



Full length article



## Climate change rivals fertilizer use in driving soil nitrous oxide emissions in the northern high latitudes: Insights from terrestrial biosphere models

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## ARTICLE INFO

Handling Editor: Xavier Querol

## Keywords:

Nitrous oxide  
Northern high latitudes  
Climate change  
Anthropogenic activities  
Model ensemble estimation  
Permafrost

## 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 exposure to rapid rates of regional warming and changing hydrology due to climate change. In this study, we used an ensemble of six process-based terrestrial biosphere models (TBMs) from the Global Nitrogen/Nitrous Oxide Model Intercomparison Project (NMIP) to quantify soil N<sub>2</sub>O emissions across the NHL during 1861–2016. Factorial simulations were conducted to disentangle the contributions of key driving factors, including climate change, nitrogen inputs, land use change, and rising atmospheric CO<sub>2</sub> concentration, to the trends in emissions. The NMIP models suggest NHL soil N<sub>2</sub>O emissions doubled from 1861 to 2016, increasing on average by  $2.0 \pm 1.0$  Gg N/yr ( $p < 0.01$ ). Over the entire study period, while N fertilizer application ( $42 \pm 20$  %) contributed the largest share to the increase in NHL soil emissions, climate change effect was comparable ( $37 \pm 25$  %), underscoring its significant role. In the recent decade (2007–2016), anthropogenic sources contributed  $47 \pm 17$  % ( $279 \pm 156$  Gg N/yr) of the total N<sub>2</sub>O emissions from the NHL, while unmanaged soils contributed a comparable amount ( $290 \pm 142$  Gg N/yr). The trend of increasing emissions from nitrogen fertilizer reversed after the 1980 s because of reduced applications in non-

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<https://doi.org/10.1016/j.envint.2025.109297>

Received 24 September 2024; Received in revised form 20 December 2024; Accepted 19 January 2025

Available online 21 January 2025

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permafrost regions. In addition, increased plant growth due to CO<sub>2</sub> fertilization suppressed simulated 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. The rigorous interplay between process modeling and field experimentation will be essential for improving model representations of the mechanisms controlling N<sub>2</sub>O fluxes in the Northern High Latitudes and for reducing associated uncertainties.

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

The terrestrial nitrogen cycle in the NHL is closely related with permafrost, which underlays more than 60 % of the area (Brown et al., 1997). Although large N stocks are stored in this region (Harden et al., 2012; Hugelius et al., 2020), the associated soil N<sub>2</sub>O emissions received little attention because they were considered to be small due to limited microbial activities and low mineralization rates under low-temperature and the presence of permafrost. Permafrost constrains soil processes by limiting the depth of the active layer, influencing soil moisture dynamics, and reducing nitrogen availability. (Voigt et al., 2020). However, recent in-situ studies found that both barren and vegetated soils in the NHL can emit substantial amounts of N<sub>2</sub>O (Marushchak et al., 2011; Marushchak et al., 2021; Repo et al., 2009; Voigt et al., 2017b). For example, Marushchak et al. (2011) found that N<sub>2</sub>O emissions from an unvegetated peatland in Russia can be  $1.40 \pm 0.15$  g N<sub>2</sub>O m<sup>-2</sup> yr<sup>-1</sup>, which is higher than the average N<sub>2</sub>O emission rate (0.19 g N<sub>2</sub>O m<sup>-2</sup> yr<sup>-1</sup>) from tropical rainforest soils (Werner et al., 2011). The N<sub>2</sub>O emission during the snow-free season was  $1.2 \pm 0.3$  g N<sub>2</sub>O m<sup>-2</sup> in a pristine tundra area (Repo et al., 2009), which is as high as the rate of boreal organic croplands (0.3–1.7 g N<sub>2</sub>O m<sup>-2</sup> yr<sup>-1</sup>; Majanen et al., 2007). Meanwhile, Arctic amplification, the phenomenon that climate change is amplified in the NHL, is projected to continue in the 21st century (Christensen et al., 2013; Pithan and Mauritsen, 2014) with further implications for N<sub>2</sub>O emissions: first, a large amount of immobile N stored in permafrost becomes available for decomposition and remobilization after permafrost thawing; second, rapid warming enhances N mineralization and promotes nitrification and denitrification; and third, warming may also promote biological nitrogen fixation (BNF), increasing ecosystem N availability and thereby potentially also N<sub>2</sub>O production. Field experiments show that experimental warming can increase soil N<sub>2</sub>O emissions from the northern high latitudes by 10–460 % (Cui et al., 2018; 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.

In recent years, process-based models have been employed to investigate soil N<sub>2</sub>O emissions from northern regions. For example, Xu-Ri et al. (2012) used the DyN-LPJ model and found that N<sub>2</sub>O emissions from boreal forests and tundra are generally low (typically less than 0.2 kg N ha<sup>-1</sup> yr<sup>-1</sup>) due to temperature limitations. Lacroix et al. (2022) applied the QUINCY terrestrial biosphere model to simulate N<sub>2</sub>O emissions in permafrost regions and demonstrated that emissions could increase disproportionately with deeper soil thawing. Additionally, Del Grosso et al. (2022) highlighted that N<sub>2</sub>O emissions in cold climates are often underestimated due to the omission of freeze–thaw-induced pulses in national greenhouse gas inventories. They suggested that incorporating these dynamics into process-based models could significantly improve emission estimates. However, despite these advances, no study has comprehensively evaluated the magnitude, trends, and drivers of soil N<sub>2</sub>O emissions from the NHL using process-based models. To address these gaps, this study aims to provide a comprehensive quantification of soil N<sub>2</sub>O emissions across the NHL using an ensemble of process-based terrestrial biosphere models (TBMs) and atmospheric inversion frameworks. The main objectives of our study are to: (1) investigate the spatiotemporal variations of soil N<sub>2</sub>O emissions from 1861 to 2016 in the NHL, (2) disentangle the contributions of climate change, nitrogen inputs, land use change, and atmospheric CO<sub>2</sub> concentration to N<sub>2</sub>O emissions, and (3) assess the uncertainties associated with both bottom-up and top-down approaches in estimating N<sub>2</sub>O fluxes in this region. Specifically, we investigated NHL soil N<sub>2</sub>O emissions using six TBMs from 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). NMIP models are calibrated against a range of available datasets, including observations in the northern high latitudes (Xu et al., 2017, 2020; Xu-Ri et al., 2012; Zaehle 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 paired simulations, i.e., S1-S2, S2-S3, S3-S4, S4-S5, S5-S6, and S6-S0, were used to evaluate the simulated 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 GFED4.1 s 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 GFEDv4.1 s, 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 NHL 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 soil N<sub>2</sub>O emissions and two climate factors: temperature and precipitation. Specifically, our conceptual model assumes that precipitation and temperature not only directly affect soil N<sub>2</sub>O emissions but also indirectly affect cumulative soil N<sub>2</sub>O emissions through interactions between temperature and precipitation (Fig. S7). When conducting the PAM, we used the ensemble mean of NMIP estimations from S6 (climate only experiment) and temperature and precipitation data from CRU-NCEP. The effects of the explanatory variables on response variable were quantified using standardized path coefficients which describe the strength and sign of the relationship between two variables. We also conducted partial correlation analysis between soil N<sub>2</sub>O emissions and temperature/precipitation to explore the linkage between N<sub>2</sub>O emissions and single climatic driving factor while controlling the effects of other remaining factors. 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. When calculating the ensemble means of terrestrial biosphere models and atmospheric inversion models, each model was weighed equally, we assume that each individual model simulation is of equal value since there is no prior knowledge of model performance in estimating N<sub>2</sub>O emissions from the northern high latitudes. In this study, we use mean value ± one standard deviation to represent the best estimate and the uncertainty range.

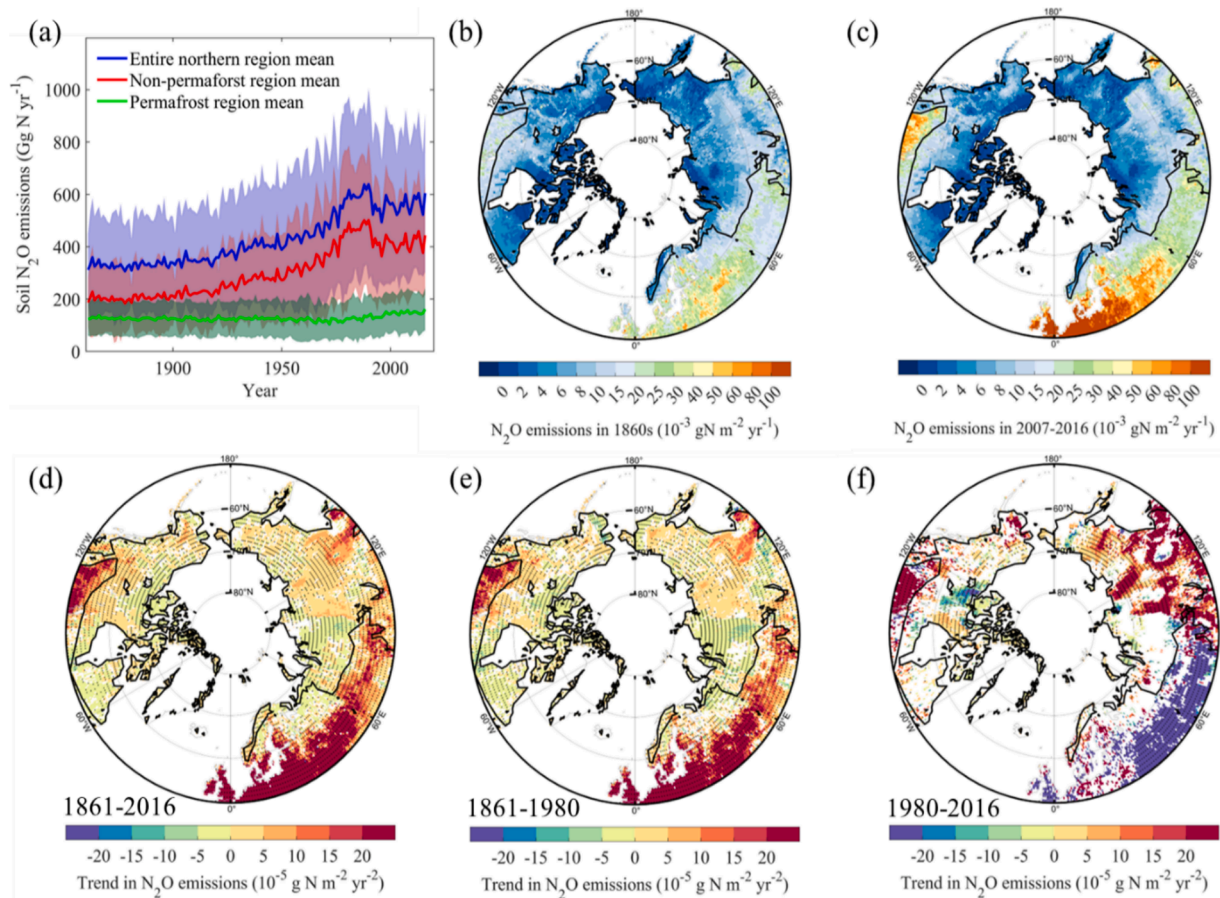
## 3. Results

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

Multi-model ensemble estimates show that soil N<sub>2</sub>O emissions from the NHL increased from 312 ± 125 Gg N/yr in 1861 to 605 ± 269 Gg N/yr in 2016 (Fig. 1a), with an average increase rate of 2.0 ± 1.0 Gg N/yr ( $p < 0.01$ ). 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 1920 s to the 1980 s, and peaked in the 1980 s. In the late 1980 s and early 1990 s, 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 1980 s, and rapidly increased thereafter. In the 1860 s, 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 flux (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  $2 \times 10^{-4} \text{ g N m}^{-2} \text{ yr}^{-1}$  (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).

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

Our results derived from model 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 1930 s and N inputs made increasing contributions from the 1940 s to the 1980 s. From the 1860 s to the 1980 s, fertilizer application contributed 53 ± 22 % to the increase in emissions, followed by atmospheric N deposition (26 ± 12 %), manure N application (15 ± 9 %), climate change (12 ± 11 %), and land use change (5 ± 7 %). The effect of increased atmospheric CO<sub>2</sub> (−10 ± 10 %) almost offset that of climate change. Since the 1980 s, 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 ± 25 %) to the increase of NHL soil emissions after N fertilizer application (42 ± 20 %). 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 ± 58 % (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 ± 17 %) (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 1980 s (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



**Fig. 1.** (a) Changes in soil  $N_2O$  emissions from the NHL as estimated by NMIP models, the shaded area indicates one standard deviation of all estimates. (b) and (c) show spatial pattern of mean annual soil  $N_2O$  emissions during the 1860s and 2007–2016, respectively. Trends in soil  $N_2O$  emissions during 1861–2016 (d), 1861–1980 (e), and 1980–2016 (f); grids with non-significant trends ( $p > 0.05$ ) were excluded, and stippling indicates where a majority of models (at least 4 out of 6) agree on the sign of the trend.

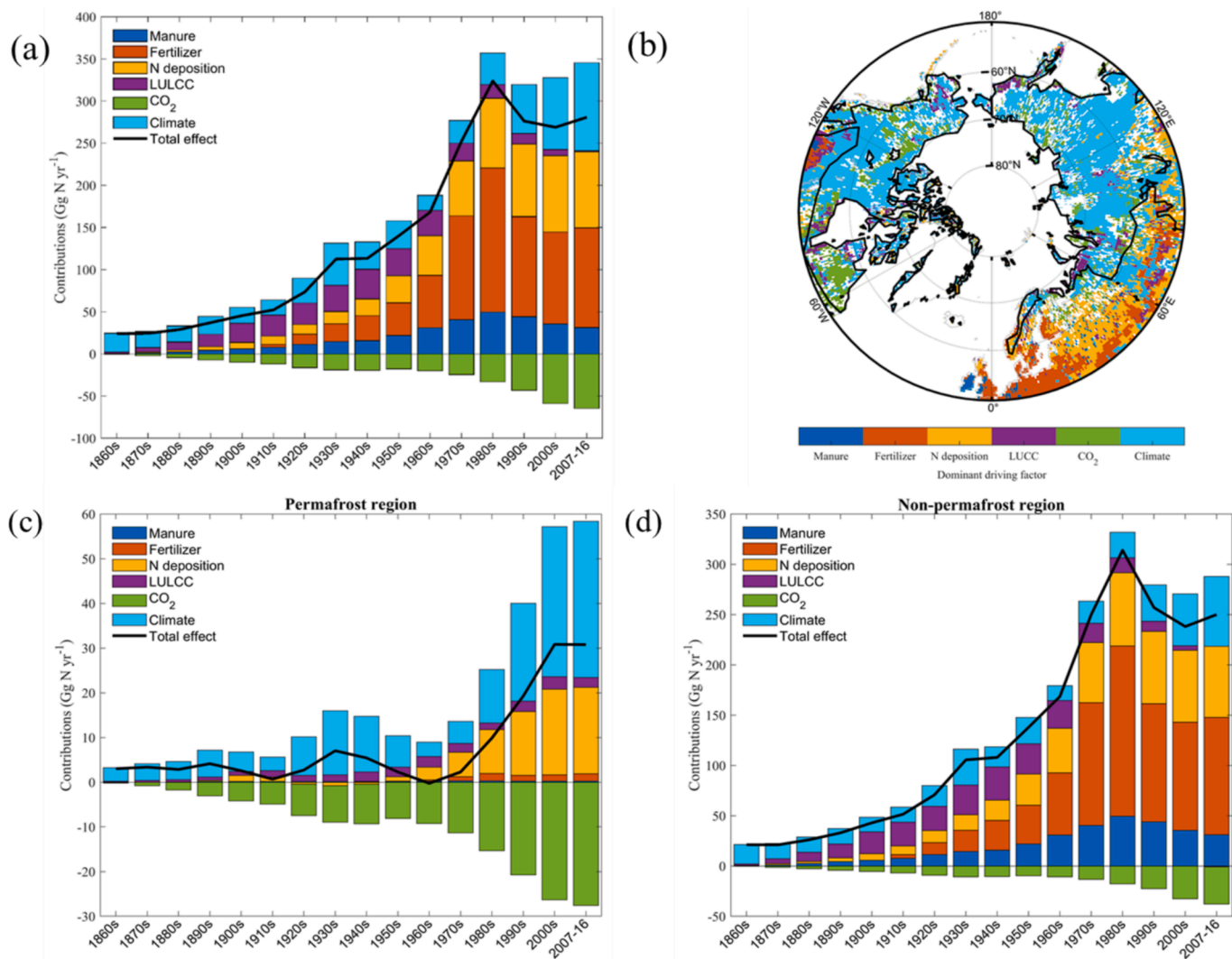
dominated by other factors were relatively small (Fig. 2b).

Temperature and precipitation changes alter soil microclimate, nutrient availability and microbial ecology, thereby influencing  $N_2O$  emissions (Dalal and Allen, 2008). For the entire NHL, both temperature and precipitation significantly increased during 1901–2016, with rates of  $0.14\text{ }^\circ\text{C}$  per decade and  $0.38\text{ mm yr}^{-1}$ , respectively (Fig. S3). According to multiple regression model results, the sensitivities of soil  $N_2O$  emissions to temperature and precipitation were  $29 \pm 21\text{ Gg N }^\circ\text{C}^{-1}$  and  $0.4 \pm 0.7\text{ Gg N mm}^{-1}$  during 1901–2016, suggesting that warming and wetting increased soil  $N_2O$  emissions by  $48 \pm 35\text{ Gg N/yr}$  and  $15 \pm 26\text{ Gg N/yr}$ , respectively. The path analysis model also suggested that warming contributed more to soil  $N_2O$  emission increases than wetting (Fig. S7). Both warming and wetting have accelerated since 1980 (Fig. S3, S8, S9), with average rates of  $0.38\text{ }^\circ\text{C}$  per decade and  $0.57\text{ mm yr}^{-2}$ , respectively. At the same time, the sensitivities of soil  $N_2O$  emissions to temperature and precipitation increased. These two factors together led to the large climate effects in the recent four decades. Soil  $N_2O$  emissions were positively correlated with temperature in most regions (Fig. S10a), because warming 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  $N_2O$  emissions in this region. Unlike temperature, the correlation between soil  $N_2O$  emissions and precipitation varied spatially (Fig. S10b), which explained why precipitation had a smaller effect than temperature on the regional total emissions even though most regions experienced significant wetting (Fig. S10d).

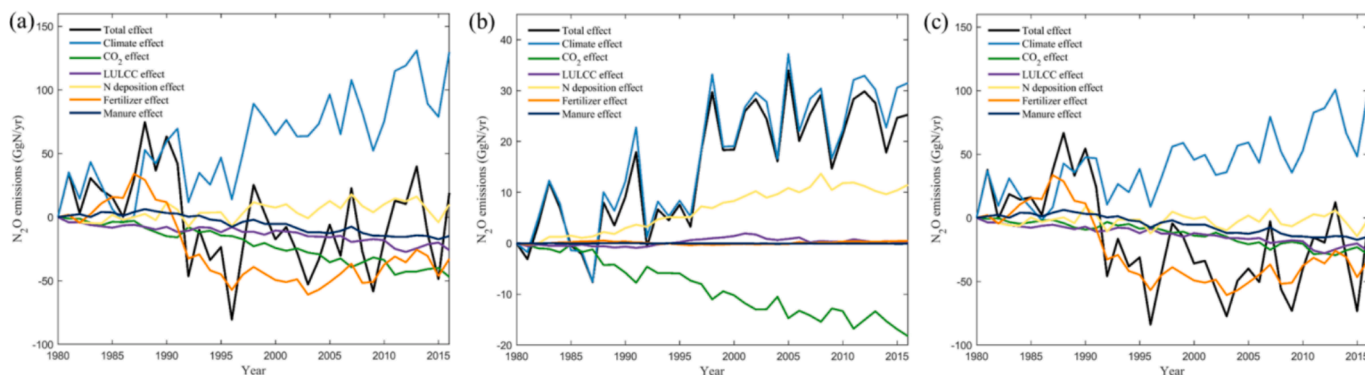
### 3.3. Declining soil $N_2O$ emissions since the 1980s

According to NMIP models, soil  $N_2O$  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  $N_2O$  emissions from the NHL decreased at an average rate of  $-1.1\text{ GgN yr}^{-1}$  ( $p < 0.05$ ) during 1980–2016 (Fig. 3a). The rapid decline in emissions during 1988–1996 was due to reduced fertilizer application (Fig. S13), after which period the negative effect of  $CO_2$  fertilization was enhanced (Fig. S14). Decreases in fertilizer use and the enhanced  $CO_2$  effect roughly cancelled increased in emissions due to climate change, resulting in a non-significant trend in emissions from non-permafrost regions after the 1990s. 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  $N_2O$  emissions fluctuated after 1998 because the positive climate effect was counteracted by combined effects of fertilizer application,  $CO_2$  and land use change.

The dominant drivers of negative effects differed between permafrost and non-permafrost regions. In permafrost regions, elevated  $CO_2$  concentration was the only factor suppressing soil  $N_2O$  emissions and counteracted more than half of the climate-induced emissions (Fig. 3b). By contrast, reduced N fertilizer application, elevated  $CO_2$  concentration and land use change jointly reduced emissions from non-permafrost regions (Fig. 3c). For the entire NHL, the atmospheric  $CO_2$ -induced decline in soil  $N_2O$  emissions surpassed the effect of reduced fertilizer



**Fig. 2.** (a) Decadal variations in the contributions of different driving factors. (b) Distribution of dominant driving factors of soil  $N_2O$  emissions during 1861–2016; grids with non-significant trends were excluded. Contributions of different driving factors to soil  $N_2O$  emissions from permafrost regions (c) and non-permafrost regions (d).



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

application over the recent decade. Elevated atmospheric  $CO_2$  significantly suppressed  $N_2O$  emissions in most northern regions (Fig. S14). Since the 1980 s, increased atmospheric  $CO_2$  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  $N_2O$  emissions. The largest

stimulation effect of  $CO_2$  on vegetation growth and nitrogen uptake occurred in the boreal forests, where the  $CO_2$ -induced suppression of  $N_2O$  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  $N_2O$  emissions due to enhanced plant growth and nitrogen uptake is also consistent

with field observations in the NHL (Gong and Wu, 2021; Groffman et al., 2011; Marushchak et al., 2011; Stewart et al., 2012).

### 3.4. 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 (192 Gg N yr<sup>-1</sup>) and biomass burning (91 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 (855 ± 267 Gg N yr<sup>-1</sup>) was generally higher than the TD estimate (668 ± 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 (include emissions from soils, biomass and other anthropogenic sources) had no significant trend during 1998–2014 ( $p > 0.05$ ). Subtracting BU estimates of 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 -8.6 to -2.4 Gg N yr<sup>-2</sup>, mean -5.7 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.

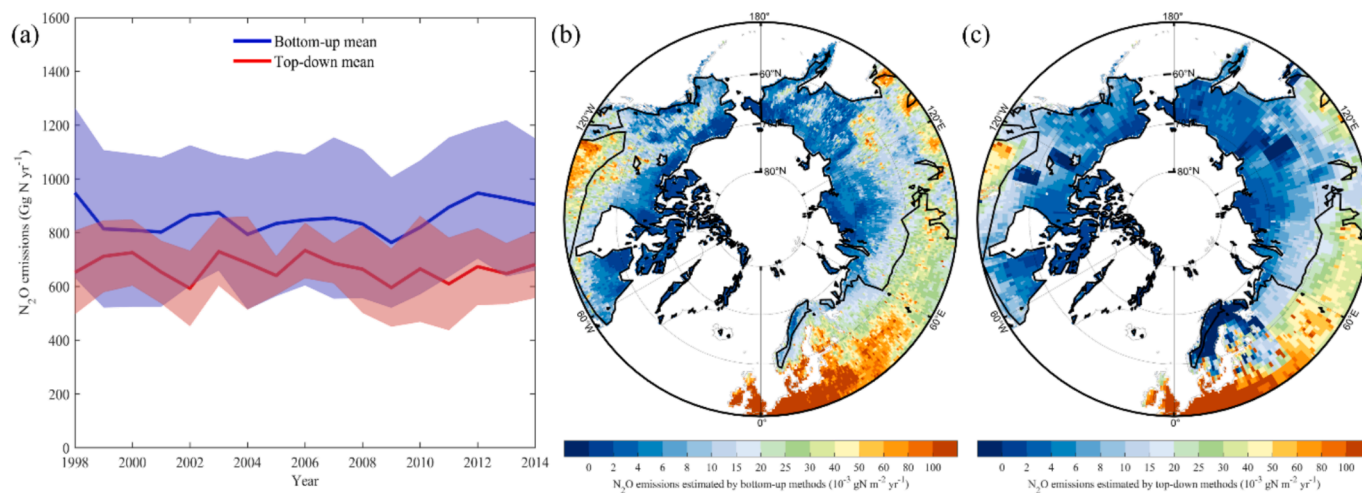
### 3.5. Comparison with empirical estimates

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 yr<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 ± 5 Gg N yr<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 yr<sup>-1</sup>), which partly reflect the usage of unified model input data.

## 4. Discussion

Our study provides a first estimate of simulated soil N<sub>2</sub>O emissions from the NHL, although large uncertainties remain in both TD and BU approaches (Fig. S17, S18). Our results are based on state-of-the-art terrestrial biosphere models; however, these estimates are subject to uncertainties arising from model parameterization, limited field observations, and 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 freeze–thaw processes which contribute to a large proportion of annual N<sub>2</sub>O emissions in northern croplands (Wagner-Riddle et al., 2017), the absence of freeze–thaw processes likely renders these models incapable of capturing the pulses of N<sub>2</sub>O emissions associated with early spring thaw (Del Grosso et al., 2022), leading to an underestimation of total soil N<sub>2</sub>O emissions. The inclusion of freezing and thawing would enable better representation of “hot moments” of soil N<sub>2</sub>O emissions in northern agricultural ecosystems (Voigt et al., 2020; Wagner-Riddle et al., 2017). A recent study suggests that high-Arctic N<sub>2</sub>O emissions are threefold higher when considering permafrost nitrogen release from permafrost thaw (Lacroix et al., 2022). NMIP models with permafrost/freezing–thaw cycles show higher sensitivity to climate change. During the study period, these models attribute an increase of 112 ± 58 Gg N/yr in soil N<sub>2</sub>O emissions to climate change, accounting for 71 ± 11 % of the total increase (Fig. S4). In contrast, models without permafrost layer and freeze–thaw cycles estimate a smaller increase of 103 ± 51 Gg N/yr (25 ± 8 % of total increase). These findings suggest that missing permafrost layers and freeze–thaw dynamics may lead to an underestimation of the reaction of soil N<sub>2</sub>O emissions to climate change, underscoring the importance of including the permafrost layer and freeze–thaw cycles in terrestrial biosphere models. 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



**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.

important for process-based TBMs to explicitly consider peatland thermal, hydrological, and biogeochemical processes.

Soil N<sub>2</sub>O emissions from non-permafrost regions are largely controlled by fertilizer and manure applications. According to NMIP models, the emission factors of fertilizer and manure in non-permafrost regions during 1980–2016 were  $1.4 \pm 0.7\%$  and  $1.7 \pm 0.5\%$ , respectively. Both factors were positively correlated with temperature and precipitation, suggesting positive interactions between nitrogen additions and climate change (Tian et al., 2020). NMIP models show varying effects of land use and land cover change on soil N<sub>2</sub>O emissions, ranging from  $-57$  to  $45$  Gg N/yr during 2007–2016 (Fig. S4), while the ensemble mean remained close to zero ( $1$  Gg N/yr, Fig. 2a). These uncertainties arise from differences in how each model represents land use distribution and land conversion processes (see Section S1 of the supplementary material). Although all NMIP models used the same cropland area and nitrogen fertilizer input data, variations in crop cultivation, management practices, the classification, distribution, and parameterization of natural vegetation, nitrogen pool allocation after vegetation conversion contributed to the model divergence. Despite these uncertainties in the land use effect ( $-57$  to  $45$  Gg N/yr during 2007–2016), our conclusion that nitrogen fertilizer application ( $120 \pm 60$  Gg N/yr during 2007–2016) and climate change ( $108 \pm 55$  Gg N/yr during 2007–2016) were the dominant and counteracting drivers of NHL soil N<sub>2</sub>O emissions remains robust. Although the NMIP models can simulate transitions between different biomes, such as from boreal forests to peatlands and wetlands, they do not account for transitions from terrestrial ecosystems to aquatic ecosystems, including streams, rivers and lakes. This restricts the models' ability to capture changes in N<sub>2</sub>O emissions due to the potential expanding of wet environments under climate change. As permafrost thaw and changing hydrology create wetter conditions, N<sub>2</sub>O emission hotspots may shift to wet environments. Using estimates from DLEM-TAC (Yao et al., 2020; Tian et al., 2023), which accounts for both small streams and large rivers, we calculated that N<sub>2</sub>O emissions from rivers and streams in the NHL were  $29 \pm 2$  Gg N yr<sup>-1</sup> during 2007–2016, equivalent to about 5 % of soil N<sub>2</sub>O emissions. These findings suggest that aquatic systems, while currently a smaller contributor, may play a larger role in the regional N<sub>2</sub>O budget as warming continues. Therefore, integrating terrestrial and aquatic N<sub>2</sub>O dynamics into future model frameworks will be crucial for accurately assessing the evolving N<sub>2</sub>O budget in the NHL.

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; Thompson et al., 2014; Rona L. Thompson et al., 2019). 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 atmospheric N<sub>2</sub>O concentration monitoring networks and launching (regular) aircraft campaigns in the NHL will help improve model reliability by providing additional constraints for atmospheric inversion models, reducing biases, improving parameterization, and refining uncertainty estimates (Thompson et al., 2019; Bisht et al., 2021; Stell et al., 2022).

NMIP model 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). By 2016, NHL soil N<sub>2</sub>O emissions had increased by  $293 \pm 158$  Gg N/yr compared to 1861, a value 63 % higher than the combined N<sub>2</sub>O emissions from energy, transportation, and industry in 2016, as estimated by EDGAR V6.0. 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 to 2016 when warming was strongest, the NHL contributed 14 % of global climate effect enhancing soil N<sub>2</sub>O emissions. Our analysis indicates that the ten warmest years in the NHL during 1901–2016 all occurred after 1990, with nine of these years occurring after 2003. This trend reflects the increasing frequency of extreme warm years in recent decades. During the ten warmest years, climate change increased N<sub>2</sub>O emissions by an average of  $98$  Gg N yr<sup>-1</sup>, with the highest increase of  $139$  Gg N yr<sup>-1</sup> in 2016. These findings suggest that the rising frequency of extreme warming events, particularly under high-emission scenarios, will likely stimulate substantial increases in 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, additional processes such as abrupt permafrost collapse (thermokarst) and wildfires may further amplify N<sub>2</sub>O emissions. Permafrost collapse exposes previously frozen organic matter, increasing nitrogen availability and enhancing nitrification and denitrification, which results in higher N<sub>2</sub>O emissions (Abbott et al., 2015; Takakai et al., 2008). Studies suggest that thermokarst landscape can become significant sources of N<sub>2</sub>O (Hashemi et al., 2024; Schulze et al., 2023). Wildfires can increase inorganic nitrogen availability, thus stimulate N<sub>2</sub>O emissions (Guo et al., 2024; Köster et al., 2017). Moreover, post-fire conditions can also accelerate permafrost thaw, which alters soil thermal and moisture regimes, further enhancing N<sub>2</sub>O emissions. Given the increasing frequency and intensity of thermokarst and wildfires under future scenarios, their combined effects may significantly contribute to NHL 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). Future N<sub>2</sub>O emission projections in the NHL are influenced by the interplay of multiple factors, including rising temperatures, increasing frequency of extreme climate events, permafrost thaw, land-cover changes, and wildfires. To reduce uncertainties and improve confidence in future emission projections, it is essential to integrate more extensive field observations, refine model parameterizations, and enhance the representation of critical processes including abrupt permafrost thaw, wildfires, and transitions from dry soils to wet environments. 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. The statistical methods indicate that warming in the recent four decades largely stimulated soil NO<sub>2</sub> emissions. 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). 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. While NMIP models provided valuable insights into nitrogen

cycle and N<sub>2</sub>O emissions in terrestrial ecosystems, they did not provide outputs related to permafrost shrinkage and thaw, because NMIP project primarily focused on terrestrial nitrogen fluxes and did not request permafrost-related variables as model outputs. Since permafrost thaw can significantly alter soil conditions, moisture regimes, and microbial activities- all of which influence N<sub>2</sub>O emissions, we propose that future NMIP simulations include outputs for permafrost extent, thaw depth, and associated land-cover transitions. Incorporating these variables would enable a more comprehensive understanding of how permafrost dynamics affect N<sub>2</sub>O emissions.

## 5. Conclusion

This study provides a comprehensive assessment of soil N<sub>2</sub>O emissions across the NHL from 1861 to 2016, emphasizing the important role of these regions in global N<sub>2</sub>O dynamics. Using an ensemble of process-based terrestrial biosphere models and atmospheric inversion frameworks, we quantified historical trends and identified the key drivers of N<sub>2</sub>O emissions. The results show that soil N<sub>2</sub>O emissions from the NHL have doubled since 1861, and climate change played a role nearly equivalent to fertilizer use in driving the increase in NHL soil N<sub>2</sub>O emissions, highlighting the profound influence of warming and hydrological changes on regional nitrogen cycling, particularly in the context of Arctic amplification. Our study identified regional differences in emission trends: non-permafrost regions experienced a peak in the 1980s, followed by a decline linked to reduced nitrogen inputs and enhanced plant nitrogen uptake due to CO<sub>2</sub> fertilization; in contrast, emissions from permafrost regions have continued to rise due to rapid warming. These divergent trends illustrate the growing importance of permafrost regions in the global N<sub>2</sub>O budget. More field experiments and long-term observational networks are crucial for elucidating the key mechanisms and processes controlling soil N<sub>2</sub>O emissions in the NHL and will lay a robust foundation for improving models and reducing uncertainties.

## CRedit authorship contribution statement

**Naiqing Pan:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis, Data curation, Conceptualization. **Hanqin Tian:** Writing – review & editing, Funding acquisition, Conceptualization. **Hao Shi:** Writing – review & editing. **Shufen Pan:** Writing – review & editing. **Josep G. Canadell:** Writing – review & editing. **Jinfeng Chang:** Writing – review & editing. **Philippe Ciais:** Writing – review & editing. **Eric A. Davidson:** Writing – review & editing. **Gustaf Hugelius:** Writing – review & editing. **Akihiko Ito:** Writing – review & editing. **Robert B. Jackson:** Writing – review & editing. **Fortunat Joos:** Writing – review & editing. **Sebastian Lienert:** Writing – review & editing. **Dylan B. Millet:** Writing – review & editing. **Stefan Olin:** Writing – review & editing. **Prabir K. Patra:** Writing – review & editing. **Rona L. Thompson:** Writing – review & editing. **Nicolas Vuichard:** Writing – review & editing. **Kelley C. Wells:** Writing – review & editing. **Chris Wilson:** Writing – review & editing. **Yongfa You:** Writing – review & editing. **Sönke Zaehle:** Writing – review & editing.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgements

This study was resulted from NMIP (global N/N<sub>2</sub>O Model Inter-comparison Project) co-sponsored by Global Carbon Project and International Nitrogen Initiative. This work contributes to the REgional Carbon Cycle Assessment and Processes-2 of the Global Carbon Project. H.T., N. Pan, S. Pan acknowledge funding support from NSF (grant nos.

1903722, 1922687); PKP is partly funded by the Arctic Challenge for Sustainability phase II (ArCS-II; JPMXD1420318865) Projects of the Ministry of Education, Culture, Sports, Science and Technology (MEXT); KCW and DBM acknowledge support from NASA (Grant #NNX17AK18G) and NOAA (Grant #NA13OAR4310086).

## 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). GFED4.1 s 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.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2025.109297>.

## Data availability

Data will be made available on request.

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