



1 **Nitrous oxide emissions from pan-Arctic terrestrial ecosystems: A**  
2 **process-based biogeochemistry model analysis from 1969 to 2019**  
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11

12 **Abstract:** Nitrous oxide (N<sub>2</sub>O) is a potent greenhouse gas with radiative forcing 265-298 times  
13 stronger than that of carbon dioxide (CO<sub>2</sub>). Increasing atmospheric N<sub>2</sub>O burden also contributes  
14 to stratospheric ozone depletion. Recent field studies show N<sub>2</sub>O emissions from the Arctic  
15 ecosystems have increased due to warming. To date, the emissions across space and time have  
16 not been adequately quantified. Here we revised an extant process-based biogeochemistry  
17 model, the Terrestrial Ecosystem Model (TEM) to incorporate more detailed processes of soil  
18 biogeochemical nitrogen (N) cycle, permafrost thawing effects, and atmospheric N<sub>2</sub>O uptake in  
19 soils. The model is then used to analyze N<sub>2</sub>O emissions from pan-Arctic terrestrial ecosystems.  
20 We find that both regional N<sub>2</sub>O production and net emissions increased from 1969 to 2019, with  
21 production ranging from 1.2 - 1.3 Tg N yr<sup>-1</sup> and net emissions from 1.1 - 1.2 Tg N yr<sup>-1</sup>  
22 considering the permafrost thaw effects. Soil N<sub>2</sub>O uptake from the atmosphere was 0.1 Tg N yr<sup>-1</sup>  
23 with a small interannual variability. Atmospheric N deposition significantly increased N<sub>2</sub>O  
24 emission by 31.5 ± 3.1%. Spatially, terrestrial ecosystems act as net sources or sinks ranging



25 from -12 to 700 mg N m<sup>-2</sup> yr<sup>-1</sup> depending on temperature, precipitation, soil characteristics, and  
26 vegetation types in the region.

## 27 1. Introduction

28 Global climate change entails rising temperatures, a change in precipitation patterns, and  
29 a more frequent occurrence of extreme weather events. The common consensus is that  
30 temperature increase will be most pronounced in the northern high latitudes (Overland et al.,  
31 2013). Pan-Arctic land areas are predicted to warm up to 5.6–12.4 °C by the end of this century  
32 (IPCC, 2013), leading to widespread permafrost degradation and thaw (Borge et al., 2017; Jones  
33 et al., 2016; Romanovsky et al., 2010) and substantial changes in ecosystem functioning (Grosse  
34 et al., 2016), accelerating decomposition of permafrost soil organic matter (SOM) and  
35 stimulating greenhouse gas (GHG) emissions (Yang et al., 2010; Sierra et al., 2015; Voigt et al.,  
36 2017). Permafrost soils are also large nitrogen (N) reservoirs (Post et al., 1985), with a  
37 conservative estimate of 67 billion tons of total N in the upper 3 m (Harden et al., 2012). The  
38 storage of organic nitrogen supplies plenty of substrates for nitrification and denitrification.

39 While past studies on greenhouse gas emissions from the Arctic have mainly focused on  
40 carbon-based compounds, recent studies show that Arctic soils might also be a significant source  
41 of N<sub>2</sub>O from peatlands (Repo et al., 2009; Marushchak et al., 2011), and high N<sub>2</sub>O production  
42 potential in soils after permafrost thaw (Elberling et al., 2010). Elevated soil N<sub>2</sub>O concentrations  
43 have been observed in upland tundra due to thawing permafrost (Abbott & Jones, 2015).  
44 Furthermore, rising temperatures also promote N<sub>2</sub>O emissions from tundra ecosystems (Vigot et  
45 al., 2017), which cover large areas of the Arctic. The great unknown, however, is how  
46 permafrost thaw affects N<sub>2</sub>O emissions by potentially unlocking the vast N stocks currently  
47 stored in Arctic soils (Harden et al., 2012). Although Arctic N<sub>2</sub>O fluxes are low on a mass basis,



48 they are important since N<sub>2</sub>O is a more potent GHG with radiative forcing of 265-298 times  
49 stronger than that of CO<sub>2</sub> (IPCC, 2013). N<sub>2</sub>O contributes nearly 10% of the total anthropogenic  
50 radiative forcing and is among the key chemicals destroying the stratospheric ozone layer  
51 (Ravishankara et al., 2009).

52 Soil temperature, moisture, acidity, soil organic matter (SOM) content, C/N ratio, and  
53 plant growth have been identified as the key regulators of Arctic N<sub>2</sub>O emissions (Marushchak et  
54 al., 2011; Voigt et al., 2017; Smith et al. 1998). Soil temperature involves in both organic and  
55 inorganic processes in the N cycle. Soil moisture determines the rates of gas destruction via  
56 oxidation. High water content leads to the low oxygen condition, which is associated with  
57 reducing environment, whereas less water leads to an oxidizing environment. Soil acidity  
58 controls the chemical reactions involving H<sup>+</sup> or OH<sup>-</sup>, both of which influence the activity of  
59 microorganisms and enzymes (Kunhikrishnan et al., 2016). Traditionally, N<sub>2</sub>O emissions  
60 originating from areas of high latitudes are viewed as an insignificant source (Martikainen et al.  
61 1993, Potter et al. 1996) due to a slow mineralization rate under low temperature, humid  
62 conditions, and low atmospheric deposition of nitrogen (Dentener et al. 2006). In pan-Arctic  
63 terrestrial ecosystems, the shortage of mineral N used to be one of the main reasons for the low  
64 nitrous oxide (N<sub>2</sub>O) emissions measured from tundra soils (Ludwig et al., 2006; Ma et al., 2007).  
65 However, in recent decades, the warming climate and loss of permafrost may disrupt the past  
66 closed Arctic N cycling and cause significant export of inorganic N (Frey et al. 2007;  
67 McClelland et al. 2007). Such patterns may result from the introduction of previously frozen N  
68 into the cycling pool due to gradual degradation of permafrost that deepens the zone of  
69 seasonally thawed soils, or from direct effects of temperature on microbial processes.



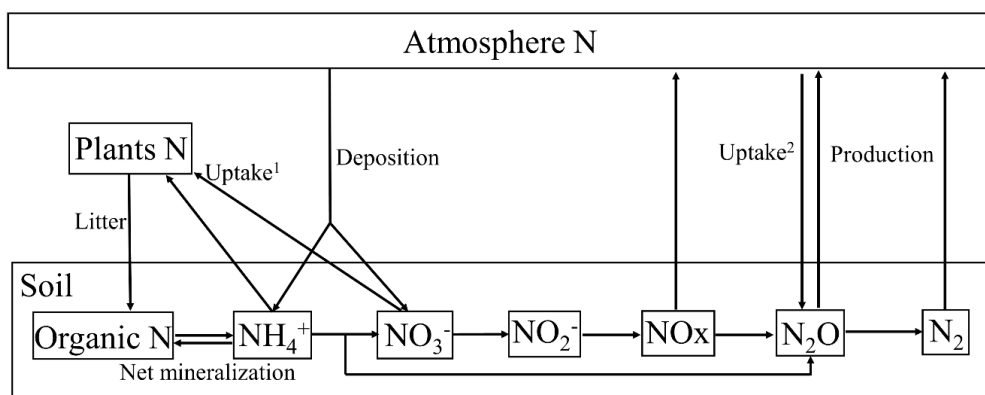
70 To date, a group of process-based biogeochemistry models have been used to quantify  
71 N<sub>2</sub>O fluxes, including a version of Terrestrial Ecosystem Model (TEM) (Qin et al. 2014,2015;  
72 Yu, 2016; Yu and Zhuang 2019), the Community Land Model, Carbon and Nitrogen cycles  
73 (CLM-CN) (Saikawa et al. 2013), the daily Century (DayCent) model (Parton et al. 1998, Del  
74 Grosso et al. 2005), and the Denitrification/ Decomposition (DNDC) Model (Li 1992). However,  
75 none of them has included the Arctic permafrost thawing effects on N<sub>2</sub>O emission and N<sub>2</sub>O  
76 uptake from atmosphere in modeling. Here we further develop the TEM based on the version of  
77 the model in Yu (2016) and Yu and Zhuang (2019) by incorporating the effects of permafrost  
78 thawing on nitrogen mineralization and the soil N<sub>2</sub>O uptake from the atmosphere. The model is  
79 then parameterized and verified and applied to quantify N<sub>2</sub>O emissions from pan-Arctic  
80 terrestrial ecosystems from 1969 to 2019.

81

## 82 2 Method

### 83 2.1 Model Description

84 TEM is a global-scale biogeochemical model designed to quantify the cycling of carbon  
85 (C) and nitrogen (N) in terrestrial ecosystems (Melillo et al., 1993; Zhuang et al., 2003; Yu,  
86 2016; Yu and Zhuang., 2019). The major processes of nitrogen (N) dynamic module were  
87 inherited from Yu and Zhuang (2019), including the effect of physical conditions on both  
88 nitrification and denitrification, as well as the principles of the stoichiometry of carbon and  
89 nitrogen dynamics in soils. Here we revised the N cycling algorithms in TEM by incorporating  
90 the loss of nitrogen through gas emissions, the uptake of N<sub>2</sub>O from the atmosphere, and  
91 additional inputs of organic nitrogen and carbon resulting from permafrost thawing (Fig. 1).



92

93 Figure 1. Nitrogen cycle and N<sub>2</sub>O production in the revised Terrestrial Ecosystem Model. Net  
 94 mineralization: the difference between mineralization (organic N mineralized to inorganic N)  
 95 and mobilization (inorganic N to organic N); Litter: organic N from plant litters; Uptake<sup>1</sup>:  
 96 inorganic N uptake by plants; Deposition: Atmospheric deposition of N; Production: N<sub>2</sub>O  
 97 production in soils; Uptake<sup>2</sup>: atmospheric N<sub>2</sub>O uptake in soils.  
 98

99 The net N<sub>2</sub>O emission is calculated as the difference between N<sub>2</sub>O production (N<sub>P</sub>) in  
 100 soils and soil N<sub>2</sub>O uptake (N<sub>U</sub>) from the atmosphere. We include N<sub>2</sub>O generated from  
 101 nitrification (N<sub>N</sub>) and denitrification (N<sub>DN</sub>) in the total N<sub>2</sub>O production:

$$102 \quad N_E = N_P - N_U \quad (1)$$

$$103 \quad N_P = N_N + N_{DN} \quad (2)$$

104 The amount of N<sub>2</sub>O uptake is calculated according to Fick's law of gas diffusion (Eq. 3),  
 105 where C<sub>air</sub> and C<sub>soil</sub> represent N<sub>2</sub>O concentration in air and soils, respectively.

$$106 \quad N_U = (C_{air} - C_{soil}) \times k_s \quad (3)$$

107 We use atmospheric N<sub>2</sub>O concentrations of 331.1 ppb. k<sub>s</sub> represents the diffusion  
 108 coefficient. The N<sub>2</sub>O diffusion coefficient used in this study is 1.26 × 10<sup>-6</sup> m<sup>2</sup> s<sup>-1</sup> (van Bochove,  
 109 Bertran, & Caron, 1998).

110 The N<sub>P</sub> from nitrification was determined by a fraction of the nitrification rate k<sub>n</sub>, which



111 is 0.02 in the DayCent model (William et al. 1998, Delgrosso et al. 2005), and 0.0006 in the  
112 DNDC Model (Li 1992). In this study, we varied  $k_n$  within the range of 0.0006 to 0.02 and  
113 calibrated it using observational data. The  $NI_P$  from denitrification,  $k_{dn}$ , is the fraction of  
114 denitrified N lost as  $N_2O$  fluxes, calculated based on the ratio of  $N_2$  to the sum of  $N_2$  and  $N_2O$   
115 ( $N_2/(N_2+N_2O)$ ) from denitrification, which is influenced by soil moisture, pH and electron level.  
116 The effects of electron donor to the substrate ( $f_{nc}$ ) and water function to  $N_2$  ratio ( $f_m$ ) are from Del  
117 Grosso et al (2000), in which the pH function for  $N_2/N_2O$  ratio ( $f_{ph_{rn2}}$ ) is derived from the  
118 SWAT model (Wagena et al, 2017):

119

$$120 \quad N_N = k_n \times f_{nit} \quad (4)$$

$$121 \quad N_{DN} = k_{dn} \times f_{deni} \quad (5)$$

$$122 \quad k_{dn} = \frac{1}{1+r_{n2}} \quad (6)$$

$$123 \quad r_{n2} = f_{nc} \times f_m \times f_{ph_{rn2}} \quad (7)$$

$$124 \quad f_{ph_{rn2}} = \begin{cases} 0.1, & (pH \leq 4.5) \\ \frac{1}{1470 \times e^{-1.1 \times pH}}, & (pH > 4.5) \end{cases} \quad (8)$$

$$125 \quad f_{nc} = \begin{cases} k_1 \times e^{(-0.8 \times \frac{no3}{co2})}, & (0.16 \times k_1 \leq k_1 \times e^{(-0.8 \times \frac{no3}{co2})}) \\ 0.16 \times k_1, & (0.16 \times k_1 > k_1 \times e^{(-0.8 \times \frac{no3}{co2})}) \end{cases} \quad (9)$$

$$126 \quad k_1 = \begin{cases} 1.7, & ((38.4 - 350 \times \text{diff}) < 1.7) \\ 38.4 - 350 \times \text{diff}, & ((38.4 - 350 \times \text{diff}) \geq 1.7) \end{cases} \quad (10)$$

$$127 \quad f_m = \begin{cases} 0.1, & ((0.015 \times \text{wfps} - 0.32 < 0.1) \\ 0.015 \times \text{wfps} - 0.32, & ((0.015 \times \text{wfps} - 0.32 \geq 0.1) \end{cases} \quad (11)$$

128 To assess the impact of permafrost thaw on N mineralization, we account for the  
129 additional nitrogen and carbon input due to permafrost thaw depth changes. The thawing depth  
130 in the current year was compared with the maximum thawing depth in previous years. If the



131 thawing depth in the current year exceeded the maximum thawing depth in past years, the  
 132 thawing depth difference was considered as the thawing depth change, and the organic N and C  
 133 between two depths were added to soil organic N and C pools. We based our calculation on a  
 134 soil thermal model, detailed information can be found in Liu et al. (2022) and Zhuang et al.  
 135 (2001). Due to lacking permafrost deep layer profile data of C and N, we assumed they are  
 136 uniformly distributed in vertical (Wang et al., 2020), with nitrogen density of  $2 \text{ kg m}^{-3}$  (Strauss et  
 137 al., 2022) and C density of  $29 \text{ kg m}^{-3}$  (Mishra et al., 2021), and half of the total N and C ( $1 \text{ kg m}^{-3}$   
 138 and  $14.5 \text{ kg m}^{-3}$ ) are organic that will undergo N mineralization and release.

139

## 140 2.2 Model parameterization and verification

141 We conducted model parameterization using observation data from 22 sites across the  
 142 pan-Arctic region, including 4 wet tundra sites, 12 boreal forest sites, and 6 alpine tundra sites  
 143 (Table.1).

144 Table 1: Site description for  $\text{N}_2\text{O}$  emission observations that are used for model  
 145 parameterization and verification.

Site	Classification	Location	Country	Latitude	Longitude	Soil bulk density ( $\text{g cm}^{-3}$ )	pH	Time	Reference
W1	Wet Tundra	Inner Mongolia	China	N 49.3	E 119.9	1.2	4.9	2008 -5 to 2009 -9	Liu et al (2017)
W2	Wet Tundra	Yakutsk, eastern Siberia	Russia	N 62.5	E 129.5	1.0	8.0	2004 -6 to 2005 -9	Takakai et al. (2008)
W3	Wet Tundra	Seida	Russia	N 67	E 63	0.0	3.7	2012 -07 to	Voigt et al. (2017)



								2013 -09	
W4	Wet Tundra		Canada	N 48.5	W 58.5	0.1	4. 5	2015 -6 to 2016 -9	Gong et al. (2019)
B1	Boreal Forest		Russia	N 64.5	E 100	1.4	5. 8	2005 -6 to 2007 -9	Morishita et al. (2014)
B2	Boreal Forest	Joensuu, Eastern Finland	Finland	N 62.5	E 29.5	1.6	4. 3	2007 -3 to 2007 -9	Maljanen et al. (2010)
B3	Boreal Forest	New Hampshire	USA	N 44.0	W 72	1.3	3. 9	1997 -12 to 2000 -4	Groffman et al. (2006)
B4	Boreal Forest	Daxing'an Mountain	China	N 53.5	E 122.5	1.0	5. 6	2016 -5 to 2016 -12	Wu et al. (2019)
B5	Boreal Forest	Ballyholly	Ireland	N 52.0	W 8.5	1.3	3. 4	1993 -11 to 1995 -7	Butterbach-Bahl et al. (1998)
B6	Boreal Forest	Yakutsk, Eastern Siberia	Russia	N 62.5	E 129.5	1.1	7. 9	2004 -6 to 2005 -9	Takakai et al. (2008)
B7	Boreal Forest	Great Hing'an Mountains	China	N 53	E 123	14.0	4. 6	2013 -5 to 2015 -10	Cui et al. (2018)
B8	Boreal Forest	Mid-boreal Upland Ecoregion of central Saskatchewan	Canada	N 53.5	W 105.5	1.1	5. 7	2006 -5 to 2007 -10	Matson et al. (2009)





B9	Boreal Forest	Mont St. Hilaire	Canada	N 45.6	W 73.2	0.7	4.7	2006 -5 to 2008 -5	Ullah & Moore (2011)
B10	Boreal Forest	Southern Finland	Finland	N 61.3	E 25	1.2	3.7	2000 -6 to 2003 -8	Maljanen et al. (2006)
B11	Boreal Forest	Southern Lower Saxony	Germany	N 51.6	E 10.1	1.3	4.5	1999 -4 to 2000 -3	Teepe et al. (2004)
B12	Boreal Forest	Hoglwald Forest	Germany	N 48.5	E 11.2	1.0	3.6	1995 -1 to 1997 -12	Luo t al. (2013)
A1	Dry Tundra	Yakutsk, Eastern Siberia	Russia	N 62.3	E 129.5	1.4	8.3	2004 -6 to 2005 -9	Takakai et al. (2008)
A2	Alpine Tundra	Changbai Mountain	China	N 42.5	E 128.0	0.8	7.3	2011 -7 to 2013 -9	Zhou et al. (2016)
A3	Alpine Meadow	Qinghai–Tibetan Plateau	China	N 31.4	E 92.0	1.0	5.3	2015 -5 to 2016 -12	Yan et al. (2018)
A4	Alpine Meadow	Tibetan Plateau	China	N 37.5	E 101.5	0.8	8.4	2013 - 2014	Du et al.(2016)
A5	Upland Grassland	Daun	Germany	N 50.2	E 6.8	1.3	5.1	1997 -4 to 1998 -3	Anger et al. (2003)
A6	Upland Grassland	French Massif Central region	France	N 45.7	E 3.0	1.2	6.2	2007 -4 to 2009 -3	Cantarel et al. (2011)

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The N<sub>2</sub>O emission observational data was organized on a monthly basis over the measurement period. Data of soil density and pH were obtained from the same publications or



149 relevant publications associated with the same site, or from the global soil bulk density map  
150 (Global Soil Data Task, 2000) and Global Database of Soil Properties (Carter and Scholes.,  
151 2000). Meteorological data including air temperature, water vapor pressure, precipitation, and  
152 cloudiness were collected from literature or the Climate Research Unit (CRU TS v. 4.05) (Harris  
153 et al. 2020).

154 We use PEST (V17.2 for Linux) for parameterization (<https://pesthhomepage.org/>) of  
155 nitrification and denitrification parameters for major ecosystem types in the region (Table 2).

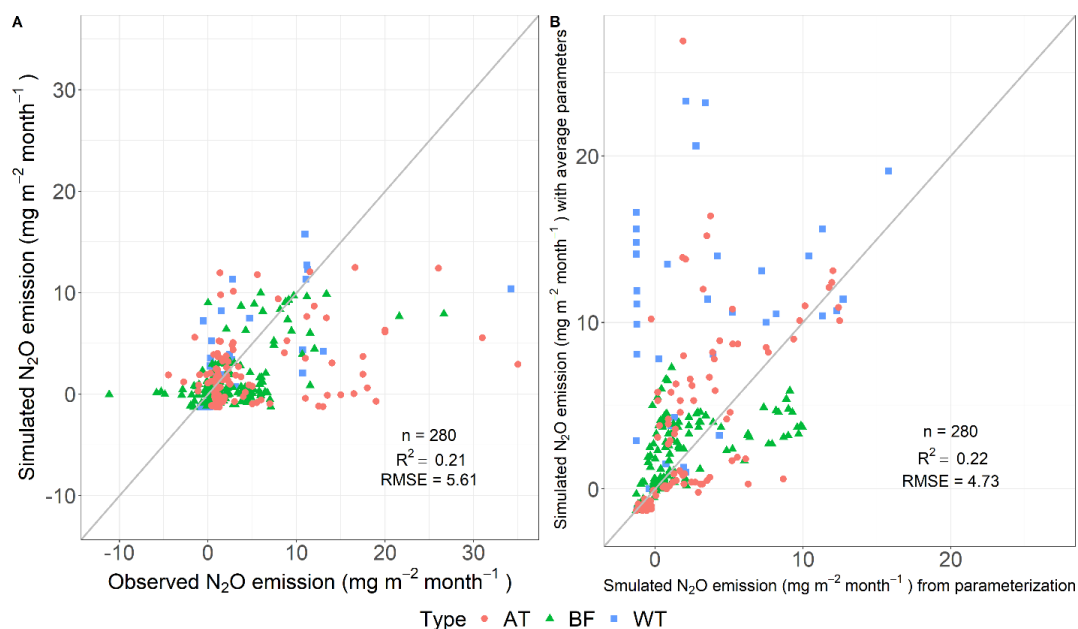
156 The parameters are obtained through minimizing the difference between simulated and observed  
157 N<sub>2</sub>O emissions (Figure 2).

158 Table 2. Parameter values in nitrification and denitrification used in this study (Average ±  
159 standard error).

	K1	Kmax	fno3k	fco2k	kn
Boreal Forest	0.2 ± 0.05	0.21 ± 0.07	1.23 ± 0.1	0.08 ± 0.01	0.016 ± 0.001
Wet Tundra	0.19 ± 0.1	0.0003 ± 0.0003	0.62 ± 0.36	0.075 ± 0.02	0.0055 ± 0.005
Alpine Tundra	0.23 ± 0.07	0.13 ± 0.07	0.94 ± 0.16	0.08 ± 0.02	0.012 ± 0.004

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163 Figure 2. Comparison between the simulated and observed N<sub>2</sub>O emissions (A) and simulated  
164 N<sub>2</sub>O emissions using averaged parameters and simulated N<sub>2</sub>O emissions (B) for alpine tundra  
165 (AT), boreal forest (BF) and wet tundra (WT). The grey line represents y=x.  
166

### 167 2.3 Regional extrapolation

168 To get the spatially and temporally explicit estimates of N<sub>2</sub>O emission at the regional  
169 scale, we used the data of land cover, soils, and climate from various sources at a spatial  
170 resolution of 0.5° latitude × 0.5° longitude to drive TEM. We used climate forcing data  
171 including the monthly CRU TS v. 4.05 data during 1969–2019. Data of soil density and pH were  
172 obtained from the global soil bulk density map (Global Soil Data Task, 2000) and the Global  
173 Database of Soil Properties (Carter and Scholes., 2000). The regional N deposition data is  
174 sourced from re-gridded model results from GEOS-Chem (Ackerman et al., 2018). We assumed  
175 half of the inorganic N deposition is NH<sub>4</sub><sup>+</sup>. Gaps between modeled years were filled with the  
176 average values between two time periods.



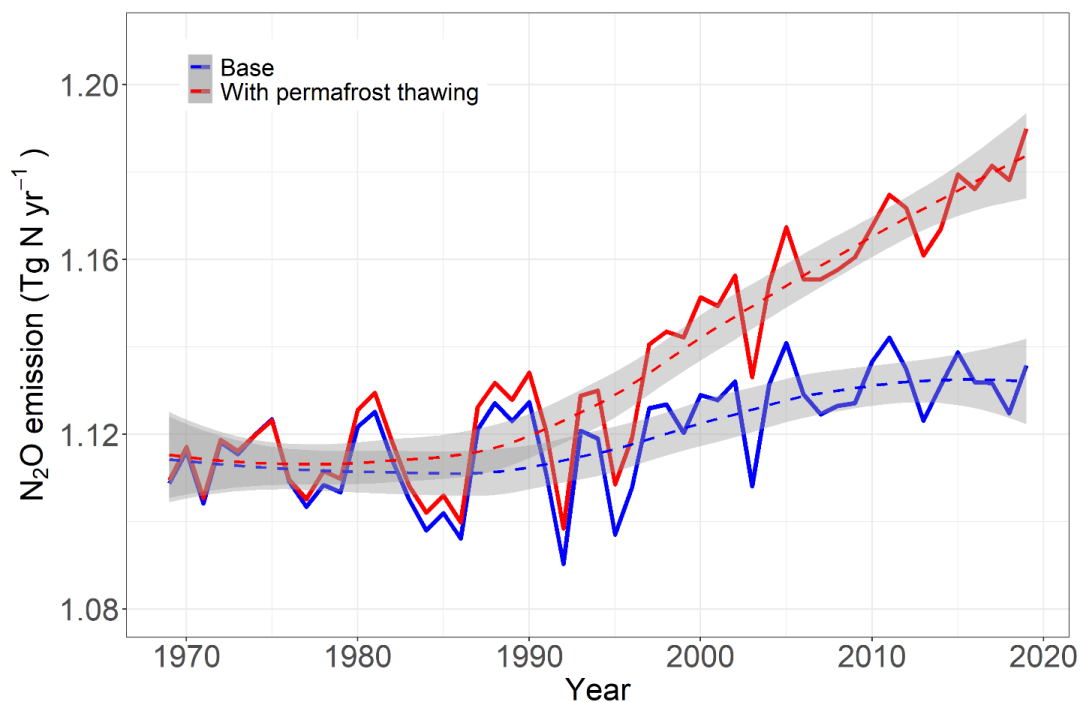
177 We also conducted model sensitivity test of N<sub>2</sub>O emissions, nitrification, and  
178 denitrification by increasing or decreasing temperature by 3 °C and increasing or decreasing  
179 precipitation by 30% for each grid while keeping all other meteorological input the same, from  
180 1969 to 2019.

181

## 182 3 Results

### 183 3.1 Annual emissions of N<sub>2</sub>O

184 TEM estimated that the annual N<sub>2</sub>O emissions in pan-Arctic region increased from 1969  
185 to 2019, with the highest value of 1.14 Tg N yr<sup>-1</sup> in 2011 and the lowest value of 1.09 Tg N yr<sup>-1</sup>  
186 in 1992 without considering the effect of permafrost thawing (hereafter, referred to as base  
187 simulation). When permafrost thawing effects were included (hereafter, referred to as permafrost  
188 thawing simulation), the emissions were highest in 2019 (1.19 Tg N yr<sup>-1</sup>) and lowest in 1992 (1.1  
189 Tg N yr<sup>-1</sup>) (Figure 3). The difference between base simulation and permafrost thawing  
190 simulation increases with the largest difference in the last year of our simulation (2019, 4.8%).



191

192 Figure 3. Annual N<sub>2</sub>O emissions from pan-Arctic terrestrial ecosystems from 1969 to 2019 in  
193 base simulation (blue) and permafrost thawing simulation (red). The dashed curves are locally  
194 estimated scatterplot smoothing (LOOSE) lines.

195

196

Our calibration provides lower and upper bounds of parameters for each ecosystem type  
197 in the pan-arctic region. Based on these parameter values, our model estimates that regional N<sub>2</sub>O  
198 emissions range from 0.82 to 0.87 Tg N yr<sup>-1</sup> and 1.4 to 1.46 Tg N yr<sup>-1</sup> for the periods of 1969 to  
199 2019 (Appendix A, Figure A1).

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### 201 3.2 Spatial variability of N<sub>2</sub>O emissions

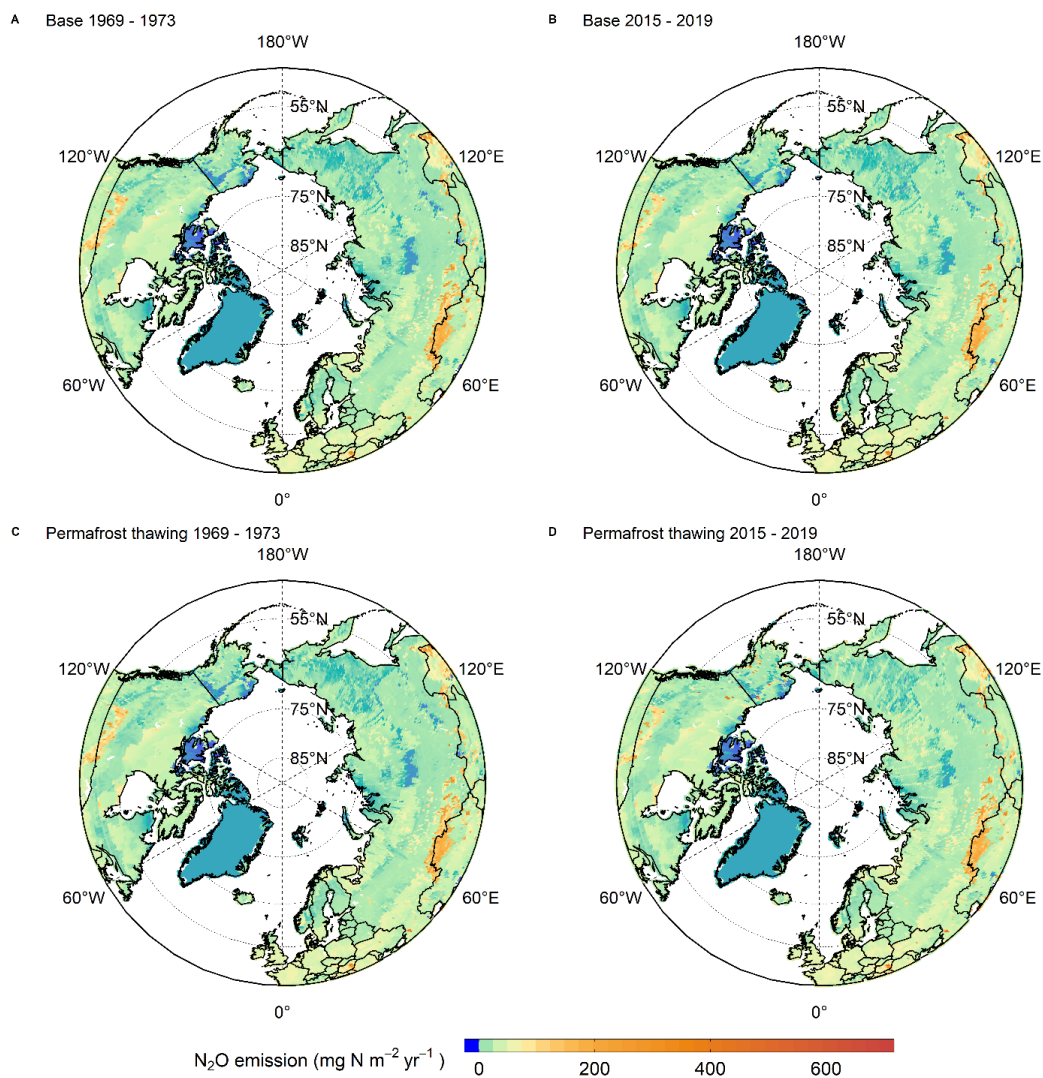
202 Spatially, in the base simulation, the N<sub>2</sub>O emissions range from -12.41 mg N m<sup>-2</sup> yr<sup>-1</sup> to  
203 686.02 mg N m<sup>-2</sup> yr<sup>-1</sup> in 1969 -1973 period, and from -12.35 mg N m<sup>-2</sup> yr<sup>-1</sup> to 686.81 mg N m<sup>-2</sup>  
204 yr<sup>-1</sup> in 2015 - 2019 period. The ecosystems with net sink of N<sub>2</sub>O (N<sub>2</sub>O net emission < 0.0) are  
205 mainly scattered in higher latitude regions (>65°), while the hotspots for net N<sub>2</sub>O sources are



206 concentrated in the lower latitude regions ( $<65^\circ$ ) (Figure 4). The general spatial pattern remains  
207 relatively stable from 1969 to 2019. However, over this period, the net sink areas of  $\text{N}_2\text{O}$  reduced  
208 from 3.08% to 2.54% in the region.

209         Considering permafrost thawing effects, simulated  $\text{N}_2\text{O}$  emission ranges from  $-12.41 \text{ mg}$   
210  $\text{N m}^{-2} \text{ yr}^{-1}$  to  $686.02 \text{ mg N m}^{-2} \text{ yr}^{-1}$  in 1969-1973 period, from  $-10.59 \text{ mg N m}^{-2} \text{ yr}^{-1}$  to  $703.42 \text{ mg}$   
211  $\text{N m}^{-2} \text{ yr}^{-1}$  in the 2015-2019 period. Similar to the base simulation, the general spatial pattern  
212 does not have an obvious change from 1969 to 2019. However, from 1969 to 2019, the  
213 proportion of net sink grid cells of  $\text{N}_2\text{O}$  decreases from 3.07% to 1.95% in the region.

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215

216 Figure 4. Averaged net annual  $\text{N}_2\text{O}$  emissions in the pan-Arctic from 1969 to 1973 (A) and from  
217 2015 to 2019 (B) of the base simulation and the simulation considering permafrost thawing  
218 effects (C and D). Blue indicates the net  $\text{N}_2\text{O}$  sink (negative values).

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### 227 3.3 Model sensitivity

228 Table 3. Sensitivity of N<sub>2</sub>O emissions, nitrification rate, and denitrification in the pan-Arctic  
229 region to changing temperature and precipitation between 1969 and 2019.

230

	Temperature +3 °C	Temperature -3 °C	Precipitation + 30%	Precipitation - 30%
N <sub>2</sub> O emissions (%)	21.2 ± 0.47	-20.9 ± 0.42	2.69 ± 0.53	-2.22 ± 0.5
Nitrification rate (%)	9.4 ± 0.36	-16.6 ± 0.27	-0.24 ± 0.19	-0.73 ± 0.24
Denitrification rate (%)	25.0 ± 0.68	-22.7 ± 0.47	0.55 ± 0.62	0.54 ± 0.41

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232

233 Model sensitivity analysis shows that summed N<sub>2</sub>O emissions are highly sensitive to  
234 changes in temperature ( $\pm 3$  °C) but less sensitive to precipitation change ( $\pm 30\%$ ). In general,  
235 higher temperature and precipitation levels encourage N<sub>2</sub>O emissions in the region, while lower  
236 temperature and precipitation inhibit the emission. Regional nitrification rate increases by about  
237 10% while denitrification increases about 25% under 3°C temperature increase, leading to an  
238 N<sub>2</sub>O emission increase by 21% (Table 3). Conversely, decreasing temperature by 3°C results in a  
239 similar magnitude decrease of N<sub>2</sub>O emissions and denitrification rate, but nitrification tends to  
240 decrease more. The total nitrification and denitrification rates exhibit smaller sensitivity to  
241 precipitation. Precipitation changes by 30% did not induce dramatic change in both summed  
242 nitrification rate and denitrification rate, thus, the regional net N<sub>2</sub>O emissions in values.  
243 Spatially, pan-Arctic region shows various responses to the changes in precipitation in  
244 nitrification and denitrification rates. Nitrification has an optimal value of soil moisture, thus soil  
245 moisture differences in the region result in large different emission responses under both higher  
246 (+30%) and lower (-30%) precipitation conditions. In addition to the effect of precipitation on  
247 soil moisture, denitrification is also influenced by nitrification since nitrification provides  
248 substrate NO<sub>3</sub><sup>-</sup> for denitrification, leading to variation in denitrification rate at different grid cells



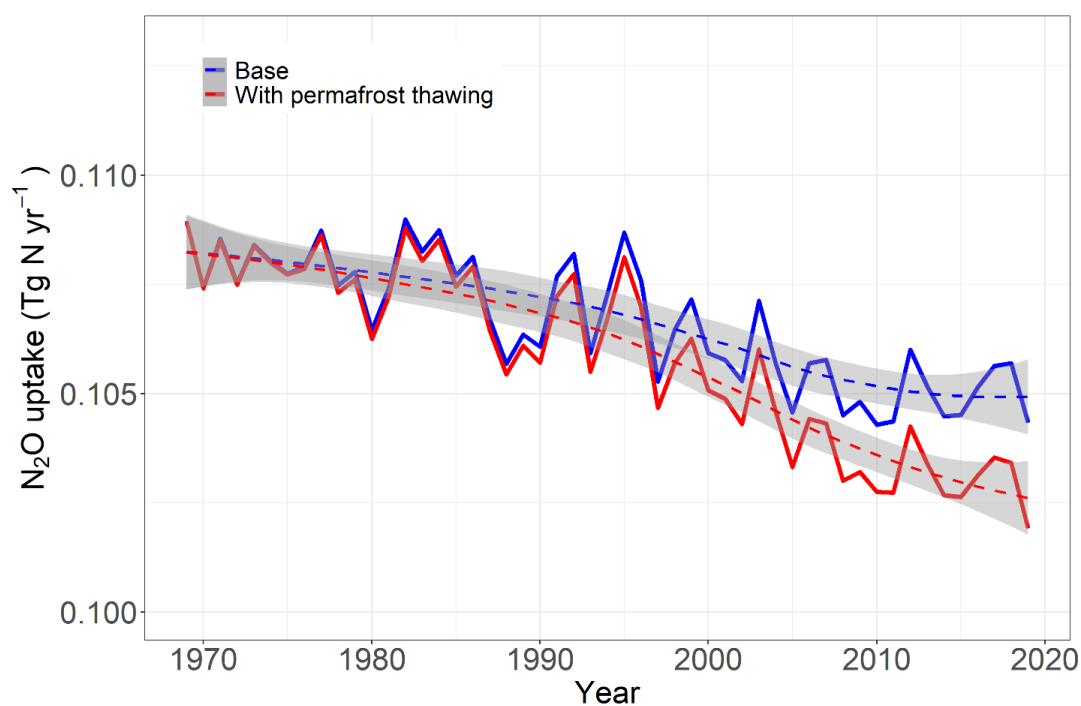


249 in the region.

## 250 4 Discussion

### 251 4.1 The role of soil N<sub>2</sub>O uptake in net emissions

252 Wetlands and peatlands are considered as the major N<sub>2</sub>O sinks (Schlesinger 2013). Soil  
253 water or oxygen content, mineralization, temperature, pH, concentrations of electron donors and  
254 acceptors influence soil N<sub>2</sub>O uptake (Wen et al., 2016; Chapuis-Lardy et al., 2007). Negative  
255 N<sub>2</sub>O fluxes were observed at various ecosystem sites in the pan-Arctic (Takakai et al., 2008;  
256 Gong et al., 2019; Morishita et al 2014; Groffman et al., 2006; Wu et al., 2019; Butterbach-Bahl  
257 et al., 1998; Cui et al., 2018; Matson et al., 2009; Ullah and Moore 2011; Teepe et al., 2004a;  
258 Zhou et al., 2016; Cantarel et al., 2011; Goldberg & Gebauer 2009).



259

260 Figure 5. Annual N<sub>2</sub>O uptake in pan-Arctic terrestrial ecosystems from 1969 to 2019 in base



261 (blue) and permafrost thawing simulations (red). The dashed curves are locally estimated  
262 scatterplot smoothing (LOOSE) lines.  
263

264 In our simulation, the total N<sub>2</sub>O uptake in pan-Arctic terrestrial ecosystems slightly  
265 declines in both base and permafrost thawing simulations (Figure 5) over the simulation period.  
266 The highest value of N<sub>2</sub>O uptake (0.11 Tg N yr<sup>-1</sup>) under base simulation was estimated in 1982  
267 and the lowest value of 0.1 Tg N yr<sup>-1</sup> in 2010, while permafrost simulation has the highest value  
268 in 1969 of 0.11 Tg N yr<sup>-1</sup> and the lowest value in 2019 of 0.1 Tg N yr<sup>-1</sup>. N<sub>2</sub>O uptake accounts for  
269 8.28 - 8.92% of the total production and 9.14 - 9.24% of the total emission in base simulation,  
270 7.8 - 8.86% of total production and 8.57- 9.85% of the total emission in permafrost thawing  
271 simulation. N<sub>2</sub>O uptake mostly occurs in winter when soils have the lowest production of N<sub>2</sub>O  
272 with the maximum N<sub>2</sub>O uptake reaching 0.05 mg N m<sup>-2</sup> day<sup>-1</sup>. Previous study suggests that the  
273 median uptake potential is 4 μg N m<sup>-2</sup> h<sup>-1</sup> and global consumption is less than 0.3 Tg N yr<sup>-1</sup>  
274 (Schlesinger 2013). The projected sink is about 5% of the currently estimated global net N<sub>2</sub>O  
275 fluxes from soils to the atmosphere (Schlesinger 2013). N<sub>2</sub>O uptake in the pan-Arctic region  
276 plays a more significant role than globally.

277 In our model, the N<sub>2</sub>O uptake is calculated based on constant N<sub>2</sub>O concentration in the  
278 atmosphere. However, the soil released N<sub>2</sub>O increases, leading to higher N<sub>2</sub>O concentrations in  
279 the atmosphere, consequently, increasing the N<sub>2</sub>O uptake from the atmosphere. If considering the  
280 increasing atmospheric N<sub>2</sub>O concentration, the total N<sub>2</sub>O uptake amount may be altered. Current  
281 simulation only considered the physical process of diffusion because the biochemical processes  
282 have not been well understood. However, N<sub>2</sub>O is consumed in several reactions of nitrification  
283 (Wrage et al., 2004), and under anaerobic conditions, incomplete denitrification produces N<sub>2</sub>O  
284 whereas the terminal step of denitrification (i.e., the reduction of N<sub>2</sub>O to N<sub>2</sub> with the absence of



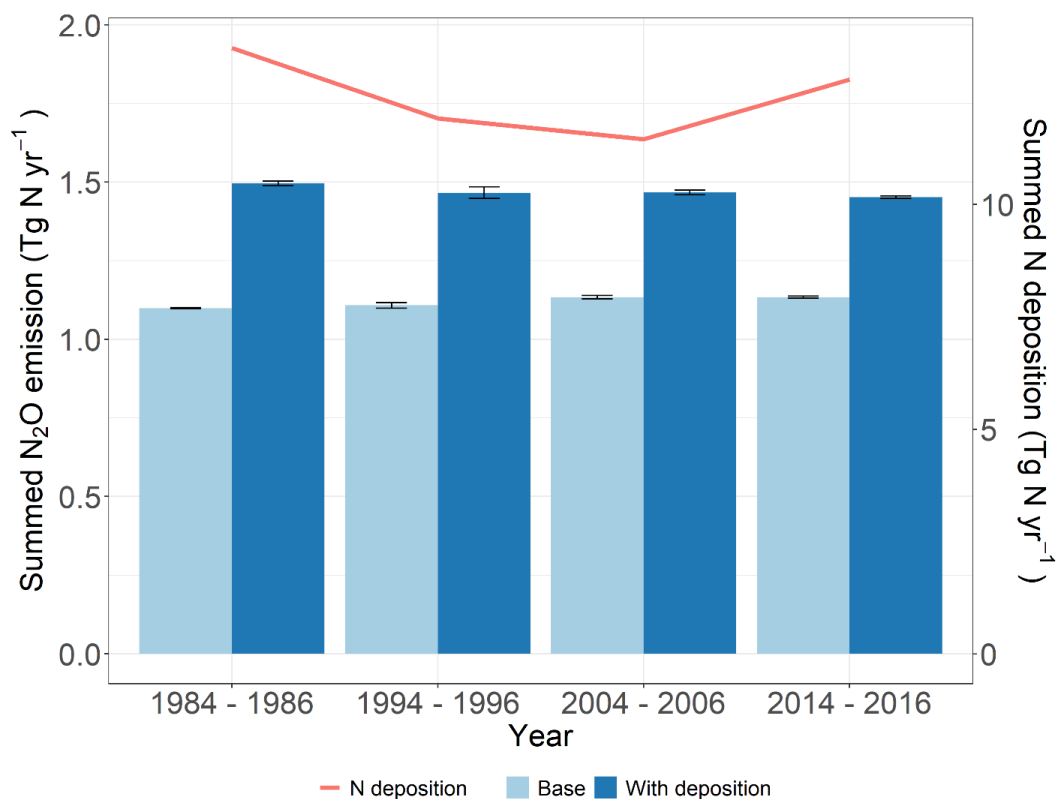
285 *nosZ* gene) consumes N<sub>2</sub>O (Wen et al., 2016, Shan et al., 2021). In addition, within the soil  
286 profile and in the soil air-filled pores, N<sub>2</sub>O can be further reduced to N<sub>2</sub> during its transport to the  
287 soil surface (Chapuis-Lardy et al., 2007; Wen et al., 2016, Yang & Silver., 2016). We expect an  
288 increase in N<sub>2</sub>O uptake by taking these processes into account.

289

#### 290 4.2 The role of N deposition in regional N<sub>2</sub>O emissions

291 In field observations, nitrogen deposition has been shown to induce a substantial increase  
292 of N<sub>2</sub>O emissions of around 95% in dry alpine meadow (Yan et al., 2018), grasslands (Du et al.,  
293 2021), boreal and temperal forests (Deng et al., 2020). In our simulation, the impact of N  
294 deposition on N<sub>2</sub>O emissions varies at different sites depending on initial conditions and the  
295 amount of N deposition.

296



297

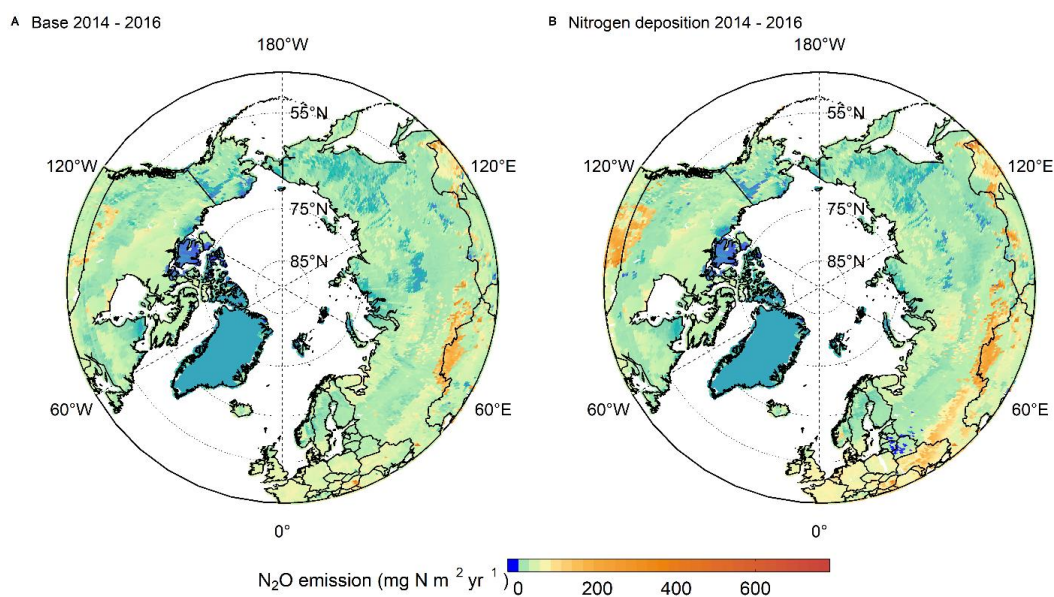
298 Figure 6. Modeled annual net N<sub>2</sub>O emissions from pan-Arctic terrestrial ecosystems from 1984  
299 to 2016 with and without considering N deposition effects. Red line represents the total N  
300 deposition in pan-Arctic region.

301

302

Our simulation indicates that nitrogen deposition from the atmosphere leads to a  
303 significant increase ( $31.5 \pm 3.1\%$ ,  $p < 0.001$ ) in the total N<sub>2</sub>O emissions compared with the  
304 simulation without N deposition (Figure 6). With N deposition from the atmosphere, although  
305 the spatial distribution of net N<sub>2</sub>O sink and the source remains the same, there are more grid cells  
306 in lower latitudes ( $< 60^\circ$ ) that have high N<sub>2</sub>O emissions ( $> 100 \text{ mg N m}^{-2} \text{ yr}^{-1}$ , Figure 7), generally  
307 following the spatial pattern of nitrogen deposition (Ackerman et al., 2018).

308



309

310 Figure 7. Estimated net N<sub>2</sub>O emissions from pan-Arctic terrestrial ecosystems from 2014 to 2016  
311 with (B) and without (A) considering N deposition effects. Blue indicates net sink (negative  
312 values).

313

314

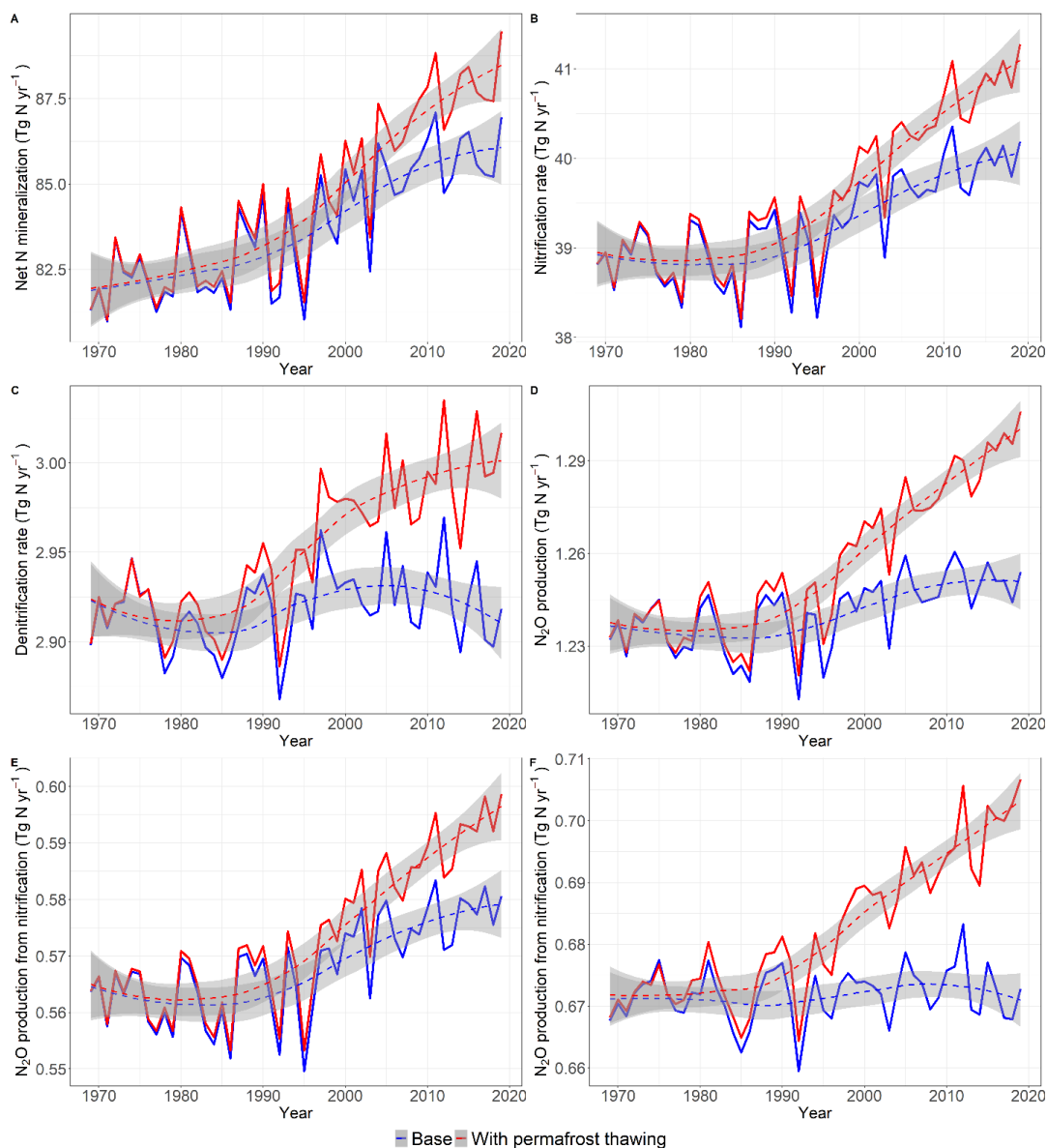
#### 315 4.3 The role of permafrost thawing

316

317 Permafrost thawing adds more soil organic N for mineralization, consequently, increasing

318 the amount of inorganic N availability, resulting in higher nitrification and denitrification rate

319 (Figure 8A, B, C).



320

321 Figure 8. Simulated N mineralization (A), nitrification rate (B), denitrification rate (C), total  $\text{N}_2\text{O}$   
 322 production (D),  $\text{N}_2\text{O}$  production from nitrification (E) and  $\text{N}_2\text{O}$  production from denitrification in  
 323 base (blue) and permafrost thawing simulations (red) from 1969 to 2019. The dashed curves are  
 324 locally estimated scatterplot smoothing (LOOSE) lines.

325

326

In the base simulation, regional nitrification and production of  $\text{N}_2\text{O}$  from nitrification

327

show an upward trend from 1969 to 2019, whereas the denitrification rate slightly decreases and



328 N<sub>2</sub>O production from denitrification remains relatively stable (Figure 5). Higher soil moisture  
329 favors denitrification (Klemetsson et al 1988). Decreasing average soil moisture (Figure A2)  
330 may decrease the denitrification rate from 2000 to 2019. In contrast, in the permafrost thawing  
331 simulation, the denitrification rate increases from 2000 to 2019. Although lower soil moisture  
332 inhibits the denitrification rate, higher nitrification rate provides more NO<sub>3</sub><sup>-</sup> for denitrification,  
333 making up for the loss caused by lower soil moisture. The higher N<sub>2</sub>O production from both  
334 nitrification and denitrification contributes to the higher total N<sub>2</sub>O production.

335         Similar to the simulation of the annual N<sub>2</sub>O emissions (N<sub>2</sub>O production - N<sub>2</sub>O uptake) in  
336 the pan-Arctic region, the N<sub>2</sub>O production generally increased from 1969 to 2019 (Figure 6, D),  
337 with the highest value of 1.26 Tg N yr<sup>-1</sup> in 2011 and the lowest value of 1.21 Tg N yr<sup>-1</sup> in 1992 in  
338 the base simulation, and the highest in 2019 (1.31 Tg N yr<sup>-1</sup>) and lowest in 1992 (1.22 Tg N yr<sup>-1</sup>)  
339 in the permafrost thawing simulation. The difference of N<sub>2</sub>O production between base simulation  
340 and permafrost thawing simulation increases, with the largest difference in the last year of our  
341 simulation (2019, 4.14%). The averaged maximum permafrost thawing depth in the pan-Arctic  
342 region increased 0.2 m in our simulation, meaning the organic nitrogen and carbon stored in the  
343 0.2 m permafrost was released. Average temperature in the pan-Arctic region increases from  
344 -4.37 °C to -1.37 °C during 1969 - 2019 according to CRU Ts v. 4.05 data. The increasing  
345 thawing depth from 1969 to 2019 is small. However, the Arctic region is predicted to warm up to  
346 5.6–12.4 °C and the permafrost is expected to have 3 meters thawing depth (Schuur et al., 2011)  
347 for 47-61% area by 2100, which will significantly intensify N<sub>2</sub>O emissions from the region.

348

#### 349 4.4 Study limitations

350         There are several limitations to this study. First, current parameterization is based on a



351 limited amount of observational data. Obtaining additional observations could provide a more  
352 complete understanding of the spatial and temporal variability of key parameters that influence  
353 N<sub>2</sub>O emissions, such as soil moisture, temperature, and N availability.

354         Second, the uncertainty of input soil data may overestimate N<sub>2</sub>O emissions. Specifically,  
355 the input of soil bulk density data has a median bulk density value of 1.39 g cm<sup>-3</sup>, which differs  
356 from our observation sites with the median bulk density of 1.13 g cm<sup>-3</sup>. Similarly, the input of  
357 soil pH data has a median value of 6.04 while the median value at observation sites is 4.9. These  
358 differences in bulk density and pH between input data at observation sites may bias our model  
359 results.

360         Third, high N<sub>2</sub>O emissions during spring thawing and later fall have been observed. Two  
361 sources have been proposed to contribute to the enhanced N<sub>2</sub>O emissions upon permafrost  
362 thawing, including the physical release of N<sub>2</sub>O produced throughout the winter and trapped  
363 under frozen surface layers and the emission of newly produced N<sub>2</sub>O. While early studies  
364 suggested that the physical release of accumulated N<sub>2</sub>O from subsurface soil layers was the  
365 primary mechanism contributing to spring thaw emissions (Risk et al., 2013), most current  
366 studies favor the newly produced N<sub>2</sub>O as microorganisms remain active during both periods  
367 (Teepe et al., 2004b; Röver et al., 1998, Risk et al., 2013) because the timing and amount of N<sub>2</sub>O  
368 release during thawing do not correspond to the amount of N<sub>2</sub>O trapped in soils (Wagner-Riddle  
369 et al., 2008; Furon et al., 2008). These “shoulder season” emissions have not been well modeled  
370 as the model requires considering the snow dynamics and soil physical models need to be driven  
371 with daily data to capture these final temporal scale dynamics. The time step in current model is  
372 not fine enough to account for these freeze-thaw processes. In current modeling, the N<sub>2</sub>O





373 emission mainly happens in the growing season, with N<sub>2</sub>O emission in winter being negligible.

374 This may cause an underestimate of annual N<sub>2</sub>O emissions.

375 Fourth, while our model simulates nitrogen mineralization by taking into account soil  
376 nitrogen availability and environmental factors; recent studies also suggested that soil carbon to  
377 nitrogen (C/N) ratio may also play a role in influencing nitrification (Bengtsson et al., 2003;  
378 Elrys et al., 2021). This ratio affects the abundance of ammonia-oxidizing bacteria (AOB) and  
379 archaea (AOA) (Xiao et al., 2021) and denitrification (Aulakh et al., 2000; Wang et al., 2023).  
380 Denitrification prefers a certain range of C/N ratio (Lee et al., 2019). Missing this stoichiometric  
381 control factor in our model may introduce uncertainties to our estimates.

382 Finally, our simulation has not included N loading effects caused by dynamics of  
383 groundwater, nearby rivers and wetlands, which may significantly influence soil N availability in  
384 soils and enhance N<sub>2</sub>O emissions (Zhang et al., 2022). The omission of this factor may have led  
385 to an underestimation of N<sub>2</sub>O emissions. The increasing level of N loading caused by  
386 anthropogenic activities may increase the total N<sub>2</sub>O emissions and change their spatial patterns.  
387 Further research is needed to investigate the extent to which N loading affects N<sub>2</sub>O emissions  
388 from the region.

389

## 390 5 Conclusions

391 We use a process-based biogeochemistry model to quantify the regional N<sub>2</sub>O emissions  
392 considering the effects of N<sub>2</sub>O uptake, thawing permafrost, and N deposition. Our simulations  
393 show there is an increasing trend in regional net N<sub>2</sub>O emissions from 1969 to 2019, ranging from  
394 1.1 to 1.19 Tg N yr<sup>-1</sup> considering the permafrost thawing effects. Spatially, annual N<sub>2</sub>O emissions



395 or soil N<sub>2</sub>O uptake range from -12 to around 700 mg N m<sup>-2</sup> yr<sup>-1</sup>, with the negative values  
396 indicating an N<sub>2</sub>O sink, exhibiting significant spatial variabilities in both N<sub>2</sub>O emissions and soil  
397 uptake in this region. Nitrogen deposition from the atmosphere leads to a significant increase  
398 (31.49 ± 3.09 %) in the emission. Our results suggest that in the future, the pan-Arctic terrestrial  
399 ecosystem might act as an even larger N<sub>2</sub>O source with more warming, permafrost thawing, and  
400 higher N deposition.

401

402

403 Code and data availability. The TEM codes, outputs, and samples of running directory can be  
404 accessed via the Purdue University Research Repository [doi:10.4231/KZ5W-DC21](https://doi.org/10.4231/KZ5W-DC21)

405 Author contributions. YY was responsible for model revision, model simulation and paper  
406 writing. QZ was responsible for project design and paper revision. BZ was responsible for model  
407 revision support and paper revision. NS was responsible for project design support and paper  
408 revision.

409 Competing interests. The contact author has declared that neither of the authors has any  
410 competing interests.

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413

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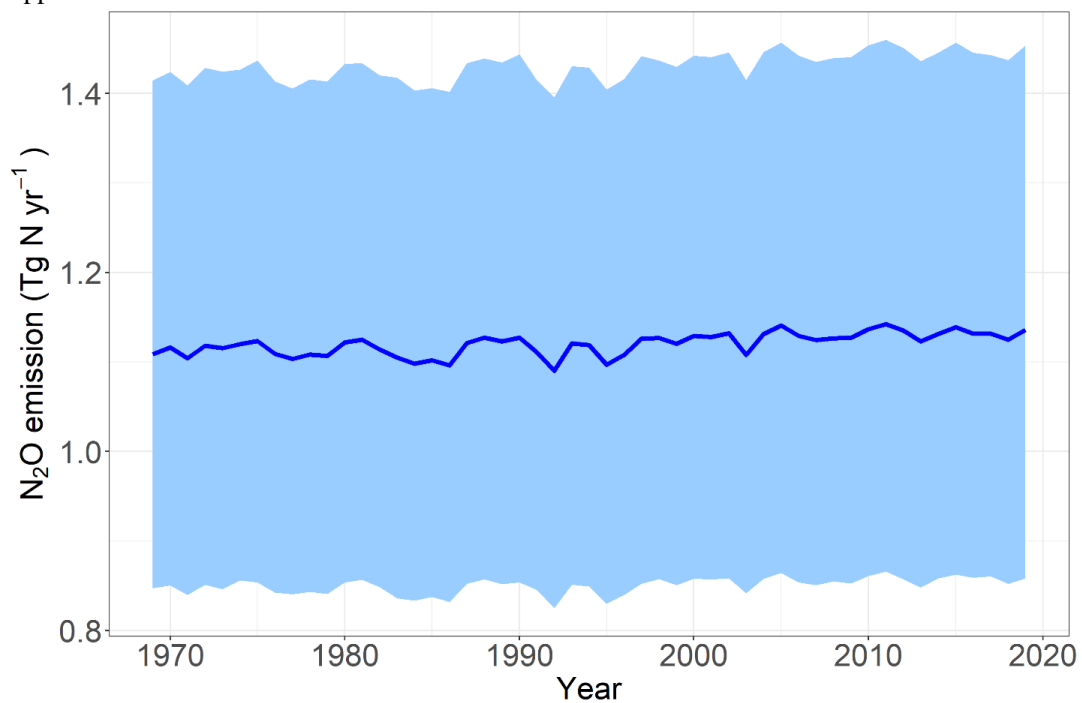
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724 Appendix A



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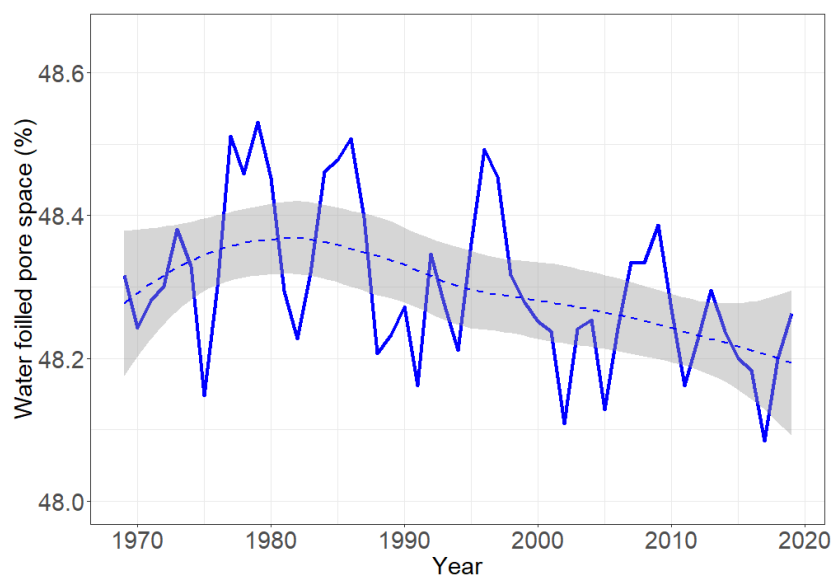
726 Figure A1. Annual N<sub>2</sub>O emissions from pan-Arctic terrestrial ecosystems from 1969 to 2019 in  
727 base scenario with boundaries.

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734 Figure A2. Average soil water filled pore space in the pan-Arctic terrestrial ecosystems from  
735 1969 to 2019.

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