

# **Soil nitrous oxide emissions across the northern high latitudes**

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## Abstract

Nitrous oxide (N<sub>2</sub>O) is the most important stratospheric ozone-depleting agent based on current emissions and the third largest contributor to increased net radiative forcing. Increases in atmospheric N<sub>2</sub>O have been attributed primarily to enhanced soil N<sub>2</sub>O emissions. Critically, contributions from soils in the Northern High Latitudes (NHL, >50°N) remain poorly quantified despite their vulnerability to permafrost thawing induced by climate change. An ensemble of six terrestrial biosphere models suggests NHL soil N<sub>2</sub>O emissions doubled since the preindustrial 1860s, increasing on average by  $2.0 \pm 1.0 \text{ Gg N yr}^{-1}$  ( $p < 0.01$ ). This trend reversed after the 1980s because of reduced nitrogen fertilizer application in non-permafrost regions and increased plant growth due to CO<sub>2</sub> fertilization suppressed emissions. However, permafrost soil N<sub>2</sub>O emissions continued increasing attributable to climate warming; the interaction of climate warming and increasing CO<sub>2</sub> concentrations on nitrogen and carbon cycling will determine future trends in NHL soil N<sub>2</sub>O emissions.

## Key Points

1. N<sub>2</sub>O emissions from northern high latitudes during 1997-2014 are estimated at 0.5–1.3 Tg N yr<sup>-1</sup>, and soil was the largest source.
2. Northern high latitudes soil N<sub>2</sub>O emissions increased from  $0.3 \pm 0.1 \text{ Tg N yr}^{-1}$  in 1861 to  $0.6 \pm 0.3 \text{ Gg N yr}^{-1}$  in 2016.
3. Climate change stimulated soil N<sub>2</sub>O emissions, while the increased atmospheric CO<sub>2</sub> concentration suppressed emissions.

## Plain Language Summary

Soils in the Northern High Latitudes (NHL) store large amounts of nitrogen, providing rich substrates for the emissions of nitrous oxide (N<sub>2</sub>O) which is a potent greenhouse gas and ozone-depleting substance. The NHL has experienced rapid climate warming in recent decades, however, to what extent climate and other environmental factors have affected soil N cycling and N<sub>2</sub>O emissions in the NHL remain poorly quantified. This study has provided the first quantification of the magnitudes and spatiotemporal variations of soil N<sub>2</sub>O emissions across the NHL and showed that the NHL contributed about 8% of the increase in global soil N<sub>2</sub>O emissions since pre-industrial period (the 1860s). Our results further reveal that changes in climate and atmospheric CO<sub>2</sub> concentration not only largely affected historical variations in soil N<sub>2</sub>O emissions from the NHL but also will determine their future trends. Our study suggests the need to better understand climate and CO<sub>2</sub> controls on soil N<sub>2</sub>O emissions and nitrogen cycling across the NHL and to improve their representation in earth system models.

## 1 Introduction

Nitrous oxide (N<sub>2</sub>O) emissions have received increasing attention, because N<sub>2</sub>O is the most important stratospheric ozone-depleting agent based on current emissions [Ravishankara *et al.*, 2009] and the third largest contributor to net radiative forcing by greenhouse gases [Canadell *et al.*, 2021; Etminan *et al.*, 2016]. The large amount of nitrogen additions to soils since the preindustrial period has significantly increased the atmospheric N<sub>2</sub>O burden [Canadell *et al.*, 2021; Tian *et al.*, 2020]. Denitrification and nitrification are two primary soil processes controlling N<sub>2</sub>O production, which are regulated by multiple factors such as temperature, water availability, acidity, substrate availability and microbial diversity [Butterbach-Bahl *et al.*, 2013; Rees *et al.*, 2013].

91 Over the past 40 years, the northern high latitudes, usually defined as the region north of 50°N  
92 [Watts *et al.*, 2012], have experienced climate warming at a rate faster than anywhere else on Earth  
93 [Rantanen *et al.*, 2022], a trend expected to continue in the coming decades [Masson-Delmotte *et*  
94 *al.*, 2021]. Therefore, there is an urgent need to understand and quantify how changes in climate  
95 and other environmental factors since the pre-industrial era have affected soil N<sub>2</sub>O emissions from  
96 the NHL and thus have shaped the strength of climate-biogeochemical feedback.

97 The terrestrial nitrogen cycle in the NHL is closely related with permafrost, which underlays more  
98 than 60% of the area [Brown *et al.*, 1997]. Although large N stocks are stored in this region [Harden  
99 *et al.*, 2012; Hugelius *et al.*, 2020], the associated soil N<sub>2</sub>O emissions have received little attention  
100 because they were considered to be small due to limited microbial activity and low mineralization  
101 rates under low-temperature and waterlogged conditions [Voigt *et al.*, 2020]. However, recent in-  
102 situ studies found that both barren and vegetated soils in the NHL can emit substantial amounts of  
103 N<sub>2</sub>O [Marushchak *et al.*, 2011; Marushchak *et al.*, 2021; Repo *et al.*, 2009; Voigt *et al.*, 2017b].  
104 Meanwhile, Arctic amplification, the phenomenon that climate change is amplified in the NHL, is  
105 projected to continue in the 21st century [Christensen *et al.*, 2013; Pithan and Mauritsen, 2014]  
106 with further implications for N<sub>2</sub>O emissions: first, a large amount of immobile N stored in  
107 permafrost becomes available for decomposition and remobilization after permafrost thawing;  
108 second, rapid warming enhances N mineralization and promotes nitrification and denitrification;  
109 and third, warming may also promote biological nitrogen fixation (BNF), increasing ecosystem N  
110 availability and thereby potentially also N<sub>2</sub>O production. Field experiments also confirm that  
111 warming can significantly increase N<sub>2</sub>O emissions from permafrost-affected soils [Cui *et al.*, 2018;  
112 Voigt *et al.*, 2017b; Wang *et al.*, 2017].

Another influential factor for N<sub>2</sub>O emissions in the NHL is the atmospheric CO<sub>2</sub> concentrations. Elevated atmospheric CO<sub>2</sub> concentrations do not have significant direct effects on reactive N flows controlling N<sub>2</sub>O production, but can indirectly affect soil N<sub>2</sub>O emissions by changing plant nitrogen uptake and root exudates due to enhanced plant growth [Usyskin-Tonne *et al.*, 2020]. On one hand, elevated atmospheric CO<sub>2</sub> promotes plant growth and thus more absorption of soil mineral N, restricting N<sub>2</sub>O production [Tian *et al.*, 2019]. On the other hand, it may stimulate denitrification-derived N<sub>2</sub>O emissions by increasing plant biomass and hence carbon substrate availability [Kammann *et al.*, 2008]. Additionally, elevated CO<sub>2</sub> can affect soil moisture by improving plant water-use efficiency, which can increase anaerobic conditions that stimulate denitrification [Butterbach-Bahl *et al.*, 2013]. Such contrasting effects of elevated CO<sub>2</sub> concentrations on N<sub>2</sub>O emissions have been observed in field experiments [Dijkstra *et al.*, 2012; Liu *et al.*, 2018; X Sun *et al.*, 2018] but the magnitude of the CO<sub>2</sub> effect on northern soil N<sub>2</sub>O emissions remains poorly understood.

Here, we investigated NHL soil N<sub>2</sub>O emissions using six process-based terrestrial biosphere models (TBMs) from the global N<sub>2</sub>O Model Intercomparison Project (NMIP) [Tian *et al.*, 2018]. Using factorial simulation experiments, we quantified the contributions of different driving factors, particularly climate change and rising atmospheric CO<sub>2</sub>, to the variations in soil N<sub>2</sub>O emissions during 1861-2016. Statistical methods were further employed to disentangle the effects of temperature and precipitation on soil N<sub>2</sub>O emissions. We also compared bottom-up (BU, including process-based TBMs for soil emissions and emission factor approaches for non-soil emissions) estimates of N<sub>2</sub>O emissions with those of three atmospheric inversion frameworks (top-down, TD) [Rona L. Thompson *et al.*, 2019] to investigate the uncertainties in current estimates of N<sub>2</sub>O emissions from the NHL.

## 2 Materials and methods

### 2.1 Data sources

#### 2.1.1 Soil N<sub>2</sub>O emissions

An ensemble estimate of soil N<sub>2</sub>O emissions from the NHL was derived from simulations by the six TBMs that participated in the NMIP: (1) DLEM [Tian *et al.*, 2015], (2) LPJ-GUESS [Olin *et al.*, 2015], (3) LPX-Bern [Joos *et al.*, 2020], (4) O-CN [Zaehle *et al.*, 2011], (5) ORCHIDEE-CNP [Goll *et al.*, 2017; Y Sun *et al.*, 2021], and (6) VISIT [Inatomi *et al.*, 2010]. Each model performed a subset of seven simulations (S0-S6) to quantify N<sub>2</sub>O emissions from both agricultural and natural soils, and to disentangle the effects of multiple environmental factors on N<sub>2</sub>O emissions (Table S1). The differences between pairs of simulations, i.e. S1-S2, S2-S3, S3-S4, S4-S5, S5-S6, and S6-S0, were used to evaluate the effects of manure N, mineral N fertilizer, atmospheric N deposition, land use and land cover change (LULCC), atmospheric CO<sub>2</sub> concentration, and climate, respectively. More information about the model simulation protocol and forcing data can refer to Tian *et al.* [2018]. Among the six NMIP models, LPJ-GUESS and LPX-Bern have dedicated permafrost modules and consider freeze-thaw processes; O-CN lacks an explicit permafrost representation but describes freeze-thaw cycles; the other models have no explicit representation of the permafrost layer or freeze-thaw processes.

#### 2.1.2 Fire-induced N<sub>2</sub>O emissions and non-soil anthropogenic N<sub>2</sub>O emissions

N<sub>2</sub>O emissions from biomass burning were from the GFED4.1s dataset. N<sub>2</sub>O emissions from non-soil anthropogenic sources were obtained from EDGAR 6.0 [Crippa *et al.*, 2019]. EDGAR non-soil anthropogenic emissions were combined with GFED biomass burning emissions and with

NMIP soil emissions to constitute BU estimates of total N<sub>2</sub>O emissions, aiming to make comparison with TD estimates.

### **2.1.3 Top-down N<sub>2</sub>O emission estimates**

Three independent atmospheric inversion models were used: GEOS-Chem [Wells *et al.*, 2018], INVICAT [Wilson *et al.*, 2014] and MIROC4-ACTM [Patra *et al.*, 2018; Patra *et al.*, 2022]. GEOS-Chem and INVICAT used the same prior estimates: soil emissions from the O-CN model, biomass burning emissions from GFEDv4.1s, and non-soil anthropogenic emissions from EDGAR v4.2FT2010. The MIROC4-ACTM prior used natural soil emissions from the VISIT model, and all anthropogenic emissions from EDGAR 4.2. The MIROC4-ACTM prior included agricultural burning but did not explicitly include wildfire emissions. All models used the Bayesian inversion framework to find the optimal emissions that provide the best agreement to observed N<sub>2</sub>O mixing ratios while being coupled to an atmospheric transport model.

## **2.2 Statistical methods**

The path analysis model (PAM) was used to investigate how climatic factors affected permafrost soil N<sub>2</sub>O emissions. PAM can deal with complex relationships among multiple independent and dependent variables, and disentangle direct and indirect effects of the explanatory variables on the response variable [Alwin and Hauser, 1975; You and Pan, 2020]. Here, we developed the conceptual model by specifying the relationships between climatic factors and soil N<sub>2</sub>O emissions and considering the interactions between these factors. We also conducted partial correlation analysis between soil N<sub>2</sub>O emissions and temperature/precipitation. The temporal sensitivities of



soil N<sub>2</sub>O emissions to temperature and precipitation were fitted using a multiple regression model. The Mann–Kendall test was used to assess the significance of trends in N<sub>2</sub>O emissions.

### 3 Results

#### 3.1 Spatiotemporal variations of soil N<sub>2</sub>O emissions since the 1860s

Multi-model ensemble estimates show that soil N<sub>2</sub>O emissions from the NHL increased from 312±125 Gg N yr<sup>-1</sup> in 1861 to 605±269 Gg N yr<sup>-1</sup> in 2016 (Fig.1a), with an average increase rate of 2.0±1.0 Gg N yr<sup>-1</sup> ( $p<0.01$ ). Soil N<sub>2</sub>O emissions from non-permafrost regions dominated the temporal variations of total NHL emissions, which were relatively stable over the first five decades, then rapidly increased from the 1920s to the 1980s, and peaked in the 1980s. In the late 1980s and early 1990s, northern soil N<sub>2</sub>O emissions drastically decreased and fluctuated afterwards. Meanwhile, soil N<sub>2</sub>O emissions from permafrost regions showed different temporal dynamics; they remained relatively stable before the 1980s, and rapidly increased thereafter. In the 1860s, the highest emission density occurred in Central Europe. During 1861-2016, soil N<sub>2</sub>O emissions from most regions significantly increased. In the recent decade (2007-2016), Western Europe had the highest emission density (Fig.1b-d), and more than half of the soil N<sub>2</sub>O emissions were from croplands (Fig. S2). During 1861-1980, the fastest increase in N<sub>2</sub>O emissions occurred in Western and Central Europe where the average increase exceeded  $2\times 10^{-4}$  g N m<sup>-2</sup> yr<sup>-1</sup> (Fig.1e). However, trends in soil N<sub>2</sub>O emissions have largely changed since 1980, with emissions significantly decreasing in Eastern Europe and Russia but rapidly increasing in Siberia and Southern Canada (Fig.1f).

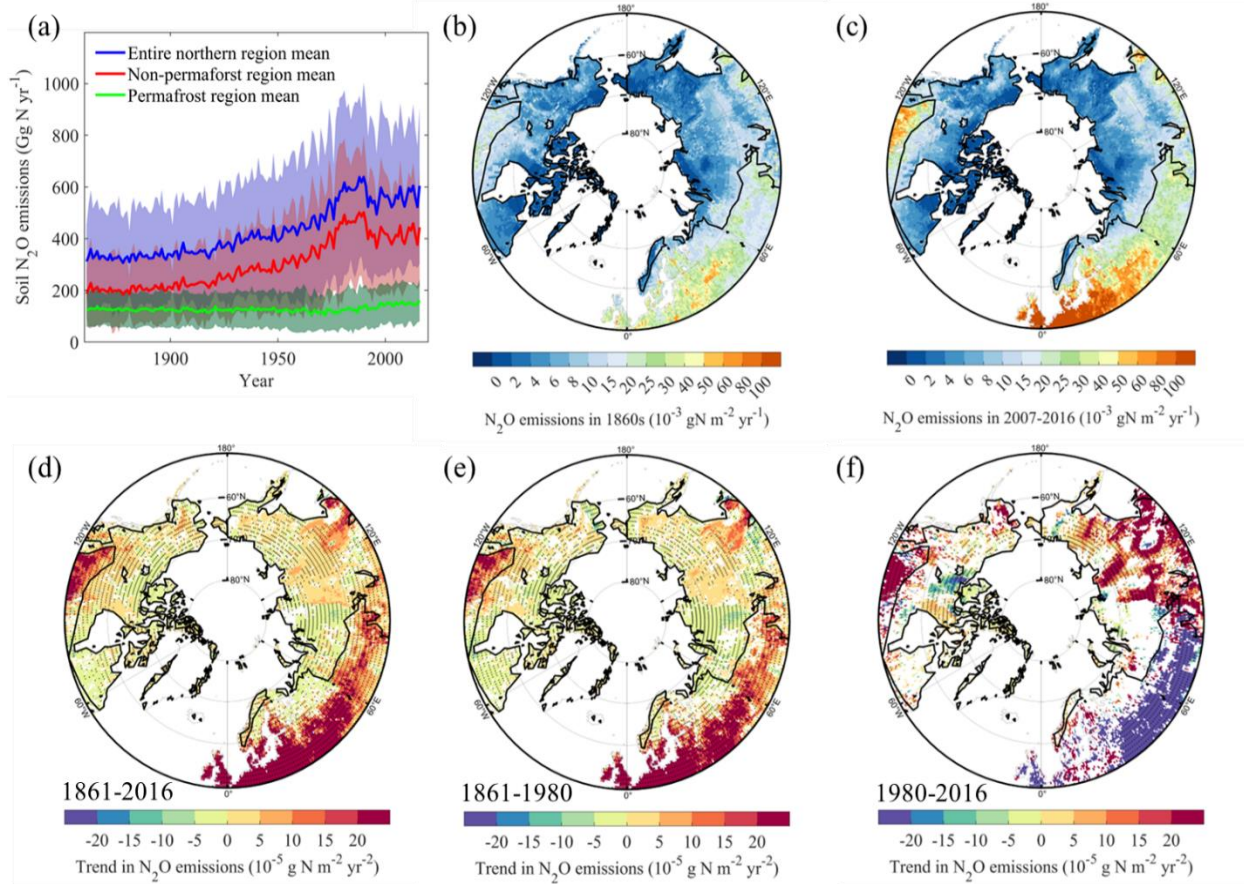


Fig. 1: (a) Changes in soil N<sub>2</sub>O emissions from the NHL, the shaded area indicates one standard deviation of all estimates. (b) and (c) show spatial pattern of mean annual soil N<sub>2</sub>O emissions during the 1860s and 2007-2016, respectively. Trends in soil N<sub>2</sub>O emissions during 1861-2016 (d), 1861-1980 (e), and 1980-2016 (f); grids with non-significant trends ( $p \geq 0.05$ ) were excluded, and stippling indicates where a majority of models (at least 4 out of 6) agree on the sign of the trend.

### 3.2 Contributions of different driving factors to soil N<sub>2</sub>O emissions during 1861-2016

Our results derived from factorial simulations suggested that increasing atmospheric CO<sub>2</sub> concentrations reduced NHL soil N<sub>2</sub>O emissions, while the other five factors stimulated N<sub>2</sub>O emissions (Fig. 2a). Climate change played a dominant role in stimulating N<sub>2</sub>O emissions before

the 1930s and N inputs made increasing contributions from the 1940s to the 1980s. From the 1860s to the 1980s, fertilizer application contributed 53% to the increase in emissions, followed by atmospheric N deposition (26%), manure N application (15%), climate change (12%), and land use change (5%). The effect of increased atmospheric CO<sub>2</sub> (-10%) almost offset that of climate change. Since the 1980s, the role of anthropogenic N inputs in stimulating N<sub>2</sub>O emissions weakened gradually; by contrast, drastic warming and wetting made climate change increasingly important (Fig. S3). Over the entire study period, climate change made the second largest contribution (37%) to the increase of NHL soil emissions after N fertilizer application (42%). Climate change had a larger relative contribution to the emission increase in permafrost regions (Fig. 2c) than in non-permafrost regions. During 1861-2016, climate change contributed 114% (partly offset by the negative CO<sub>2</sub> effect) to the emission increase in permafrost regions, which was stronger than in non-permafrost regions (28%) (Fig. 2d). All individual models agreed that climate change made a larger relative contribution to emission increases in permafrost regions than in non-permafrost regions, and that the effects of climate change have increased since the 1980s (Fig. S4-6). In most northern regions, trends in soil N<sub>2</sub>O emissions were dominated by climate change; fertilizer only dominated trends in Western Europe and some intensive agricultural lands over Eastern Europe, Russia, and south Canada, while atmospheric N deposition dominated trends in part of Central and Eastern Europe. Regions dominated by other factors were relatively small (Fig. 2b).

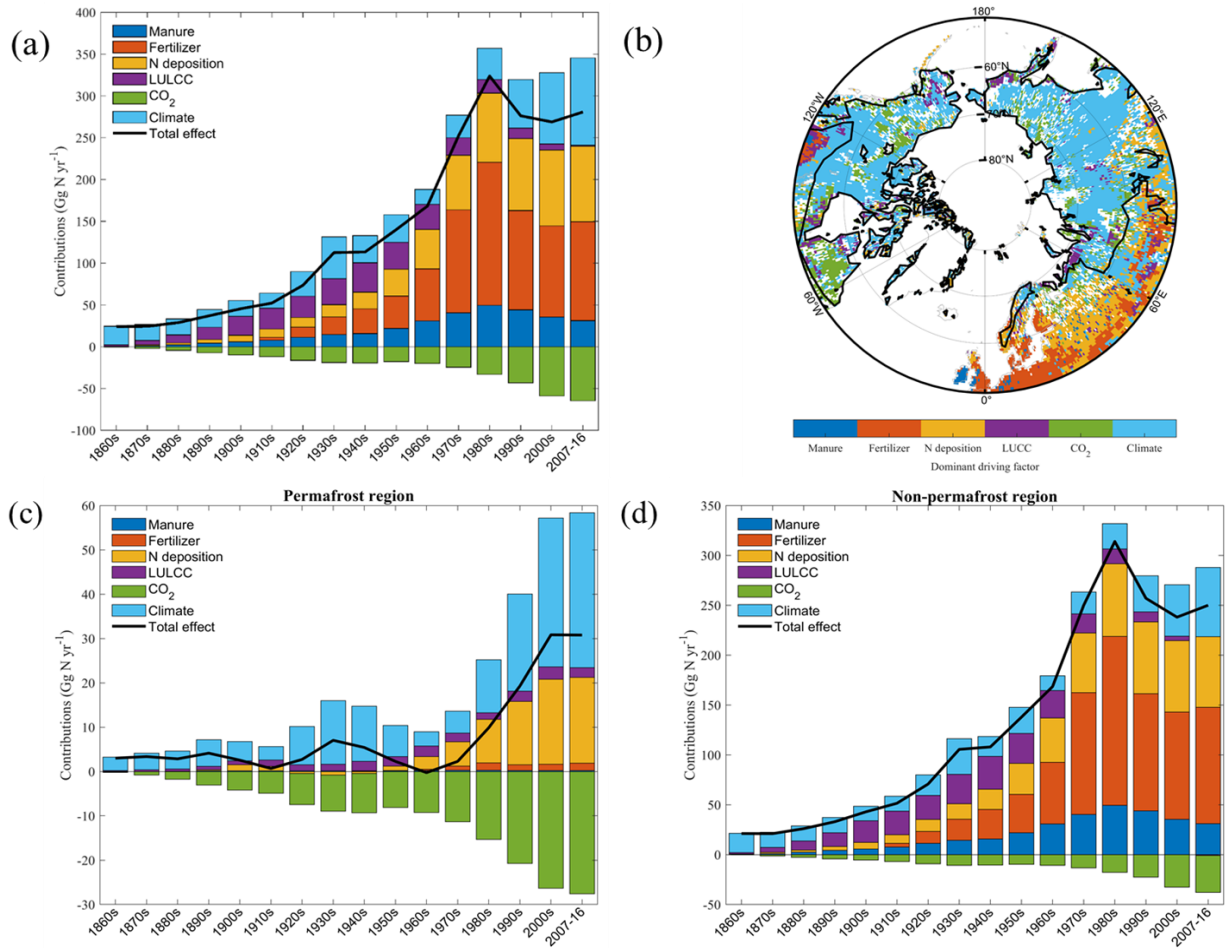


Fig. 2: (a) Decadal variations in the contributions of different driving factors. (b) Distribution of dominant driving factors of soil N<sub>2</sub>O emissions during 1861-2016; grids with non-significant trends were excluded. Contributions of different driving factors to soil N<sub>2</sub>O emissions from permafrost regions (c) and non-permafrost regions (d).

### 3.3 Effects of temperature and precipitation on soil N<sub>2</sub>O emissions

Temperature and precipitation changes alter soil microclimate, nutrient availability and microbial ecology, thereby influencing N<sub>2</sub>O emissions [Dalal and Allen, 2008]. For the entire NHL, both temperature and precipitation significantly increased during 1901-2016, with rates of 0.14 °C per decade and 0.38 mm yr<sup>-1</sup> (10% total increase since the 1900s), respectively (Fig. S3). According

to multiple regression model results, the sensitivities of soil N<sub>2</sub>O emissions to temperature and precipitation were  $29 \pm 21$  Gg N °C<sup>-1</sup> and  $0.4 \pm 0.7$  Gg N mm<sup>-1</sup> during 1901-2016, suggesting that warming and wetting increased soil N<sub>2</sub>O emissions by  $48 \pm 35$  Gg N yr<sup>-1</sup> and  $15 \pm 26$  Gg N yr<sup>-1</sup>, respectively. The path analysis model also suggested that warming contributed more to soil N<sub>2</sub>O emission increases than wetting (Fig. S7). Both warming and wetting have accelerated since 1980 (Fig. S3, S8, S9), with average rates of 0.38 °C per decade and 0.57 mm yr<sup>-2</sup>, respectively. At the same time, the sensitivities of soil N<sub>2</sub>O emissions to temperature and precipitation increased to  $38 \pm 22$  Gg N °C<sup>-1</sup> and  $1.2 \pm 0.8$  Gg N mm<sup>-1</sup>, respectively. These two factors together led to the large climate effects in the recent four decades.

Soil N<sub>2</sub>O emissions were positively correlated with temperature in most northern regions (Fig. S10a). Compared with the 1901-1980 period, warming after 1980 was more pronounced and prevalent (Fig. S8-9), which enhanced biological N fixation and net N mineralization and further promoted nitrification and denitrification (Fig. S11). During the study period, most of the NHL experienced significant warming (Fig. S10c), indicating that warming universally stimulated N<sub>2</sub>O emissions in this region. Recent manipulation experiments also suggest that warming can significantly increase soil N<sub>2</sub>O emissions from the NHL [Cui *et al.*, 2018; Voigt *et al.*, 2017b; Wang *et al.*, 2017]. Unlike temperature, the correlation between soil N<sub>2</sub>O emissions and precipitation varied spatially (Fig. S10b). Although a large area of the NHL experienced significant wetting (Fig. S10d), the positive effects of wetting on emissions from Eastern Europe, central Canada and Siberia were partly counteracted by the negative effects in Northern Europe and northwestern Russia, which explained why precipitation had a smaller effect than temperature on the regional total emissions.

### 3.4 Declining soil N<sub>2</sub>O emissions since the 1980s

Soil N<sub>2</sub>O emissions from the NHL rapidly increased before the 1980s, however, declined thereafter. Although total BNF over the NHL increased since 1980 (Fig. S12), the ensemble mean of soil N<sub>2</sub>O emissions from the NHL decreased at an average rate of -1.1 GgN yr<sup>-1</sup> ( $p < 0.05$ ) during 1980-2016 (Fig. 3a). The rapid decline in emissions during 1988-1996 was due to reduced fertilizer application, after which period the negative effect of CO<sub>2</sub> fertilization was enhanced (Fig. S14). The most pronounced decline occurred in Eastern Europe and Russia (Fig. 1f), mainly caused by the sharp decrease in external nitrogen inputs due to the collapse of the Soviet Union (Fig. S14). Concurrently, soil emissions from Siberia and Southern Canada significantly increased, due to climate change and nitrogen enrichment, respectively (Fig. S14). Soil N<sub>2</sub>O emissions fluctuated after 1998 because the positive climate effect was counteracted by combined effects of fertilizer application, CO<sub>2</sub> and land use change.

The dominant drivers of negative effects differed between permafrost and non-permafrost regions. In permafrost regions, elevated CO<sub>2</sub> concentration was the only factor suppressing soil N<sub>2</sub>O emissions and counteracted more than half of the climate-induced emissions (Fig. 3b). By contrast, reduced N fertilizer application, elevated CO<sub>2</sub> concentration and land use change jointly reduced emissions from non-permafrost regions (Fig. 3c). For the entire NHL, the atmospheric CO<sub>2</sub>-induced decline in soil N<sub>2</sub>O emissions surpassed the effect of reduced fertilizer application over the recent decade. Elevated atmospheric CO<sub>2</sub> significantly suppressed N<sub>2</sub>O emissions in most northern regions (Fig. S14). Since the 1980s, increased atmospheric CO<sub>2</sub> concentrations stimulated terrestrial gross primary production (Fig. S15a, c), thus enhancing plant nitrogen uptake (Fig. S15b, d) and reducing the availability of soil inorganic nitrogen, which finally suppressed N<sub>2</sub>O emissions.

The largest stimulation effect of CO<sub>2</sub> on vegetation growth and nitrogen uptake occurred in the boreal forests, where the CO<sub>2</sub>-induced suppression of N<sub>2</sub>O emissions was the most pronounced. Enhanced vegetation growth in the NHL has been reported in previous studies [Berner *et al.*, 2020; Myers-Smith *et al.*, 2020; Virkkala *et al.*, 2021]. Reduced N<sub>2</sub>O emissions due to enhanced plant growth and nitrogen uptake is also consistent with field observations in the NHL [Gong and Wu, 2021; Marushchak *et al.*, 2011; Stewart *et al.*, 2012].

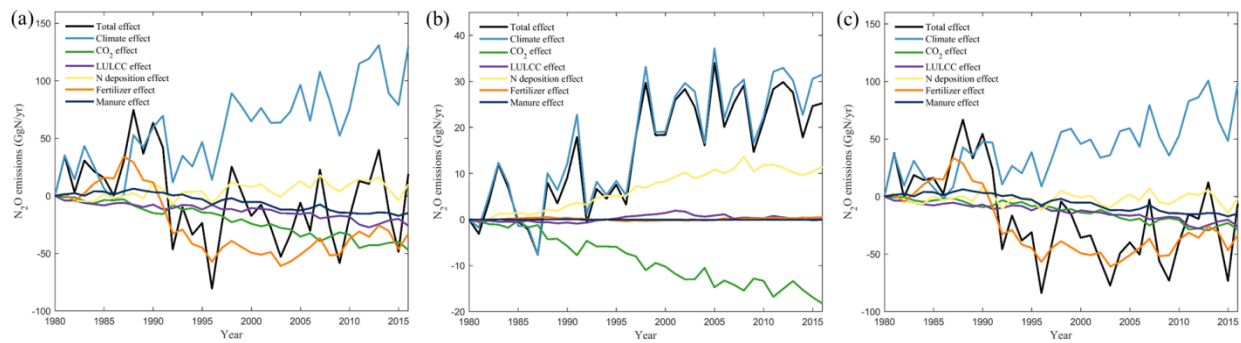


Fig. 3: Contributions of different driving factors in the entire NHL (a), permafrost regions (b), and non-permafrost regions (c) during 1980-2016.

### 3.5 Comparison with TD estimates

Using the current N<sub>2</sub>O observation network, TD models estimate total N<sub>2</sub>O emissions with its spatial distribution across the land but cannot well quantify the contributions of different sources. With the aim of comparing BU estimates with TD estimates, we added N<sub>2</sub>O emissions from soil, biomass burning and non-soil anthropogenic sources (Fig. S16) together to constitute BU estimates of total N<sub>2</sub>O emissions. According to the resulting BU estimates, soil was the largest source of N<sub>2</sub>O emissions in the NHL (mean value: 572 Gg N yr<sup>-1</sup> during 1998-2014), followed by non-soil anthropogenic sources (280 Gg N yr<sup>-1</sup>) and biomass burning (143 Gg N yr<sup>-1</sup>). Both BU and TD

approaches indicated similar spatial emission patterns (Fig. 4), but the ensemble mean of total BU estimate ( $995 \pm 267$  Gg N yr<sup>-1</sup>) was substantially higher than the TD estimate ( $668 \pm 134$  Gg N yr<sup>-1</sup>) for the overlapping 1998-2014 period. Both TD and BU approaches revealed that the total N<sub>2</sub>O emissions had no significant trend during this period ( $p > 0.05$ ). Removing N<sub>2</sub>O emitted by biomass burning and non-soil anthropogenic sources from the TD estimates, the remaining N<sub>2</sub>O exhibited a decreasing trend during 1998-2014 (from -10.0 to -3.2 Gg N yr<sup>-2</sup>, mean -7.3 Gg N yr<sup>-2</sup>), implying that the TD models also suggest a decreasing trend in NHL soil N<sub>2</sub>O emissions.

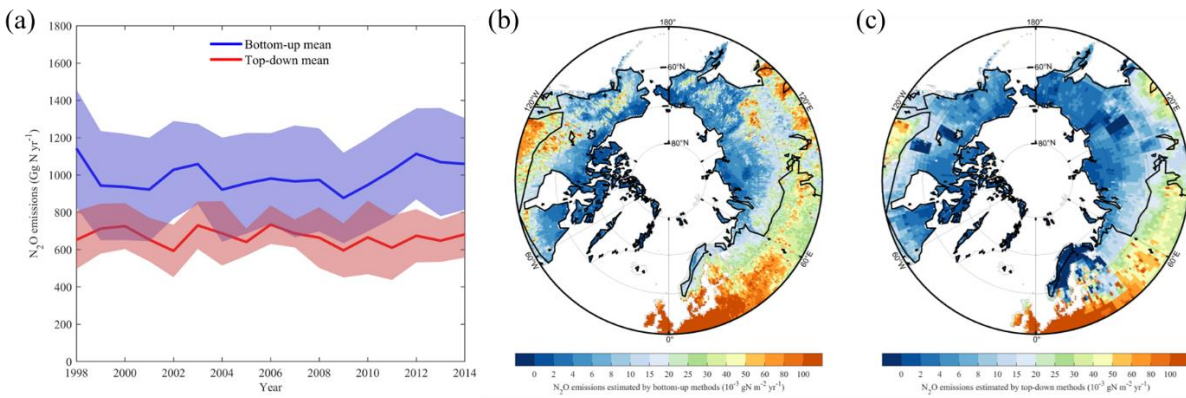


Fig. 4: (a) Comparison between TD and BU estimates of total N<sub>2</sub>O emissions, the lines represent the ensemble means and the shaded areas indicate one standard deviation of model estimates. Spatial pattern of total N<sub>2</sub>O emissions estimated by BU (b) and TD (c) approaches.

### 3.6 Comparison with empirical estimates

Based on site-level observation data, Voigt *et al.* [2020] estimated soil N<sub>2</sub>O emissions from permafrost regions using a simple extrapolation method, and proposed that peatlands had the highest N<sub>2</sub>O emissions among natural permafrost ecosystems. However, these extrapolation-based estimates have large uncertainties, with the implied annual soil N<sub>2</sub>O emissions from the NHL ranging from 140 to 1030 Gg N<sup>-1</sup>. In particular, estimates based on mean fluxes are an order of



magnitude larger than those based on median fluxes because of several N<sub>2</sub>O emission hot spots. Combining observed peatland annual fluxes and peatland distribution maps, *Hugelius et al.* [2020] estimated a much smaller northern peatland source of  $22 \pm 5$  Gg N·y<sup>-1</sup>, with only half of that peatland area being permafrost. This suggests a smaller source than the estimates of *Voigt et al.* [2020]. NMIP estimates of soil N<sub>2</sub>O emissions from the permafrost regions are close to the lower-limit of estimates by *Voigt et al.* [2020], and have smaller uncertainty range (0.11-0.26 Tg N<sup>-1</sup>, mean 0.17 Tg N<sup>-1</sup>), which partly reflect the usage of unified model input data. Soil N<sub>2</sub>O emissions from non-permafrost regions are largely controlled by fertilizer and manure applications. According to NMIP models, the average emission factors of fertilizer and manure in non-permafrost regions during 1980-2016 were 1.4% and 1.7%, respectively. Both factors were positively correlated with temperature and precipitation, suggesting positive interactions between nitrogen additions and climate change [*Tian et al.*, 2020].

#### 4 Discussion

Our study provides a first estimate of soil N<sub>2</sub>O emissions from the NHL, although large uncertainties remain in both TD and BU approaches (Fig. S17, S18). Since the process-based models used in this study were driven by the same input data, differences were mainly induced by missing or uncertain representation of important processes such as seasonal freeze-thaw cycles and permafrost thaw [*Risk et al.*, 2013], BNF [*Meyerholt et al.*, 2020] and reactive N flows through ecosystems [*Butterbach-Bahl et al.*, 2013], and critical information such as timing and frequency of fertilizer application [*Nishina et al.*, 2017]. Several NMIP models do not include an explicit permafrost layer or freeze-thaw processes; inclusion of such factors would enable better representation of “hot spots” and “hot moments” soil N<sub>2</sub>O emissions in the NHL [*Voigt et al.*, 2020;

Wagner-Riddle *et al.*, 2017]. Current process-based TBMs also have insufficient representation of the upland thermokarst formation [Yang *et al.*, 2018] and fine-grained landscape structure of arctic ecosystems (e.g., landscape elements that are ultra-emitters of N<sub>2</sub>O such as non-vegetated organic soil). Integrating sub-grid scale information and processes into models may provide a solution for fine-grained physical-hydrological modelling. As revealed by Voigt *et al.* [2020], peatlands have the highest N<sub>2</sub>O emission rate in permafrost regions. It is thus important for process-based TBMs to explicitly consider peatland thermal, hydrological, and biogeochemical processes.

TD estimates have a stronger dependence on the prior fluxes in NHL where atmospheric N<sub>2</sub>O measurements are sparse [Nevison *et al.*, 2018; Rona Louise Thompson *et al.*, 2014; Rona L. Thompson *et al.*, 2019]. In this study, the average prior N<sub>2</sub>O flux employed in the TD models (846±141 Gg N yr<sup>-1</sup>) was lower than our BU estimates (955±267 Gg N yr<sup>-1</sup>). These low prior N<sub>2</sub>O fluxes, as well as lower TD emissions in summer compared to the BU estimates, are the likely causes of the lower TD estimates (Fig. S18). Differing prior N<sub>2</sub>O fluxes between the inversions (see methods) also lead to somewhat varying inversion estimates. Using the ensemble mean NMIP soil emission estimates as prior for the TD inversions may improve model agreement. The total prior ocean flux also has important impacts on the magnitude of the terrestrial flux. However, there have been few observational constraints on the ocean source until recently [Patra *et al.*, 2022]. The sparseness of atmospheric observations over both land and ocean north of 50°N and systematic model errors in stratosphere-troposphere exchange increase the uncertainty in TD estimates. Building denser regional N<sub>2</sub>O monitoring networks and launching (regular) aircraft campaigns in the NHL will help better constrain inversion models [Bisht *et al.*, 2021].

Our results suggest that the NHL contributed approximately 8% of the increase in global soil N<sub>2</sub>O emissions during 1861-2016 [Tian *et al.*, 2019]. Warming and wetting stimulated NHL soil N<sub>2</sub>O emissions, while elevated CO<sub>2</sub> concentrations suppressed emissions (through increased plant growth and larger uptake of soil N), findings that are in line with field observations [Cui *et al.*, 2018; Dijkstra *et al.*, 2012; Gong and Wu, 2021; Marushchak *et al.*, 2011; Voigt *et al.*, 2017a]. From 1980-2016 when warming was strongest, the NHL contributed 14% of global climate effect enhancing soil N<sub>2</sub>O emissions. Under the SSP370 and SSP585 scenarios, CMIP6 climate models predict that the mean temperature of the NHL will increase by 6.2 (4.1-9.8) °C and 7.8 (5.5-12.1) °C, respectively, during 2015-2100; the mean precipitation will increase by 96 (65-177) mm yr<sup>-1</sup> and 129 (51-206) mm yr<sup>-1</sup>, respectively (Fig. S19). If the sensitivities of soil N<sub>2</sub>O emissions to temperature and precipitation in the future are consistent with historical values, future climate change alone will substantially increase NHL soil N<sub>2</sub>O emissions. However, atmospheric CO<sub>2</sub> concentrations also rapidly increase under SSP370 and SSP585 scenarios (Fig. S20), potentially offsetting a significant fraction of the positive climate effect if arctic vegetation continues to take up more carbon and nitrogen with elevated CO<sub>2</sub>. Uncertainties arise regarding the degree of recycling of that extra nitrogen uptake in soils by mineralization. The magnitude of the future CO<sub>2</sub> effect is also highly uncertain [Walker *et al.*, 2021], and how it will affect future northern N<sub>2</sub>O emissions requires further study. Reconstructions from ice cores show that global N<sub>2</sub>O emissions increased over the last deglaciation when the climate warmed, CO<sub>2</sub> increased, and land carbon inventories grew in size, providing evidence for a net positive relationship between past warming, CO<sub>2</sub>, land carbon, stocks, and N<sub>2</sub>O emissions at the global scale [Fischer *et al.*, 2019; Joos *et al.*, 2020].

Since the NMIP project did not design simulation experiments to separate the effects of temperature and precipitation on soil N<sub>2</sub>O emissions, we used statistical methods to explore these relationships. However, the collinearity between temperature and precipitation variations may undermine the reliability of the inferred sensitivities of soil N<sub>2</sub>O emissions to temperature and precipitation. Future model intercomparison projects need to design simulations to disentangle the effects of temperature and precipitation.

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## **Data availability statement**

EDGAR 6.0 dataset is available at [https://edgar.jrc.ec.europa.eu/dataset\\_ghg60](https://edgar.jrc.ec.europa.eu/dataset_ghg60). GFED4.1s dataset is available at <https://www.geo.vu.nl/~gwerf/GFED/GFED4/>. Soil N<sub>2</sub>O emissions, terrestrial GPP and plant nitrogen uptake estimated by NMIP models and top-down N<sub>2</sub>O emission are available at <https://datadryad.org/stash/share/isclqpURaZ5GJLLok3LCvjBrQ20ybXX7M3dQzuVWFCK>

## **Author contributions**

H.T. initiated and designed this research, N.P. conducted data analysis and synthesis, N.P. and H.T. drafted the manuscript. All co-authors contributed to the writing and development of the manuscript.

## Competing interests

The authors declare no competing interests.

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