



## Effects of fertilization on soil CH<sub>4</sub> and N<sub>2</sub>O fluxes in young Norway spruce stands

Charlotta Håkansson<sup>a,\*</sup>, Per-Ola Hedwall<sup>b</sup>, Monika Strömgren<sup>c</sup>, Magnus Axelsson<sup>a</sup>, Johan Bergh<sup>a</sup>

<sup>a</sup> Linnaeus University, Faculty of Technology, Department of Forestry and Wood Technology, SE-351 95 Växjö, Sweden

<sup>b</sup> Swedish University of Agricultural Sciences, Southern Swedish Forest Research Centre, SE-230 53 Alnarp, Sweden

<sup>c</sup> Swedish University of Agricultural Sciences, Department of Soil and Environment, SE-750 07 Uppsala, Sweden

### ARTICLE INFO

#### Keywords:

Climate change mitigation  
Forest management  
GHG  
Nitrogen availability

### ABSTRACT

Climate change mitigation strategies have increased the demand for wood products, resulting in an urgent need to increase wood production. One approach is to fertilize forest land, but this can influence greenhouse gas (GHG) fluxes within the ecosystem. The aim of this study was to examine the effects of forest N fertilization on soil CH<sub>4</sub> and N<sub>2</sub>O fluxes in young Norway spruce (*Picea abies* (L.) Karst.) stands in southern Sweden. The gas fluxes were measured using flow-through non-steady-state dark chambers. In the first, long-term, experiment, half of the stand was fertilized twice (once in 2014 and once in 2016) with 150 kg ha<sup>-1</sup> of N, and gas flux measurements were taken throughout 2014–2017. In the second, dose, experiment, 0, 150, 300, or 450 kg ha<sup>-1</sup> of N was added to the stand in April 2016, and gas flux measurements were taken during April–December 2016. The dose experiment showed that the sink strength of CH<sub>4</sub> decreased with increasing amounts of N; the long-term experiment indicated that repeated fertilization decreased the CH<sub>4</sub> sink strength over time. Additionally, the long-term experiment indicated that, while significantly higher N<sub>2</sub>O emissions were recorded in the fertilization years, this was not detected in subsequent years, suggesting the effect to be short-lived. In the dose experiment, fertilization tended to increase the N<sub>2</sub>O emissions relative to the amount of fertilizer. However, despite the significant effects of fertilization on these GHGs, the summed fluxes were a fraction of the net uptake of C at the sites, as recorded in another study. These findings suggest that fertilizing forest land with commercial NP or NPK fertilizers corresponding to 150 kg ha<sup>-1</sup> of N, the level used in operational forestry in Sweden today, can be conducted without changing CH<sub>4</sub> and N<sub>2</sub>O fluxes to any great extent.

### 1. Introduction

Climate change caused by rising levels of carbon dioxide (CO<sub>2</sub>) and other greenhouse gases (GHGs), such as methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O), in the atmosphere, is one of the greatest challenges of our time, and the risks of rising temperatures caused by GHG emissions are critical to life on Earth (IPCC, 2013). Trees sequester CO<sub>2</sub> from the atmosphere through photosynthesis, and wood is used by humans for a variety of products, including as a substitute for fossil fuels and energy-intensive materials. Independently of whether a forest is used to store carbon (C), or its wood utilized to replace fossil fuel or C-intensive materials, the growth rate of the forest is important to maximize its potential to contribute to climate change mitigation (Lundmark et al., 2014). High growth rates provide more opportunities for capturing and

storing CO<sub>2</sub> in the trees or supplying more wood that can be used as a substitution resource (Poudel et al., 2012).

Forest management impacts forest growth using tools such as seedling improvement, choice of tree species, and thinning regimes. However, the impact of most silvicultural tools on forest growth is slow. One of the few ways to enhance tree growth quickly is nitrogen (N) fertilization. In Sweden, most fertilization of forest land comprises one or more applications of 150 kg ha<sup>-1</sup> of N during a rotation period (Hedwall et al., 2014) in middle-aged and older boreal forests. This method of fertilization can enhance stem growth by 13–20 m<sup>3</sup> ha<sup>-1</sup> during a 10-year period (Nohrstedt, 2001), while more intensive fertilization regimes can result in much larger increases in wood production and thus further increase the potential to mitigate climate change. For example, biannual fertilization with 125–150 kg ha<sup>-1</sup> of N in young forest stands

\* Corresponding author.

E-mail address: [charlotta.hakansson@lnu.se](mailto:charlotta.hakansson@lnu.se) (C. Håkansson).

<https://doi.org/10.1016/j.foreco.2021.119610>

Received 29 April 2021; Received in revised form 31 July 2021; Accepted 5 August 2021

Available online 13 August 2021

0378-1127/© 2021 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

can result in incremental increases of  $7 \text{ m}^3 \text{ ha}^{-1} \text{ yr}^{-1}$  over a six-year period (Bergh et al., 2008).

To understand the potential impact of forest fertilization on climate change, it is essential we understand how soil N levels affect GHG fluxes (Vuichard et al., 2019). Adding N via fertilization causes changes in the N cycle of the forest soil, which, in turn, may affect fluxes in  $\text{CH}_4$  and  $\text{N}_2\text{O}$ , and cause reduced uptake or increased emissions of these gases (Aronson and Helliiker, 2010; Liu and Greaver, 2009). The global warming potential (GWP) of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  is 34 and 298 times higher, respectively, than the warming potential of  $\text{CO}_2$  (Myhre et al., 2013; Bodelier and Steenbergh, 2014). In addition, the atmospheric lifetime of ca 12 years for  $\text{CH}_4$  and 120 years for  $\text{N}_2\text{O}$  (Schimel et al., 1996) indicates that what we do today may influence the atmosphere for more than our lifetime. The trade-offs between higher volumes of wood and increased C sequestration after fertilization, and possible changes in GHG fluxes, therefore need to be addressed (Liu and Greaver, 2009).

Mineral forest soils are generally sinks for  $\text{CH}_4$  (Gundersen et al., 2012; Ullah et al., 2009), and there are indications that the sink strength has increased during the last few decades as a result of climate warming (Yu et al., 2017). The uptake of  $\text{CH}_4$  and levels of nitrate ( $\text{NO}_3^-$ ) in the soil are positively correlated (Jang et al., 2006). N fertilization enhances the  $\text{CH}_4$  uptake when the dose is  $\leq 100 \text{ kg ha}^{-1} \text{ yr}^{-1}$  of N (Aronson and Helliiker, 2010). With higher doses, however, N fertilization tends to decrease  $\text{CH}_4$  uptake (Bodelier and Steenbergh, 2014; Jassal et al., 2011).

Mineral forest soils generally act as sources of  $\text{N}_2\text{O}$ , even though the levels are usually low ( $\leq 200 \text{ kg ha}^{-1} \text{ yr}^{-1}$  of  $\text{CO}_2$  eqv) (Gundersen et al., 2012; Aurangojeb et al., 2017).  $\text{N}_2\text{O}$  fluxes are highly variable in both time and space (Gundersen et al., 2012), and studies of fertilization effects on mineral soils have inconsistently shown that N fertilization enhances  $\text{N}_2\text{O}$  emissions, decreases the uptake of  $\text{N}_2\text{O}$  or no significant effects (Liu and Greaver, 2009; Jassal et al., 2011; Papen et al., 2001; Siljanen et al., 2020).

Few studies on  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions from soils after fertilization have been conducted in managed forest ecosystems (Shrestha et al., 2015). The majority of such studies have been based on peatland sites (Matson et al., 2009; Maljanen et al., 2010), and there is a particular lack of  $\text{N}_2\text{O}$  flux data (Siljanen et al., 2020). Hence, the objective of this study was to investigate how the fertilization of young Norway spruce (*Picea abies* (L.) Karst.) stands on mineral soils affected  $\text{CH}_4$  and  $\text{N}_2\text{O}$  fluxes. To achieve this, two experiments using chamber measurements were set up in southern Sweden. The hypothesis was that the forest soil  $\text{CH}_4$  and  $\text{N}_2\text{O}$  fluxes would be marginally influenced by N fertilization with the standard application of  $150 \text{ kg ha}^{-1}$  of N from a commercial NP or NPK fertilizer, while larger applications would turn the forest soils from  $\text{CH}_4$  sinks to sources, and lead to an increase in  $\text{N}_2\text{O}$  emissions.

## 2. Material and methods

### 2.1. The site

The site was located in the boreonemoral zone of southwest Sweden, at Toftaholm (N 57°0', E 14°3'), close to Ljungby. The climate at the site is humid continental, with a mean annual temperature of 7.0 °C and mean annual precipitation of ca 750 mm during the years 2007–2016 (SMHI, 2017). The soil is mainly mesic sandy moraine, with a bedrock of mainly acid granite with some ultrabasic rock, and very little variation in terrain elevation. Prior to fertilization, the mean C/N ratio in the top 0–10 cm did not differ between the two stands used in the experiment ( $24.3 \pm 0.6$  and  $24.5 \pm 0.6$  ( $\pm$ SE) in fertilized and unfertilized stands, respectively). Soil scarification and planting of the area with three-year-old seedlings of Norway spruce was carried out in 2005 and 2006. The stands were mixed, with naturally regenerated birch (*Betula pendula* and *Betula pubescens*) and planted spruce in the proportion of 60/40.

### 2.2. Experimental design

Two experiments, the long-term experiment ( $150 \text{ kg ha}^{-1}$  of N applied every second year, studied 4 years) and the dose experiment (0, 150, 300, and  $450 \text{ kg ha}^{-1}$  of N applied once, studied 7 months) (Tables 1 and 2), were established to study how N fertilization influences soil emissions of the GHGs  $\text{CH}_4$  and  $\text{N}_2\text{O}$ . The long-term experiment, supplemented a study of the effects of fertilization on ecosystem-level C fluxes and took place in two Norway spruce stands, one fertilized and one (the control) unfertilized. Fertilization,  $150 \text{ kg ha}^{-1}$  of N plus other nutrients in granular form, was applied at a stand scale ( $>20 \text{ ha}$ ) using a helicopter, in April of every second year (2014 and 2016) (Table 1). Within each fertilized and control area, 10 rectangular collars of galvanized steel covered with plastic foil (Table 2) were placed at representative points to measure GHG fluxes from the soil surface. To ensure that the correct amount of fertilizer was applied within the collars, the fertilizer for each collar was weighed and distributed manually at the time of aerial fertilization.

The dose experiment took place in a third (unfertilized) area, adjacent to the two areas used for the long-term experiment, with similar site characteristics regarding soil and land-use history. A complete block design was used, with six blocks placed 4–10 m apart, each with four circular plots ( $\varnothing 3 \text{ m}$ ). At the center of each plot, a circular collar was inserted permanently into the soil to measure  $\text{CH}_4$  and  $\text{N}_2\text{O}$  fluxes. Four fertilization treatments (0, 150, 300, and  $450 \text{ kg ha}^{-1}$  of N) were allocated randomly to the plots within each block. Fertilizer (YaraBela N27, 27 % N, and 2.4 % Mg) was distributed evenly by hand across each plot in April 2016. The collar was covered during this procedure and fertilized separately to ensure the correct amount of fertilizer was applied within the collar.

### 2.3. $\text{CH}_4$ and $\text{N}_2\text{O}$ fluxes from the soil-surface

In the long-term experiment,  $\text{CH}_4$  and  $\text{N}_2\text{O}$  flux measurements were taken every second week from June to October, and monthly for the rest of the year, for the period 2014–2017. In the dose experiment, measurements were taken during 2016, every second week from mid-April, then once a week from the end of May to October, and once a month to November.

For each  $\text{CH}_4$  and  $\text{N}_2\text{O}$  flux measurement, a flow-through non-steady-state dark chamber was attached on top of the collar, ensuring an airtight seal (Table 2). To obtain a sequence of measurements from each collar, four air samples were collected at 10–20 min intervals without moving the chamber (Strömngren et al., 2016). In the dose experiment, all samples from one block were extracted within an hour; in the long-term experiment, all samples from five of the ten plots in both treatments were extracted within two hours. The first air sample was collected ten minutes after attachment to minimize the systematic errors in estimated soil  $\text{N}_2\text{O}$  concentrations that occur when attaching a chamber to a collar (Pihlatie et al., 2014), and to avoid the effects of chamber placement (Christiansen et al., 2011). All samples were collected in 22-ml glass vials. To ensure a representative sample, a pump circulated the air between the chamber and the vial for 30 s before sample extraction. In the long-term experiment, the larger chambers had a fan installed to ensure air circulation.

The  $\text{CH}_4$  and  $\text{N}_2\text{O}$  content of the air samples was analyzed in a gas chromatograph (GC) (Clarus 500, PerkinElmer, Waltham, Massachusetts, USA), equipped with a flame ion detector (FID) for  $\text{CH}_4$  and an electron capture device (ECD) for  $\text{N}_2\text{O}$  analysis, as well as an automatic head-space injector (TurboMatrix HS 110, PerkinElmer, Waltham, Massachusetts, USA).

In association with the air sampling, soil moisture and soil temperature were measured at a depth of 5 and 10 cm respectively, within 50 cm of the collar, using a ThetaProbe soil moisture sensor (ML2x, Delta-T, Cambridge, UK) and a temperature sensor (STP1, PP-systems, Hitchin, UK), respectively.

**Table 1**

Fertilizer applied in the long-term experiment for two years of fertilization. Nutrient content is presented as the total amount supplied per hectare (kg) and as percentage weight (%).

Year	Trade name	N	P	K	S	B	Mg	Ca	Se
2014	Skog-can + Yara Superfosfat P20 (kg)	150	111		6.67	0.11	13.3	27.8	
	Skog-can + Yara Superfosfat P20 (%)	27	20	–	1.2	0.02	2.4	5	–
2016	YaraMila 23–3-8/S + B (kg)	150	19.6	52.2	19.6	0.13	–	–	0.01
	YaraMila 23–3-8/S + B (%)	23	3	8	3	0.02	–	–	0.0015

**Table 2**

Comparison of the basic materials and methods for the two experiments. The demarcation between summer and winter was set to 15 April and 15 October, respectively.

	Long-term experiment	Dose experiment
Planted (year)	2005 + 2006	2005
Year of fertilization treatment	2014 + 2016	2016
Fertilizer granulate	SkogCan + P20 (2014) YaraMila 23–3-8/S + B (2016)	YaraBela N27
Treatments (kg ha <sup>-1</sup> of N)	0, 150	0, 150, 300, 450
Method of fertilizing the stand/plot	helicopter	manually Ø 3 m
Method of fertilizing the collar	manually	manually
Collar material	galvanized steel	PVC
Collar (cm <sup>2</sup> )	1880 (34,5 * 54,5 cm)	263 (Ø 18,3 cm)
Chamber (cm <sup>3</sup> )	47,500 (36,5*56,5*23 cm)	4640 (Ø18,3*16,7 cm)
Airtight seal collar-chamber	water	rubber gasket
CH <sub>4</sub> & N <sub>2</sub> O sampling, chamber with a fan	yes	no
Period of measurement	Jan. 2014 - Dec. 2017	Apr. 2016 - Nov. 2016
Number of measurements (summer:winter)	46:16	22:2
CH <sub>4</sub> & N <sub>2</sub> O sampling, minutes after attaching the chamber	10, 20, 40, 50	10, 20, 30, 40
Estimation of vegetation coverage per plot	no	yes

#### 2.4. N availability in soil

Using PST-1 ion-exchange capsules (UNIBEST International, Walla Walla, Washington, USA), the relative amount of available N in the form of NO<sub>3</sub> and NH<sub>4</sub> was measured. Two capsules were buried 10 cm deep within 50 cm of each collar in late April 2016. At the end of November 2016, they were excavated and sent for NO<sub>3</sub>-N and NH<sub>4</sub>-N (mg L<sup>-1</sup>) analysis in KCl extract. The analysis was performed in an AutoAnalyzer 3 Spectrophotometer (Omniprocess, Solna, Sweden). Two capsules from one of the 150 kg ha<sup>-1</sup> of N plots were missing.

#### 2.5. Data analysis

CH<sub>4</sub> and N<sub>2</sub>O fluxes were calculated from the linear slope of concentration against time using all the analyzed gas samples except for a few with obvious errors, for example, because of leaking vials. A linear fit was chosen, even though it is known to underestimate GHG fluxes, because the low levels measured would not tolerate the higher uncertainty that exponential models confer (Pihlatie et al., 2013). The performance of the chambers had been tested previously (Clough et al., 2020) in a chamber-comparison campaign with known N<sub>2</sub>O fluxes (Pihlatie et al., 2014), where the larger chambers tended to underestimate (15%) and the smaller chambers overestimate the fluxes (7%) when calculated by linear fit.

The CH<sub>4</sub> and N<sub>2</sub>O fluxes were corrected for air temperature and air pressure, and the eventual difference in active chamber volume caused by different heights of collar insertion, before further analysis, and two outliers were removed. The regional mean air temperature for the

measurement days was calculated from hourly values between 9 and 18 for the station Ljungby A (SMHI, 2017).

To calculate CO<sub>2</sub> equivalents, GWP values of 34 and 298 were used for CH<sub>4</sub> and N<sub>2</sub>O, respectively. To calculate CO<sub>2</sub> equivalents per treatment and year for the long-term experiment, the means of the CO<sub>2</sub> equivalents per treatment and date of measurement were used. Each date represented a period corresponding to half the period between the previous and next measurement date, except the first and last date, which represented the period from the start and end of the year, respectively, as well as half the period to the nearest measurement date. For the dose experiment, the measurement period was used to calculate CO<sub>2</sub> equivalents instead of the full year.

Treatment effects on CH<sub>4</sub> and N<sub>2</sub>O fluxes were tested using linear mixed models (LMM) using R 4.0.2 (R Core and Team, 2020) and the lme function in the nlme package (Pinheiro et al., 2020), followed by an anova test (the anova function in R) of the models to obtain a P-value for each explanatory variable. For both experiments, the individual flux measurements (per measurement occasion and collar) were used as the response variable. For the long-term experiment, treatment, year and the interaction between treatment and year were fixed effects, while collar identity was a random effect. In the case of a significant interaction effect, the model was reformulated to estimate coefficients and P-values for the treatment effect within a year. For the dose experiment, treatment was a fixed effect, and collar nested within a block a random effect. The measurement date was weighted to allow for heteroscedasticity.

### 3. Results

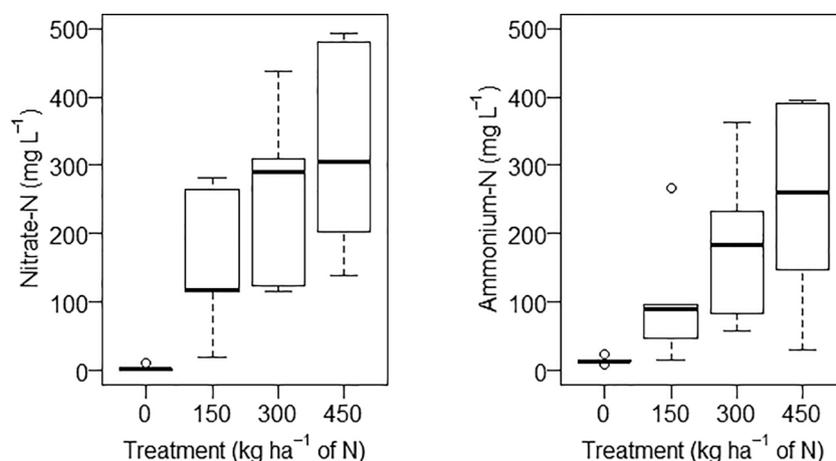
#### 3.1. N availability in soil

In the dose experiment, analyses of the ion-exchange capsules show that fertilization had a significant effect on both NO<sub>3</sub><sup>-</sup> (P < 0.001) and NH<sub>4</sub><sup>+</sup> (P = 0.0004) levels in the soil (Fig. 1). The mean level of NO<sub>3</sub>-N in the control (0 kg ha<sup>-1</sup> of N) plots was 3 mg L<sup>-1</sup> (in capsules KCl extract) and rose in correlation with the amount of fertilizer added (150, 300 and, 450 kg ha<sup>-1</sup> of N), with means of 160, 261 and, 321 mg L<sup>-1</sup> respectively. The mean level of NH<sub>4</sub>-N in the control plots was 14 mg L<sup>-1</sup> and rose in correlation with the amount of fertilizer added (150, 300 and, 450 kg ha<sup>-1</sup> of N), with means of 103, 184 and, 247 mg L<sup>-1</sup> respectively. The levels of available N after fertilization varied between 2 and 516 mg L<sup>-1</sup>.

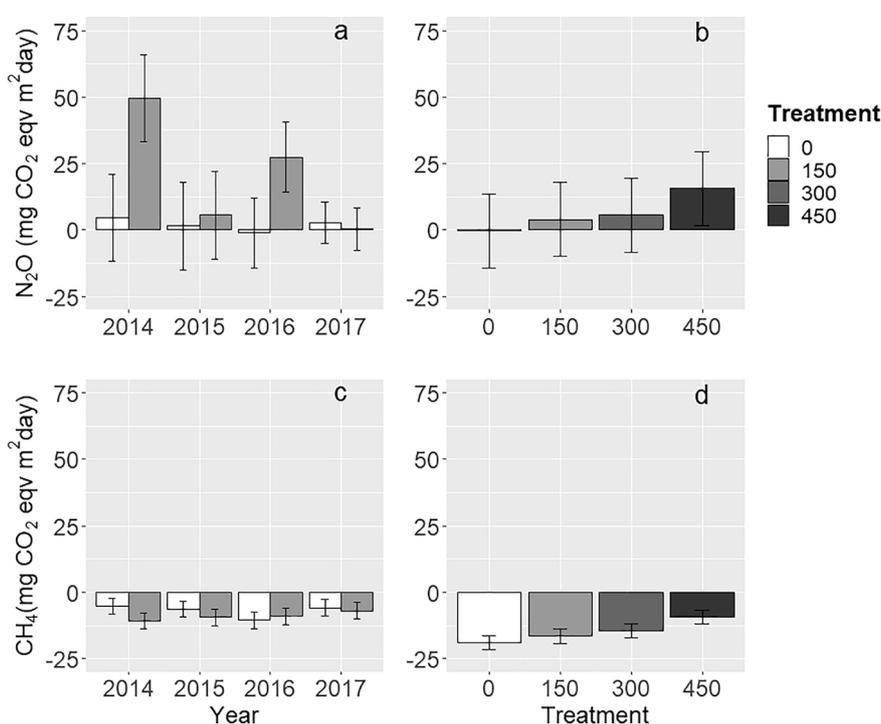
#### 3.2. CH<sub>4</sub> and N<sub>2</sub>O fluxes from the soil-surface

In the long-term experiment, the LMM anova indicated significant effects of fertilization (P = 0.022), year (P < 0.001), and the interaction between fertilization and year (P < 0.001), on CH<sub>4</sub> flux. In the first year of fertilization (2014), the CH<sub>4</sub> uptake was significantly higher (P = 0.013) in the fertilized treatment compared with the control, but not so in the years 2015–2017 (Fig. 2c). It is worth emphasizing that fertilization was applied prior to the growth season in years 2014 and 2016 but was not applied in the years 2015 and 2017.

In the dose experiment, the LMM anova indicated significant effects of fertilization on CH<sub>4</sub> flux (P < 0.001), with a negative trend between the uptake of CH<sub>4</sub> and the dose of N (Fig. 2d). The effect was significant



**Fig. 1.** Relative N availability in soil, as NO<sub>3</sub>-N, and NH<sub>3</sub>-N, for each treatment in the dose experiment, presented as the means of two ion-exchange capsules per plot, with a total of twelve capsules per treatment.



**Fig. 2.** Predicted flux levels for CH<sub>4</sub> (c, d) and N<sub>2</sub>O (a, b) in the long-term experiment (a, c) and dose experiment (b, d). Error bars correspond to a 95% confidence interval. Treatments are presented as kg ha<sup>-1</sup> of N. Positive values indicate gas emission from, and negative values imply a gas uptake in, the soil.

after adding 300 and 450 kg ha<sup>-1</sup> of N ( $P = 0.025$  and  $P = 0.001$  respectively), but not after adding 150 kg ha<sup>-1</sup> of N ( $P = 0.206$ ).

In the long-term experiment, the LMM anova showed significant effects of fertilization ( $P = 0.012$ ) and the interaction between fertilization and year ( $P < 0.003$ ) on N<sub>2</sub>O flux, but not year ( $P < 0.944$ ). N<sub>2</sub>O emissions were significantly increased in the fertilized treatment compared with the control during the two years of fertilization ( $P < 0.001$  in 2014 and  $P = 0.003$  in 2016), but not in 2015 ( $P = 0.737$ ) or 2017 ( $P = 0.669$ ), indicating that fertilization caused elevated emissions of N<sub>2</sub>O only in the years the stand was fertilized (Fig. 2a).

In the dose experiment, there was a tendency towards higher emissions of N<sub>2</sub>O with increasing additions of fertilizer (Fig. 2b); however, the fluxes were very low and the LMM anova did not show any significant effects of fertilization ( $P = 0.505$ ).

The CH<sub>4</sub> and N<sub>2</sub>O flux measurements were recalculated as CO<sub>2</sub> equivalents (kg ha<sup>-1</sup> yr<sup>-1</sup>) for both the long-term and dose experiments

**Table 3**

Yearly CH<sub>4</sub> and N<sub>2</sub>O fluxes as CO<sub>2</sub> equivalents for the long-term experiment. Calculated from the mean flux for all plots and dates of measurement, including 95% confidence intervals (CI). Each measurement date represents half the period before and half the period after the next date of measurement, except for the beginning and the end of the year, for which the first and last dates represent the full start and end period, respectively.

Year	CH <sub>4</sub> (kg ha <sup>-1</sup> yr <sup>-1</sup> of CO <sub>2</sub> eqv)				N <sub>2</sub> O (kg ha <sup>-1</sup> yr <sup>-1</sup> of CO <sub>2</sub> eqv)			
	Control		Fertilized		Control		Fertilized	
	Mean	CI	Mean	CI	Mean	CI	Mean	CI
2014	-16	11	-37	15	62	122	178	116
2015	-22	12	-31	15	-2	89	-14	65
2016	-29	7	-24	13	-14	37	48	103
2017	-17	9	-24	4	20	44	14	43

(Table 3 and Table 4, respectively). The flux levels were low in both experiments, and in both control and fertilized treatments.

#### 4. Discussion

The effects of N fertilization on soil CH<sub>4</sub> and N<sub>2</sub>O fluxes were examined using chamber measurements from mixed stands of young spruce and birch on mineral soils in southern Sweden. Our study adds to the scant literature on fluxes of these potent GHGs from mineral soils, as previous work has primarily been carried out on organogenic soils (Maljanen et al., 2010; Matson et al., 2009; Ojanen et al., 2019; Shrestha et al., 2015; Siljanen et al., 2020), and provides new information about how CH<sub>4</sub> and N<sub>2</sub>O fluxes are affected by forest fertilization of mineral soils.

##### 4.1. CH<sub>4</sub> fluxes

Large amounts of N ( $\geq 300$  kg ha<sup>-1</sup>) in single applications significantly decreased the uptake of CH<sub>4</sub> (Fig. 2d, Table 3, Table 4), albeit at low levels. This is in accordance with earlier studies, which have shown that fertilization decreases oxidation of CH<sub>4</sub> and correspondingly decreases the uptake or increase in emissions (Aronson and Helliker, 2010; Gundersen et al., 2012; Le Mer and Roger, 2001; Liu and Greaver, 2009; Shrestha et al., 2015), even if these effects are short-lived (<1 year) (Börjesson and Nohrstedt, 2000). In the dose experiment treatment with the lowest N addition, no significant effect was observed on the CH<sub>4</sub> flux, which agrees with an earlier study on mineral soil that found no significant effect on CH<sub>4</sub> uptake with an N supply of 200 kg ha<sup>-1</sup> (Maljanen et al., 2006).

The results of the long-term experiment possibly corroborate the minor effect of fertilization at the lowest N dose on CH<sub>4</sub> fluxes, except during the first year, when a higher CH<sub>4</sub> uptake was observed in the fertilized compared with the unfertilized treatment (Fig. 2c, Table 3). The dose fertilization treatment was not replicated in the long-term experiment, so it is difficult to distinguish a treatment effect from a possible initial difference between the stands. The CH<sub>4</sub> uptake with the fertilized treatment decreased over the years, which could potentially be attributed to the repeated fertilizer treatment but could also represent natural yearly variation. However, as the CH<sub>4</sub> uptake was low for both treatments throughout the long-term experiment, we suggest that a single dose of  $\leq 150$  kg ha<sup>-1</sup> of N does not affect the forest's CH<sub>4</sub> balance to any great extent in boreal and cold-temperate coniferous forest ecosystems.

##### 4.2. N<sub>2</sub>O fluxes

Fertilization had a significant effect on N<sub>2</sub>O emissions in the long-term experiment, with higher N<sub>2</sub>O emissions from the fertilized stand than the control stand in years with fertilization, but not in the years after fertilization (Fig. 2a). This is in accordance with previous studies

**Table 4**

CH<sub>4</sub> and N<sub>2</sub>O fluxes as CO<sub>2</sub> equivalents for the measurement period (April–November) of the dose experiment. Calculated from the mean flux for all plots and dates of measurement, including 95% confidence intervals (CI). Each measurement date represents half the period before and half the period after the next date of measurement, except for the beginning and the end of the period, for which the first and last dates represent the full start and end period, respectively.

Treat (kg ha <sup>-1</sup> of N)	CH <sub>4</sub> (kg ha <sup>-1</sup> period <sup>-1</sup> of CO <sub>2</sub> eqv)		N <sub>2</sub> O (kg ha <sup>-1</sup> period <sup>-1</sup> of CO <sub>2</sub> eqv)	
	Mean	CI	Mean	CI
0	-37	4	-4	38
150	-32	10	4	38
300	-28	6	28	42
450	-17	5	33	34

indicating that the effect of fertilization is short-lived (1 year) (Jassal et al., 2010). The increase in N<sub>2</sub>O emissions in the long-term experiment corresponded to 120 and 60 kg ha<sup>-1</sup> yr<sup>-1</sup> of CO<sub>2</sub> eqv in the years with fertilization, which is considerably less than the emissions recorded by Maljanen et al. (2006) (400 kg ha<sup>-1</sup> yr<sup>-1</sup> of CO<sub>2</sub> eqv or 1.2–1.4 kg ha<sup>-1</sup> yr<sup>-1</sup> of N<sub>2</sub>O) after the addition of 200 kg ha<sup>-1</sup> of N in a boreal Norway spruce forest on moraine soil.

For the dose experiment, there was a tendency towards a positive relationship between the amount of fertilizer and N<sub>2</sub>O emissions, which partly confirms our hypothesis, and in accordance with the study by Bowden et al. (1991), who applied 50 and 150 kg ha<sup>-1</sup> yr<sup>-1</sup> of N, and Cheng et al. (2016), who applied nine levels (0–140 kg ha<sup>-1</sup> yr<sup>-1</sup>) of N addition. The latter study found a significant increase in N<sub>2</sub>O flux after adding >60 kg ha<sup>-1</sup> yr<sup>-1</sup> of N. Studies of agricultural land indicate an exponential increase in N<sub>2</sub>O emissions with increasing N addition (Shcherbak et al., 2014). However, to confirm the tendency shown by our dose experiment, and to relate the results to agricultural land, more detailed studies of N<sub>2</sub>O fluxes at higher levels of fertilization, and repeated levels of fertilization, are needed.

For national reporting of N<sub>2</sub>O emissions caused by forest fertilization, an emission factor is often used that assumes 1% of the added N is emitted as N<sub>2</sub>O-N (IPCC, 2006). In the long-term experiment, where 150 kg ha<sup>-1</sup> of N was added on each occasion, this implies that N<sub>2</sub>O corresponding to 700 kg ha<sup>-1</sup> of CO<sub>2</sub> eqv would be emitted. This estimate is much higher than the values of 8 to 120 kg ha<sup>-1</sup> of CO<sub>2</sub> eqv obtained in our study (Table 3). The emission factors varied between 0.01% and 0.17%, depending on the experiment and year, and were lower than those reported for three spruce experiments by Gundersen et al. (2012), which varied between 0.13% and 0.67%. This indicates that the currently used emission factor can generate large overestimations when applied to conventional forest fertilization in northern conditions. A possible explanation for this discrepancy is that northern forest ecosystems are generally N limited (Tamm, 1991), while the IPCC emission factor was derived from a few studies on agricultural land (IPCC, 2006) with considerably larger N availability.

##### 4.3. Chamber measuring methods

Flux levels tend to be underestimated by chamber measurements (Pumpanen et al., 2004), especially in low-volume chambers (Pihlatie et al., 2013; Christiansen et al., 2011). Furthermore, flow-through non-steady-state chambers usually underestimate N<sub>2</sub>O flux when linear calculation models are used (Pihlatie et al., 2014). After completion of our fieldwork, attempts to rectify chamber measurements of CH<sub>4</sub> and N<sub>2</sub>O were published (Pavelka et al., 2018; Clough et al., 2020), identifying the need to know the site, purpose of the study, and the behavior of the chamber in use, to make results more comparable across sites and experiments. Both types of chambers used in this study were flow-through non-steady-state chambers and fluxes were calculated using a linear relationship, hence there was a risk of underestimating the flux values, although differences between the experiments could also be a consequence of the different chambers used. Both types of chambers have been experimentally tested against a known flux: flux values from smaller chambers were slightly overestimated, while the values from larger chambers were underestimated (Pihlatie et al., 2014). These differences are, however, of minor importance in this study, where the focus is on the relative effects of fertilization, not the gas levels per se (Rochette and Eriksen-Hamel, 2008).

N<sub>2</sub>O fluxes tend to vary greatly in both time and space (Parkin, 1987; Groffman et al., 2009; Gundersen et al., 2012), and by using chamber measurements short peaks in emissions may go unregistered and lead to underestimations of flux values. However, the measurements were conducted once a week or once a month and throughout the year, hence with different moisture and temperature conditions, which helped mitigate the risk of underestimations.

#### 4.4. Comparing CH<sub>4</sub> and N<sub>2</sub>O emissions with other sources and sinks

A cubic meter of stem wood corresponds to the uptake and storage of ca 1.4 Mg of CO<sub>2</sub> if other parts of the trees are included (Pettersson et al., 2012). A one-time fertilization with 150 kg ha<sup>-1</sup> of N thus facilitates an increased CO<sub>2</sub> uptake in the stand of approximately 20 Mg ha<sup>-1</sup> through increased growth (Bergh et al., 2020). When fertilizing forest land, N<sub>2</sub>O and CO<sub>2</sub> emissions into the atmosphere also arise from the production of the fertilizer (9 kg CO<sub>2</sub> eqv kg<sup>-1</sup> of N applied) and spreading by helicopter (0.022 kg CO<sub>2</sub> emission kg<sup>-1</sup> fertilizer) (Sathre et al., 2010), hence adding 150 kg ha<sup>-1</sup> of N results in an increase in CO<sub>2</sub> emissions of approximately 1.4 Mg ha<sup>-1</sup>. However, these emissions, as well as the level of N<sub>2</sub>O emission (<180 kg ha<sup>-1</sup> yr<sup>-1</sup> of CO<sub>2</sub> eqv) and CH<sub>4</sub> uptake (<17 kg ha<sup>-1</sup> yr<sup>-1</sup> of CO<sub>2</sub> eqv) after fertilization in the long-term experiment, are low (≤1%) compared with both the estimated increased uptake in biomass and the net uptake of ≈ 18 Mg ha<sup>-1</sup> yr<sup>-1</sup> of CO<sub>2</sub> measured at the site in 2014 (Grelle et al., in prep.).

#### 4.5. Conclusions

This study indicates that fertilizing forest land in Sweden with ≤ 150 kg ha<sup>-1</sup> of N, to increase wood production and meet the future demand for forest products, does not significantly decrease the uptake of CH<sub>4</sub> nor the increase in N<sub>2</sub>O emissions. However, further studies on CH<sub>4</sub> and N<sub>2</sub>O fluxes in boreal forest mineral soils are needed to further our understanding of the long-term effects of continued forest fertilization. This study also shows that the IPCC emission factors greatly overestimate the real N<sub>2</sub>O flux after fertilization in this kind of forest, creating the need to develop more accurate emission factors for the fertilization of forests.

#### CRedit authorship contribution statement

**Charlotta Håkansson:** Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Visualization. **Per-Ola Hedwall:** Conceptualization, Methodology, Formal analysis, Investigation, Data curation, Writing – review & editing, Visualization, Funding acquisition. **Monika Strömberg:** Conceptualization, Methodology, Formal analysis, Resources, Data curation, Writing – review & editing, Funding acquisition. **Magnus Axelsson:** Investigation, Writing – review & editing. **Johan Bergh:** Conceptualization, Methodology, Writing – review & editing, Supervision, Funding acquisition.

#### 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.

#### Acknowledgments

Jan Fiedler is acknowledged for valuable help with gas analysis and Inga Bödeker for great assistance with fieldwork during the first season. Swedish Energy Agency, SLU Partnership Alnarp, Carl Tryggers Foundation (13:448, 15:474, Formas, Yara, Södra.

#### References

Aronson, E., Helliker, B., 2010. Methane flux in non-wetland soils in response to nitrogen addition: A meta-analysis. *Ecology* 91, 10.

Aurangzeb, M., Klemetsson, L., Rütting, T., He, H., Weslien, P., Banzhaf, S., Kasimir, Å., 2017. Nitrous oxide emissions from Norway spruce forests on drained organic and mineral soil. *Can. J. For. Res.* 47, 1482–1487.

Bergh, J., Egnell, G., Lundmark, T., 2020. Skogens kolbalans och klimatet. In: Fries, C. (Ed.), Skogsskötselserien. Swedish Forest Agency.

Bergh, J., Nilsson, U., Grip, H., Hedwall, P.-O., Lundmark, T., 2008. Effects of frequency of fertilisation on production, foliar chemistry and nutrient leaching in young Norway spruce stands in Sweden. *Silva Fennica* 42, 13.

Bodelier, P.L.E., Steenbergh, A.K., 2014. Interactions between methane and the nitrogen cycle in light of climate change. *Current Opin. Environ. Sustainab.* 9–10, 11.

Bowden, R., Melillo, J.M., Steudler, P., Aber, J., 1991. Effects of nitrogen additions on annual nitrous oxide fluxes from temperate forest soils in the northeastern United States. *J. Geophys. Res.* 96, 8.

Börjesson, G., Nohrstedt, H.-Ö., 2000. Fast recovery of atmospheric methane consumption in a Swedish forest soil after single-shot N-fertilization. *For. Ecol. Manage.* 134, 6.

Cheng, S., Wang, L., Fang, H., Yu, G., Yang, X., Li, X., Si, G., Geng, J., He, S., Yu, G., 2016. Nonlinear responses of soil nitrous oxide emission to multi-level nitrogen enrichment in a temperate needle-broadleaved mixed forest in Northeast China. *CATENA* 147, 8.

Christiansen, J.R., Korhonen, J.F.J., Juszczak, R., Giebel, M., Pihlatie, M., 2011. Assessing the effects of chamber placement, manual sampling and headspace mixing on CH<sub>4</sub> fluxes in a laboratory experiment. *Plant Soil* 343, 171–185.

Clough, T.J., Rochette, P., Thomas, S.M., Pihlatie, M., Christiansen, J.R., Thorman, R.E., 2020. Global research alliance N<sub>2</sub>O chamber methodology guidelines: Design considerations. *J. Environ. Qual.* 49, 1081–1091.

Grelle, A., Hedwall, P.-O., Strömberg, M., Håkansson, C., Bergh, J. in prep. From source to sink – recovery of the carbon balance in young forests.

Groffman, P.M., Butterbach-Bahl, K., Fulweiler, R.W., Gold, A.J., Morse, J.L., Stander, E. K., Tague, C., Tonitto, C., Vidon, P., 2009. Challenges to incorporating spatially and temporally explicit phenomena (hotspots and hot moments) in denitrification models. *Biogeochemistry* 93, 29.

Gundersen, P., Christiansen, J., Alberti, G., Uggemann, N., Castaldi, S., Gasche, R., Kitzler, B., Klemetsson, L., Lobo-Do-vale, R., Moldan, F., Utting, T., Schleppe, P., Weslien, P., Zechmeister-Boltenstern, S., 2012. The response of methane and nitrous oxide fluxes to forest change in Europe. *Biogeosciences* 9, 14.

Hedwall, P.-O., Gong, P., Ingerslev, M., Bergh, J., 2014. Fertilization in northern forests – biological, economic and environmental constraints and possibilities. *Scand. J. For. Res.* 29, 301–311.

IPCC, 2006. 2006 IPCC Guidelines for national Greenhouse Gas Inventories. In: Eggleston, S., Buendia, M., Miwa, K., Ngara, T., Tanabe, K. (Eds.), IPCC/OECD/IEA/IGES. Hayama, Japan.

IPCC, 2013. Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge, United Kingdom and New York, NY, USA, Cambridge University Press.

Jang, I., Lee, S., Hong, J.-H., Kang, H., 2006. Methane oxidation rates in forest soils and their controlling variables: a review and a case study in Korea. *Ecol. Res.* 21, 6.

Jassal, R.S., Black, T.A., Roy, R., Ethier, G., 2011. Effect of nitrogen fertilization on soil CH<sub>4</sub> and N<sub>2</sub>O fluxes, and soil and bole respiration. *Geoderma* 162, 5.

Jassal, R.S., Black, T.A., Trofymow, J.A., Roy, R., Nesic, Z., 2010. Soil CO<sub>2</sub> and N<sub>2</sub>O flux dynamics in a nitrogen-fertilized Pacific Northwest Douglas-fir stand. *Geoderma* 157, 118–125.

le Mer, J., Roger, P., 2001. Production, oxidation, emission and consumption of methane by soils: A review. *Eur. J. Soil Biol.* 37, 25.

Liu, L., Greaver, T.L., 2009. A review of nitrogen enrichment effects on three biogenic GHGs: the CO<sub>2</sub> sink may be largely offset by stimulated N<sub>2</sub>O and CH<sub>4</sub> emission. *Ecol. Lett.* 12, 15.

Lundmark, T., Bergh, J., Hofer, P., Lundström, A., Nordin, A., Poudel, B.C., Sathre, R., Taverna, R., Werner, F., 2014. Potential Roles of Swedish Forestry in the Context of Climate Change Mitigation. *Forests* 22.

Maljanen, M., Jokinen, H., Saari, A., Strömmer, R., Martikainen, P., 2006. Methane and nitrous oxide fluxes, and carbon dioxide production in boreal forest soil fertilized with wood ash and nitrogen. *Soil Use Manag.* 22, 7.

Maljanen, M., Sigurdsson, B., Gudmundsson, J., Oskarsson, H., Huttunen, J.T., Martikainen, P., 2010. Greenhouse gas balances of managed peatlands in the Nordic countries – present knowledge gaps. *Biogeosciences* 7, 28.

Matson, A., Pennock, D., Bedard-Haughn, A., 2009. Methane and nitrous oxide emissions from mature forest stands in the boreal forest, Saskatchewan, Canada. *Forest Ecol. Manage.* 258, 11.

Myhre, G., Shindell, D., Bréon, F.-M., Collins, W., Fuglestvedt, J., Huang, J., Koch, D., Lamarque, J.-F., Lee, D., Mendoza, B., Nakajima, T., Robock, A., Stephens, G., Takemura, T., Zhang, H., 2013. Anthropogenic and Natural Radiative Forcing. In: Stocker, T.F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P.M. (Eds.), Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.

Nohrstedt, H.-Ö., 2001. Response of Coniferous Forest Ecosystems on Mineral Soils to Nutrient Additions: A Review of Swedish Experiences. *Scand. J. For. Res.* 16, 20.

Ojanen, P., Penttillä, T., Tolvanen, A., Hotanen, J.-P., Saarimaa, M., Nousiainen, H., Minkkinen, K., 2019. Long-term effect of fertilization on the greenhouse gas exchange of low-productive peatland forests. *For. Ecol. Manage.* 432, 13.

Papen, H., Daum, M., Steinkamp, R., Butterbach-Bahl, K., 2001. N<sub>2</sub>O- and CH<sub>4</sub>-fluxes from soils of a N-limited and N-fertilized spruce forest ecosystem of the temperate zone. *J. Appl. Botany* 75, 5.

Parkin, T.B., 1987. Soil Microsites as a Source of Denitrification Variability. *Soil Sci. Soc. Am. J.* 51, 6.

Pavelka, M., Acosta, M., Kiese, R., Altirir, N., Bruemmer, C., Crill, P., Darenova, E., Fuß, R., Gielen, B., Graf, A., Klemetsson, L., Lohila, A., Longdoz, B., Lindroth, A., Nilsson, M., Marañón-Jiménez, S., Merbold, L., Montagnani, L., Peichl, M., Kutsch, W.L., 2018. Standardisation of chamber technique for CO<sub>2</sub>, N<sub>2</sub>O and CH<sub>4</sub> fluxes measurements from terrestrial ecosystems. *Int. Agrophys.* 32, 34.

Pettersson, H., Holm, S., Ståhl, G., Alger, D., Fridman, J., Lehtonen, A., Lundström, A., Mäkipää, R., 2012. Individual tree biomass equations or biomass expansion factors

- for assessment of carbon stock changes in living biomass – A comparative study. *For. Ecol. Manage.* 270, 7.
- Pihlatie, M., Mäki, M., Korhikoski, M., Bosco, S., Brummer, C., Cade, S., S.-Carter, M., Darenova, E., Delorme, J.-P., Dementriades, T., Dusek, J., Galkowski, M., Görres, C., Halmesmäki, E., Hermann, A., Hupp, J., Hurkuck, M., Jordan, S., Juszcak, R., Lee, M., Liu, C., Moffat, A., Merbold, L., Nickerson, N., Pumpanen, J., Smith, J., Strömngren, M., Volpi, I., Korhonen, J.F. 2014. Inter-comparison of N<sub>2</sub>O chambers using laser absorption spectrometry: quantification of systematic errors. Report Series in Aerosol Science 157. Helsinki, Finland: Finnish Association for Aerosol Research FAAR.
- Pihlatie, M.K., Christiansen, J.R., Aaltonen, H., Korhonen, J.F.J., Nordbo, A., Rasilo, T., Benanti, G., Giebels, M., Helmy, M., Sheehy, J., Jones, S., Juszcak, R., Klefoth, R., Lobo-Do-vale, R., Rosa, A.P., Schreiber, P., Serça, D., Vicca, S., Wolf, B., Pumpanen, J., 2013. Comparison of static chambers to measure CH<sub>4</sub> emissions from soils. *Agric. For. Meteorol.* 171, 13.
- Pinheiro, J., Bates, D., Debroy, S., Sarkar, D., Team, R.C. 2020. nlme: Linear and Nonlinear Mixed Effects Models. R package version 3.1-149.
- Poudel, B.C., Sathre, R., Bergh, J., Gustavsson, L., Lundstrom, A., Hyvonen, R., 2012. Potential effects of intensive forestry on biomass production and total carbon balance in north-central Sweden. *Environ. Sci. Policy* 15, 106–124.
- Pumpanen, J., Kolari, P., Ilvesniemi, H., Minkinen, K., Vesala, T., Niinistö, S., Lohila, A., Larmola, T., Morero, M., Pihlatie, M., Janssens, I., Yuste, J.C., Grünzweig, J.M., Reth, S., Subke, J.-A., Savage, K., Kutsch, W., Østregren, G., Ziegler, W., Anthoni, P., Lindroth, A., Hari, P., 2004. Comparison of different chamber techniques for measuring soil CO<sub>2</sub> efflux. *Agric. For. Meteorol.* 123, 159–176.
- R Core Team 2020. R: A language and environment for statistical computing. In: R CORE TEAM (Ed.) R Foundation for Statistical Computing. Vienna.
- Rochette, P., Eriksen-Hamel, N., 2008. Chamber Measurements of Soil Nitrous Oxide Flux: Are Absolute Values Reliable? *Soil Science Society of America Journal - SSSAJ* 72, 12.
- Sathre, R., Gustavsson, L., Bergh, J., 2010. Primary energy and greenhouse gas implications of increasing biomass production through forest fertilization. *Biomass Bioenergy* 34, 10.
- Schimel, D., Alves, D., Enting, I., Heimann, M., Joos, F., Raynaud, D., Wigley, T., Prather, M., Derwent, R., Ehhalt, D., Fraser, P., Sanhueza, E., Zhou, X., Jonas, P., Charlson, R., Rodhe, H., Sadasivan, S., Shine, K., Fouquart, Y., Ramaswamy, V., Solomon, S., Sprinivasan, J., Albritton, D., Derwent, R., ISAKSEN, I., LAL, M., Wuebbles, D. 1996. Radiative Forcing of Climate Change. In: Houghton, J., Meira Fihlo, L. G., Callander, B., Harris, N., Kattenberg, A., Maskell, K. (Eds.), *Climate change 1995: The Science of Climate Change*. Cambridge University Press, Cambridge.
- Shcherbak, I., Millar, N., Robertson, G.P., 2014. Global metaanalysis of the nonlinear response of soil nitrous oxide (N<sub>2</sub>O) emissions to fertilizer nitrogen. *PNAS* 111, 16.
- Shrestha, R.K., Strahm, B.D., Sucre, E.B., 2015. Greenhouse gas emissions in response to nitrogen fertilization in managed forest ecosystems. *New Forest.* 46, 167–193.
- Siljanen, H.M.P., Welti, N., Voigt, C., Heiskanen, J., Biasi, C., Martikainen, P.J., 2020. Atmospheric impact of nitrous oxide uptake by boreal forest soils can be comparable to that of methane uptake. *Plant Soil* 18.
- SMHI. 2017. Ladda ner meteorologiska observationer [Online]. Available: <https://www.smhi.se/klimatdata/meteorologi/ladda-ner-meteorologiska-observationer#param=airtemperatureInstant,stations=period,selectedDate=2017-12-01,stationid=63510> [Accessed 31.03.2017 2017].
- Strömngren, M., Hedwall, P.O., Olsson, B.A., 2016. Effects of stump harvest and site preparation on N<sub>2</sub>O and CH<sub>4</sub> emissions from boreal forest soils after clear-cutting. *For. Ecol. Manage.* 8.
- Tamm, C.O., 1991. Nitrogen in Terrestrial Ecosystems: Questions of Productivity, Vegetational Changes and Ecosystem Stability. Springer-Verlag, Berlin.
- Ullah, S., Frasier, R., Pelletier, L., R.Moore, T., 2009. Greenhouse gas fluxes from boreal forest soils during the snow-free period in Quebec, Canada. *Can. J. Forest Res.* 39, 14.
- Vuichard, N., Messina, P., Luysaert, S., Guenet, B., Zaehle, S., Ghattas, J., Bastrikov, V., Peylin, P., 2019. Accounting for carbon and nitrogen interactions in the global terrestrial ecosystem model ORCHIDEE (trunk version, rev 4999): multi-scale evaluation of gross primary production. *Geosci. Model Dev.* 12, 29.
- Yu, L., Huang, Y., Zhang, W., Li, T., Sun, W., 2017. Methane uptake in global forest and grassland soils from 1981 to 2010. *Sci. Total Environ.* 607–608, 1163–1172.