to capture the changes of response. Conducting experiments with multiple N addition levels will facilitate a comprehensive understanding of changes in biogeochemical cycles and the underlying mechanisms. Hypothesized response patterns of soil variables controlled by additive or subtractive interactions (Fig. 5) may provide reference for setting N addition levels in experiments.

Moreover, it should be noted that the calculated response factors showed high degrees of uncertainty, due to the limited experimental data available. Additional experiments are required, especially in boreal forests which are sensitive to future climate and N deposition change (Fleischer et al., 2015; Galloway et al., 2004). On the basis, researchers will be able to reduce uncertainties in the global forest soil CH_4 budget under spatially and temporally varying N loads deposited from the atmosphere.

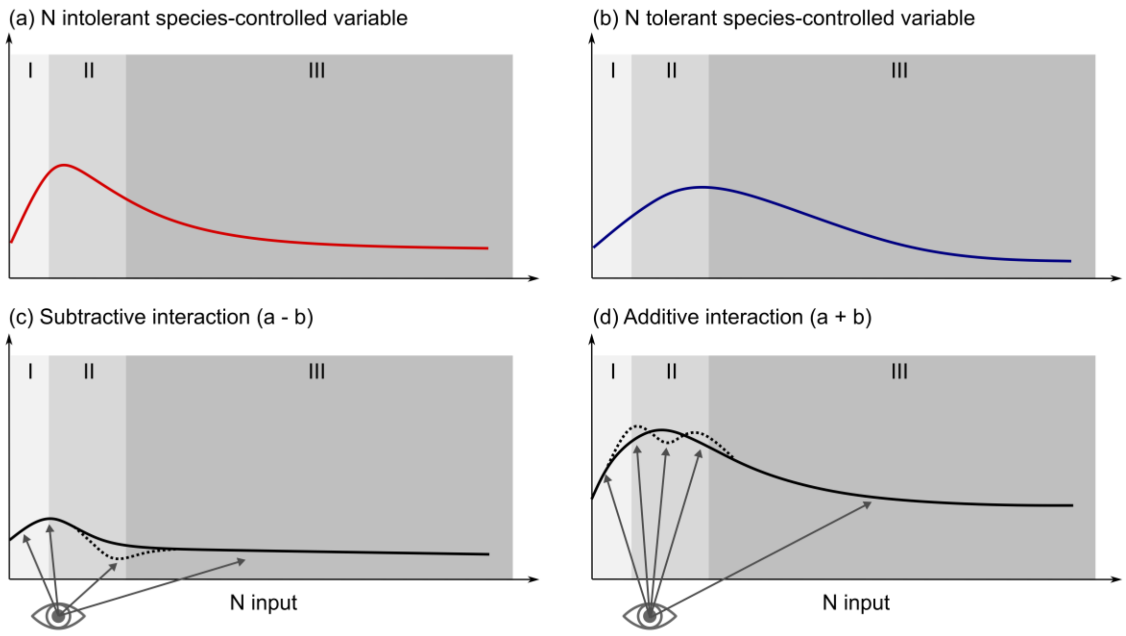

Fig. 5. Hypothesized effects of N input on variables controlled by N-tolerant or intolerant species, and their interactions. (a) Effect of N input on N intolerant species-controlled variables (such as CH_4 oxidation rate); (b) Effect of N input on N tolerant species-controlled variables (such as CH_4 production rate); (c) Effect of N input on variables controlled by subtractive interactions between different species (such as soil CH_4 uptake rate, which is the difference between CH_4 oxidation rate and CH_4 production rate); (d) Effect of N input on variables controlled by additive interactions between different species (such as soil respiration rate, which is the sum of plant-root respiration and the respiration of various soil microbes). In panels c and d, dashed curves illustrate the alternative responses of the interaction-controlled variables to N input, depending on the relative importance of the participating species for their interactions; arrows indicate critical stages and transitions in the response curves, which should ideally be captured in experiments aiming to fully reveal the changes in responses.

4.2 Effects of N deposition and forest N status on soil CH_4 flux

The global map presented in Figure 4 illustrates the distribution of soil CH_4 uptake, with hotspots predominantly located in temperate forests. This can be attributed to favorable conditions in temperate forests, such as optimal soil moisture levels for aeration and suitable temperatures for enhanced methanotrophic activity, both of which promote CH_4 uptake by soils (Castro et al., 1995). Meanwhile, soils in central Amazon rainforest, tropical forests in Southeast Asia, and boreal forests in Siberia and northwestern Canada were predicted to be CH_4 sources, which is consistent with field observations (Melling et al., 2005; Pangala et al., 2017; Rask et al., 2002). The net emission of CH_4 is probably caused by submerged soils widespread in these regions, which favors methanogenesis and hinders methanotrophy. The estimated global budget for CH_4 uptake by forest soils in this study is 11.2 $TgCH_4\ yr^{-1}$. This aligns well with estimations from previous studies using data extrapolation or modeling approaches (as shown in Fig. 6; (Curry, 2007; Dutaur & Verchot, 2007; Potter et al., 1996; Ridgwell et al., 1999; Steudler et al., 1989; L. J. Yu et al., 2017; Zhuang et al., 2013).

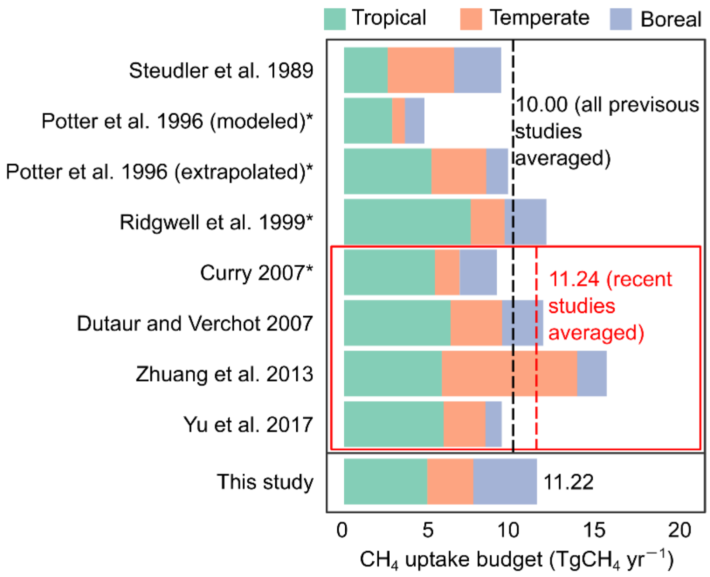

Fig. 6. Comparison of the estimated global forest soil CH_4 uptake budgets from previous studies with the findings of this study. It should be noted that the global forest area used in four earlier studies (~6×10^9 ha) significantly exceeded the currently accepted value (~4×10^9 ha). To facilitate accurate comparison, we rectified the estimates to account for the differences in forest area. The rectified estimates are indicated with asterisks (*).

N deposition impacts the capacity of forest soils to absorb atmospheric CH_4. N deposition enhances plant growth in N-limited ecosystems, leading to increased root exudates, which adds to the substrates and anoxic microsites for methanogenesis. Moreover, deposited N may stimulate the activity of methanogens, thereby accelerating the rate of CH_4 production. The produced CH_4 can either diffuse into the soil or be released into the near-surface atmosphere through tree stems and leaves. Elevated CH_4 concentrations promote methanotrophy, the process of CH_4 oxidation (Carmichael et al., 2014; Covey & Megonigal, 2019; Le Mer & Roger, 2001). Methanotrophs present in near-surface soils can be stimulated by atmospheric N deposition, further enhancing CH_4 oxidation. On the other hand, long-term high N deposition can drive forests towards a state of N saturation (Aber et al., 1998; Ågren & Bosatta, 1988). Additional N input to N-saturated forests may suppress plant and microbial activities, leading to a decrease in the rate of CH_4 oxidation. Furthermore, the deposited ammonium may compete with CH_4 for oxidants, further reducing CH_4 uptake by soils (Schnell & King, 1994).

Despite of the mechanistic relevance between N saturation status and soil CH_4 uptake, previous studies were unable to separately analyze the N effect on R_{CH4} in N-limited and N-saturated forests, owing to the lack of site-level N status information or a global map of the N saturation status of forests. We innovatively determined the N limitation or saturation status of global forest ecosystems using the sensitivity of soil N_2O emissions to N deposition as an indicator, which was estimated from soil N_2O emission data measured in global N addition experiments (Fig. 2a; see Supporting Text S1 for details). Globally-distributed, coordinated manipulative experiments can help reveal spatially-varying sensitivities of ecosystems to environmental changes, facilitating biogeochemical and global change studies.

4.3 Suppressing effect of N deposition on global soil CH_4 uptake depends on forest N status

Findings in this study (Fig. 2e,f; Fig. 3) suggest that the current level of N deposition (< 40 kgN ha^{-1} yr^{-1} in the majority of forests) primarily stimulates soil CH_4 uptake in N-limited forests whereas suppressing soil CH_4 uptake in N-saturated forests. Globally, we revealed that N deposition decreased forest soil CH_4 uptake. However, the extent of this suppression effect varies across different biomes depending on the N limitation or saturation status of the forests.

The most pronounced suppression effect was observed in temperate forests (Table 1), likely due to the transition of many forests in this region from a N-limited to a N-saturated status caused by N deposition. At this stage, N input suppresses CH_4 uptake (refer to Supporting Fig. S5). In contrast, tropical forests naturally exist in or near N saturation (Lu et al., 2021; Matson et al., 2002), resulting in a weakening response of R_{CH4} to additional N input. Boreal forests, mostly N-limited by nature, exhibit a stimulated CH_4 uptake in response to N deposition (Supporting Fig. S5).

It is important to note that maximizing soil CH_4 uptake might suggest maintaining a relatively high N deposition level around the transition point between Stage I and Stage II. However, this approach should consider the potential acceleration of N_2O emissions resulting from N deposition (Cen et al., 2022). Further research is required to evaluate whether maintaining a relatively high N deposition rate can effectively reduce combined greenhouse gas emissions from soils. Additionally, the shift between N-limited and N-saturated status driven by N deposition can lead to systematic changes in the structure and function of plant and microbial communities. This can at least transiently reduce ecosystem resilience and increase environmental risks such as species invasion. Therefore, maintaining forest ecosystems near the threshold for N status change may not be a feasible strategy for climate change mitigation.

To ensure long-term environmental health, it is crucial to regulate and address N pollution. Western Europe and the eastern United States have witnessed a decrease in atmospheric N deposition due to the reduction in anthropogenic N emissions (Ackerman et al., 2019). Similarly, China has stabilized its N deposition through effective N pollution control measures (G. Yu et al., 2019), although many parts of the country are still under a relatively high level of N deposition (> 20 kgN ha^{-1} yr^{-1}). These countries and regions have experienced significant anthropogenic impacts, leading to large areas of N-saturated forests. With continued efforts to control N pollution effectively, a decline in N deposition is anticipated. Consequently, soils in N-saturated forests in these counties and regions are likely to absorb more atmospheric CH_4, thus contributing to global warming mitigation.

In this study, we computed the response factors of soil CH_4 flux to N input by utilizing data from global N addition experiments. We quantified the impact of N deposition on soil CH_4 uptake in forests worldwide. It is important to note that the majority of the experiments were conducted over a short-term period (approximately 85% of the data in the CH_4_exp dataset comprised forest sites where N addition experiments lasted no longer than 2 years). Therefore, the derived response factors primarily reflect the short-term influence of N deposition on soil

CH_4 flux. They may not provide insights into the long-term adaptation of plants and microbes to altered N deposition regimes. Consequently, our results should be interpreted as the short-term direct effect of N deposition on soil CH_4 uptake. If future research aims to estimate or predict the influence of N deposition on soil CH_4 uptake over a long period of time (e.g., on a centennial scale), additional observational data from long-term experiments will be necessary. These data should encompass the adaptive changes in soil microbial communities (especially methanogens and methanotrophs), as well as the quantity and quality of plant root exudates.

5 Conclusions

Using compiled data from N additon experiments in global forests, we validated a “stimulating-suppressing-weakening” (“three stages”) response pattern of soil CH_4 uptake to N input, which could generalize the diverse effects of N input on soil CH_4 flux in N-limited and N-saturated forests. On the basis, we quantified that on global level, current level of N deposition suppressed forest soil CH_4 uptake by ~3%. The suppressing effect, however, differs among biomes, because of the different proportions of N-saturated forests in different biomes. Our findings imply that by controlling N pollution and reducing N deposition, soil CH_4 uptake in N-saturated forests (mostly in tropical and temperate biomes) are expected to increase, potentially mitigating global warming. Due to the limitations of available data, our result could only show the short-term effect of N deposition on global forest soil CH_4 flux. In the future when more long-term experimental data become available, researchers could further study the adaptations of methanogens and methanorophs to long-term N addition, thus improving predictions of N deposition-induced change in the global methane budget.

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Open Research

All the source data and R code used for data analysis in this study have been uploaded as supporting information for peer review purposes, which will be archived in a publicly accessible repository prior to publication.

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PUBLICATIONS

Global Biogeochemical Cycles

Supporting Information for

Suppression of nitrogen deposition on global forest soil CH_4 uptake depends on nitrogen status

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Introduction

The uploaded Data Set S1 (CH_4_exp dataset in main text) was used to derive the response factors of soil CH_4 flux to N input in global forests; Data Set S2 (CH_4_obs dataset in main text) was used to estimate the soil CH_4 fluxes in global forests; Data Sets S3–S7 were used to classify the N-limited and N-saturated forests on global level; Data Set S8 contains environmental factors (MAT, MAP, soil texture, etc.) for global estimations; Data Set S9 contains global forest soil CH_4 budgets reported in previous studies. The data analysis process and produced figures can be replicated with the uploaded R script (Code S1).

Text S1. Nitrogen saturation status of global forests indicated by sensitivity of soil N_2O emission to N deposition.

Globally, human-induced increase in atmospheric N deposition is changing forests from a nitrogen-limited to nitrogen-saturated status. In N-limited forests, plants and microbes utilize N conservatively for a lower proportion of input N to be leaked from tight N cycling processes (Chapman et al., 2006; Van Der Heijden et al., 2008). However, when forests become N-saturated, input N exceeds the N demand of plants and microbes, leading to excessive utilization of N, and thus, the N cycle becomes more open (Hietz et al., 2011). Therefore, a higher proportion of input N is lost via leaching or gaseous emission (Aber et al., 1989). This implies that increased gaseous N emissions (N_2O, NO, and N_2) per unit of N deposition (i.e., higher sensitivity of gaseous N emissions to N deposition) may indicate forests reaching N saturation (Aber et al., 1998). Coincidently, studies have measured nitrous oxide (N_2O) greenhouse gas emissions under different N input levels since the 1980s in global forests, using a controlled experiment design and standard sampling method (Holland et al., 1999). The accumulated experimental data provide an opportunity to quantify the sensitivity of N_2O emissions to N deposition in various forests, and indicate the N limitation or saturation status of global forests.

Gathering data

To quantify the sensitivity of soil N_2O emissions to N deposition (s_N), we compiled soil N_2O emission data observed in N addition experiments conducted in global forests. On 03/30/2022, we searched for papers and theses published before 01/01/2022 from the Web of Science Core Collection database (www.webofscience.com) and China National Knowledge Infrastructure Theses and Dissertations Database (https://oversea.cnki.net/kns?dbcode=CDMD), using the following keywords: "forest" AND "greenhouse gas" OR "N_2O" OR "nitrous oxide". The retrieved 7422 papers and 718 theses were then refined manually based on the following criteria: (i) experimental N addition was conducted in forest ecosystem; (ii) literature recorded the location, time, and dose of the experiment(s); (iii) soil N_2O flux was observed in experimental sites and measured using gas chromatograph technique (Holland et al., 1999). As a result, the compiled "N_2O_exp" dataset (Data Set S3) contained 553 observations from 102 sites worldwide (Fig. S7).

Similarly, we compiled data on the soil N_2O emission rates of global forests observed under natural conditions. We refined from the same papers and theses as above, using a different set of criteria: (i) no nutrients, including N, were artificially added to the forest site so the site only received naturally deposited N; (ii) literature recorded the location, and time of flux measurement; (iii) soil N_2O flux was observed in the field and measured using gas chromatograph technique (Holland et al., 1999). The compiled "N_2O_obs" dataset (Data Set S4) contained 246 observations from 140 sites worldwide (Fig. S7).

In addition, we compiled data on total N loss (N leaching and gaseous N emission combined), N leaching, and change in soil N pool, from N addition experiments in global forests. We searched in the aforementioned databases using the following keywords: "forest" AND "nitrogen addition" OR "fert*" AND "nitrogen loss" OR "nitrogen leaching" OR "nitrogen budget". Retrieved 2693 papers and theses were then refined based on the following criteria: (i) literature recorded the location, time, and dose of experimental N addition in forests; (ii) total N loss rate, N leaching rate, or change rate of soil N pool was observed or estimated in the experiments. The compiled “Ncycle_exp” dataset (Data Set S5) contained 169 observations from 37 sites (Fig. S7).

To analyze the relationship between s_N and N saturation status, we compiled data on field-observed N-limited and N-saturated forests indicated by N leaching. On 10/31/2022, we searched for literature in the aforementioned databases using the following keywords: “forest” AND “leaching” AND “nitrogen limit*” OR “nitrogen saturat*”. Retrieved 823 papers and theses were then refined based on the following criteria: (i) literature recorded whether the forest was N-limited or N-saturated, and its location; (ii) literature used nitrogen leaching as an indicator of N limitation or saturation status. The compiled “Nleach” dataset (Data Set S6) contains 136 observations from 92 sites worldwide (Fig. S6). We also used data on field-observed N-limited and N-saturated ecosystems indicated by plant growth response to N input (“NuLi” dataset; Data Set S7) from a published database by Du et al. (2020). It covers 106 sites worldwide, 65 of which are forest sites (Fig. S6).

Moreover, we extracted auxiliary information from the literature on environmental factors (including mean annual temperature, MAT; mean annual precipitation, MAP; mean annual N deposition rate, N_{depo}; coefficients of temporal variation, MAT.cv, MAP.cv, and N_{depo}.cv; soil sand content, soil clay content, and other soil properties) for the forest sites in the datasets. However, the literature did not provide the necessary auxiliary information for all sites; therefore, spatial datasets were used to fill in the missing data based on the location of the sites. Global temperature and precipitation datasets were obtained from the Climatic Research Unit, University of East Anglia (https://crudata.uea.ac.uk/cru/data/hrg/cru_ts_4.03/). The soil C:N ratio was obtained from a published database(Shangguan et al., 2014). Other soil properties were obtained from the HWSD dataset (https://www.fao.org/soils-portal/data-hub/soil-maps-and-databases/harmonized-world-soil-database-v12/en/). N deposition rate and forest cover data were from published databases (Ackerman et al., 2019; Liu et al., 2020). The forest biome map was derived from the Global Forest Monitoring project (Hansen et al., 2010).

Quantifying the sensitivity of soil N_2O emissions to N deposition

Under low N input, the soil N_2O emission rate responds almost linearly to N input, whereas high N input may induce non-linear responses (Aber et al., 1998; D.-G. Kim et al., 2013). High N input may change ecosystem properties, leading to a deviation from the natural response of ecosystems to environmental change. Therefore, we used a linear model (Eq. S1) to define and quantify the sensitivity (s_N) of soil N_2O emissions to N deposition (or low N input), for s_N to reflect ecosystem properties (i.e., N saturation status).

$$R_{N2O} = s_N \times N_{depo} + R_0 \qquad \text{(Eq. S1)}$$

where R_{N2O} is the soil N_2O emission rate (kgN_2O-N ha^{-1} yr^{-1}), N_{depo} is the atmospheric N deposition rate (kg N ha^{-1} yr^{-1}), s_N is the sensitivity of soil N_2O emission to N deposition, quantified as soil N_2O emission per unit of low N input (kgN_2O-N kgN^{-1}), and R_0 is the background soil N_2O emission rate when there is no N deposition or artificial N addition (kgN_2O-N ha^{-1} yr^{-1}).

A segmented regression analysis on N_2O_exp dataset showed that there is one change point in the linear relationship between N input rate and R_{N2O}, which is 174.70 ± 19.73 kgN ha^{-1} yr^{-1}. That is in line with change points estimated or used in previous studies (Bouwman et al., 2002; Hoben et al., 2011; M. Lu et al., 2022; McSwiney & Robertson, 2005; Shcherbak et al., 2014). Conservatively, experimental data with N addition rates not exceeding 150 kg N ha^{-1} yr^{-1} were used as "low N input" data in further analysis. The N deposition rates in global forests were lower than the level (Ackerman et al., 2019). For all the low-N input data in the N_2O_exp dataset, we aggregated them to 0.5° × 0.5° grids based on their coordinates to match the spatial resolution with environmental factors and reduce random errors in sampling. A linear model (Model: R_{N2O} ~ N input rate) was built for each grid with low-N input data. The slope of the linear model was the estimated s_N of the grid (Table S3).

Based on the estimated s_N of all grids and the corresponding environmental factors, we built a generalized linear model to simulate s_N (Table S4). In addition, another generalized linear model was built to simulate R_0.

To validate s_N, we firstly used the modeled s_N, together with the modeled R_0 and N_{depo} datasets, to estimate R_{N2O} (Eq. S1). The estimated R_{N2O} values were compared with R_{N2O} observations (N_2O_obs dataset) and indirectly validated the intermediate variable s_N (Fig. S8). In addition, s_N was validated using a second approach. The sensitivity of N loss to N input (c_1), the sensitivity of N leaching to N input (c_2), and the end-product ratio of nitrification and denitrification processes (c_3) were either derived from the Ncycle_exp dataset or extracted from the literature; s_N was then calculated from these parameters (Eq. S2).

$$s_N = c_3 \times (c_1 - c_2) \tag{Eq. S2}$$

The limited observations allowed us to calculate s_N on a biome scale (Fig. S7), which was then compared with the biome-mean value of the modeled s_N to validate it. The good agreement also validated the modeled s_N (r = 0.998).

Determining N saturation status of global forests using s_N

We tested whether s_N can distinguish between N-limited and N-saturated forests using data from forests having field-observed N saturation status data. First, we combined Nleach and NuLi datasets to enlarge the sample size and derive a universal classification. Excluding three duplicate sites in both datasets, the combined dataset had 154 sites with field-observed N saturation status (86 N-limited and 68 N-saturated sites).

We modeled the s_N of the 154 sites using environmental factors (Table S4). We then analyzed the s_N of N-limited and N-saturated forests and verified if there were significant differences on the global and biome scales. In Western Europe, North America, and East Asia, where there were abundant sites, we also compared the s_N of forests with N-limited or N-saturated status on a regional scale. The mean s_N was significantly different on global and regional scales ($p < 0.001$; Fig. S9), proving that s_N can indicate N limitation or saturation status in forests.

Then we calculated an optimal threshold for s_N using data from 154 sites with field-observed N saturation status and s_N information. The bootstrap method accounted for the different sample sizes of N-limited and N-saturated sites (Davison & Hinkley, 1997). Specifically, from the 154 sites, we randomly sampled 10 N-limited and 10 N-saturated sites and selected a cutoff value for their s_N at a precision of 0.0001 kgN_2O-N kgN^{-1}. Sites in which s_N were above the cutoff value were classified as "N-saturated," and the rest were classified as "N-limited." The classified N saturation status of the sites was compared with field observations to determine the accuracy of the classification, which was calculated as the proportion of sites accurately classified into the same category as that observed. All possible cutoff values were tested, and the one with the highest classification accuracy was the “optimal” cutoff value. Random sampling and detection of optimal cutoff values were repeated 5000 times, during which some optimal cutoff values were detected more frequently than others. The optimal threshold for s_N in all samples was the most frequently detected optimal cutoff value, which was 0.0143 kgN_2O-N kgN^{-1}.

The N saturation status of global forests was determined based on the optimal threshold. Forests with s_N above the threshold were classified as N-saturated, and the rest were classified as N-limited. The accuracy of the classification was higher than 70% on global and regional scales (Fig. S6). Based on the classification, we produced a rasterized map of N-limited and N-saturated forests (0.5° × 0.5° resolution) in ArcGIS (ESRI, 2011).

Text S2. Inferring the variation of methane production and oxidation rates from the variation of observed methane fluxes

Soil CH_4 flux observed on the soil-air interface is codetermined by methane production (methanogenesis) and oxidation rates (Eq. S3). However, it has been difficult to disentangle the responses of methane production and oxidation to N input, because of the limited ability to separately observe methanogenesis and methane oxidation processes in the field. Here, we inferred the variation of methane production and oxidation rates from the variation of observed methane fluxes. This could further support the "three stage" hypothesis we proposed.

$$R_{CH4} = R_{CH4\_prod} - R_{CH4\_oxid} \qquad \text{(Eq. S3)}$$

where R_{CH4} is the observed soil CH_4 flux (kg ha^{-1} yr^{-1}), positive R_{CH4} value means methane emission, whereas negative R_{CH4} value means methane uptake; $R_{\mathrm{CH4\_prod}}$ is methane production rate (kg ha^{-1} yr^{-1}); R_{CH4_oxid} is methane oxidation rate (kg ha^{-1} yr^{-1}).

The change in methane production and oxidation rates could hardly be inversely calculated from the R_{CH4} values. Here, we inferred the change in $R_{\mathrm{CH4\_prod}}$ and R_{CH4_oxid} by analyzing the mean values and standard deviations of R_{CH4}.

Firstly, the standard deviation of R_{CH4} could be calculated from that of R_{CH4_prod} and R_{CH4_oxid} (not considering the interaction between R_{CH4_prod} and R_{CH4_oxid}; Eq. S4).

$$SD(R_{CH4}) = \sqrt{SD(R_{CH4\_prod})^2 + SD(R_{CH4\_oxid})^2} \qquad \text{(Eq. S4)}$$

Usually, when the expected value of a variable becomes higher, its observations will be more dispersed. This is because the random errors in the observations are often proportional to their values. That is to say, statistical dispersion of R_{CH4_prod} and R_{CH4_oxid} (as indicated by their standard deviations) should be positively related to their mean values.

Therefore, the decrease in the standard deviation of R_{CH4} under high N input (Fig. S2) may result from: (1) R_{CH4_prod} decreased under high N input, and R_{CH4_oxid} didn't change or slightly increased; (2) R_{CH4_oxid} decreased under high N input, and R_{CH4_prod} didn't change or slightly increased; (3) both R_{CH4_prod} and R_{CH4_oxid} decreased under high N input.

Meanwhile, we observed that the mean values of R_{CH4} remained nearly unchanged under high N input (Fig. S1a), which may result from: (i) both R_{CH4_prod} and R_{CH4_oxid} increased under high N input; (ii) both R_{CH4_prod} and R_{CH4_oxid} decreased under high N input; (iii) both R_{CH4_prod} and R_{CH4-_oxid} remained constant under high N input.

Combining the two evidences (standard deviation and mean values of R_{CH4} under high N input), it can be inferred that only hypotheses (3) and (ii) can be true at the same time. That is, both R_{CH4_prod} and R_{CH4_oxid} decreased under high N input.

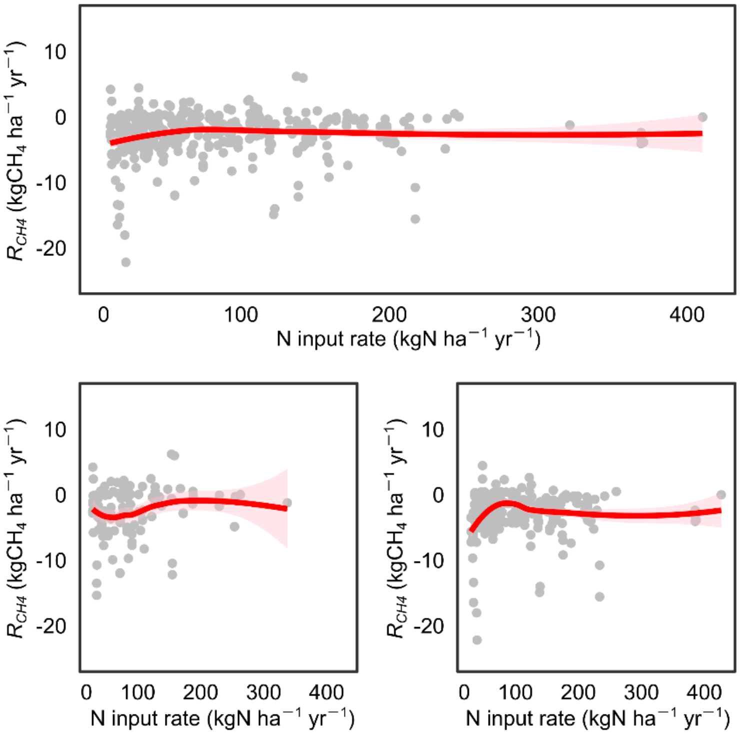


Fig. S1. Locally weighed regression ("LOWESS") model on soil CH_4 emission rate and N input rate. (a) Using all observations compiled from global N addition experiments, the N input rates of which were no greater than 400 kgN ha^{-1} yr^{-1} (n = 448). The few but variable observations on soil CH_4 fluxes at sites where N input rates were above 400 kgN ha^{-1} yr^{-1} (n = 17) were not used in further analysis. (b) LOWESS model constructed using data from N-limited sites and also where N addition experiments have been conducted for no more than 3 years when CH_4 emissions were observed (n = 131); (c) LOWESS model constructed using data from N-saturated forests, or data from sites where N addition experiments have been conducted for more than 3 years before observing the CH_4 fluxes (n = 317). Pink shadings represent the standard errors of the fitted models.

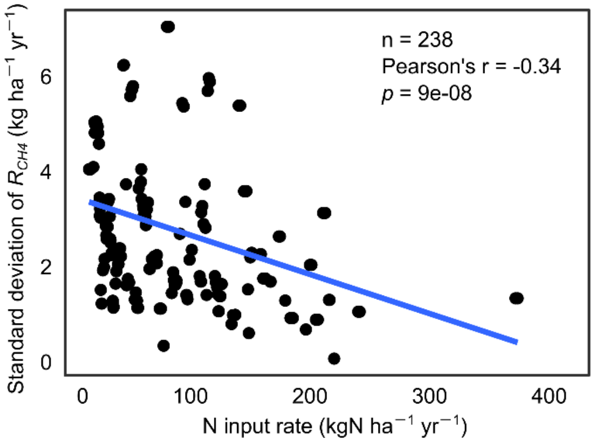


Fig. S2. Standard deviation of soil methane flux (*R_{CH4}*) was negatively correlated to N input rate. Data corresponding to N input levels above 400 kgN ha^{-1} yr^{-1} were not included in this analysis, because the very limited observations may not sufficiently reveal the statistical distribution of *R_{CH4}*. There were 238 unique N input rates that was no greater than 400 kgN ha^{-1} yr^{-1}. In practice, standard deviation was calculated for *R_{CH4}* corresponding to each N input rate, and N input rates less than 2 kgN ha^{-1} yr^{-1} in difference (e.g., standard deviation of *R_{CH4}* corresponding to 5 kgN ha^{-1} yr^{-1} was calculated using observations whose N input rates were within the range of 3 to 7 kgN ha^{-1} yr^{-1}). That was to make sure that there were sufficient observations for each N input level.

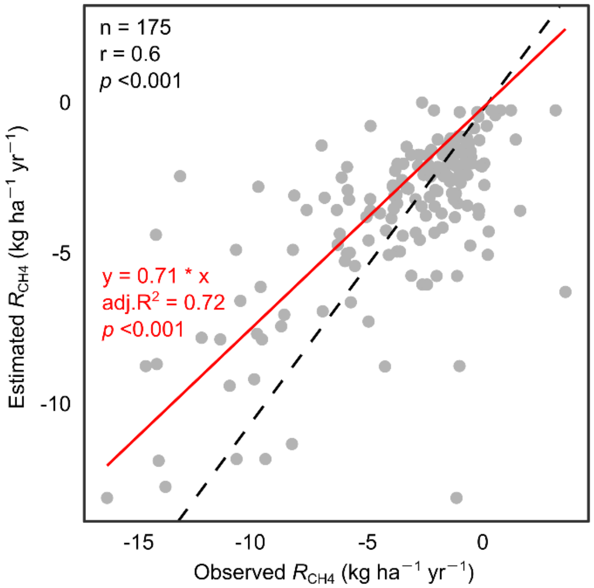


Fig. S3. Comparing observed soil CH_4 flux (R_{CH4}) in testing dataset with that estimated using averaged outputs from 1,000 random forest regression models. The red line and font indicate the fitted linear model on estimated and observed R_{CH4} values.

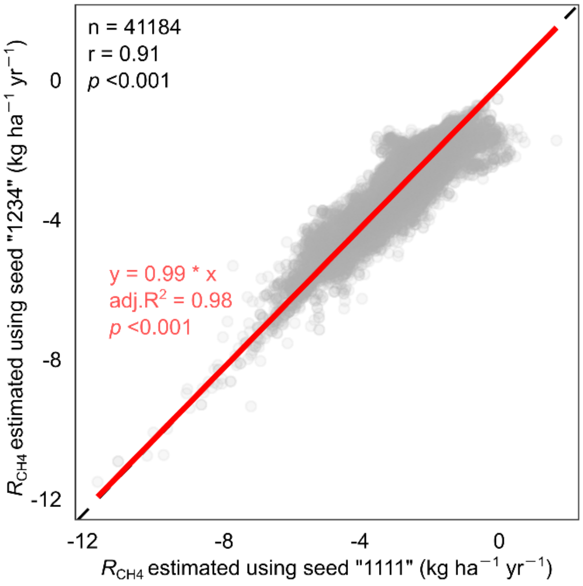


Fig. S4. Comparing soil CH_4 flux (R_{CH4}) estimated from different models built out of different training datasets. The sampling of observations to form a training (or testing) dataset was randomized by using different “seeds”. Each seed corresponds to a determined set of samples, and different seeds lead to different samples. In this study, we randomly used seeds “1111” and “1234” for sampling. This analysis was to ensure that the estimated R_{CH4} values were not dependent on which data were used for training and testing the models, so that the derived spatial pattern of R_{CH4} was robust on grid level.

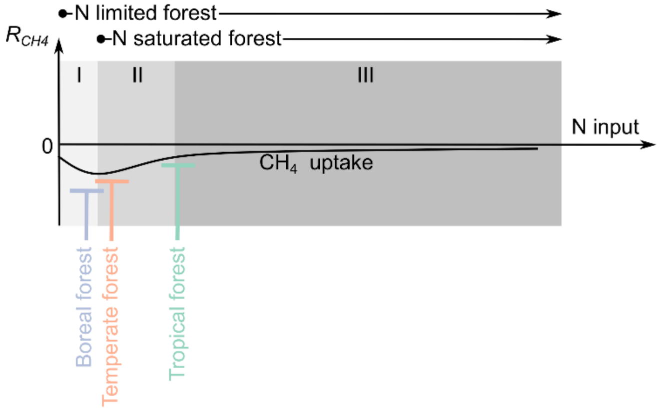
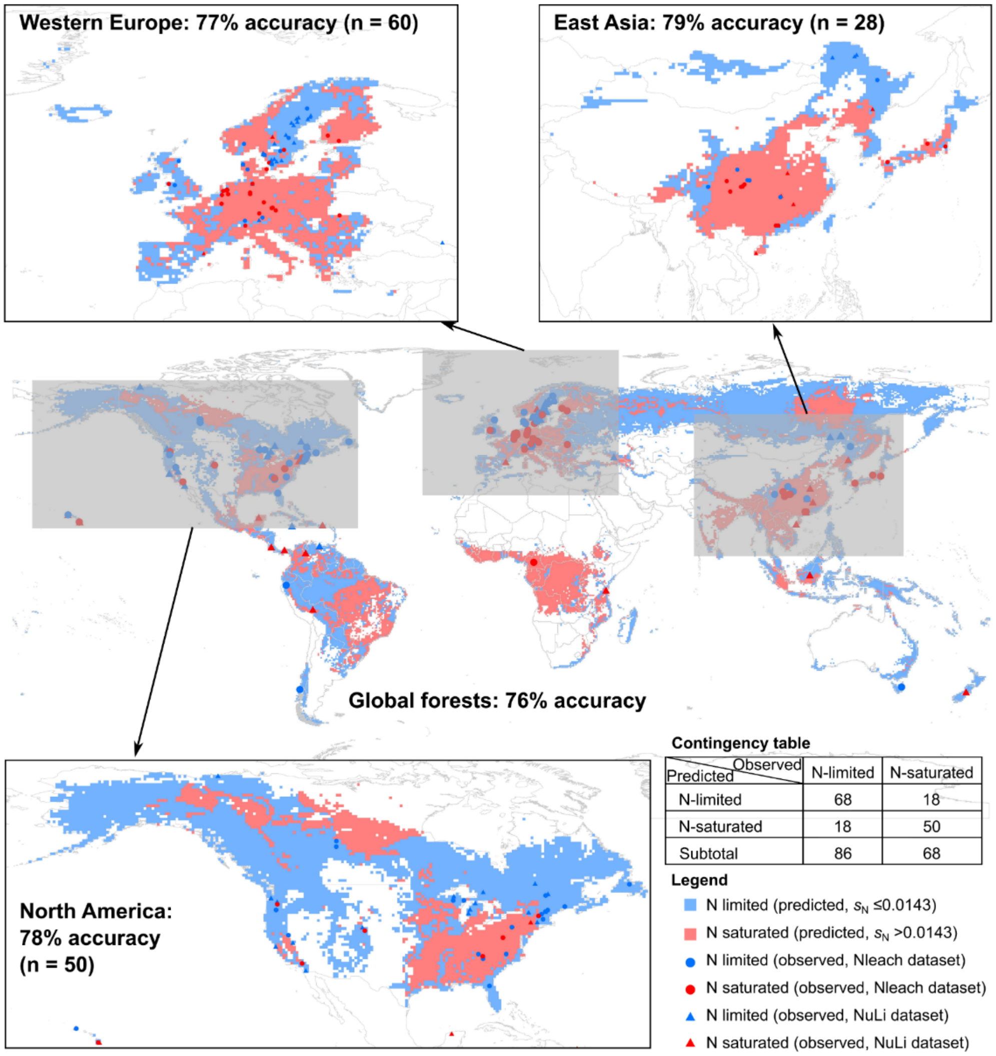


Fig. S5. Various forests are at different “stages” (in the stimulating-suppressing-weakening “three stages” framework), in accordance with the overall effects of N deposition on soil CH_4 fluxes in the forests.



Contingency table

Observed Predicted	N-limited	N-saturated
N-limited	68	18
N-saturated	18	50
Subtotal	86	68

Fig. S6. Classified N-limited and N-saturated forests based on the sensitivity of soil N_2O emission to N deposition (s_N) compared with field-observed N limitation or saturation status, with extra details in regions where field-observations were more abundant.

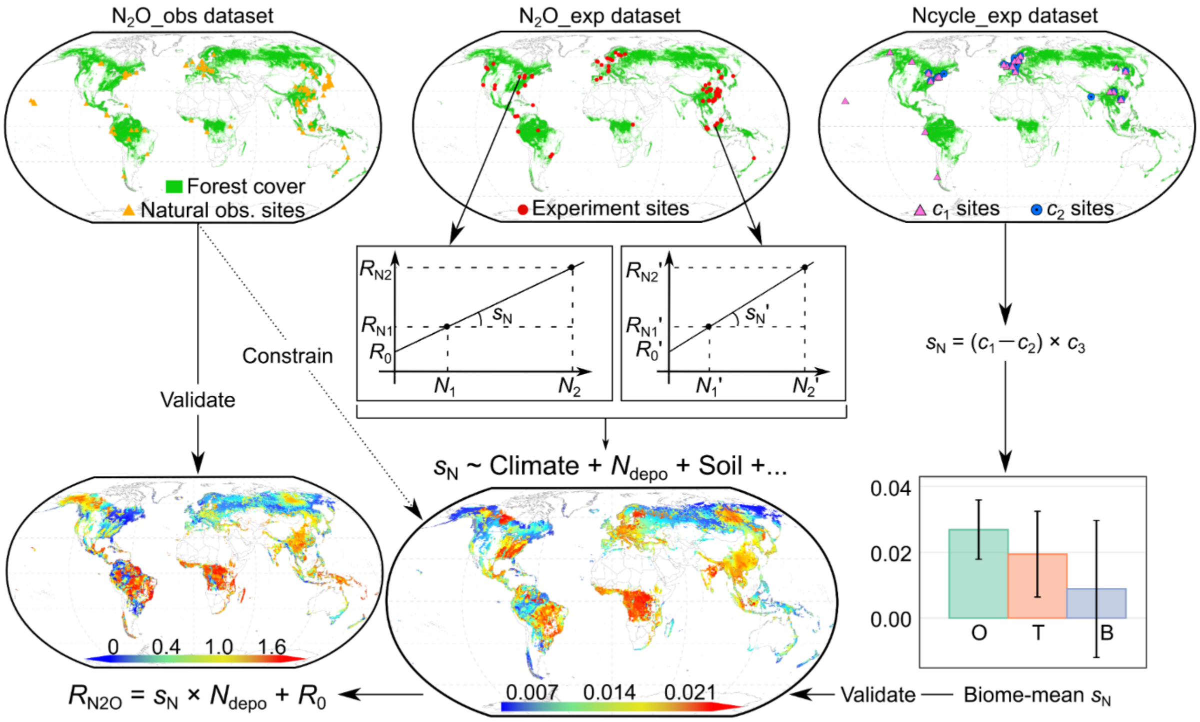


Fig. S7. Workflow illustrating the quantification and validation of the sensitivity of soil N_2O emission to N deposition (s_N) of global forests. N_1 and N_2 are different rates of low N input, and R_{N1} and R_{N2} are the corresponding soil N_2O emission rates. N_{depo}: N deposition rate (kgN ha^{-1} yr^{-1}); R_0: background soil N_2O emission rate (kgN_2O-N ha^{-1} yr^{-1}); c_1: sensitivity of N loss to N deposition (kgN kgN^{-1}); c_2: sensitivity of N leaching to N deposition (kgN kgN^{-1}); c_3: ratio of N_2O to other gaseous end-products from nitrification and denitrification processes (kgN_2O-N kgN^{-1}). O: Tropical; T: Temperate; B: Boreal.

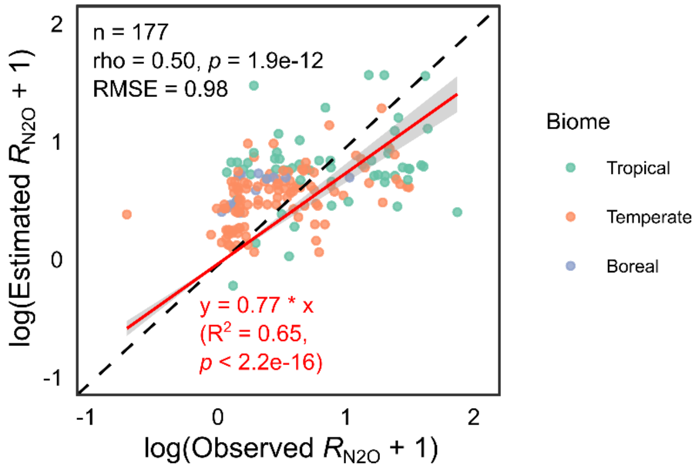


Fig. S8. Comparing estimated and observed soil N_2O emission rates (R_{N2O}). Observations were aggregated to 0.5°×0.5° grids to match with the spatial resolution of the environmental factors. Each point represents a grid-year. Points of different colors represent grid-years in different biomes. Dashed black line is the 1:1 line. The red line and fonts show a linear regression model on estimated and observed R_{N2O}.

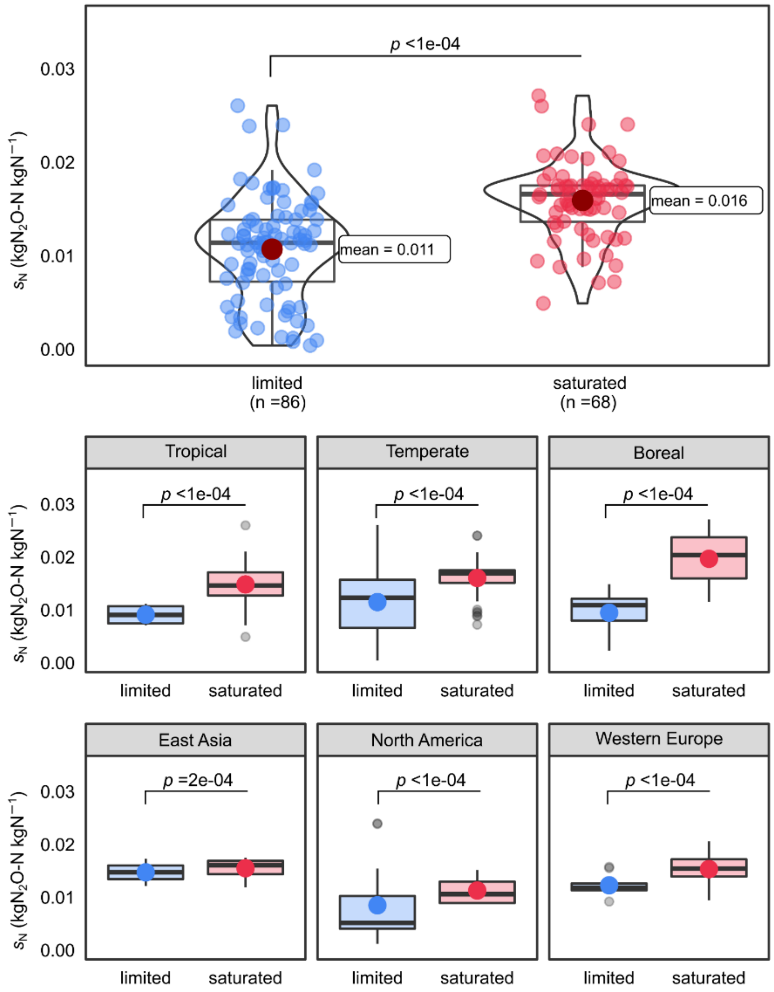


Fig. S9. Comparing the sensitivity of soil N_2O emission to N deposition (s_N) of N-limited and N-saturated forests on global and regional scales.

Table S1. Parameters of segmented linear regression models on soil CH_4 flux (R_{CH4}) and N input rate.

No.	Model (R_{CH4} ~ N input rate)	Parameters
①	y = -0.037*x – 2.45	n = 53, R^2 = 0.01, p = 0.44
②	y = 0.045*x – 5.75	n = 49, R^2 = 0.06, p = 0.09
③	y = -0.004*x – 0.73	n = 29, R^2 = 0.00, p = 0.80
④	y = 0.096*x – 5.28	n = 121, R^2 = 0.10, p = 0.0003
⑤	y = -0.006*x – 1.53	n = 196, R^2 = 0.03, p = 0.02

Table S2. Parameters of the constructed random forest regression models.

Model	R_{CH4} ~ MAT + MAT.cv + MAP + MAP.cv + N_{depo} + N_{depo}.cv + Sand + Clay + s_N	
Parameters	mtry	3
	ntree	1000
	Number of runs	1000

R_{CH4}: soil CH_4 emission rate; MAT: mean annual temperature; MAP: mean annual precipitation; N_{depo}: mean annual N deposition; Sand: soil sand content; Clay: soil Clay content; MAT.cv, MAP.cv and N_{depo}.cv are the corresponding coefficients of temporal variation; s_N: sensitivity of soil N_2O emission to N deposition, which indicates soil N limitation or saturation status. The predictors were selected based on mechanistic relevance and data availability.

Table S3. Linear models on soil N_2O emission rate (R_{N2O}) and N input rate (model: R_{N2O} ~ N input rate) built with low N input data (N addition rate ≤ 150 kgN ha^{-1} yr^{-1}) from global forest experiment sites, and the derived sensitivity (s_N) of soil N_2O emission to N deposition and background N_2O emission rate (R_0).

No.	Longitude range	Latitude range	Biome	s_N	R_0	n	adj.R^2	p value	References
1	(19,19.5)	(64,64.5)	Boreal	0.002	0.045	2	NA	NA	(Rutting et al., 2021)
2	(30.5,31)	(62.5,63)	Boreal	0.025	5.132	4	0.14	0.347	(Regina et al., 1998)
3	(22.5,23)	(62,62.5)	Boreal	0.013	0.538	2	NA	NA	(Ojanen et al., 2019)
4	(8,8.5)	(58.5,59)	Boreal	0.026	0.343	6	0.57	0.052	(Sitaula et al., 1995a, 1995b)
5	(-3.5,-3)	(55.5,56)	Temperate	0.02	0.258	6	0.18	0.224	(U. M. Skiba et al., 1998)
6	(-3,-2.5)	(55.5,56)	Temperate	0.006*	-0.009	6	0.73	0.019	(U. Skiba et al., 1999; U. M. Skiba et al., 1998)
7	(1.5,2)	(52.5,53)	Temperate	0.004	0.233	2	NA	NA	(U. M. Skiba et al., 1998)
8	(9.5,10)	(51.5,52)	Temperate	0.042*	0.51	10	0.48	0.015	(Borken et al., 2002; Brumme & Beese, 1992; Marife D Corre et al., 2003)
9	(128.5,129)	(47,47.5)	Boreal	0.015	0.777	11	0.02	0.300	(He, 2015; L. Song et al., 2017; Tian et al., 2018)
10	(8.5,9)	(47,47.5)	Temperate	0.003	-0.062	4	0.63	0.134	(Krause et al., 2013)
11	(-80.5,-80)	(43.5,44)	Temperate	0.009	1.374*	4	0.79	0.073	(Lutes et al., 2016)
12	(-72.5,-72)	(43,43.5)	Temperate	0.012	-0.216	2	NA	NA	(M. S. Castro et al., 1992)
13	(141,141.5)	(43,43.5)	Temperate	0.025	1.647	2	NA	NA	(Y. S. Kim et al., 2012)
14	(-72.5,-72)	(42.5,43)	Temperate	0.001	0.074	6	0.05	0.323	(Richard D. Bowden et al., 1991)
15	(128,128.5)	(42,42.5)	Temperate	0.01	0.67	2	NA	NA	(Geng et al., 2017)
16	(127.5,128)	(41.5,42)	Temperate	0.029	2.287	13	0.11	0.141	(Bai et al., 2014; Cheng et al., 2016; B. Peng et al., 2021)
17	(-80.5,-80)	(41.5,42)	Temperate	0.003	0.217	2	NA	NA	(R. D. Bowden et al., 2000)
18	(-4,-3.5)	(40,40.5)	Temperate	0.001*	0.026*	4	0.95	0.017	(Lafuente et al., 2020)
19	(112,112.5)	(36.5,37)	Temperate	0.056	2.754	3	0.98	0.068	(H. Yu, 2019)
20	(111,111.5)	(31.5,32)	Temperate	0.013**	0.483	27	0.28	0.003	(Zhaolan Lin, 2013; Zhaolan Lin et al., 2012; R. Wang, 2012; Xu et al., 2017)
21	(110,110.5)	(31.5,32)	Temperate	0.023	-0.31	4	0.54	0.166	(Pan, 2013)
22	(120.5,121)	(30.5,31)	Temperate	0.017	1.135	4	0.51	0.181	(Tu & Zhang, 2018)
23	(119.5,120)	(30,30.5)	Temperate	0.003	1.238***	16	0.01	0.308	(X. Chen, 2014; X. Chen et al., 2014; Ziwen Lin, 2019; X. Z. Song et al., 2020; Z. Wang, 2014)
24	(120,120.5)	(30,30.5)	Temperate	0.012**	0.834*	12	0.64	0.001	(J. Zhang, 2013; J. Zhang et al., 2013)
25	(106.5,107)	(29.5,30)	Temperate	0.025*	0.875*	3	1	0.018	(Xie et al., 2018)

26	(115.5,116)	(29.5,30)	Temperate	0.012	2.025	6	0.14	0.248	(C. Li et al., 2019)
27	(116.5,117)	(28,28.5)	Temperate	0.013	0.16	2	NA	NA	(Fan et al., 2020)
28	(118,118.5)	(27,27.5)	Tropical	0.015	1.948	9	0.12	0.190	(S. Chen, 2012)
29	(115,115.5)	(26.5,27)	Tropical	0.026***	-0.092	54	0.47	<0.001	(Dang, 2015; X. Li, 2017; X. Y. Li et al., 2015; Sun & Zhang, 2015; J. Wang, 2016; L. Wang, 2015; L. Wang et al., 2016; Y. Wang, 2015; Y. S. Wang et al., 2016; L. Zhang, 2013)
30	(117,117.5)	(26,26.5)	Tropical	0.007	0.5	3	0.55	0.313	(Wu, 2018)
31	(118,118.5)	(25.5,26)	Tropical	0.012	0.601	4	0.33	0.257	(Yuan, 2016)
32	(113,113.5)	(23.5,24)	Tropical	0.014	-0.226	3	0.77	0.220	(Cai, 2013)
33	(112.5,113)	(23,23.5)	Tropical	0.027*	0.19	22	0.15	0.041	(H. Chen et al., 2016; Gao et al., 2017; Mo et al., 2006)
34	(112.5,113)	(22.5,23)	Tropical	0.004	1.919***	14	0.11	0.129	(W. Zhang et al., 2014)
35	(106.5,107)	(22,22.5)	Tropical	0.012***	-0.038	8	0.84	0.001	(Hong, 2015)
36	(107,107.5)	(22,22.5)	Tropical	0.043*	-0.089	10	0.51	0.013	(R. Li et al., 2014, 2015; Yang, 2015; Kai Zhang et al., 2015)
37	(107.5,108)	(22,22.5)	Tropical	0.007**	0.589**	4	0.98	0.007	(K. Zhang et al., 2017)
38	(101,101.5)	(21.5,22)	Tropical	0.037	2.101*	9	0.18	0.144	(Yan, 2006; Zhou et al., 2016)
39	(110.5,111)	(21,21.5)	Tropical	0.018	3.195	3	0.69	0.256	(F. M. Wang et al., 2014)
40	(-80,-79.5)	(9,9.5)	Tropical	0.021**	0.674	8	0.71	0.005	(M. D. Corre et al., 2014; Koehler et al., 2009)
41	(-82.5,-82)	(8.5,9)	Tropical	0.019	1.063	8	0.32	0.083	(M. D. Corre et al., 2014; Koehler et al., 2009)
42	(116.5,117)	(6,6.5)	Tropical	0.007**	0.517**	10	0.61	0.005	(Hall et al., 2004)
43	(31.5,32)	(1.5,2)	Tropical	0.018***	1.756***	4	1	0.001	(Tamale et al., 2021)
44	(102,102.5)	(-1.5,-1)	Tropical	0.022**	0.919*	7	0.84	0.002	(Aini et al., 2015)
45	(-79.5,-79)	(-4,-3.5)	Tropical	0.005	0.135	3	0.44	0.356	(Muller et al., 2015)
46	(-79,-78.5)	(-4.5,-4)	Tropical	0.006	0.471	3	0.5	0.333	(Muller et al., 2015)
47	(-79.5,-79)	(-4.5,-4)	Tropical	0.006	-0.11	3	0.95	0.106	(Muller et al., 2015)

* $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$; NA, not applicable

Table S4. Generalized linear models on environmental factors and the sensitivity (s_N) of soil N_2O emission to N deposition and the background N_2O emission rate (R_0).

	Estimate	**SE**	**t**	***p***
Refined model on s_N[†] (Deviance explained = 91.1%, n=46)				
Clay	4.77E-03	1.83E-03	2.605	0.013*
Sand	3.15E-03	9.20E-04	3.419	0.001**
log(N_{depo})	2.01E-02	1.14E-02	1.769	0.085
Clay × log(N_{depo}.cv)	2.13E-03	9.35E-04	2.282	0.028*
Sand × log(N_{depo}.cv)	1.17E-03	3.82E-04	3.056	0.004**
Clay × Sand	-1.90E-04	6.94E-05	-2.735	0.009**
Clay × Sand × log(N_{depo}.cv)	-1.14E-04	3.66E-05	-3.112	0.003**
Refined model on R_0[‡] (Deviance explained = 43.2%, n = 45)				
log(N_{depo}.cv)	1.99E-01	9.56E-02	2.084	0.043*
MAT × Sand × Clay	3.04E-06	5.99E-07	5.072	0.000***
MAP × MAP.cv × log(N_{depo})	-8.31E-04	2.91E-04	-2.854	0.007**

MAT: mean annual temperature; MAP: mean annual precipitation; N_{depo}: mean annual N deposition; Sand: soil sand content; Clay: soil clay content.

[†] s_N ~ (Clay + Sand + log(N_{depo}) + Clay × log(N_{depo}.cv) + Sand × log(N_{depo}.cv) + Clay × Sand + Clay × Sand × log(N_{depo}.cv))^2

[‡] R_0 ~ EXP(log(N_{depo}.cv) + MAT × Sand × Clay + MAP × MAP.cv × log(N_{depo})) – 0.5

* p <0.05; ** p <0.01; *** p <0.001

Data Set S1. (separate file)

Compiled dataset on soil CH_4 flux from N addition experiments in global forests (CH_4_exp dataset in main text).

Data Set S2. (separate file)

Compiled data on soil CH_4 flux under natural conditions in global forests (CH_4_obs dataset in main text).

Data Set S3. (separate file)

Compiled dataset on soil N_2O emission rate from N addition experiments in global forests (N_2O_exp dataset in Text S1).

Data Set S4. (separate file)

Compiled data on soil N_2O emission rate under natural conditions in global forests (N_2O_obs dataset in Text S1).

Data Set S5. (separate file)

Compiled dataset on N loss rate, N leaching rate and change rate of soil N pool from N addition experiments in global forests (Ncycle_exp dataset in Text S1).

Data Set S6. (separate file)

Compiled dataset on global forest N saturation status (limited or saturated) indicated by N leaching rate (Nleach dataset in Text S1).

Data Set S7. (separate file)

An existing dataset from Du et al. (2020) on global forest N saturation status (limited or saturated) indicated by plant growth response to N input (NuLi dataset in Text S1).

Data Set S8. (separate file)

Data on environmental factors (MAT, MAP, N deposition rate, etc.) in global forests, extracted from spatial datasets mentioned in Methods section.

Data Set S9. (separate file)

Global forest soil methane budgets estimated in previous studies.

Code S1. (separate file)

R code script used to carry out the data analysis processes, and produce the figures.